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Accumulation of Natural Uranium Isotopes in Malaysian Harbour Sediment

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Abstract

Sediment core samples were collected at eight stations in Peninsular Malaysia (PM) and East Malaysian (EM) harbour waters to estimate the sources of uranium deposits. The range of average activity of ^{234}U was from 35.31 ± 4.39 Bq/kg to 67.34 ± 5.65 Bq/kg, while ^{238}U was from 31.25 ± 4.50 Bq/kg to 81.27 ± 7.48 Bq/kg. The range of activities for ^{230}Th and ^{232}Th were also recorded during this study with values from 38.24 ± 3.62 Bq/kg to 150.83 ± 13.86 Bq/kg and 28.08 ± 3.26 Bq/kg to 119.59 ± 14.58 Bq/kg, respectively. The accumulation rate of uranium in harbours were calculated using a simple box model as 0.66 x 10^{-3} Bq/cm^2/year to 6.14 × 10^{-3} Bq/cm^2/year and 3.98 × 10^{-3} Bq/cm^2/year to 17.82 × 10^{-3} Bq/cm^2/year for authigenic and lithogenic fractions of uranium, respectively. In Peninsular Malaysia, results indicated an 86% fraction of lithogenic uranium, while East Malaysian harbours only registered a 61% fraction of authigenic uranium. This result is because PM has a high input of base granitic rock and East Malaysia is affected by the western Pacific Ocean as well as shows by aluminum, zircon and organic carbon.

Keywords: Uranium; Sediment; Harbour; Lithogenic; Authigenic

Introduction

Harbours are usually located in estuaries and have large inputs from land through river flow and runoff which contains various pollutants such as trace elements, radionuclides, etc. Most of the pollutants (i.e., natural radionuclides) are accumulated into harbour sediments as a result of scavenging processes that occur in water column [1-3]. The harbour also behaves like a reservoir which traps and buries all anthropogenic and natural pollutants [4]. Furthermore, harbours which are usually located at the end of rivers might be influenced by eroded material transported from land into the harbour area. Naturally occurring radionuclides present in a harbour ecosystem might come from river effluents of which their distribution is related to their back rock sediments and terrestrial origin [5,6]. In addition, the distribution and concentration levels of radionuclides in harbour also has oceanic influences i.e., authigenic uranium [7] and anthropogenic sources i.e., phosphorites or phosphatic fertilizers [8,9].

The origin and formation of uranium 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 [10]. In addition, lithogenic uranium is commonly found incorporated into minerals of detrital origin such as clay minerals, monazite and zirconium [5]. Non-lithogenic uranium in the water column can be co-precipitated with organic matter and iron oxide [11-13]. 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 [14] and Sc [15]. Authigenic uranium is part of non-lithogenic uranium but is explicitly formed within the sediment and can be estimated based on non-lithogenic uranium [10]. The origin of the uranium can also be predicted through various proxies such as $^{234}U/^{238}U$, $^{230}Th/^{234}U$ and $^{230}Th/^{238}U$ [5,16-19]. In this study, we highlight the role of organic carbon and lithogenic elements (i.e., Al and Zr) as proxies to predict the origin of uranium before accumulation in the harbour.

In Malaysian waters, the monitoring of pollutant concentration levels i.e., heavy metals is conducted by the local authorities or government agencies [20-22]. However, the concentration and behavior of natural radionuclides i.e., uranium in harbour sediment is not well documented by previous researchers. This kind of study is interesting due to the special characteristics of Malaysian harbours which are located at marginal sea areas and are also influenced by granitic rock processes [23,24]. Therefore, the main objective of this manuscript is to investigate the origin of uranium in sediment harbour and also to develop an accumulation model for uranium in Malaysian harbours.

Materials and Methods

Sampling location

Eight harbours located on Peninsular Malaysia (PM) and East Malaysia (EM) coasts were selected to investigate the accumulation of natural uranium. Generally, the texture of sediment on the east coast of PM is silt loam [25], while in the west coast of PM is alluvial deposits [26] but geographically, the base substrate of PM shows extensive granitic rock composition compared to East Malaysia [24].

Sediment core samples of 1.5 m length were collected at eight stations from the selected harbour locations using a gravity corer and slicing at 3 cm intervals. These were then kept refrigerated until further analysis (Figure 1 and Table 1). Basic water quality parameters such as temperature, salinity, conductivity, dissolved oxygen, pH and turbidity were measured using the calibrated water quality multi-parameter AAQ-1183H manufactured by Alec Electronics Co. Ltd.

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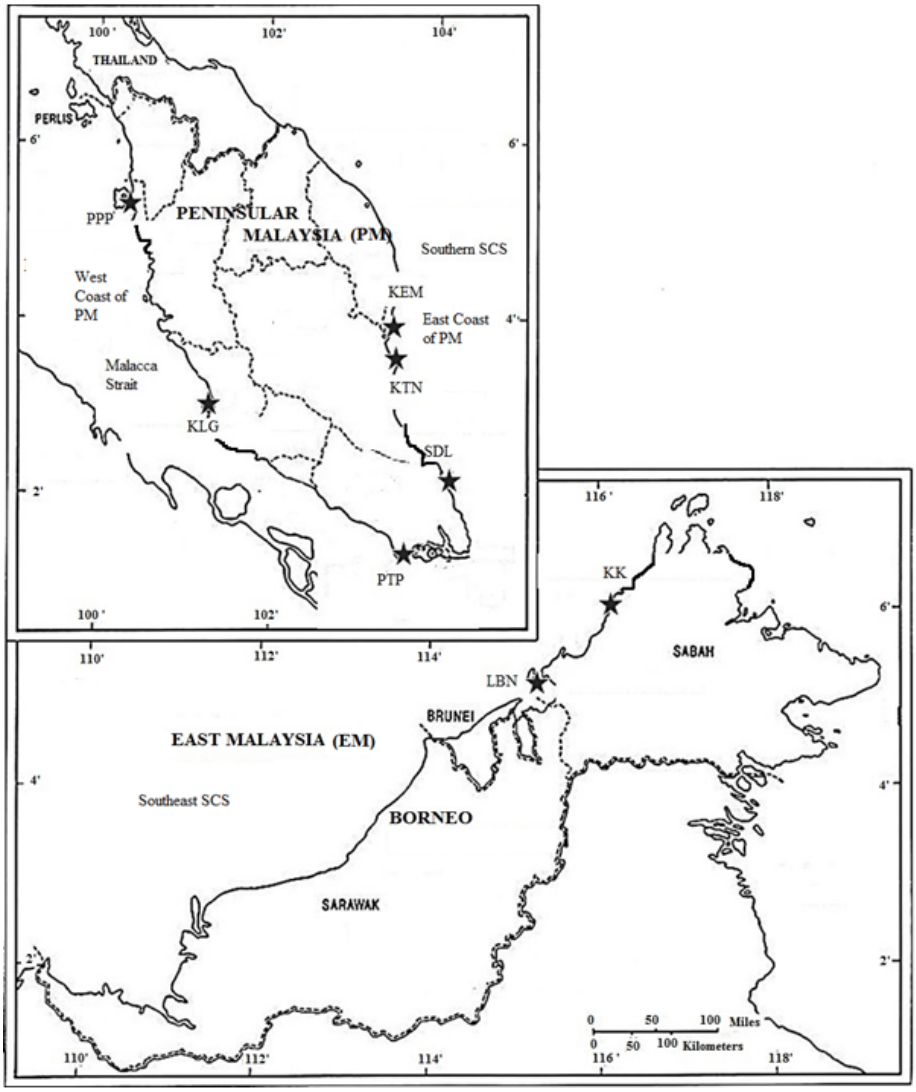

Figure 1: Harbour sediment sampling stations in the west and east coasts of Peninsular Malaysia (PM) and west coast of East Malaysia (EM).

Location	Harbours Name	Sampling date	Latitude (°N)	Longitude (°E)	Water depth (m)	DO (mg/L)	pH	Salinity (psu)
Peninsular Malaysia (PM)	Pulau Pinang (PPP)	22-Jan-13	05°24'39.0"	100°21'39.5"	11	5.04	8.30	30.04
	Klang (KLG)	15-Jan-13	03°00'55.8"	101°21'12.7"	2.3	3.39	7.83	28.60
	Tanjung Pelepas (PTP)	09-Jan-13	01°20'17.5"	103°32'46.4"	10	4.63	8.12	29.74
	Kuantan (KTN)	25-Apr-13	03°58'20.5"	103°26'22.9"	15	-	-	-
	Kemaman (KEM)	20-Jan-13	04°14'45.9"	103°28'13.0"	16	5.82	8.34	31.24
	Sedili (SDL)	10-Jan-13	01°55'54.3"	104°06'39.8"	3.0	4.25	7.31	14.70
East Malaysia (EM)	Labuan (LBN)	03-Dec-12	05° 16' 01"	115° 14' 46"	10	4.30	7.85	30.35
	Kota Kinabalu (KK)	06-Dec-12	06° 00' 53"	116° 04' 57"	15	4.03	8.12	32.85

The (-) is unrecorded

Table 1: Sampling location and physical parameters of water column at Malaysian harbours.

Uranium and thorium isotopes analyses

Published analytical procedures by previous researchers [14] were applied to digest our sediment samples. Briefly, about 0.25 g of dried sediments were spiked with ^{232}U and ^{229}Th tracers (2 dpm/ml) as a chemical yield, and mixed with acids of (HNO_3: HF) (2:1v/v) for 2 h. About 2 ml of perchloric acid ($HClO_4$) and 0.5 g of boric acid (H_3BO_3) were then added to remove excess fluoride. The cation column of Bio-Rad AG 50W × 4 (200-400 mesh) were used to purify uranium and thorium isotopes from other matrices. The purified fractions of uranium and thorium were electrodeposited onto a silver disc for 2 h at 200-250 mA with 15 V using ethanol as a medium. The activities of ^{232}U, ^{234}U, ^{238}U, ^{229}Th, ^{232}Th and ^{230}Th were measured using the Alpha Analyst Spectroscopy system with a silicon surface barrier detector by Canberra,

Inc., with Apex-Alpha software. The silicone surface barrier detector was placed in stand-alone nucleus alpha spectroscopy with a negligible background (0.0003 cpm) and a counting efficiency of 11–13%.

Sequential extraction analysis

About 1.5 g of surface sediment samples (0-3 cm) were performed through the sequential extraction analyses to estimate the lithogenic activity of uranium and thorium isotopes [27]. Briefly, the sequential process usually uses strong extractants to obtain various chemical fractions, which are U-Th bound with Fe–Mn oxyhydroxides (F1); U-Th bound with organic matter (F2) and lastly U-Th bound with lithogenic particles (F3). For F1 fraction, the elements were leached with 30 ml of 0.1 M $NH_2OH.HCL$ in 25% acetic acid heated 60°C for 1 h in a water bath. While, for the F2 fraction, the elements were leached by 3 ml concentrated H_2O_2 heated on a hot plate at <90°C for 2 h and finally the F3 fractions was digest completely with mixing of HNO_3, HCL, H_2O_2 and HF(3:2:2:1 ratio v/v) to estimate the lithogenic uranium and thorium isotopes. The sequential extraction analyses were performed to determine the activities of lithogenic uranium and thorium isotopes in the top (0-3 cm) sediment of the samples.

Trace elements and organic carbon analysis

Trace elements were also analyzed using published analytical procedures; about 150 mg of sediment were digested with total digestion procedures with a mix of concentrated HF, HNO_3 and HCl acid (1:2:1 v/v ratio) in a Teflon beaker for 2 h [28,29]. Then the concentration of trace elements such as Al, Ca, Ba and Zr were measured using an Inductive Couple Mass Spectroscopy (ICP-MS) Perkin Elmer Elan 9000. The content of organic matters was also measured using published methodology [30,31].

Method validation

The analytical procedures for uranium isotope analyses were validated with a NIST standard reference material (SRM IAEA-315), while trace elements with SRM 1646a and SY-4 Diorite Gneiss (Table 2). All the trace elements results had a good yield recovery except for Ca and Zr because the formation of calcium with fluoride ion might have been generated from the hydrofluoric acid during the digestion process. While low recovery was obtained for Zr, this might be related to the type of SRM SYS4-diorite gneiss which is from the rock; the digestion procedures applied in this study were only suitable for normal marine sediments [28]. The average recovery for radionuclide is within the acceptable values (80-100%).

Results and Discussion

Distribution and origin of uranium isotopes in harbour sediment

The average activities of ^{234}U and ^{238}U in harbours sediment in PM ranged from 49.52 ± 5.55 Bq/kg to 63.70 ± 5.65 Bq/kg and 44.21 ± 5.83 Bq/kg to 81.27 ± 7.48 Bq/kg, respectively. On the other hand, in EM harbours sediment ranged from 35.31 ± 4.39 Bq/kg to 39.57 ± 4.94 and 31.25 ± 4.50 to 41.19 ± 5.25 Bq/kg. Average activity concentrations of ^{230}Th and ^{232}Th in PM harbours ranged from 89.71 ± 6.45 Bq/kg to 119.59 ± 14.98 Bq/kg and 91.11 ± 6.41 Bq/kg to 150.83 ± 13.86 Bq/kg, respectively. But the activity of ^{230}Th and ^{232}Th in EM harbour ranged from 38.24 ± 3.62 Bq/kg 60.73 ± 4.52 Bq/kg and 28.08 ± 3.26 Bq/kg to 35.18 ± 3.20 Bq/kg, respectively. The overall data for uranium and thorium isotopes and heavy metals in marine sediment harbours are shows in Appendix's 1 and 2, respectively. In general, concentrations of uranium and thorium isotopes obtained from PM harbours are higher compared to EM harbours indicating more U-Th-rich minerals in the drainage basin of Peninsular Malaysia. Furthermore, activity concentrations of thorium isotopes in sediment samples from eastern and western coasts of Peninsular Malaysia can be considered high compared to other locations such as the Gulf of Thailand [18], the eastern coast of the Thai gulf [32] and northeastern Taiwan [6].

The vertical profiles of uranium and thorium isotopes at the surface layer (~30 cm) from all harbours sediment showed a zig zag trend (Figure 2), which might be due to the re-suspension process that frequently takes place in harbours sediment. Previous studies show that chemicals deposited in harbour sediment are frequently disturbed by re-suspension processes [4]. Frequent shipping activities also result in more internal currents, potentially making surface sediment release significant quantities of pollutants such as uranium and thorium isotopes, from the harbour floor [4]. The vertical profile of uranium in PM harbours sediment shows consistent values but do not show any significant trend except at KLG and SDL harbours. At KLG harbours, the trend shows slightly increasing values of ^{234}U and ^{238}U as well as a ratio value of $^{234}U/^{238}U$ from bottom to the top with a range value of 0.96 to 1.27 as shown in Table 3. This suggests that this area receives continuous uranium input from the water column which is due to the reduction process of U(VI) to U(IV). The sampling stations are located adjacent to the Klang estuary which is subject to a great deal of environmental stress [20,21,33]. The polluted sediment transported by the rivers show very high oxygen demand resulting from low Dissolved Oxygen (DO) values in bottom water at this area. Previous studies show that the value of DO in the bottom water is <0.5 mg/L and sometimes goes down to zero, indicating anoxic conditions [33]. Our results show that surface water at KLG harbour stations have a lower DO content compared to other areas (3.39 mg/L) which indicates possible anoxic conditions in this area. It has been well documented that in low DO scenarios, enrichment of uranium is mostly by chemical reduction of U(VI) to U(IV) [34,35].

Aside from KLG harbour, SDL harbour also showed increasing trends of ^{234}U as well as $^{234}U/^{238}U$ ratio from bottom to surface sediment with values of 0.94 (45-48 cm) to 2.19 (3-6 cm). Table 3 shows the

Elements	Values	Actual values	Recovery (%)	Types of SRM
^{234}U	9.24-15.40	12.01 (Bq/kg)	76.98-128.34 (93.14)	SRM IAEA-315
^{238}U	6.70-11.19	12.10 (Bq/kg)	60.00-93.30 (80.14)	SRM IAEA-315
^{232}Th	8.57-14.55	13.01(Bq/kg)	65.93-111.95 (86.23)	SRM IAEA-315
^{230}Th	8.78-12.56	12.01(Bq/kg)	73.19-104.67 (94.07)	SRM IAEA-315
Al	2.13-2.31	2.30 (%)	92.60-100.43 (98.00)	SRM 1646a
Ca	0.31-0.34	0.52%	60.00-65.31 (62.25)	SRM 1646a
Ba	175.39-195.91	210 (mg/kg)	83.52-93.30 (89.25)	SRM 1646a
Zr	231.27-286.77	517 (mg/kg)	44.71-55.47 (48.30)	SY-4 Diorite Gneiss

Table 2: Recovery obtained from the various types of standard reference material SRM (n=5).

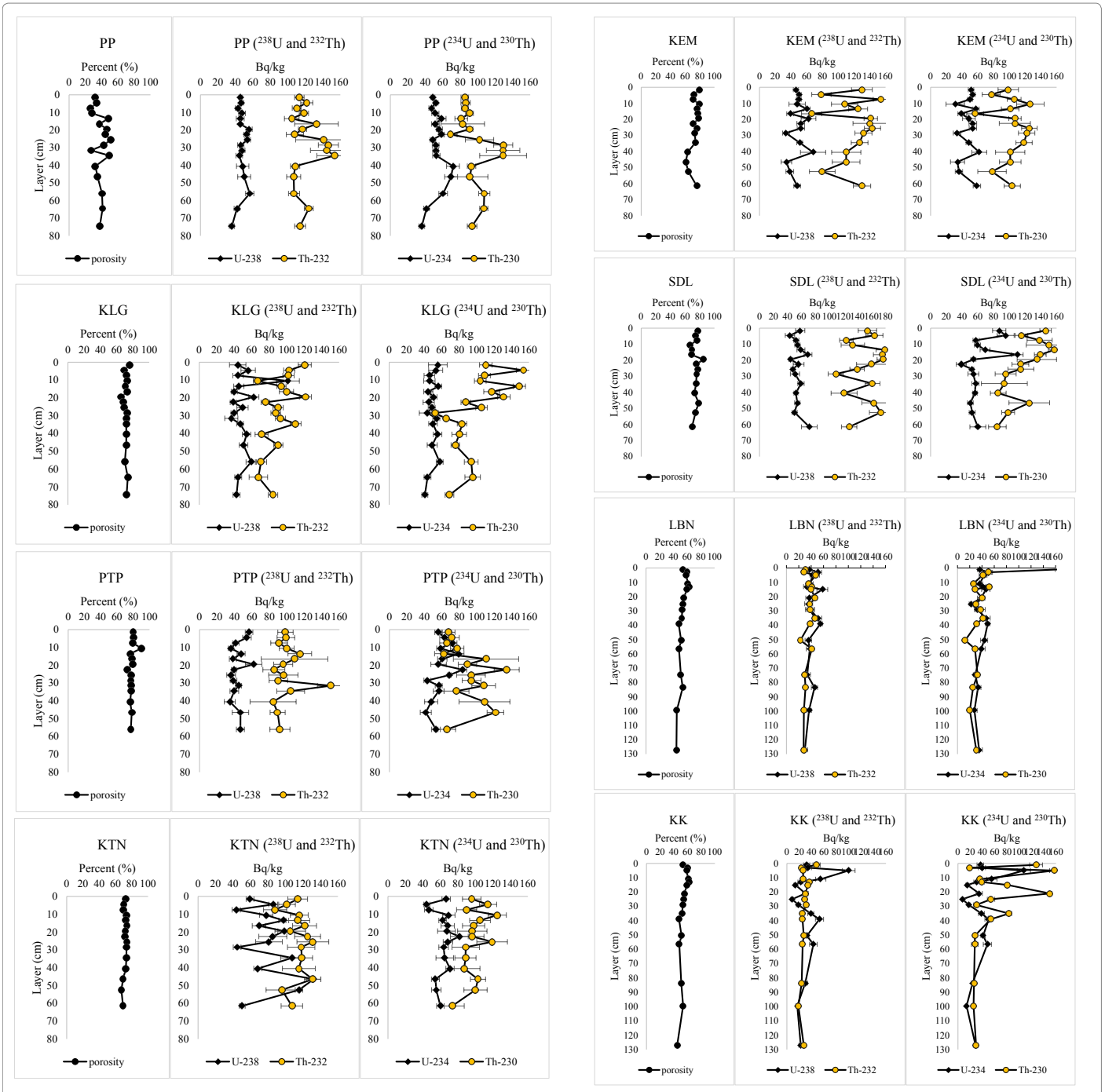

Figure 2: Vertical trends of porosity, ^{238}U, ^{232}Th, ^{234}U and ^{230}Th in marine sediment cores from Malaysian harbours.

^{234}U/^{238}U activity ratio exhibiting a marked disequilibrium with greater than unity at the surface area demonstrating the presence of authigenic ^{234}U in the sediment; this is mainly attributed to deposition by adsorption and/or adhesion onto settling particles. The SDL harbour has been characterized as having a high value of Ba/Zr (2.16 ± 0.43 to 72.82 ± 12.08) indicating the dominance of particulate barite ($BaSO_4$) compared to other harbours. This is evidence of large freshwater input into this sampling station. Furthermore, SDL harbour is located at an area of high freshwater input with low salinity (14.7 psu) and low pH value (7.31) compared to other harbour areas. Previous studies reported that in most surface freshwater under oxic conditions with pH value 5-9, U is in a free and soluble form and the uranyl ion UO_2^{2+} can form complexes with other ions in freshwater such as hydroxyls or carbonates, phosphates, fluorides, chlorides and also with some organic compounds such as humic acids then subsequently deposit them onto the sediment floor [36].

The origin of uranium and thorium isotopes in PM harbours is slightly different at EM harbours; most stations show lithogenic origins of uranium and thorium isotopes. The activities of ^{232}Th > ^{238}U in PM harbours compared to EM harbours (LBN and KK) are shown in

Harbour	Layer (cm)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{232}Th/^{238}U$	Ca/Al	Ba/Zr	OC%
PP	0-3	1.06 ± 0.11	0.75 ± 0.05	2.46 ± 0.16	0.062 ± 0.04	1.39 ± 0.23	9.33
	3-6	1.10 ± 0.17	0.71 ± 0.06	2.59 ± 0.21	0.057 ± 0.02	2.04 ± 0.34	8.65
	6-9	1.10 ± 0.20	0.78 ± 0.05	2.58 ± 0.21	0.056 ± 0.03	2.18 ± 0.57	9.54
	9-12	1.08 ± 0.12	0.77 ± 0.05	2.49 ± 0.23	0.054 ± 0.01	2.11 ± 0.37	10.23
	12-15	1.29 ± 0.14	0.78 ± 0.09	2.3 ± 0.29	0.069 ± 0.03	2.10 ± 0.43	9.51
	15-18	1.13 ± 0.10	0.62 ± 0.22	2.92 ± 0.55	0.053 ± 0.03	2.06 ± 0.45	10.6
	18-21	1.01 ± 0.11	0.78 ± 0.05	2.1 ± 0.16	0.049 ± 0.03	1.92 ± 0.36	10.34
	21-24	1.11 ± 0.08	0.64 ± 0.09	2.05 ± 0.22	0.053 ± 0.03	2.17 ± 0.33	10.41
	24-27	0.90 ± 0.07	0.72 ± 0.2	2.61 ± 0.61	0.053 ± 0.02	2.07 ± 0.33	10.38
	27-30	1.12 ± 0.20	0.88 ± 0.1	3.17 ± 0.35	0.051 ± 0.03	2.37 ± 0.46	10.59
	33-36	1.09 ± 0.09	0.89 ± 0.18	3.03 ± 0.44	0.05 ± 0.02	2.08 ± 0.14	10.18
	39-42	1.19 ± 0.13	0.84 ± 0.21	3.46 ± 0.54	0.052 ± 0.02	1.80 ± 0.26	8.60
	45-48	1.48 ± 0.18	0.86 ± 0.06	2.24 ± 0.35	0.076 ± 0.04	1.68 ± 0.26	9.55
	51-54	1.39 ± 0.17	0.85 ± 0.31	2.13 ± 0.68	0.049 ± 0.03	1.14 ± 0.25	9.48
	60-63	1.07 ± 0.13	1.00 ± 0.08	1.90 ± 0.20	0.051 ± 0.03	1.22 ± 0.36	9.55
	69-72	0.99 ± 0.13	0.86 ± 0.05	2.97 ± 0.26	0.058 ± 0.03	1.10 ± 0.18	9.70
	78-81	1.00 ± 0.13	0.82 ± 0.07	3.17 ± 0.31	0.047 ± 0.01	1.02 ± 0.13	9.69
Harbour	Layer (cm)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{232}Th/^{238}U$	Ca/Al	Ba/Zr	OC%
KLG	0-3	1.27 ± 0.27	0.92 ± 0.08	2.74 ± 0.61	0.29 ± 0.21	1.16 ± 0.16	6.98
	3-6	0.95 ± 0.17	1.49 ± 0.12	1.84 ± 0.29	0.22 ± 0.15	1.04 ± 0.43	5.81
	6-9	1.04 ± 0.16	1.07 ± 0.09	2.3 ± 0.34	0.27 ± 0.22	0.90 ± 0.22	6.68
	9-12	0.45 ± 0.07	1.56 ± 0.15	0.66 ± 0.1	0.18 ± 0.11	0.86 ± 0.36	6.56
	12-15	1.24 ± 0.12	1.58 ± 0.12	2.08 ± 0.29	0.17 ± 0.10	0.70 ± 0.55	5.77
	15-18	1.12 ± 0.16	1.18 ± 0.13	2.49 ± 0.3	0.15 ± 0.07	2.09 ± 0.93	6.87
	18-21	0.81 ± 0.12	1.08 ± 0.09	1.95 ± 0.21	0.23 ± 0.12	1.94 ± 0.27	5.22
	21-24	1.16 ± 0.17	1.16 ± 0.10	1.93 ± 0.22	0.18 ± 0.08	2.84 ± 0.86	5.23
	24-27	0.98 ± 0.13	1.17 ± 0.11	1.82 ± 0.27	0.22 ± 0.12	2.04 ± 0.27	6.95
	27-30	1.09 ± 0.25	0.6 ± 0.06	2.18 ± 0.25	0.25 ± 0.16	2.77 ± 0.22	6.10
	33-36	1.47 ± 0.16	0.70 ± 0.04	2.53 ± 0.54	0.25 ± 0.14	2.40 ± 0.45	5.30
	39-42	1.08 ± 0.12	0.76 ± 0.07	2.36 ± 0.24	0.15 ± 0.05	2.61 ± 0.55	7.14
	45-48	1.02 ± 0.11	1.12 ± 0.17	1.32 ± 0.18	0.12 ± 0.03	3.21 ± 0.45	7.13
	51-54	0.97 ± 0.13	0.84 ± 0.07	1.78 ± 0.2	0.16 ± 0.06	2.38 ± 0.48	6.89
	60-63	0.97 ± 0.11	1.33 ± 0.16	1.19 ± 0.16	0.18 ± 0.10	3.73 ± 2.36	7.33
	69-72	0.98 ± 0.12	1.41 ± 0.26	1.52 ± 0.27	0.21 ± 0.13	2.44 ± 0.32	7.94
	78-81	0.96 ± 0.14	0.82 ± 0.08	1.98 ± 0.22	0.28 ± 0.04	2.11 ± 0.34	7.83
Harbour	Layer (cm)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{232}Th/^{238}U$	Ca/Al	Ba/Zr	OC%
PTP	0-3	0.99 ± 0.09	0.68 ± 0.1	1.73 ± 0.2	0.12 ± 0.06	2.12 ± 0.35	10.97
	3-6	1.16 ± 0.13	0.72 ± 0.12	1.82 ± 0.25	0.13 ± 0.08	2.06 ± 0.4	10.38
	6-9	1.74 ± 0.20	0.72 ± 0.12	2.19 ± 0.3	0.15 ± 0.09	2.5 ± 0.29	9.65
	9-12	1.62 ± 0.23	0.78 ± 0.1	2.78 ± 0.34	0.13 ± 0.08	2.86 ± 0.35	10.27
	12-15	1.66 ± 0.21	0.55 ± 0.11	2.41 ± 0.37	0.16 ± 0.13	2.41 ± 0.58	10.04
	15-18	1.58 ± 0.19	1.02 ± 0.50	2.85 ± 1.04	0.22 ± 0.20	2.29 ± 0.37	10.00
	18-21	0.91 ± 0.17	0.93 ± 0.15	1.55 ± 0.29	0.19 ± 0.16	2.53 ± 0.53	9.96
	21-24	2.13 ± 0.17	1.58 ± 0.29	2.16 ± 0.36	0.19 ± 0.16	2.25 ± 0.55	9.72
	24-27	1.90 ± 0.14	0.97 ± 0.24	2.65 ± 0.59	0.14 ± 0.07	2.20 ± 0.41	9.48
	27-30	1.12 ± 0.11	1.04 ± 0.17	2.32 ± 0.35	0.19 ± 0.18	2.19 ± 0.33	9.44
	33-36	1.27 ± 0.21	0.72 ± 0.12	3.36 ± 0.42	0.15 ± 0.12	2.58 ± 0.52	9.52
	39-42	1.42 ± 0.19	0.74 ± 0.18	2.62 ± 0.52	0.16 ± 0.12	2.25 ± 0.33	9.60
	45-48	1.38 ± 0.25	1.29 ± 0.53	2.43 ± 0.88	0.20 ± 0.18	2.31 ± 0.27	9.32
	51-54	0.89 ± 0.16	1.36 ± 0.17	1.91 ± 0.42	0.16 ± 0.13	2.57 ± 0.55	9.77
	60-63	1.14 ± 0.15	0.71 ± 0.14	1.97 ± 0.31	0.21 ± 0.19	2.23 ± 0.40	9.46
Harbour	Layer (cm)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{232}Th/^{238}U$	Ca/Al	Ba/Zr	OC%
KTN	0-3	1.12 ± 0.1	0.84 ± 0.12	1.92 ± 0.24	0.2 ± 0.02	0.99 ± 0.12	6.05
	3-6	0.52 ± 0.05	1.12 ± 0.14	1.18 ± 0.13	0.26 ± 0.03	1.4 ± 0.25	5.01
	6-9	1.07 ± 0.15	1.02 ± 0.20	2.02 ± 0.37	0.25 ± 0.03	1.91 ± 0.4	5.12
	9-12	0.9 ± 0.14	1.08 ± 0.13	1.49 ± 0.2	0.26 ± 0.04	1.66 ± 0.26	6.43
	12-15	0.64 ± 0.08	0.92 ± 0.15	1.17 ± 0.15	0.21 ± 0.03	1.63 ± 0.34	6.10
	15-18	0.98 ± 0.13	0.80 ± 0.14	1.75 ± 0.28	0.25 ± 0.03	1.52 ± 0.38	5.14
	18-21	0.68 ± 0.12	0.91 ± 0.22	1.07 ± 0.19	0.24 ± 0.04	1.79 ± 0.36	5.58
	21-24	0.96 ± 0.13	0.77 ± 0.14	1.47 ± 0.32	0.22 ± 0.02	1.53 ± 0.43	5.48

	24-27	0.85 ± 0.09	0.91 ± 0.18	1.62 ± 0.38	0.24 ± 0.03	2.15 ± 0.51	4.77
	27-30	1.43 ± 0.13	0.75 ± 0.17	2.64 ± 0.44	0.21 ± 0.02	2.04 ± 0.28	4.30
	33-36	0.60 ± 0.13	0.75 ± 0.13	1.1 ± 0.18	0.27 ± 0.05	1.63 ± 0.29	4.81
	39-42	1.04 ± 0.11	0.76 ± 0.20	1.68 ± 0.3	0.28 ± 0.03	2.02 ± 0.36	4.42
	45-48	0.41 ± 0.05	0.79 ± 0.09	0.99 ± 0.09	0.27 ± 0.03	1.51 ± 0.28	3.39
	51-54	0.48 ± 0.07	1.04 ± 0.24	0.83 ± 0.16	0.23 ± 0.03	1.95 ± 0.32	4.69
	60-63	1.19 ± 0.14	0.68 ± 0.15	2.13 ± 0.28	0.28 ± 0.04	2.20 ± 0.31	5.20
Harbour	Layer (cm)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{232}Th/^{238}U$	Ca/Al	Ba/Zr	OC%
KEM	0-3	1.11 ± 0.13	0.76 ± 0.13	2.82 ± 0.35	0.12 ± 0.02	4.47 ± 0.55	8.24
	3-6	1.07 ± 0.11	0.98 ± 0.21	1.57 ± 0.26	0.08 ± 0.01	3.31 ± 0.55	7.58
	6-9	1.00 ± 0.10	0.69 ± 0.07	3.09 ± 0.28	0.07 ± 0.01	5.33 ± 0.82	7.29
	9-12	0.66 ± 0.25	1.17 ± 0.24	2.26 ± 0.6	0.06 ± 0.01	8.87 ± 1.53	7.94
	12-15	0.96 ± 0.09	0.81 ± 0.12	2.08 ± 0.24	0.13 ± 0.02	4.69 ± 0.90	5.95
	15-18	1.00 ± 0.14	0.85 ± 0.23	1.7 ± 0.38	0.15 ± 0.03	5.67 ± 1.18	7.37
	18-21	0.78 ± 0.16	0.76 ± 0.07	2.27 ± 0.37	0.08 ± 0.01	3.67 ± 0.63	8.09
	21-24	1.03 ± 0.11	0.76 ± 0.18	2.68 ± 0.48	0.1 ± 0.02	5.82 ± 0.91	6.13
	24-27	1.02 ± 0.14	0.87 ± 0.09	2.73 ± 0.31	0.1 ± 0.01	4.42 ± 0.81	6.63
	27-30	1.00 ± 0.17	0.92 ± 0.11	3.92 ± 0.59	0.14 ± 0.03	4.47 ± 0.82	7.73
	33-36	0.95 ± 0.1	0.93 ± 0.11	2.49 ± 0.28	0.09 ± 0.01	3.91 ± 0.28	7.74
	39-42	0.91 ± 0.15	0.92 ± 0.23	1.62 ± 0.47	0.12 ± 0.01	4.98 ± 0.63	5.26
	45-48	1.00 ± 0.27	0.92 ± 0.19	3.22 ± 0.81	0.14 ± 0.01	4.72 ± 0.74	5.30
	51-54	0.94 ± 0.14	0.98 ± 0.3	2.07 ± 0.5	0.18 ± 0.03	4.99 ± 0.88	5.00
	60-63	1.23 ± 0.15	0.80 ± 0.10	2.72 ± 0.33	0.14 ± 0.02	3.29 ± 0.51	8.47
Harbour	Layer (cm)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{232}Th/^{238}U$	Ca/Al	Ba/Zr	OC%
SDL	0-3	1.5 ± 0.16	0.95 ± 0.06	2.65 ± 0.32	0.05 ± 0.01	3.32 ± 0.49	8.99
	3-6	2.19 ± 0.29	0.7 ± 0.06	3.77 ± 0.61	0.06 ± 0.01	23.4 ± 3.77	8.21
	6-9	1.1 ± 0.12	1.11 ± 0.19	2.37 ± 0.34	0.07 ± 0.01	72.82 ± 12.05	8.73
	9-12	1.1 ± 0.15	1.12 ± 0.25	2.46 ± 0.32	0.11 ± 0.01	23.09 ± 4.44	4.97
	12-15	1.17 ± 0.16	0.88 ± 0.06	3.02 ± 0.32	0.07 ± 0.01	65.41 ± 14.15	4.10
	15-18	1.59 ± 0.16	0.79 ± 0.05	2.53 ± 0.23	0.11 ± 0.02	52.95 ± 12.63	3.23
	18-21	1.21 ± 0.13	0.77 ± 0.18	3.94 ± 0.6	0.06 ± 0.01	4.67 ± 0.96	10.26
	21-24	0.7 ± 0.12	0.71 ± 0.08	2.87 ± 0.44	0.09 ± 0.02	4.05 ± 0.83	8.49
	24-27	1.09 ± 0.1	0.82 ± 0.15	2.94 ± 0.38	0.07 ± 0.01	4.48 ± 0.95	7.57
	27-30	1.07 ± 0.14	0.88 ± 0.16	2.15 ± 0.38	0.04 ± 0.01	3.52 ± 0.71	7.73
	33-36	0.96 ± 0.16	0.58 ± 0.21	2.73 ± 0.53	0.06 ± 0	3.42 ± 0.4	5.90
	39-42	1.08 ± 0.1	0.71 ± 0.1	2.32 ± 0.23	0.08 ± 0.01	7.32 ± 1.05	8.10
	45-48	0.94 ± 0.11	0.77 ± 0.2	3.03 ± 0.51	0.05 ± 0.01	3.55 ± 0.62	8.82
	51-54	1.05 ± 0.13	0.57 ± 0.05	3.45 ± 0.3	0.04 ± 0	4.88 ± 1.11	9.58
	60-63	0.84 ± 0.16	0.66 ± 0.1	1.8 ± 0.32	0.07 ± 0.01	2.16 ± 0.43	6.60
Harbour	Layer (cm)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{232}Th/^{238}U$	Ca/Al	Ba/Zr	OC%
LBN	0-2	0.97 ± 0.12	5.41 ± 0.59	0.82 ± 0.12	0.53 ± 0.06	1.71 ± 0.21	4.62
	2-4	0.97 ± 0.14	1.79 ± 0.32	0.55 ± 0.08	0.52 ± 0.06	0.69 ± 0.12	4.27
	4-6	1.03 ± 0.16	0.86 ± 0.15	1.13 ± 0.17	0.56 ± 0.07	1.13 ± 0.24	4.61
	10-12	0.94 ± 0.17	0.72 ± 0.1	0.94 ± 0.15	0.63 ± 0.09	1.91 ± 0.38	4.90
	12-14	1.23 ± 0.2	1.25 ± 0.13	1.24 ± 0.23	0.59 ± 0.07	2.16 ± 0.52	4.72
	14-16	0.8 ± 0.14	0.71 ± 0.09	0.69 ± 0.12	0.58 ± 0.07	1.32 ± 0.36	4.91
	20-22	1.05 ± 0.2	0.87 ± 0.09	1.27 ± 0.21	0.62 ± 0.09	4.66 ± 0.93	3.96
	24-26	0.62 ± 0.12	0.8 ± 0.11	1.05 ± 0.21	0.44 ± 0.03	6.59 ± 1.13	3.29
	28-30	0.83 ± 0.14	0.98 ± 0.28	0.99 ± 0.24	0.5 ± 0.06	4.83 ± 0.82	4.16
	34-36	0.93 ± 0.14	0.91 ± 0.08	0.91 ± 0.13	0.59 ± 0.06	14.03 ± 2.55	4.11
	38-40	0.9 ± 0.09	0.83 ± 0.09	0.69 ± 0.07	0.62 ± 0.1	8.33 ± 0.78	3.92
	49-52	1.23 ± 0.19	0.56 ± 0.15	0.63 ± 0.13	0.68 ± 0.06	4.59 ± 0.71	3.73
	55-58	1.03 ± 0.19	0.7 ± 0.08	1.09 ± 0.18	0.67 ± 0.08	5.53 ± 1.06	3.58
	73-76	0.87 ± 0.14	1.1 ± 0.15	0.88 ± 0.16	0.6 ± 0.08	1.13 ± 0.23	3.23
	82-87	0.74 ± 0.12	0.83 ± 0.1	0.68 ± 0.09	0.69 ± 0.09	1.42 ± 0.25	3.47
	97-102	0.76 ± 0.14	0.72 ± 0.13	0.77 ± 0.11	0.8 ± 0.07	0.66 ± 0.1	3.39
	122-142	1.19 ± 0.2	1.11 ± 0.14	0.93 ± 0.16	0.84 ± 0.15	1.09 ± 0.15	3.06
	142-147	1.34 ± 0.22	1.09 ± 0.14	0.52 ± 0.07	1.13 ± 0.15	1.24 ± 0.15	2.59
Harbour	Layer (cm)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{232}Th/^{238}U$	Ca/Al	Ba/Zr	OC%

KK	0-2	1.14 ± 0.21	2.66 ± 0.39	1.5 ± 0.30	0.74 ± 0.09	0.47 ± 0.07	4.50
	2-4	1.14 ± 0.2	0.84 ± 0.17	0.67 ± 0.14	0.7 ± 0.09	0.46 ± 0.08	4.20
	4-6	1.08 ± 0.14	6.09 ± 0.72	0.26 ± 0.04	0.7 ± 0.06	0.65 ± 0.1	4.54
	10-12	1.01 ± 0.16	1.35 ± 0.18	0.49 ± 0.09	0.52 ± 0.05	1.28 ± 0.22	4.83
	12-14	1.4 ± 0.25	1.07 ± 0.15	1.64 ± 0.34	0.56 ± 0.06	1.23 ± 0.22	4.66
	14-16	1.11 ± 0.2	2.41 ± 0.2	2.4 ± 0.47	0.65 ± 0.1	2.92 ± 0.52	4.84
	20-22	1.16 ± 0.22	4.93 ± 0.5	1.02 ± 0.19	0.94 ± 0.17	2.75 ± 0.48	3.90
	24-26	0.84 ± 0.12	1.9 ± 0.33	3.32 ± 0.92	0.91 ± 0.19	1.87 ± 0.31	2.42
	28-30	0.96 ± 0.16	0.95 ± 0.17	1.69 ± 0.35	1.09 ± 0.19	1.86 ± 0.41	2.92
	34-36	1 ± 0.16	3.4 ± 0.44	0.64 ± 0.11	1.1 ± 0.21	0.83 ± 0.18	3.32
	38-40	0.96 ± 0.14	2.11 ± 0.28	0.48 ± 0.08	1.59 ± 0.17	1.41 ± 0.18	3.52
	49-52	1.27 ± 0.13	1.02 ± 0.15	0.84 ± 0.12	1.68 ± 0.12	1.44 ± 0.38	3.42
	55-58	1.12 ± 0.17	1.08 ± 0.25	0.6 ± 0.11	1.53 ± 0.16	1.3 ± 0.33	3.49
	82-87	0.81 ± 0.11	1.09 ± 0.17	0.81 ± 0.14	1.43 ± 0.18	1.8 ± 0.25	3.23
	97-102	0.84 ± 0.12	1.37 ± 0.26	1.1 ± 0.23	1.2 ± 0.17	1.68 ± 0.24	2.89
	122-142	1.28 ± 0.19	1.08 ± 0.16	1.22 ± 0.21	1.07 ± 0.17	1.59 ± 0.25	n.d
	142-147	1.4 ± 0.19	2.09 ± 0.3	0.97 ± 0.14	1.15 ± 0.19	1.83 ± 0.33	n.d

Note: n.d: Not determined, due to less amount of samples to analyses

Table 3: Activity ratios of radionuclide, trace elements isotopes and organic carbon in Malaysian harbour sediments.

Figure 2. Table 3 shows the ratio of $^{232}Th/^{238}U$ where most harbours in PM show high values of >1 suggesting greater abundance of thorium in the PM earth crust due to the influence of basement granitic rock. Peninsular Malaysia shows extensive granitic rock compare to other areas in Southeast Asia [24]. Thorium is highly concentrated in granitic rock with values ranging from 8-33 ppm while the value of Th/U is 3.5-6.3 [37]. During the weathering process, thorium and uranium in the tetravalent state can be adsorbed onto surface minerals of granitic rock such as apatite, monazite and zircon then transport from the rivers to the oceans [37,38]. These assumptions have been supported with good correlation observed between ^{238}U and Zr in all sediment cores from the east coast of PM ($r=0.323$, $p<0.05$) (Table 4) suggesting that uranium might be adsorbed onto Zr minerals in granitic rock, then transported and deposited in sediment harbour. Furthermore most of the activity ratios of $^{234}U/^{238}U$ in the east coast of PM such as at KEM and KTN harbours are <1 with the average activity ratio of both locations being <1 (Table 3). This suggests that these areas receive high lithogenic input where most ^{238}U originates from the river [39,40]. Good correlation is also observed between $^{234}U/^{238}U$ with lithogenic particles such as Al ($r=0.399$, $p<0.01$) in the west coast of PM (Table 3) suggesting the influence of clay minerals such as aluminosilicate in the distribution of uranium in the west coast of PM. Previous studies show that Al can be used as a proxy to investigate the influence of aluminosilicate as a carrier for radionuclide elements [41].

The ratio value of $^{230}Th/^{232}Th$ also supports the above assumptions. The low value of $^{230}Th/^{232}Th<1$ observed in PM compared to EM indicates lithogenic origins while high values of $^{230}Th/^{232}Th$ were observed at the EM coast (LBN and KK) (Table 3) with average values of 1.18 ± 0.16 and 2.14 ± 0.32 respectively, suggesting more authigenic inputs of thorium from the water column deposited in EM harbours [5,41-43]. High inputs of thorium in PM are due to the detrital input of granitic rock. This suggestion is further supported by high values of ^{226}Ra and ^{228}Ra detected on the east coast of Peninsular Malaysia; ranging between 46.23 and 121.49 Bq/kg, suggesting the dominance of granitic input to coastal areas of Peninsular Malaysia [44].

Harbour sediments in EM i.e., LBN and KK shows a dominance of biogenic origin as characterized by high values of Ca/Al (0.52 ± 0.05-1.68 ± 0.12) due to the deposition of calcium carbonate in these areas (Table 3). Most of the uranium and thorium in EM are of authigenic origin as printing by $^{234}U/^{238}U$, $^{232}Th/^{238}U$ and $^{230}Th/^{232}Th$. LBN and KK harbours which are located at the EM show almost similar trends of $^{234}U/^{238}U$ ratios which indicates that uranium at the EM coast (LBN and KK) might share similar geochemical behaviors of uranium where the precipitation of uranium to sediment might be influenced by similar particles of variable composition. In addition, surface water of these locations are well oxygenated, therefore authigenic uranium should be in U(VI) as uranyl ion UO_2^{2+}. The U(VI) will co-precipitate with other particles and be deposited onto the sediment. Good correlation was observed between ^{238}U and OC in all stations from EM harbours (Table 4) indicating the OC bound uranium is a significant source of authigenic uranium in EM harbours.

Accumulation of uranium in Malaysian harbours

Uranium accumulation models have been developed to estimate the accumulation of lithogenic and authigenic uranium in Malaysian harbour sediment. The activity of lithogenic and authigenic uranium is calculated from equations 1 through 3 [14,45]. Normalization to the ^{232}Th activities was performed because ^{232}Th is a reliable indication of the presence of lithogenic origin [45,46].

$$U(total)=U(litho)+U(aut) \quad (1)$$

$$U(aut)=U(total)-U(litho) \quad (2)$$

$$U(litho)=\text{lithogenic fraction} \times {}^{232}Th(total) \quad (3)$$

Where, the U(aut) is Authigenic U, the U(total) is Total ^{238}U obtained from sample, the lithogenic fraction is $(^{238}U/^{232}Th)_{litho}$ of which the value of ^{238}U and ^{232}Th is obtained from the lithogenic fraction during the sequential extraction process, and the ^{232}Th(total) is Total ^{232}Th is obtained from samples.

The lithogenic fraction $(^{238}U/^{232}Th)_{litho}$ was obtained from the activities of ^{238}U and ^{232}Th at the residual phase, which was in turn obtained from sequential extraction analysis. The residual phase consists of primary minerals which are of lithogenic origin [47]. The results of ^{238}U and ^{232}Th in the residual phase are shown in Table 5. The result shows that the ratio of $(^{238}U/^{232}Th)_{litho}$ in the west coast of PM is 0.45 ± 0.09; east coast of PM is 0.46 ± 0.08; and EM is 0.45 ± 0.01. The result of $(^{238}U/^{232}Th)_{litho}$ obtained from this study is lower compared to ratios obtained by Anderson et al. [2] from the deep sea (0.8 ± 0.2) and also from Sakaguchi et al. from offshore bottom sediment which is 0.65 ± 0.14. Low values of $(^{238}U/^{232}Th)_{litho}$ are because of the dominance of ^{232}Th in these areas.

The accumulation rates of authigenic uranium are estimated from the following equation 4 [14,45]:

Accumulation rate (AR)=U(aut) × S × ρ (4)

Where AR is the accumulation rate of authigenic U (Bq/cm^2/year), Uaut=authigenic U (Bq/kg), S=Sedimentation rate (cm/year), ρ=Dry densities (g/cm^3).

Dry density was calculated using an equation estimated from porosity values [48]. The value of the sedimentation rate was obtained from Yusoff [49]. Assuming a steady state in the lithogenic/authigenic U ratio, this ratio must be equal to their flux ratio in the sediment. Thus, the accumulation rate of lithogenic uranium can be obtained by using equation 5 [50].

Accumulation rate of U(litho)/Accumulation rate of U(aut)=U(litho))/(U(aut) (5)

The results show that the range of accumulation rates of authigenic uranium in Malaysian harbours are between 0.66 10^{-3} Bq/cm^2/year to 6.14 × 10^{-3} Bq/cm^2/year. as shown in Table 6. On the other hand, the range of accumulation rates of lithogenic uranium is from 3.98 × 10^{-3} Bq/cm^2/year to 17.82 × 10^{-3} Bq/cm^2/year. A box model has been developed to illustrate the deposition of uranium in harbour sediment as shown in Figures 3 and 4. This model shows that in the west and east coasts of PM, lithogenic uranium supply were estimated to be 96% and 86% of the total uranium, respectively. High accumulation of lithogenic uranium in the west coast compared to the east coast of PM is because of the high sedimentation rates detected in the east coast of PM. The highest accumulation of authigenic uranium was detected at EM followed by the east then west coast of PM with the value 61%, 14% and 4%, respectively.

Low accumulation rates of authigenic uranium at PM coasts (0.66-1.87 × 10^{-3} Bq/cm^2/year) compared to EM coasts (6.14 × 10^{-3} Bq/cm^2/year) might also be related to the rapid sedimentation and mixing processes that occur in PM. These kinds of processes might dilute the authigenic uranium added to the sediments and explain why little authigenic uranium is observed in the sediment cores [51]. The accumulation of authigenic uranium at EM (6.14 × 10^{-3} Bq/cm^2/year) is higher compared to other areas such as the Okinawa trough (1.69-1.74 × 10^{-3} Bq/cm^2/year) which might be due to marginal seas characteristics where these areas might receive input form the Western Pacific ocean. Furthermore, the accumulation rate of authigenic uranium at EM (6.14 × 10^{-3} Bq/cm^2/year) is almost similar to the rate of authigenic uranium burial in sediments (up to 6.67 × 10^{-3} Bq/cm^2/year) from the Pacific Ocean [2]. We can generally conclude that uranium located in PM harbours receive a high input of lithogenic uranium due to basement granitic rock while the EM coast receives more authigenic input with effluence rom the western Pacific Ocean.

Conclusion

Harbour sediment cores were collected from PM and EM coasts. We investigated the origin of uranium and thorium isotopes in Malaysian harbour sediment and developed a model to estimate the accumulation rate of lithogenic and authigenic uranium in Malaysian harbour sediment. The average activity of uranium and thorium isotopes at PM harbours are greater than at EM harbours. The sources of uranium were successfully predicted with the activity ratio supported by Pearson correlation analysis. Most of the uranium in PM harbours are of lithogenic origin influenced by basement granitic rock while the EM coast is dominated by authigenic origin effluence from the Western Pacific ocean. A box model suggests high accumulation rates of lithogenic uranium in PM with more than 86% of lithogenic uranium deposited at PM harbours. Meanwhile, the west coast of EM is dominated by authigenic uranium estimated at 61% of total uranium.

Pearson's Correlations				
		Al	Zr	OC
^{234}U/^{238}U (west coast of PM)	Pearson Correlation	**0.399****		
	Sig. (2-tailed)	0.004		
	N	50		
^{238}U (east coast of PM)	Pearson Correlation		**0.323***	
	Sig. (2-tailed)		0.028	
	N		46	
^{238}U (EM)	Pearson Correlation			0.**598****
	Sig. (2-tailed)			0
	N			32
**Correlation is significant at the 0.01 level (2-tailed).				
*Correlation is significant at the 0.05 level (2-tailed).				

Table 4: Pearson correlation of uranium with Al, Zr and organic carbon (OC) in all sediment cores from PM and EM harbours.

Area	Harbour name	$^{238}U_{litho}$	$^{232}Th_{litho}$	$(^{238}U/^{232}Th)_{litho}$	$(^{238}U/^{232}Th)_{litho}$
West coast of PM	PP	23.92 ± 3.26	63.35 ± 5.69	0.38 ± 0.05	0.45 ± 0.09
	KLG	43.74 ± 3.75	104.83 ± 9.51	0.42 ± 0.05	
	PTP	27.78 ± 3.63	50.32 ± 6.75	0.55 ± 0.09	
East coast of PM	KTN	40.39 ± 5.67	76.88 ± 11.39	0.53 ± 0.10	0.46 ± 0.08
	KEM	28.73 ± 3.72	80.10 ± 7.47	0.36 ± 0.05	
	SDL	50.67 ± 5.56	101.61 ± 12.46	0.50 ± 0.07	
EM	LB	12.66 ± 1.75	28.30 ± 4.02	0.45 ± 0.08	0.45 ± 0.01
	KK	13.60 ± 1.72	29.51 ± 3.49	0.46 ± 0.07	

Table 5: Ratio of $(^{238}U/^{232}Th)_{litho}$ obtained from the residual phase in sequential extraction analyses.

Box	Area	Total U (Bq/kg)	$^{238}U/^{232}Th$	^{232}Th (Bq/kg)	U(litho) (Bq/kg)	U(aut) (Bq/kg)	Sedimentation rate (cm/year)	Bulk density (g/cm^3)	AR of U (total)	AR of U (auth)	AR of U (litho)	Aut U	Litho U
									(Bq/cm^2/year) × 10^{-3}			(%)	(%)
A	West coast of PM	47.36 ± 3.12	0.45 ± 0.09	101.51 ± 17.22	45.68 ± 9.58	1.68 ± 0.37	0.39 ± 0.06	1.00 ± 0.12	18.47 ± 0.13	0.66 ± 0.19	17.82 ± 0.19	4	96
B	East coast of PM	61.87 ± 17.11	0.46 ± 0.08	116.04 ± 20.22	53.38 ± 10.92	8.5 ± 2.92	0.31 ± 0.05	0.72 ± 0.10	13.59 ± 0.65	1.87 ± 0.76	11.72 ± 0.55	14	86
C	West coast of EM	36.22 ± 7.02	0.45 ± 0.01	31.63 ± 5.02	14.23 ± 2.27	21.99 ± 5.51	0.25 ± 0.04	1.14 ± 0.17	10.12 ± 1.81	6.14 ± 2.05	3.98 ± 1.67	61	39

Note: Litho: Lithogenic, Aut: Authgenic, AR: Accumulation Rate. Total U obtained from the average activities of ^{238}U from each area, ratio of ^{238}U/^{232}Th obtained from residual phase in sequential extraction analyses, Lithogenic U is calculated from the equation 3 while accumulation rate (AR) are calculated by the equation 4. Value of sedimentation rate obtained from Yusoff [49].

Table 6: Calculation of the deposition model of uranium.

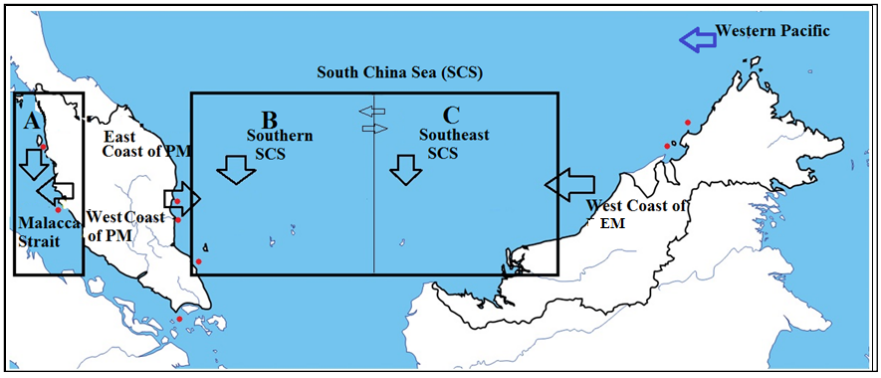

Figure 3: Illustration of uranium deposition model developed for the Malacca Strait and South China Sea (SCS). Red dots indicate sampling areas. Box A refers to the Malacca Straits located on the west coast of PM, Box B refers to the Southern SCS located on the east coast of PM and Box C refers to the Southeast SCS located on the west coast of EM.

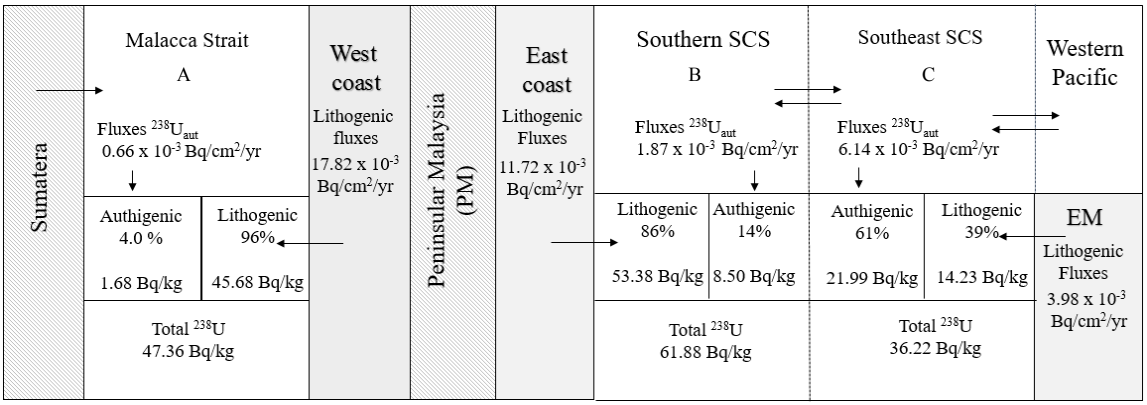

Figure 4: Schematic diagram summarizing the box-model calculations on the fluxes of lithogenic and authigenic U isotopes in the PM and EM harbours. See text for definitions of notations not indicated in the diagram. High accumulation rate (flux) of lithogenic U in PM is due to the input of basement granitic rock, while a high accumulation rate (flux) of authigenic U in EM might be due to the effluence from the Western Pacific.

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