Vertical Profiles of Natural Uranium Isotopes in Sediment Cores from Kota Kinabalu and Labuan Ports, Malaysia

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Abstract

Sediment cores were collected from Kota Kinabalu and Labuan ports to determine the distribution of natural uranium isotopes, calcium (Ca), aluminium (Al) and total organic matter (TOM). A strong statistical correlation value between $^{234}U/^{238}U$ and Ca contents were shown at Kota Kinabalu ($r^2 = 0.811$), and Labuan ($r^2 = 0.833$) ports. Good statistical correlation $r^2 > 0.8$ was also obtained between uranium isotopes and TOM at Kota Kinabalu port. Most activity ratio values of $^{234}U/^{238}U$ found in sediment cores from Kota Kinabalu port were more than 1.0; the major input of uranium at Kota Kinabalu port may be related to the sorptive process of authigenic uranium that occurs in the water column. In this case, the mobile U(VI) was reduced to immobile U(IV) by organic matter and scavenged to the sediment with co-precipitation of organic matter and calcium carbonate. On the other hand, a low activity ratio of $^{234}U/^{238}U$ (≤ 1) from Labuan was of detrital origin and also showed positive statistical correlation between calcium and aluminium.

Keywords: uranium; sediment core; total organic matter (TOM); limestone; detrital

1. Introduction

Uranium has three naturally occurring isotopes, ^{234}U (progeny of ^{238}U, half-life 2.45×10^5 years), ^{235}U (half-life 7.04×10^8 years) and ^{238}U (half-life 4.47×10^9 years). The geochemical behavior and distribution of uranium in marine systems is usually influenced by the contents of suspended particles either from organic or detrital minerals (Dawood, 2010; Henderson and Anderson, 2003; Mohamed *et al*., 2006; Zheng *et al*., 2002). Generally, uranium behaves as a nearly conservative element in seawater, but it can be precipitated under chemically reducing conditions that occur in sediments. The reduction process of U(VI) to U(IV) is highly influenced by organic matter fluxes and dissolved oxygen contents in sea water (Zheng *et al*., 2002). The reduction process of U(VI) to U(IV) has received considerable attention because the oxidation state of uranium has a significantly effect on its mobility in a natural environment (Cochran *et al*., 1986; Mohamed *et al*., 2006; Toole *et al*., 1987).

In a system that has remained closed for a sufficiently long time, activities of all radionuclides are equal to unity, a condition which called secular equilibrium. However, disequlibrium between ^{238}U and ^{234}U in the marine environment is a common phenomenon. This disequilibrium originates from a preferential release of more loosely bound ^{234}U from damaged mineral lattice sites or direct recoil of ^{234}Th into surrounding media from near mineral surface boundaries (Andersen *et al*., 2009). Variations in the $^{234}U/^{238}U$ ratio have been used as a sensitive chemical indicator for identifying geochemical processes (Andersen *et al*., 2009; Cizdziel *et al*., 2005) and to estimate uranium origin (Skwarzec *et al*., 2002; Srisuksawad *et al*., 1997). A study done by Skwarzec *et al*. (2002) have shown that the value of the $^{234}U/^{238}U$ activity ratio in sediments and bottom water should be nearly the same if the reduction process from U(VI) to U(IV) was dominant in a particular area. In this case, the major origin of uranium in sediments is by the sorptive uptake of authigenic uranium from the water column. Authigenic uranium is efficiently adsorbed onto organic matter, reduced from U(VI) to U(IV) and finally scavenged to the sediment (Rosing and Frei, 2004; Zheng *et al*., 2002). A low ratio value of < 0.95 was observed at the areas that show a high detrital input of uranium such as was reported in the Gulf of Thailand (Srisuksawad *et al*., 1997) and in the Johor Strait, which is surrounded by igneous rock (Wood *et al*., 1997).

A study of uranium isotopes, especially in marine port systems, is important and relevant so as to understand the behavior and transport of chemical constituents in rivers and oceans as a result of systems. The radionuclide distribution in marine port systems might originate from sea water, lithogenic sources and anthropogenic sources such as waste. The geochemical behaviors of uranium in marine port systems are not

well documented by previous researchers. Furthermore, no published data on uranium in coastal areas, particularly from port areas in East Malaysia (i.e. Sarawak, Sabah and Labuan) has been found. This study has been designed to investigate the factors that influence the behavior of ^{234}U and ^{238}U in port systems, specifically in Labuan and Kota Kinabalu ports.

2. Methodology

2.1. Sample collection

Sediment cores measuring about 147 cm long were obtained from Kota Kinabalu and Labuan ports, Malaysia (Fig. 1). The coordinates of the exact sampling location were recorded using Geographical Positioning System (GPS). The locations for Kota Kinabalu and Labuan ports are Latitude 06°00'884"N Longitude 116°04'957"E and Latitude 05°16'028"N Longitude 115°14'779"E, respectively. The sediment cores were collected using a gravity corer with 130 kg weight, 250 cm length and 7.5 cm diameter.

2.2. Chemical analyses

2.2.1. Uranium isotopes analyses

Briefly, about 0.25 g of dried sediments were digested with a mixture of concentrated hydrofluoric acid (HF) and nitric acid (HNO$_3$) (1:2 v/v). Then 2 ml of perchloric acid (HClO$_4$) and 0.5 g of boric acid (H$_3$BO$_3$) were added to remove excess fluoride from the samples. The cation column of Bio-Rad AG 50W x 4 (200-400 mesh) was used to fractionate uranium isotopes from the sample solutions and purified using an anion column (Bio-Rad AG 1 x 8 100-200 mesh). The samples were then electrodeposited onto a silver disk for two hours at 200-250 mA with 15V before being analysed using an alpha spectroscopy (CANBERRA's Alpha Analyst) for 48 hours (Mohamed *et al*., 2006).

2.2.2. Analyses of trace elements and total organic matter

Fine grain sediments of about 1.5 g were digested with a mixture of concentrated HF, HNO$_3$ and HCl acid (1:2:1 v/v ratio) in a Teflon beaker for 2 hours using published procedures (e.g., Morelli *et al*., 2012; Akyil and Yusof, 2007). Total organic matter (TOM) was also estimated according to their loss of ignition (LOI), a method described by Heiri *et al*. (2001).

2.3. Accuracy of the methods

The analytical procedures for uranium isotope analyses were validated with a triplicate of NIST standard reference material (SRM 4357), while Ca and Al were validated with SRM 1646a (Table 1).
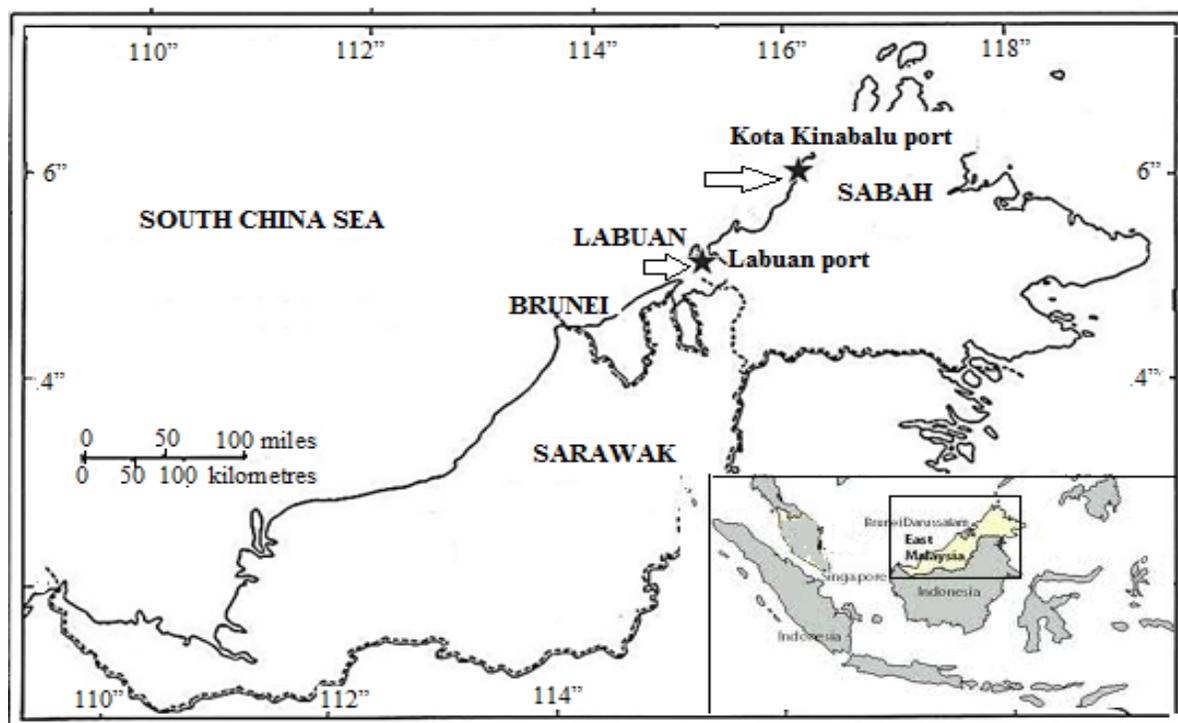
3. Results and Discussion

Figure 1. Map of the sampling stations at Kota Kinabalu and Labuan Ports, Malaysia

Table 1. Percentage of recovery obtained from the certified reference material SRM 4357 and SRM 1646a

Samples	Concentration				SRM Value				Recovery (%)			
	^{234}U (Bq/kg)	^{238}U (Bq/kg)	Ca (%)	Al (%)	^{234}U (Bq/kg)	^{238}U (Bq/kg)	Ca (%)	Al (%)	^{234}U	^{238}U	Ca	Al
Rep. 1	9.24	11.41	0.36	1.95					76.98	95.07	70.00	85.03
Rep. 2	10.54	9.10	0.38	2.12	12	12	0.52	2.30	87.84	75.86	73.76	92.49
Rep. 3	10.57	10.81	0.38	2.29					88.11	90.11	73.76	99.82
Mean	10.12	10.44	0.37	2.12	Average recovery				84.31±7.68	87.02±9.97	72.44±2.29	92.44±7.40

3.1. Geochemical of ^{234}U and ^{238}U in sediment core

The results of ^{234}U and ^{238}U in sediment cores from Kota Kinabalu and Labuan ports are presented in Table 2. The activities of ^{234}U and ^{238}U detected in the sediment core at Kota Kinabalu port ranged from 0.41±0.09 dpm/g to 6.45±0.66 dpm/g and 0.38±0.09 dpm/g to 5.99±0.63 dpm/g, respectively. While the activity of ^{234}U and ^{238}U at Labuan port ranged from 1.34±0.43 dpm/g to 3.73±0.28 and 1.52±0.31 dpm/g to 3.48±0.54 dpm/g, respectively. The profile of uranium isotopes detected at both core sediments were divided into three zones, namely zone A (0-22 cm), B (24-76 cm) and zone C (> 82 cm). The activity of ^{234}U and ^{238}U show inconsistent values at the upper layers, namely zone A (0-22 cm) and might be due to a bioturbation process occurring at the top sediment. The bioturbation process caused by benthic organisms will mix a sediment containing uranium and bring it upward toward the sediment-water interface (Zheng *et al.*, 2002). While at zone B, the activities of ^{234}U and ^{238}U starting increasing from 24 to 40 cm and decreasing from 40 cm to 76 cm. These trends (zone B) might be influenced by a major carrier of uranium in these sediments and will be discussed in the next section. The increasing trends at the bottom part of > 82 cm (zone C) suggest that there is enhanced uranium addition to deeper layers by diagenetic processes. Benthic organism activities can increase the size of the effective sediment water interface and will introduce dissolved ^{238}U and ^{234}U from overlying sea water down to the sediment (Lagauzère *et al.*, 2009). The deposits of ^{238}U and ^{234}U decrease at increased depths, but there were enhanced uranium deposits found at the bottom of the sediment (Srisuksawad *et al.*, 1997; Wood *et al.*, 1997; Yamada and Tsunogai, 1984).

3.2. Factors influencing uranium isotopic composition

Uranium content in sediment depends on variations in water chemistry, geological settings and their sources (Akyil and Yusof, 2007; Chung and Chang, 1996; Dawood, 2010). Uranium in sea water could be adsorbed directly onto organic matter and deposited into sediments through the reduction process of U(VI) to U(IV) (Rosing and Frei, 2004). Uranium can also be deposited into sediment by co-precipitation with minerals of detrital origin (Dawood, 2010; Mohamed *et al.*, 2006). This section explains several factors that have influenced uranium isotopic composition in Labuan and Kota Kinabalu ports. Figs. 2 and 3 show the correlation of TOM, Ca and Al with uranium isotopes at zone B off Kota Kinabalu and Labuan ports. The correlation of these elements was only observed at zone B as the accumulation of uranium only occurs below the zone of bioturbation (Zone A) (Zheng *et al.*, 2002), while zone C is influenced by diagenetic processes. A strong statistical correlation value of ^{234}U ($r^2 = -0.959$, $p<0.01$) and ^{238}U ($r^2 = -0.904$, $p<0.01$) was detected between uranium isotopes with TOM at Kota Kinabalu port (Fig. 2(a)). However the activities of ^{234}U and ^{238}U show no correlation with TOM at Labuan port (Fig. 3(a)). These results suggest that distributions of uranium at Kota Konabalu port are influenced by organic matter. Organic matter may act as a carrier to convert U(VI) to U(IV) in the water column and is subsequently scavenged into the sediment. Several mechanisms have been proposed for the precipitation of uranium by organic matter (Borovec *et al.*, 1979; Dawood, 2010; Min *et al.*, 2005; Porcelli *et al.*, 1997). Uranium is adsorbed very efficiently by organic matter due to its large reactive surfaces. The adsorption of uranium to different organic matter functional groups occur through ion exchange and complexation processes that result in the formation of stable U(IV) (Borovec *et al.*, 1979; Dawood, 2010). Bacteria and fungus in organic matters are also responsible for the reduction and precipitation of uranium (Min *et al.*, 2005).

A strong statistical correlation was observed between ^{234}U/^{238}U and Ca at Kota Kinabalu port ($r^2 = 0.815$, $p<0.05$) and Labuan port ($r^2 = 0.823$, $p<0.05$) as shown in Figs. 2(b) and 3(b), respectively. These results might indicate influences of calcium carbonate in the formation of uranium. Several studies have shown the effect of calcium to uranium geochemistry (Kelly *et al.*, 2007; Zhang *et al.*, 2011). Examples of these include

Table 2. Activity of ^{234}U, ^{238}U, ^{234}U/^{238}U ratio, Ca, Al and total organic matter (TOM) in sediment cores from Kota Kinabalu and Labuan ports

Zone	Layer	Kota Kinabalu					Labuan				
		^{234}U (dpm/g)	^{238}U (dpm/g)	Ca (%)	Al (%)	TOM (%)	^{234}U (dpm/g)	^{238}U (dpm/g)	Ca (%)	Al (%)	TOM (%)
Zone A	0-2	2.19 ±0.32	1.92 ±0.30	4.26 ±0.42	5.77 ±0.50	7.76 ±1.17	2.15 ±0.32	2.21 ±0.21	2.70 ±0.32	5.05 ±0.42	7.36 ±0.17
	2-4	2.35 ±0.35	2.06 ±0.32	4.04 ±0.32	5.75 ±0.32	7.24 ±1.11	2.99 ±0.33	3.08 ±0.32	2.11 ±0.17	4.07 ±0.33	7.97 ±0.09
	4-6	6.45 ±0.66	5.99 ±0.63	3.99 ±0.28	5.66 ±0.62	7.83 ±1.34	2.63 ±0.26	2.54 ±0.25	3.29 ±0.23	5.85 ±0.41	7.95 ±0.09
	8-10	3.32 ±0.51	3.28 ±0.50	3.00 ±0.15	5.79 ±0.84	8.33 ±1.38	2.16 ±0.31	2.29 ±0.30	3.51 ±0.18	5.57 ±0.28	8.45 ±0.09
	12-14	1.85 ±0.30	1.32 ±0.25	4.42 ±0.27	7.94 ±0.72	8.03 ±1.38	2.46 ±0.43	2 ±0.34	3.28 ±0.20	5.60 ±0.34	8.14 ±0.08
	14-16	0.93 ±0.14	0.84 ±0.15	3.49 ±0.28	5.35 ±0.61	8.34 ±1.18	2.77 ±0.53	3.48 ±0.54	3.63 ±0.29	6.26 ±0.50	8.41 ±0.70
	20-22	2.08 ±0.31	1.79 ±0.28	4.97 ±0.45	5.27 ±0.52	6.72 ±1.11	2.32 ±0.37	2.21 ±0.32	3.51 ±0.32	5.63 ±0.51	7.17 ±0.05
Zone B	24-26	0.43 ±0.12	0.51 ±0.13	4.48 ±0.45	4.95 ±0.34	4.17 ±0.90	1.34 ±0.40	2.16 ±0.38	2.55 ±0.25	5.82 ±0.58	7.09 ±0.23
	28-30	1.07 ±0.20	1.12 ±0.20	5.02 ±0.15	4.62 ±0.62	5.03 ±1.20	1.92 ±0.38	2.32 ±0.37	2.73 ±0.08	5.42 ±0.16	6.76 ±0.24
	34-36	2.3 ±0.30	2.31 ±0.30	5.18 ±0.36	4.71 ±0.32	5.72 ±1.43	2.81 ±0.39	3.02 ±0.39	3.63 ±0.25	6.13 ±0.43	6.16 ±0.06
	38-40	3.02 ±0.38	3.16 ±0.39	5.97 ±0.48	3.74 ±0.29	6.07 ±1.54	2.96 ±0.23	3.3 ±0.20	3.82 ±0.31	6.2 ±0.50	5.57 ±0.24
	49-52	2.47 ±0.21	1.94 ±0.18	6.22 ±0.56	3.7 ±0.23	5.90 ±1.16	2.61 ±0.34	2.13 ±0.32	4.34 ±0.39	6.37 ±0.57	5.98 ±0.24
	55-58	2.88 ±0.40	2.57 ±0.37	6.02 ±0.30	3.94 ±0.19	6.02 ±1.53	2.34 ±0.45	2.26 ±0.40	3.95 ±0.20	5.94 ±0.30	5.84 ±0.03
	73-76	0.41 ±0.09	0.38 ±0.09	5.96 ±0.36	4.44 ±0.35	4.00 ±1.28	1.75 ±0.32	2.02 ±0.35	4.04 ±0.24	6.71 ±0.40	4.47 ±0.23
Zone C	82-87	1.43 ±0.21	1.78 ±0.24	6.21 ±0.43	4.35 ±0.32	5.57 ±1.21	2.01 ±0.29	2.72 ±0.30	4.25 ±0.30	6.2 ±0.43	0.95 ±0.06
	97-102	0.85 ±0.14	1.01 ±0.20	5.74 ±0.46	4.78 ±0.26	4.98 ±1.22	1.66 ±0.28	1.52 ±0.24	4.51 ±0.36	5.62 ±0.45	0.98 ±0.70
	122-127	1.72 ±0.22	1.35 ±0.19	5.30 ±0.11	4.94 ±0.28	4.17 ±1.21	2.12 ±0.35	1.79 ±0.29	4.32 ±0.09	5.14 ±0.10	1.07 ±0.60
	142-147	2.35 ±0.22	1.68 ±0.18	5.54 ±0.44	4.80 ±0.39	0.66 ±0.12	3.73 ±0.28	2.78 ±0.27	5.74 ±0.46	5.09 ±0.41	1.05 ±0.50

a study of the effects of calcium carbonate on uranium (VI) sorption onto soil from the Oak Ridge site (0.1% calcium carbonate) and from Altamont Pass, CA, site (10% calcium carbonate) over a range of pH values. The results show that the presence of calcium carbonate in soils strongly affects U(VI) sorption. It has been proposed that the cause of this may be the formation of an aqueous $Ca_2UO_2(CO_3)_3$ complex in solution (Kelly *et al.*, 2007; Zhang *et al.*, 2011; Zheng *et al.*, 2003). Weak negative correlation ($r^2 = -0.477, p>0.05$) was observed between $^{234}U/^{238}U$ and Al at Kota Kinabalu port (Fig. 2(c)) while no correlation was observed at Labuan port. The correlations of aluminium with radionuclides in the marine environment has been used to investigate the influence of aluminosilicate as carrier for radionuclide elements (Roy-Barman *et al.*, 2005). This result indicates aluminosilicate is not major carrier phase of uranium at Kota Kinabalu and Labuan port.

3.3. Sources of uranium

The activity ratio of $^{234}U/^{238}U$ at Kota Kinabalu and Labuan ports and statistical analyses of Ca, Al and TOM were used to predict the sources of uranium.

The study of the disequilibrium ratio between the activity of ^{234}U and ^{238}U isotopes allow investigation of whether authigenic or detrital uranium dominate in the sediments (Dawood 2010; Skwarzec *et al.*, 2002). If the reduction process from U(VI) to U(IV) is dominant, we can expect the same value of $^{234}U/^{238}U$ in sediment and bottom water. This is because uranium is directly scavenged by the reduction process with no major input of uranium from other areas (Skwarzec *et al.*, 2002). Fig. 4(a) and 4(b) shows the profile ratio of $^{234}U/^{238}U$ at Labuan and Kota Kinabalu sediment cores, respectively. Most of the $^{234}U/^{238}U$ ratio at Kota Kinabalu port is greater than 1 (Fig. 4(a)) and slightly similar to the normal ratio of $^{234}U/^{238}U$ in sea water (1.14 to 1.20) (Cochran *et al.*, 1986; Sakaguchi *et al.*, 2006; Scott, 1982; Skwarzec *et al.*, 2002; Srisuksawad *et al.*, 1997). This result might indicate that the major contribution of uranium to sediments is by sorptive uptake of authigenic uranium from the water column. Mobile U(VI) have been reduced to U(IV) due to an anoxic condition at this area. This finding is supported by the high correlation of ^{234}U and ^{238}U with TOM and $^{234}U/^{238}U$ with Ca as discussed in the previous section. In an anoxic environment, pH will decrease and lead to the precipitation of Ca-carbonate (Castanier *et al.*,
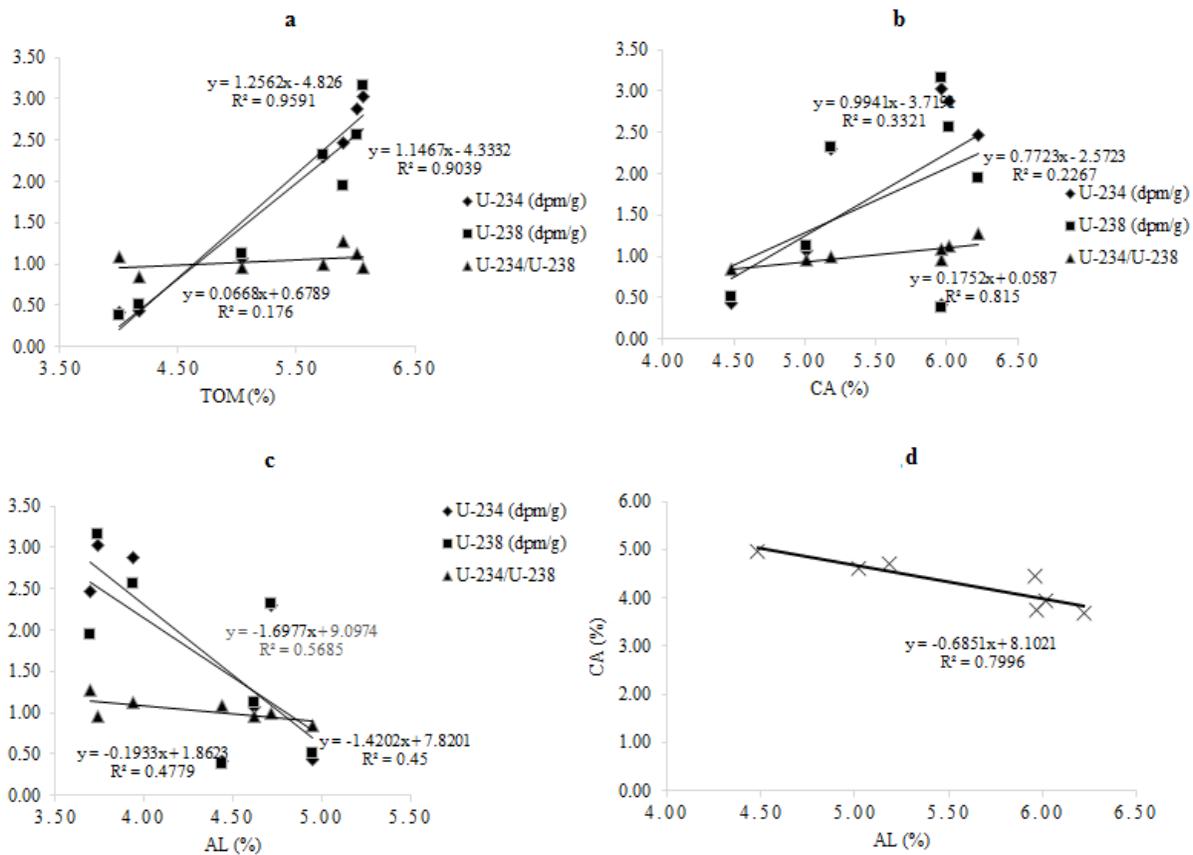

Figure 2. Correlation of a) TOM with ^{234}U, ^{238}U and $^{234}U/^{238}U$, b) Ca with ^{234}U, ^{238}U and $^{234}U/^{238}U$, c) Al with ^{234}U, ^{238}U and $^{234}U/^{238}U$ and d) Al and Ca at zone B off Kota Kinabalu port

1999) and organic matter (Bastviken *et al*., 2004). Organic matter can act as a carrier to convert U(VI) to U(IV) and scavenge directly to the sediments (Rosing and Frei, 2004). These results leads to the suggestion that major uranium contribution at Kota Kinabalu port is authigenic in origin, where mobile U(VI) has been reduced to immobile U(IV) by organic matter and scavenged to the sediment with co-precipitation of dissolved organic matter and calcium carbonate.

Most of the ratios of ^{234}U/^{238}U at Labuan port shows a less than equilibrium ratio (Fig. 4(b)). A low ratio value (<1) at Labuan port might indicate that this area received more ^{238}U input. Uranium in this area is probably of detrital origin because terregenic material is characterized by the ^{234}U/^{238}U activity ratio < 1 (Skwarzec *et al*., 2002). This suggestion is supported by the weak correlation of uranium isotopes with TOM as discussed in the previous section. An earlier study done by Ramli (1997) suggests that the activity of detrital shows a weak correlation with organic input. A high correlation of ^{234}U/^{238}U with Ca also indicates the influences of calcium carbonate in uranium distributions. However, Ca from Labuan port shows different behavior compared to Kota Kinabalu port. Ca at Labuan port shows a strong positive correlation with Al ($r^2 = 0.6099$) (Fig. 3(d)) in contrast to strong

negative correlations ($r^2 = -0.7996$) observed at Kota Kinabalu port. These results might indicate that calcium carbonate at Labuan port is of lithogenic origin because aluminium is known to be a proxy for lithogenus elements (Morelli *et al*., 2012). Calcium carbonate at Kota Kinabalu port is generated by an *in situ* process where Ca form complexes with carbonate due to anoxic conditions (Castanier *et al*., 1999). Labuan is surrounded by sedimentary rock including coal rock and limestone (Wan Hasiah *et al*., 2013). Limestone contains a high level of calcium carbonate and high ^{238}U activity which is assumed by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (1993) to be about 20 Bq/kg. Thus, based on the ratio values, geological setting and statistical values, we estimate that sources of uranium at Labuan port is of detrital origin.

4. Conclusions

The profile activities of ^{234}U and ^{238}U were successfully determined in sediment cores collected from Kota Kinabalu and Labuan ports. Statistical analyses show different behaviors of uranium isotopes at Kota Kinabalu and Labuan ports. The results from the activity ratio (^{234}U/^{238}U) coupled with statistical
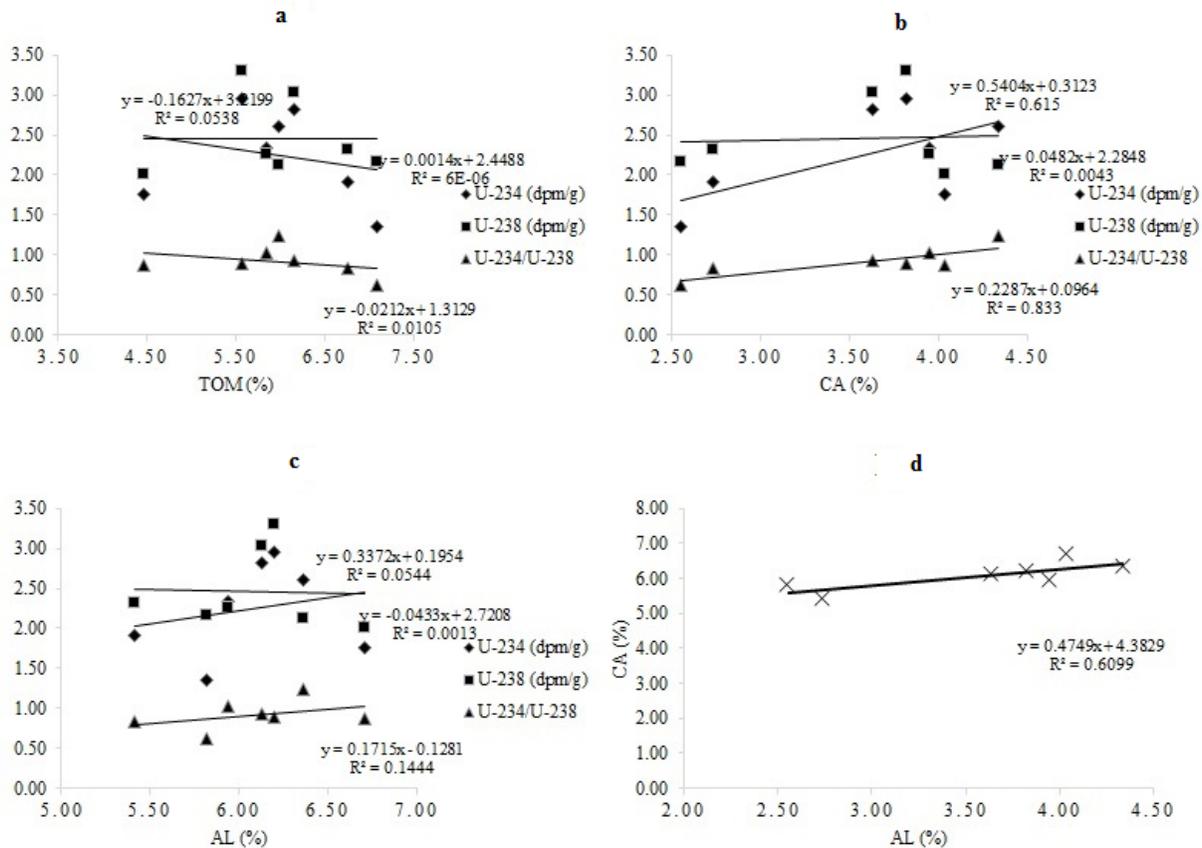

Figure 3. Correlation of a) TOM with ^{234}U, ^{238}U and ^{234}U/^{238}U, b) Ca with ^{234}U, ^{238}U and ^{234}U/^{238}U, c) Al with ^{234}U, ^{238}U and ^{234}U/^{238}U and d) Al and Ca at zone B off Labuan port
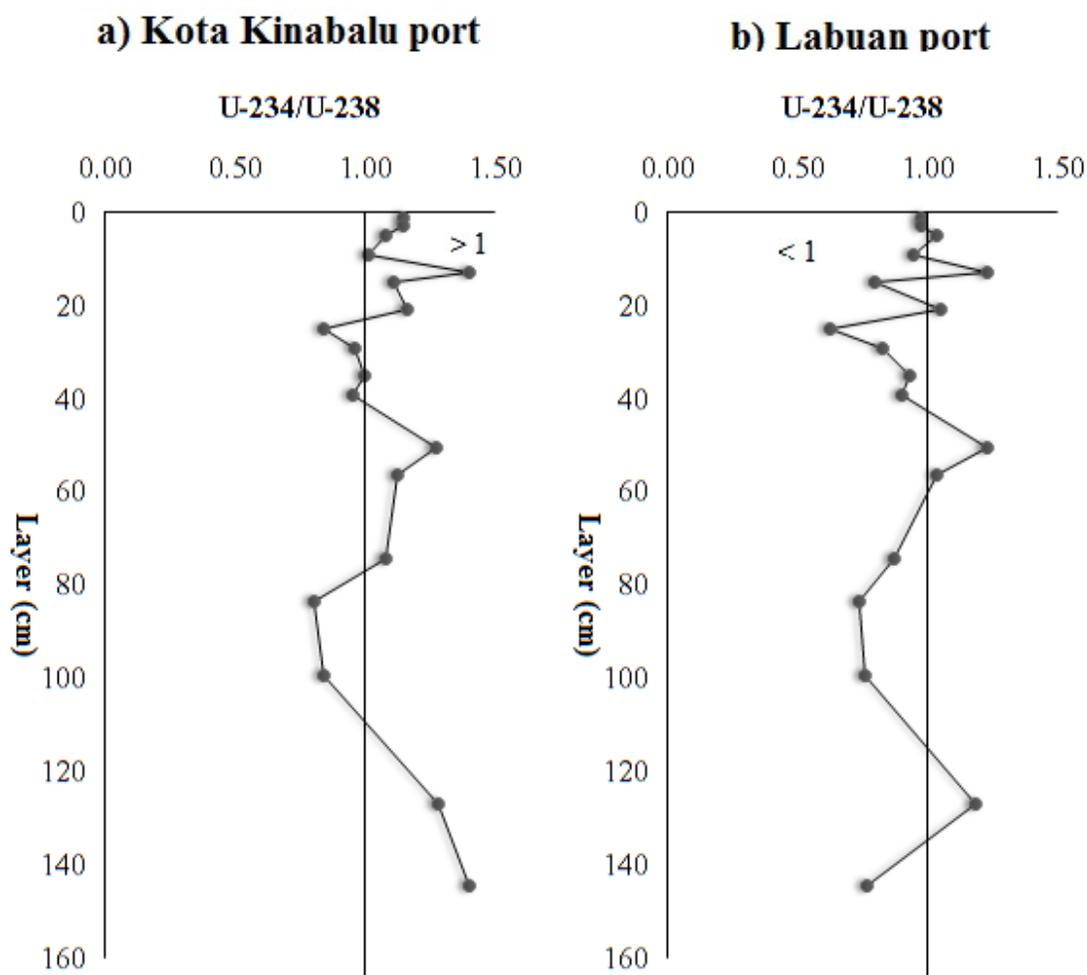
Figure 4. Activity ratio of $^{234}U/^{238}U$ in core sediments from a) Kota Kinabalu port and b) Labuan port. The black line shown is the line of equilibrium.

analyses of TOM, Ca and Al show that uranium at Kota Kinabalu port is largely of authigenic origin. U(VI) was reduced to U(IV) by organic matters and deposited into sediment with calcium carbonate. On the other hand, uranium at Labuan port is largely of detrital origin. The strong correlation between $^{234}U/^{238}U$ with Ca suggests that calcium carbonate might be the major carrier phase for uranium. These findings support the use of isotope ratio $^{234}U/^{238}U$ to determine the origin of uranium and suggests the use of $^{234}U/^{238}U$ as a proxy to investigate the flux of calcium carbonate in the marine environment.

Acknowledgements

References

Akyil S, Yusof AM. The distribution of uranium and thorium in samples taken from different polluted marine environment. Journal of Hazardous Materials 2007; 144(1-2): 564-69.

Andersen MB, Erel Y, Bourdon B. Experimental evidence for ^{234}U-^{238}U fractionation during granite weathering with implications for $^{234}U/^{238}U$ in natural waters. Geochimica et Cosmochimica Acta 2009; 73: 4124-41.

Bastviken D, Persson L, Odham G, Tranvik L. Degradation of dissolved organic matter in oxic and anoxic lake water. Limnology and Oceanography 2004; 49(1): 109-16.

Borovec Z, Kribek B, Tolar V. Sorption of uranyl by humic acids. Chemical Geology 1979; 27(1-2): 39-46.

Castanier S, Le Métayer-Levrel G, Perthuisot JP. Ca-carbonates precipitation and limestone genesis-the microbiogeologist point of view. Sedimentary Geology 1999; 126(1-4): 9-23.

Chung Y, Chang WC. Uranium and thorium isotopes in marine sediments off northeastern Taiwan. Marine Geology 1996; 133(1-2): 89-102.

Cizdziel J, Farmer D, Hodge V, Lindley K, Stetzenbach K. $^{234}U/^{238}U$ isotope ratios in groundwater from Southern Nevada: a comparison of alpha counting and magnetic sector ICP-MS. Science of the Total Environment 2005; 350(1-3): 248-60.

Cochran JK, Carey AE, Sholkovitz ER, Surprenant LD. The geochemistry of uranium and thorium in coastal marine sediments and sediment pore waters. Geochimica et Cosmochimica Acta 1986; 50(5): 663-80.

Dawood YH. Factors controlling uranium and thorium isotopic composition of the streambed sediments of the River Nile, Egypt. JAKU: Earth Science 2010; 21(2): 77-103.

Heiri O, Lotter AF, Lemcke G. Loss on ignition as a method for estimating organic and carbonate content in ediments: reproducibility and comparability of results. Journal of Paleolimnology 2001; 25(1): 101-10.

Henderson GM, Anderson RF. The U-series toolbox for paleoceanography. Reviews in Mineralogy and Geochemistry 2003; 52(1): 493-531.

Kelly SD, Kemner KM, Brooks SC. X-ray absorption spectroscopy identifies calcium-uranyl-carbonate complexes at environmental concentrations. Geochimica et Cosmochimica Acta 2007; 71(4): 821-34.

Lagauzère S, Boyer P, Stora G, Bonzom J. Effects of uranium-contaminated sediments on the bioturbation activity of *Chironomus riparius* larvae (Insecta, Diptera) and *Tubifex tubifex* worms (Annelida, Tubificidae). Chemosphere 2009; 76(3): 324-34.

Min M, Xu H, Chen J, Fayek M. Evidence of uranium biomineralization in sandstone-hosted roll-front uranium deposits, northwestern China. Ore Geology Reviews 2005; 26(3-4): 198-206.

Mohamed CAR, Ramli KNM, Ahmad Z. Distribution of ^{234}U and ^{238}U in Sungai Selangor, Peninsular of Malaysia. Journal of Applied Sciences 2006; 6(3): 562-66.

Morelli G, Gasparon M, Fierro D, Hu WP, Zawadzki A. Historical trends in trace metal and sediment accumulation in intertidal sediments of Moreton Bay, southeast Queensland, Australia. Chemical Geology 2012; 300-301: 152-64.

Porcelli D, Andersson PS, Wasserburg GJ, Ingri J, Baskaran M. The importance of colloids and mires for the transport of uranium isotopes through the Kalix River watershed and Baltic Sea. Geochimica et Cosmochimica Acta 1997; 61(19): 4095-113.

Ramli AT. Environmental terrestrial gamma radiation dose and its relationship with soil type and underlying geological formations in Pontian district, Malaysia. Applied Radiation and Isotopes 1997; 48(3): 407-12.

Rosing MT, Frei R. U-rich Archaean sea-floor sediments from Greenland-indications of > 3700 Ma oxygenic photosynthesis. Earth and Planetary Science Letters 2004; 217(3-4): 237-44.

Roy-Barman M, Jeandel C, Souhaut M, van der Loeff MR, Voege I, Leblond N, Freydier R. The influence of particle composition on thorium scavenging in the NE Atlantic ocean (POMME experiment). Earth and Planetary Science Letters 2005; 240(3-4): 681-93.

Sakaguchi A, Yamamoto M, Sasaki K, Kashiwaya K. Uranium and thorium isotope distribution in an offshore bottom sediment core of the Selenga Delta, Lake Baikal, Siberia. Journal of Paleolimnology 2006; 35(4): 807-18.

Scott MR. The chemistry of U- and Th-series nuclide in the rivers. *In*: Uranium series disequilibrium: applications to environmental problems (*Eds*: Ivanovich M, Harmon RS). Oxford University Press, New York, USA. 1982; 181-201.

Skwarzec B, Boryło A, Strumińska D. ^{234}U and ^{238}U isotopes in water and sediments of the southern Baltic. Journal of Environmental Radioactivity 2002; 61(3): 345-63.

Srisuksawad K, Porntepkasemsan B, Nouchpramool S, Yamkate P, Carpenter R, Peterson ML, Hamilton T. Radionuclide activities, geochemistry, and accumulation rates of sediments in the Gulf of Thailand. Continental Shelf Research 1997; 17(8): 925-65.

Toole J, Baxter MS, Thomson J. The behaviour of uranium isotopes with salinity change in three U.K. estuaries. Estuarine, Coastal and Shelf Science 1987; 25(3): 283-97.

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Sources and effects of ionizing radiation. United Nation sales publication, New York, USA. 1993.

Wan Hasiah A, Lee, CP, Gou P, Shuib MK, Ng TF, Albaghdady AA, Mislan MF, Mustapha KA. Coal-bearing strata of Labuan: Mode of occurrences, organic petrographic characteristics and stratigraphic associations. Journal of Asian Earth Sciences 2013; 76: 334-45.

Wood AKHJ, Ahmad Z, Shazili NAMD, Yaakob R, Carpenter R. Geochemistry of sediments in Johor Strait between Malaysia and Singapore. Continental Shelf Research 1997; 17(10): 1207-28.

Yamada M, Tsunogai S. Postdepositional enrichment of uranium in sediment from the Bering Sea. Marine Geology 1984; 54(3-4): 263-76.

Zhang F, Luo W, Parker JC, Brooks SC, Watson DB, Jardine PM, Gu B. Modeling uranium transport in acidic contaminated groundwater with base addition. Journal of Hazardous Materials 2011; 190(1-3): 863-68.

Zheng Y, Anderson RF, van Geen A, Fleisher MQ. Remobilization of authigenic uranium in marine sediments by bioturbation. Geochimica et Cosmochimica Acta 2002; 66(10): 1759-72.

Zheng Z, Tokunaga TK, Wan J. Influence of calcium carbonate on U(VI) sorption to soils. Environmental Science and Technology 2003; 37(24): 5603-08.

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