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SEDIMENTATION OF ^{238}U, ^{226}Ra AND ^{210}Po AND THEIR DISTRIBUTION IN PENINSULAR MALAYSIA

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ABSTRACT

The paper presents the systematic studies on the sedimentation activity concentrations of ^{238}U, ^{226}Ra and ^{210}Po in Peninsular Malaysia. The study was carried out to fulfil the main objectives, i.e. to quantify the sedimentation concentration of ^{238}U, ^{226}Ra and ^{210}Po in Peninsular Malaysia and to track their origin sources through their distribution in the study areas. The marine surface sediments from the east and west coast of Peninsular Malaysia were collected from 2017 – 2019 using a Ponar grab sampler. The radionuclides ^{238}U and ^{226}Ra were measured using Gamma Spectrometry System after reached secular equilibrium with their progenies, while ^{210}Po was separated using radiochemical methods and concentration was counted by Alpha Spectrometry system. The mean value of ^{238}U, ^{226}Ra and ^{210}Po concentrations in sediments were found to be 16.83 Bq/kg dw., 20.43 Bq/kg dw. and 32.66 Bq/kg dw., respectively. The result showed the variation of ^{238}U, ^{226}Ra and ^{210}Po concentrations in sediment, moreover, these radionuclides were mutually strongly correlated due to there are several factors related to environmental origin source, geochemical behavior and process, and others as well discussed. Thus, their detrital origin inputs from terrestrial were the main source to distribute them in the study area. Additionally, sand, silt and clay were also respectively played an important role as the major carrier phases to their distribution.

INTRODUCTION

The principal sources of uranium decay series radionuclide such as ^{238}U, ^{226}Ra and ^{210}Po in the marine environment are the atmospheric precipitation of terrigenous material, soil re-suspension, rock weathering, as well as river waters and fertilizers. Moreover, the concentration of these radionuclides in the natural environment particularly the marine environment is increased by human activity including industry, fossil fuel combustion, phosphate fertilizers in agriculture, and domestic and industrial sewage. On the other hand, large amounts of natural radionuclides content are produced by the modern industry such as metallurgy, oil and gas exploration and refinery, nuclear industry, ore and mineral mining and processing, as well as phosphogypsum waste dump (Andreou et al., 2012; Borylo et al., 2012; Skwarzec et al., 2010).

Uranium-238 (^{238}U) is a naturally occurring radioactive element and an alpha emitter with long half-life, $t_{1/2}$ of 4.47 x 10^9 years. In the marine environment, ^{238}U is a soluble nuclide in seawater and removed from seawater through the process of adsorption, precipitation or biological (Cochran, 1982). The origin and formation of uranium (i.e. ^{238}U) is classified into three different origins: lithogenic uranium e.g., granitoid rocks; non-lithogenic uranium e.g., uranium scavenged by particulate matter in the water column; and authigenic uranium e.g., diagenetic processes (Och et al., 2016). In addition, lithogenic uranium is commonly found incorporated into minerals of detrital origin such as clay minerals, monazite and zirconium (Dawood, 2010). Non-lithogenic uranium in the water column can be co-precipitated with organic matter and iron oxide (Zheng et al., 2002). Non-lithogenic uranium can then be distinguished from lithogenic uranium by subtracting total uranium from lithogenic uranium based on the concentration of lithogenic tracers such as ^{232}Th (Mohamed et al., 1996). Authigenic uranium is part of non-lithogenic uranium but is explicitly formed within the sediment and can be estimated based on non-lithogenic uranium (Och et al., 2016).

Radium-226 (^{226}Ra; $t_{1/2}$ = 1602 years) is a radioactive member of the ^{238}U decay series (Cochran, 1979). This radionuclide is supplied to the marine environment through river discharge, shelf and abyssal sediment release, atmospheric deposition and hydrothermal venting. Due to ^{226}Ra is much less affected by biological processes, therefore its input, mixing, and decay are the major factors controlling ^{226}Ra distributions. Other than its input, the concentrations of ^{226}Ra in the worlds' oceans are also controlled by removal balance (Bruland et al., 2014). Radium-226 is an important tracer in oceanographic issues on year time-scales in particular for ocean mixing. Moreover, this radionuclide in the marine environment is not strongly particle reactive and not scavenged from the water column into particles and behaves essentially conservatively in the oceans (Charette et al., 2001; Moore, 2000). However, this radionuclide is at most concern due to high solubility and mobility (Zal U'yun and Mei-Wo, 2012). This behavior indicating its availability in the surface sediment is mainly supplied from bottom sediments (Mohamed et al., 2010). However, ^{226}Ra is released into the water column from the sediment through thorium isotope (^{230}Th) decay and it is liberated rather from deep-sea sediments (Moore, 1997).

High activity concentrations of ^{226}Ra have been found at the water-sediment interface, where the pore-water plays as a media to transfer radium into the sediment (Cochran, 1979). This same character also belongs to sediment located in coastal regions (Key et al., 1985). The distribution of ^{226}Ra in marine sediment is mostly related to their physical, chemical and geochemical properties (Khatir et al., 1998). Ra-226 is relatively well retained in marine environments as compared with the other marine materials. Because of its diverse compositions, the uptake of radionuclides such as in marine sediments depends much on their physical and chemical properties. The distribution of these radionuclides within the marine environments can provide information on ocean circulation behavior. Furthermore, the accumulation of these radionuclides in marine environments can also allow the traceability of their origin (Noureddine and Baggoura, 1997).

Polonium-210 (^{210}Po) is an intermediate isotope within the ^{238}U decay-series and it is by far the longest-lived of the 7 naturally-occurring Po isotopes in the U and Th decay-series with a half-life of 138.376 days (Cook et al., 2018). Polonium-210 is generated via beta decay from ^{210}Bi (half-life 5.01 days) and decays to ^{206}Pb by the emission of an alpha particle. It is highly toxic (Harrison et al. 2007) and the main contributor to the natural internal radiation dose (Cherry and Heyraud, 1982). Polonium-210 is ubiquitously distributed in rocks, marine, earth's crust, atmosphere and natural waters. (Matthews et al., 2007). Naturally-occurring ^{210}Po is present in the marine environment primarily as a result of the decay of ^{210}Pb following atmospheric deposition and by the outgassing of ^{222}Rn from sediment and submarine formations such as hydrothermal vents (Boisson et al., 2001). In the water-marine environment, ^{210}Po is scavenged by suspended particulate matter by several mechanisms and accumulated in the sediments (Narayana and Rajashekara, 2010).

Furthermore, ^{210}Po 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 ^{210}Po 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. Biological processes can affect both chemical and physical mobilisation (eg. bioturbation) (Rudjord et al., 2019). Align with that, the concentration of ^{210}Po in edibles of marine organisms and sediment is very much higher than that of seawater because of biological re-concentration processes (Saiyad Musthafa and Krishnamoorthy, 2012). In other words, sediment is the primary repository of radionuclides and chemicals in the marine environment and it is a major indicator for chemicals pollution and radionuclides deposition such as for strong particle reactive of ^{210}Po.

As is well known, marine sediments are commonly used as environmental matrices in chemical and radioactive monitoring programs (Saçan et al., 2010). Moreover, the concentration and behavior of ^{238}U, ^{226}Ra and ^{210}Po in marine sediment is not well documented by previous researchers, thus this paper reports on the distribution and sedimentation concentration of those radionuclides from nearshore sediment samples in Peninsular Malaysia. This study is interesting due to the special characteristics of Malaysian waters which are located at marginal sea areas and are also influenced by granitic rock processes (Chappell et al., 2007). Therefore, the main objectives of this report are to quantify the sedimentation concentration of ^{238}U, ^{226}Ra and ^{210}Po in Peninsular Malaysia and to track their origin sources through their distribution in the study areas.

MATERIALS AND METHODOLOGY

Sampling

A total of 12 sampling points were identified to collect a sediment sample for the study of level, trends and effects of natural and anthropogenic radionuclides in the Malaysian marine environment, a part of the 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 surface sediment samples were collected from 2017 to 2019 and all the sampling points are located on the east and west coast of Peninsular Malaysia (Figure 1) and all sampling details are given in Table 1. The east coast stations are facing the South China Sea while the west coast stations are situated in the Malacca Straits and facing towards the Indonesian archipelago. The sampling points are relatively shallow with an average water depth of 13.0 – 44.7 m. These points were selected by assuming receive the input of radionuclides from the mainland of the neighbouring countries which are situated adjacent to the South China Sea and Malacca Straits.

Figure 1: Map of sediment sampling in Peninsular Malaysia

Table 1: Details (location, sampling date, coordinates and water depth) of sampling

Zone	Location	Sampling ID	Date of Sampling	Coordinates		Water Depth (m)
				Latitude, N	Longitude, E	
East Coast	Melawi	ML01	23 Sept. 2017	06° 05.151’	102° 34.468’	22.1
	Tok Bali	TB03	5 May 2018	05° 58.067’	102° 35.657’	23.4
	Pulau Perhentian	PP02	4 May 2018	05° 59.562’	102° 45.227’	35.4
	Kuala Terengganu	KT01	4 July 2018	05° 33.467’	103° 08.905’	30.7
	Marang	MG02	5 July 2018	05° 13.441’	103° 24.456’	44.7
	Redang-Bidong	RB03	9 Aug. 2018	05° 44.797’	102° 56.460’	34.8
	Dungun	DG03	3 Oct. 2018	04° 47.751’	103° 36.619’	40.5
	Paka-Kerteh	PK03	4 Oct. 2018	04° 26.346’	103° 37.934’	37.8
	Cukai-Kemaman	CK02	9 April 2019	04° 10.322’	103° 37.404’	33.5
West Coast	Pulau Sayak	PSK02	2 July 2019	05° 40.632’	100° 08.430’	22.4
	Langkawi Island	PL02	3 July 2019	06° 06.541’	100° 05.223’	25.7
	Kuala Sepetang	KS01	21 Oct. 2019	04° 48.912’	100° 20.916’	13.0

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°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 airtight container (Zal U'yun et al., 2019; Zal U'yun et al., 2020).

For gamma measurement, the ground samples were transferred into a 300 mL counting container, sealed and stored for a period in excess of 30 days to establish secular equilibrium between ^{238}U and ^{226}Ra and their respective radioactive progenies prior to gamma counting. Meanwhile for the alpha of ^{210}Po: 0.5 g of sediment sample in the 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 dry and let for a while to cool (Zal U'yun et al., 2019; Zal U'yun et al., 2020).

Spontaneous Auto-Plating

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 (HAC) 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 the plating jars 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; Zal U'yun et al., 2020).

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 and ^{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 (Zal U'yun et al., 2020):

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

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 comprises 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 its purpose (Yii et al., 2009; Zal U'yun et al., 2020). After considering the volume and the counting time of the sample, the minimum detectable activities (MDA) for thrice radionuclides of ^{238}U and ^{226}Ra were quantified at 1.0 and 0.5 Bq/kg dry wt. (dw.), respectively ((Zal U'yun et al., 2020).

Meanwhile, the measurement of ^{210}Po particles was carried out by using an alpha spectrometry system (Ortec, Ortate Plus) for 24 hours. Polonium-210 concentrations were corrected to the time of sample collection. The alpha spectrometry counting system was equipped with a 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). The MDA for ^{210}Po was quantified at 0.2 Bq/kg dw. in sediment (Yii and Zal U'yun, 2011; Zal U'yun et al., 2020).

RESULTS AND DISCUSSION

Fraction Size of Sediment

The fractions size of sediment (%) are presented in Figure 2 which are composed with the range of 1.6 – 86.4% for sand (mean: 44.0%), 13.2 – 70.5% for silt (mean: 41.9%) and 0.4 – 32.8% for clay (mean: 16.6%). This indicated there has a high content of sand and silt compared to the clay fraction found in the study area. This due to all sampling points is situated at 20 km from the mainland that there has received more input of sediment from river and soil from terrestrial.

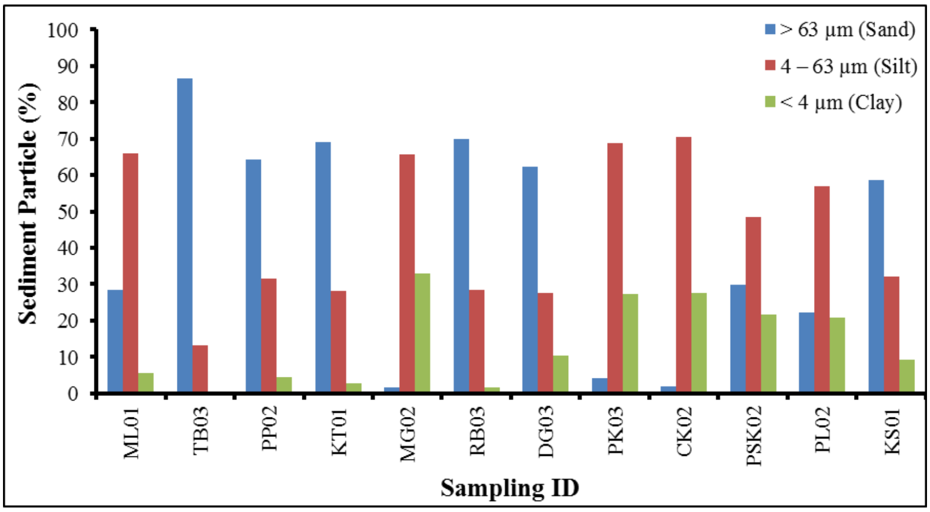


Figure 2: Fraction size of sediment

According to Wang et al., (2020) that terrigenous sediments which are contained sand, silt and clay, etc. are deposited along the coastal boundaries and 75% of marine sediments are from land particularly coarser sediments such as sand closer to coasts and finer sediments such as silt and clay at farther distances offshore. Clearly, this could be explained that seasonal variation also plays an important role to alter the textual content of sediment as a higher fraction of silt and sand was found in the study area particularly on the west coast of Peninsular Malaysia. Moreover, the shallow depth on Peninsular Malaysia coastal may lead to the re-suspension of sediment onto the seabed, thus increasing the content of small particles of silt in the study area.

Sedimentation Activity Concentration of ^{238}U, ^{226}Ra and ^{210}Po

The activity concentrations of ^{238}U, ^{226}Ra and ^{210}Po in marine surface sediment from the east and west coast of Peninsular Malaysia are summarized in Table 2. The concentrations of ^{238}U, ^{226}Ra and ^{210}Po found to be varied from 1.98 Bq/kg dw. to 31.68 Bq/kg dw. (mean: 16.83 Bq/kg dw.), 0.94 Bq/kg dw. to 39.91 Bq/kg dw. (mean: 20.43 Bq/kg dw.) and 3.63 Bq/kg dw. to 61.69 Bq/kg dw. (mean: 32.66 Bq/kg dw.), respectively. Generally, both ^{238}U and ^{226}Ra concentrations were found to be relatively lower in the east coast sediment compared to the west coast sediment.

Table 2: Locality variation of ^{238}U, ^{226}Ra and ^{210}Po in sediment of Peninsular Malaysia

Sampling ID	**Activity Concentration in Sediment (Bq/kg dw.)**			**Activity Concentration Ratio (Unitless)**		
	^{238}U	**^{226}Ra**	**^{210}Po**	**^{226}Ra/^{238}U**	**^{210}Po/^{238}U**	**^{210}Po/^{226}Ra**
ML01	31.68 ± 3.57	39.91 ± 5.21	48.21 ± 2.15	1.26	1.52	1.21
TB03	21.19 ± 2.98	31.14 ± 3.75	35.86 ± 1.60	1.47	1.69	1.15
PP02	18.60 ± 3.75	25.39 ± 3.12	51.15 ± 2.29	1.37	2.75	2.01
KT01	24.32 ± 3.24	24.58 ± 0.54	49.21 ± 2.20	1.01	2.02	2.00
MG02	16.10 ± 1.19	17.53 ± 0.69	60.42 ± 2.70	1.09	3.75	3.45
RB03	16.83 ± 3.72	21.24 ± 0.56	61.69 ± 2.76	1.26	3.67	2.90
DG03	8.80 ± 3.35	11.08 ±1.89	22.28 ± 0.99	1.26	2.53	2.01
PK03	11.42 ± 2.78	12.11 ± 1.92	26.83 ± 1.20	1.06	2.35	2.22
CK02	1.98 ± 0.29	0.94 ± 0.18	3.63 ± 0.16	0.47	1.83	3.86
PSK02	19.93 ± 1.45	22.73 ± 3.64	55.20 ± 2.46	1.14	2.77	2.43
PL02	16.26 ± 1.21	18.49 ± 2.97	4.16 ± 0.19	1.14	0.26	0.22
KS01	23.15 ± 1.88	33.21 ± 5.31	43.32 ± 1.93	1.43	1.87	1.30

High concentrations of these radionuclides in the west coast sediment probably due to receive the abundance input of these radionuclides from the mainland include neighbouring country which is situated adjacent to Malacca Straits. The relatively higher concentration of ^{238}U and ^{226}Ra in the west coast sediment could be assumed the leached inputs of those radionuclides from weathered monazite and phosphate rocks which are known to contain the high level of uranium, radium and their progeny including ^{210}Po. According to Venunathan and Narayana (2016), the radionuclides released during the weathering of these rocks are transported by rivers to the marine environment and subsequently deposited on the beach areas and most of them deposited on the sediment and uptake by the marine organism. Generally, ^{210}Po concentration found to be higher in Peninsular Malaysia waters compared to ^{238}U and ^{226}Ra. This clearly due to different geochemical behavior as a particle reactive which is affinity to particle phases following to deposit onto sediment and it has the shortest half-life lead to fast generated from its parent decay of ^{210}Pb. While, ^{210}Po found to be low in the west coast sediment than the sediment samples from the east coast. This variation could be explained that the source and geochemical behavior of this radionuclide are different from location to location. Moreover, this different distribution also due to input of ^{210}Po from terrestrial was supplied to the study area via river and run-off water, as well as sediment/soil particle from soil erosion at the terrestrial area (Zal U'yun et al., 2019). Furthermore, large amounts of natural radionuclides of ^{210}Po content in the east coast sediment due to along offshore of the east coast is actively produced by the modern industry of oil and gas exploration and refinery as well as fertilizer industries and agriculture activities.

There have good relationships with r = 0.580 and 0.651 between the concentration of ^{238}U, ^{226}Ra, and water depth in the Peninsular Malaysia marine environment (Figure 3). This due to the high concentrations of high solubility and mobility ^{238}U and ^{226}Ra is rapidly adsorbed into the sediment in the shallow water compared to deep water. The negative correlations revealed their concentrations were low because they were rapidly released from surface sediments via resuspension.

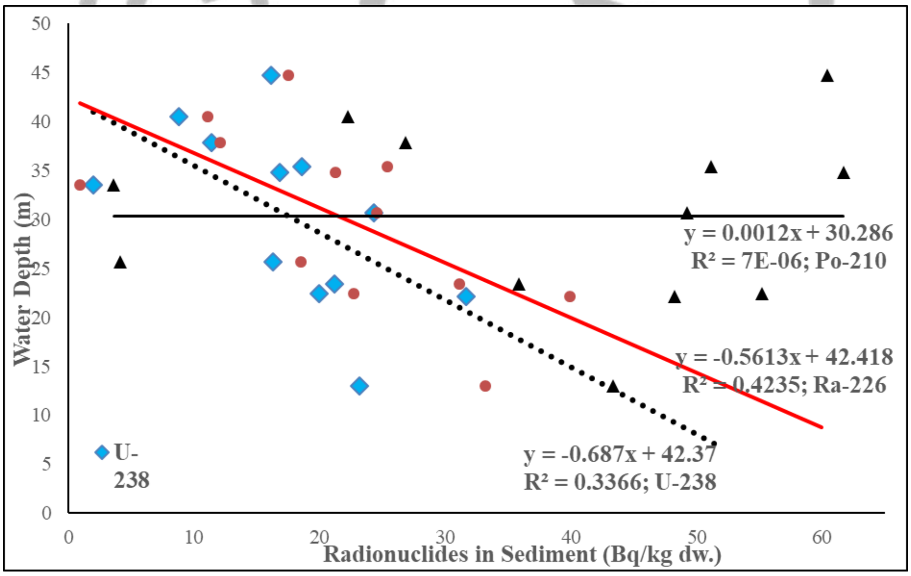


Figure 3: Relationships between ^{238}U and ^{226}Ra in the sediment and water depth

Furthermore, the variation of these radionuclides distributions probably can be related to different geochemical processes influence the interaction of dissolved radionuclide such as ^{238}U and ^{226}Ra with suspended matter and sediments. Sedimentation and re-suspension are of importance for controlling the two-way migration of radionuclide from the water column to sediments and vice-versa (Monte et al., 2005). On the other hand, the ^{238}U and ^{226}Ra concentrations in the sediment samples from the west coast were found to be higher compared to the concentration in the sediment from the east coast. This could be due to a larger percentage of silt (average: 45.8%) and clay (average: 17.3%) minerals present in the sediment samples of the west coast compared to the silt (average: 44.4%) and clay (average: 12.5%) content in the sediment samples of the east coast. As well known, radionuclides are more accumulated to silt and clay compared to coarser fraction due to the factor of large surface area per volume tendency to trap more radionuclides.

In terms of observing the distribution of the radionuclides in the sediments, the averages for 6 parameters, i.e. ^{238}U, ^{226}Ra, ^{210}Po, sand, silt and clay were classified into two major clusters by computing square Euclidean distance and the derived dendrogram is shown in Figure 4. Interpretation from this cluster analysis can be made that sand and silt are the main contributions of ^{210}Po in the sediment at the study area. Meanwhile, clay is played the main role to contribute ^{238}U and ^{226}Ra to the sediment. In other context by using similarity and distance measures in clustering of ^{238}U, ^{226}Ra and ^{210}Po in the sediment (Table 3) proved that the area which high content of sand and silt in the sediment also has a high concentration of ^{210}Po. Thus, this can be initially concluded that the ^{210}Po distribution indicated that sand and silt are the major carrier phases for polonium in the sediment at the study area. Likewise, the preferential trend for ^{238}U and ^{226}Ra accumulation in the sediment increased toward increasing clay contribution in the sediment.

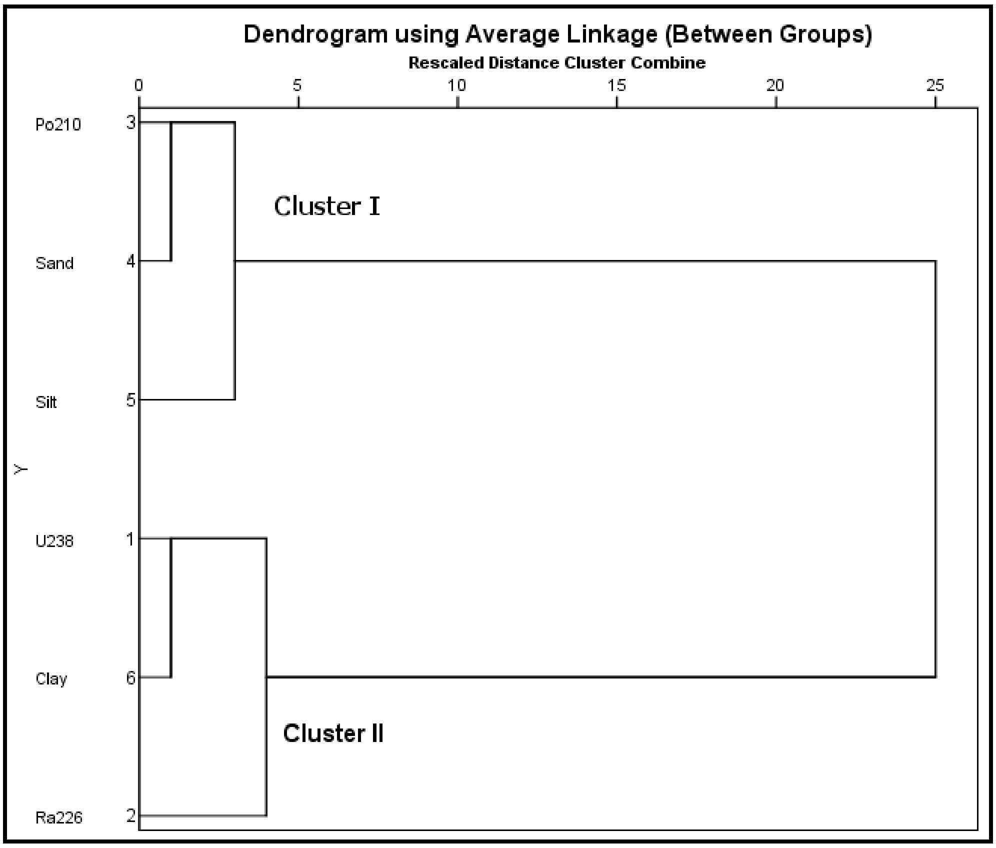


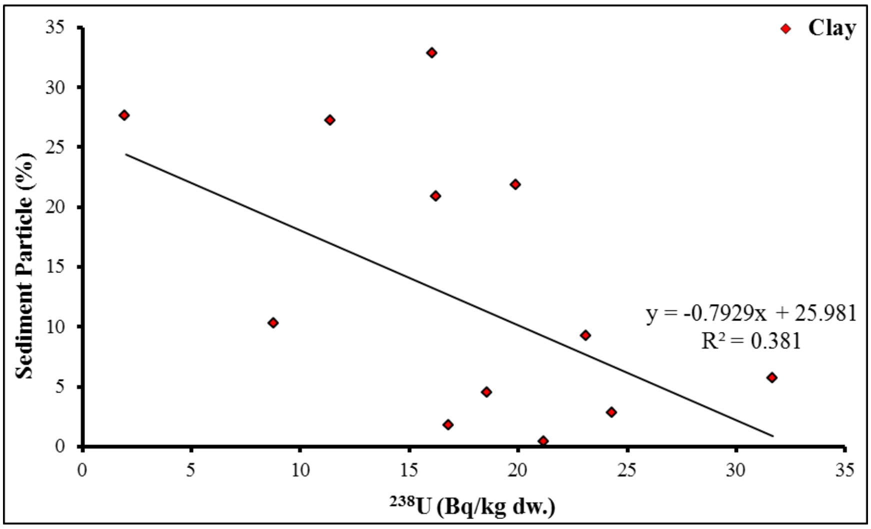
Figure 4: Dendrogram shows the clustering of ^{238}U, ^{226}Ra and ^{210}Po in the sediment

Table 3: Similarity and distance measures in clustering of ^{238}U, ^{226}Ra and ^{210}Po in the sediment

	^{238}U	**^{226}Ra**	**^{210}Po**	**Sand**	**Silt**	**Clay**
^{238}U	0	18.85	91.45	125.15	121.98	58.39
^{226}Ra	18.85	0	80.92	112.60	117.74	71.91
^{210}Po	91.45	80.92	0	101.73	109.6	122.66
Sand	125.15	112.6	101.73	0	166.36	167.04
Silt	121.98	117.74	109.60	166.36	0	115.59
Clay	58.39	71.91	122.66	167.04	115.59	0

Relationship of ^{238}U, ^{226}Ra, ^{210}Po Distribution and Sediment Fraction

The relationship of ^{238}U, ^{226}Ra, ^{210}Po distribution and sediment fraction is illustrated in Figure 5. The significant negative correlations were found between the distributions of ^{238}U, ^{226}Ra and ^{210}Po with clay content in the sediment as the Pearson's coefficient correlation, r were 0.617, 0.702 and 0.633, respectively. These correlations revealed that ^{238}U, ^{226}Ra and ^{210}Po are strongly absorbed into clay and clay can be an important carrier for transporting and widely dispersing contaminants, organic molecules and all types of ions including ^{238}U, ^{226}Ra and ^{210}Po from one area to another area (UGSG, 2021). Clearly, ^{238}U, ^{226}Ra and ^{210}Po distribution indicated that the clay is the major carrier phase for these three radionuclides in the study area. However, a negative correlation indicated their concentrations were low because they were rapidly released from the clay and dissolved in the water column through the diffusion process. Moreover, decreasing of ^{226}Ra in the sediment is related to ^{226}Ra in the ocean can be released mainly from the bottom sediments via pore water exchange and diffusion due to its high mobility and much higher concentration in the sediments compared to seawater. Meanwhile, ^{226}Ra has significantly positively correlated with the sand content showed that ^{226}Ra is associated with terrigenous sediments which is the same environmental origin with sand. These coarser sediments, i.e. sands are mainly supplied from the mainland to the marine environment and deposited along the coastal boundaries at this study area.



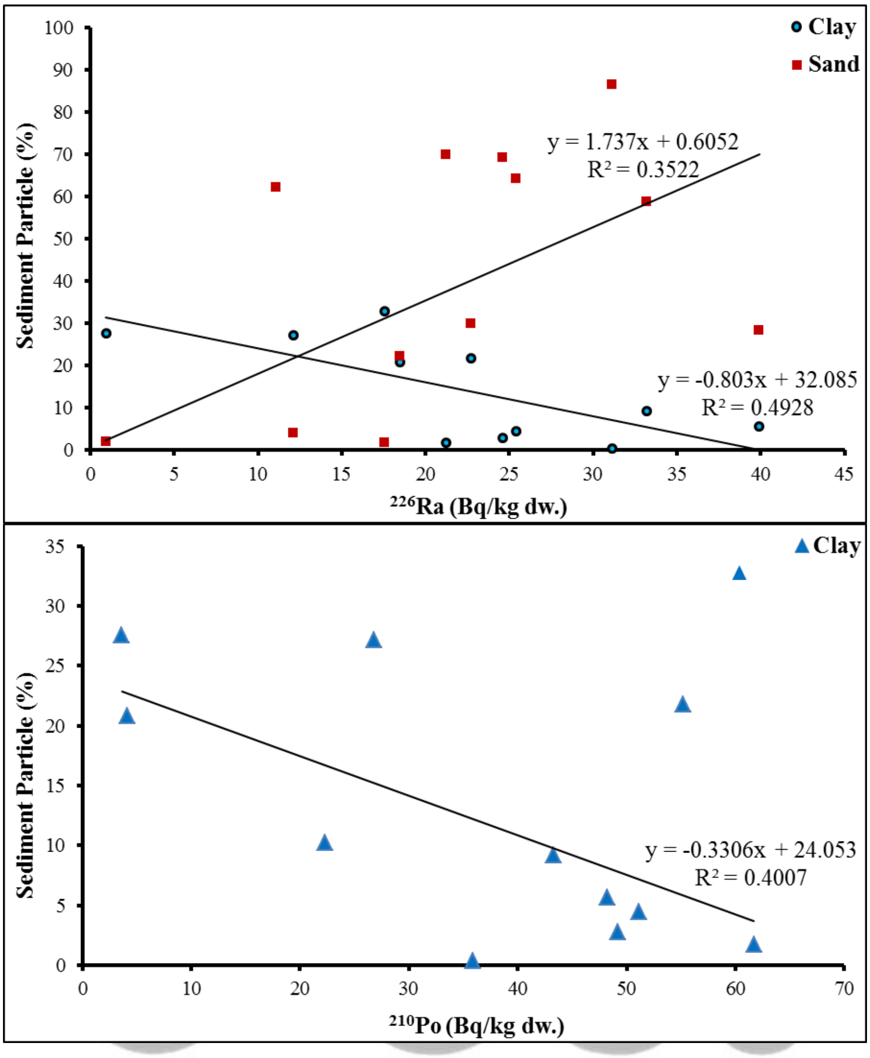


Figure 5: Correlation of ^{238}U, ^{226}Ra, ^{210}Po and their sediment fraction

Relantionship of ^{238}U-^{226}Ra, ^{238}U-^{210}Po, ^{226}Ra-^{210}Po

Generally, the variation of among radionuclide concentrations in sediment from Peninsular Malaysia was mutually strong correlated with $r = 0.961$ for ^{238}U-^{226}Ra, $r = 0.679$ for ^{238}U-^{210}Po and $r = 0.607$ for ^{226}Ra-^{210}Po, respectively (Figure 6). The main reasons could be explained for the similarities trends of these radionuclides in sediment are the same environmental of detrital origin and decay series. This indicated that ^{238}U, ^{226}Ra and ^{210}Po are from the mainland which is attached to the terrigenous sediment since the sediment is the major reservoir of these radionuclides and transport via river stream to the marine environment and deposited on the seabed. Other than that, the strong relationship within these radionuclides suggested similar behavior of chemical properties for ^{238}U, ^{226}Ra and ^{210}Po during the leaching process of the weathered monazite and phosphate rocks which are contained these radionuclides before transported to the marine environment. Despite the geochemical behaviors of each daughter radionuclide differ fundamentally from those of parent uranium, and thus, if released from the same parent mineral, they will be able to migrate and readily incorporated together in the sediment (Cook et al., 2018).

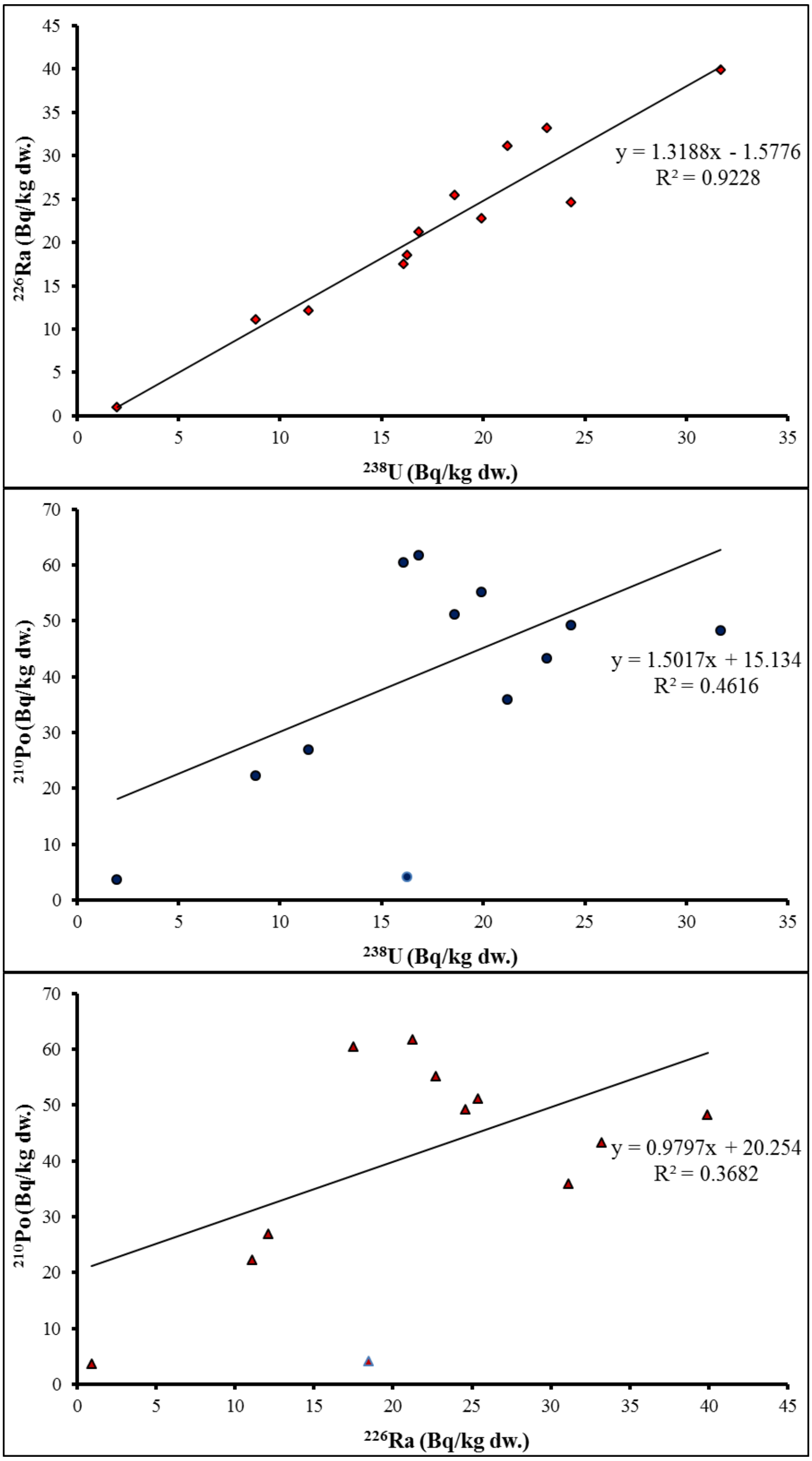


Figure 6: Relantionship of ^{238}U-^{226}Ra, ^{238}U-^{210}Po, ^{226}Ra-^{210}Po in sediment

Moreover, it is obvious the similarity of ^{238}U ^{226}Ra and ^{210}Po to preferentially in the smaller grain size fractions such as clay. A good correlation between ^{238}U-^{226}Ra is should note that both radionuclides exhibited similar behavior of high mobility and solubility in the water column before adsorbed into the sediment. However, there has less correlation between ^{238}U-

^{210}Po and ^{226}Ra-^{210}Po with $r < 0.750$ that clearly can be explained the other source and geochemical behavior of ^{238}U and ^{226}Ra are different from that of ^{210}Po. Other source of ^{210}Po present in the marine environment is primarily from atmospheric decay of ^{210}Pb following wet and dry precipitation and deposition. As ^{210}Po is a high affinity to biogenic materials and its input in the marine environment much more affected by biological processes, therefore these behaviors as a factor controlling ^{210}Po distributions. Other behavior of ^{210}Po is well known as a particle reactive whereas in a water-marine environment ^{210}Po is scavenged by suspended particulate matter by several mechanisms and accumulated in the sediments (Narayana and Rajashekara, 2010). Therefore, the scavenging process by aggregation of colloidal matter and adsorption on to particulate matter influenced the mobilization of ^{210}Po in the water column before deposit and accumulate onto surface sediment (Saili and Mohamed, 2014).

Activity Concentration Ratio of ^{226}Ra/^{238}U, ^{210}Po/^{238}U and ^{210}Po/^{226}Ra

The ratios of ^{226}Ra/^{238}U, ^{210}Po/^{238}U and ^{210}Po/^{226}Ra were found to be varied with the ranges of 0.47 – 1.47 (average: 1.16), 0.26 – 2.75 (average: 2.25) and 0.22 – 3.86 (average: 2.06), respectively (Table 2). This result obtained for ^{226}Ra and ^{238}U ratio confirm the preferential accumulation of radium in sediment; however, it can be concluded that ^{226}Ra is strong tendency to the coarse and fine fraction of sediment, i.e. sand and clay compared to ^{238}U is a tendency to the fine fraction, i.e. clay only. Contrary, the ratio of ^{226}Ra/^{238}U found to be less than unity at station CK02, probably due to the presence of the organic matters in the sediment which are able to adsorb preferentially ^{238}U. The activity ratio of ^{210}Po/^{238}U more than unity revealed that preferential trend for ^{210}Po accumulation in the sediment might be excess supplied from terrigenous source, scavenged by particulate matter in the water column and as a result of the decay of ^{210}Pb following atmospheric deposition. Moreover, input from the outgassing of ^{222}Rn in the sediment and submarine formations such as hydrothermal vents (Boisson et al., 2001).

There has less activity ratio than unity for ^{210}Po/^{238}U at station PL02 due to the presence of the organic matters in the sediment which are able to adsorb preferentially ^{238}U compare to ^{210}Po, even though both of them are affinity to organic material. A similar condition to ^{210}Po/^{226}Ra probably due to high concentrations of ^{226}Ra at the water-sediment interface, where the pore-water plays as a media to transfer radium into the sediment (Cochran, 1979) particularly at coastal region (Key et al., 1985). The fractionated sediment of ^{210}Po/^{226}Ra found to be lower than one in station PL02, might be due to preferential trend for radium accumulation in the larger grain size of sand and a fine fraction of sediment, i.e. clay compared to ^{210}Po is only preferential to clay. Other reason could be explained the presence of the interferences are strongly prevented ^{210}Po to adsorb and accumulate in the sediment. While, the result showed high activity ratio of ^{210}Po/^{226}Ra (>1) suggested that the excess input of atmospheric precipitation of ^{210}Po from the decay of ^{222}Rn as a primary source to the marine environment following the scavenging process to mobilize of ^{210}Po in the water column (Saili and Mohamed, 2014) and deposit onto sediment. Additionally, this indicated that in addition to in-situ radioactive decay of ^{210}Pb also contributes to the activity of ^{210}Po in marine sediments.

CONCLUSIONS

The activity concentrations of ^{238}U, ^{226}Ra and ^{210}Po in sediment from the east and west coast of Peninsular Malaysia found to be varied from 1.98 Bq/kg dw. to 31.68 Bq/kg dw. (mean: 16.83 Bq/kg dw.), 0.94 Bq/kg dw. to 39.91 Bq/kg dw. (mean: 20.43 Bq/kg dw.) and 3.63 Bq/kg dw. to 61.69 Bq/kg dw. (mean: 32.66 Bq/kg dw.), respectively. The variation could be explained that the source and geochemical behavior of these radionuclides are different from location to location. This also probably can be related to different geochemical processes that influence the interaction of dissolved radionuclides and particle reactive with suspended matter and sediments. Sedimentation and re-suspension are of importance for controlling the two-way migration of ^{238}U, ^{226}Ra and ^{210}Po from the water column to sediments or vice-versa. Moreover, the preferential trend for ^{238}U, ^{226}Ra and ^{210}Po accumulation in the sediment clearly depends on the content of sand, silt and clay as carrier phases on their distribution in the sediment. Thus, their detrital origin inputs from terrestrial were the main source to distribute the study area. While, other origin sources of ^{210}Po found to be excess compared to ^{238}U and ^{226}Ra were the addition inputs of its atmospheric precipitation from decay of ^{222}Rn and *in-situ* radioactive decay of ^{210}Pb in the sediment.

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