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Partitioning of ^{232}Th, ^{230}Th and ^{228}Th in the Surface Water of Northwest Peninsular Malaysia Coastal Waters as Tracer for SPM Sources

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Abstract

Natural thorium radioisotopes (^{232}Th, ^{230}Th, ^{228}Th, ^{234}Th) are well known to be particle reactive, thus they are often used to study the geochemical process of particle reactive matter. In this study, water samples were collected from Langkawi Island and Pangkor Island from six and ten sampling stations respectively, to determine the activity of ^{232}Th, ^{230}Th and ^{228}Th in dissolved (DISS) and suspended particulate matter (SPM) phases, and estimate the sources and origin of SPM into the marine environment. The results in terms of volume activity concentration (mBq/L) shows that the activity level of $^{228}Th_{DISS}$ was higher than $^{228}Th_{SPM}$ at all sampling stations possibly due to the behaviour of the ^{228}Th parent, ^{228}Ra which is easily desorbed from SPM. At several stations far from the coast; stations L4 (4.58km), P7 (3.16 km), P6 (2.1 km), P10 (1.38 km), the activity of $^{232}Th_{DISS}$ was higher compared to $^{232}Th_{SPM}$. These stations behave the same way as the open ocean in which $^{232}Th_{DISS}$ is contributed by atmospheric dust. Indeed, the partition of thorium between dissolved and SPM phase can be used to detect anthropogenic organic pollution as in station L6 in which thorium activity in dissolved phase is higher compared to in SPM phase. Moreover, the use of the $(^{232}Th/^{230}Th)_{SPM}$ ratio is useful to detect lithogenic input to the marine environment in which the ratio ranges from 0.13 to 0.5 and 0.7 to 3.16 for stations that are far and near to the coast respectively, except for station P1. However the data analysis shows that there is no clear pattern of k_d values at both sampling sites, probably due to the adsorption and desorption processes of ^{232}Th and ^{230}Th that occurs in the water column. Nevertheless, as the SPM concentration (mg/L) increases, the k_d value decreases except for the activity of ^{228}Th which shows a strong negative correlation with SPM.

Keywords: coastal water; thorium; distribution coefficient

1. Introduction

Radiogenic thorium consists of ^{234}Th, ^{230}Th, and ^{228}Th, while non-radiogenic thorium is ^{232}Th. The study of U-Th series radionuclides content in water, sediment, and suspended particulate matter (SPM) enables ones to identify the transport mechanism of nuclides from land to sea (Balakrishna *et al*., 2001). In addition, U-Th series radionuclides expecially ^{210}Po and ^{210}Pb were also studied in marine organism such as cockles (Zakri and Mohamed 2016) and seabass (Zakaria *et al*. 2013(a); 2013(b)) to determine the impact of radiation to humans health.

It is well known that Th isotopes (^{232}Th, ^{230}Th, ^{228}Th, ^{234}Th) is highly particle reactive and have a wide range of half-lives, thus it is been used as an important tracer in marine environmental studies mainly to determine the geochemical process of particle reactive matter (Bacon and Anderson, 1982; Zhang *et al*., 2004; Santschi *et al.*, 2006). There are also studies that have taken into account dissolved thorium both in the open ocean where there is less or no riverine input affecting the area (Hsieh *et al*., 2011; Hayes *et al*., 2013; Singh *et al*., 2013) and brackish water (Andersson *et al*., 1995). On the other hand, there were also several studies that did not separate ^{232}Th, ^{230}Th, and ^{228}Th in dissolved and particulate phase (Okubo *et al*., 2007).

The interphase between SPM and DISS is the colloid phase which can be both retained in the filter paper or passed through it. According to Trimble *et al*., (2004), two factors control the activity of thorium in the marine environment. These are its half-life and its scavenging rate onto SPM. On the other hand, the factors that determine the distribution of thorium in the marine environment are (1) the input of ^{228}Th, ^{230}Th and ^{232}Th from river; (2) lateral transport of ^{230}Th and ^{228}Th from the continental slope; (3) removal of dissolved thorium (^{232}Th, ^{230}Th, ^{228}Th) through the scavenging process and (4) resuspension of sediment of ^{228}Th, ^{230}Th and ^{232}Th (Zhang *et al*., 2004).

In order to utilize data on the activity of radionuclides, the disequilibrium between two radionuclides needs to be calculated. The (^{230}Th/

$^{232}Th)_{SPM}$ and $(^{228}Th/^{232}Th)_{SPM}$ activity ratios can be used to determine the input of particulate matter into the water column (Zhang *et al*., 2004). Meanwhile the particle-solution partition coefficient (k_d) is calculated to determine the equilibrium partitioning of a species between the dissolved and particulate phases (Chase *et al*., 2002).

The sampling were conducted at Langkawi Island and Pangkor Island which has different rock formation and may receive input of SPM from weathering of rock, Andaman sea or even from atmosphere. Thus, this study was conducted to determined the activity of thorium radioisotopes (^{232}Th, ^{230}Th and ^{228}Th) collected from surface seawater at Pulau Langkawi, Kedah and Pulau Pangkor, Perak both in the DISS and SPM phases and to estimate origin of SPM in the marine environment.

2. Materials and Methods

2.1 Sampling location

As stated above, sampling was carried out in the coastal waters of northwest Peninsular Malaysia; at Langkawi Island, Kedah and Pangkor Island, Perak. In terms of geology, the rock formations of Machincang, Setul, Singa, and Chuping are responsible for the existence of Langkawi Island, which has been declared a Geopark by UNESCO. The different rock formations are related to different types of rock such as granite, limestone, alluvium, etc. In this study, samples are taken from locations that have different rock formations as shown in Table 1 (Lee, 2002). Pangkor Island, on the other hand dominantly consists of alkali fieldspar granite that have become phenocrysts (Umor *et al*., 2005). Indeed, the process of rock weathering may contribute to the presence of natural uranium and thorium in the environment (Latham and Schwarcz, 1987; Andersen *et al*., 2009).

In term of location, Langkawi Island faces the Andaman Sea in the west and the Straits of Malacca in the east. While Pangkor Island is also an island in the Straits of Malacca with water depth less than 200 m. Sampling locations were plotted and presented as in Fig. 1.

2.2 Sampling

Sampling in Langkawi Island was conducted during the early southwest monsoon, while sampling at Pangkor Island was conducted at end of the southwest monsoon. Surface water samples (10 L) were collected using a bucket and stored in 12 L bottles. The in-situ parameters were measured using a calibrated portable multi-parameter (YSI Professional Plus), with a salinity range of 30.05 to 32.67 psu. The sampling dates, coordinates, surface water salinity, dissolved oxygen, pH and conductivity are presented in Table 2.

2.3 Sample analysis

Water samples (10 L) were filtered through pre-weighed Advantec Glass fiber GF-75 (0.3 μm) and acidified with 10 mL of nitric acid (HNO_3) for further analysis. The filter paper used in this study to separate dissolved and suspended particulate matter has smaller pore size when compared with previous study (eg. Lin *et al*., 2013; Pan *et al*., 2015) which used ~ 0.4 μm. The smaller pore size was used in order to maximize the collection of SPM. The filter papers containing SPM were then dried in an oven until constant weight was obtained for further analysis.

The analytical procedures for chemical separation of thorium isotopes were modified from Andersson *et al*. (1995) as shown in Fig. 2. Briefly, about 0.05 ml of ^{229}Th (36.23 dpm/ml) and 1 mL of iron chloride, $FeCl_3$ (19.59 mg/mL) were added to each sample which acts as tracer and carrier respectively.

Table 1. The nearby rock formation and type of rock/sediment at different sampling stations at Langkawi Island, Kedah

Station	Nearby rock formation	Nearby Type of rock/sediment
L1	Setul formation	Karsted limestone, vesuvianite scarn
L2	Setul formation	Karsted limestone
L3	Machincang formation (New)	Granite
L4	Machincang formation (New)	Granite, Alluvium
L5	Chuping formation	Crystalline limestone
	Singa formation	Siltstone, mudstone
		Granite, Alluvium
L6	Setul formation	Limestone, Granite

Sources: Lee (2002)

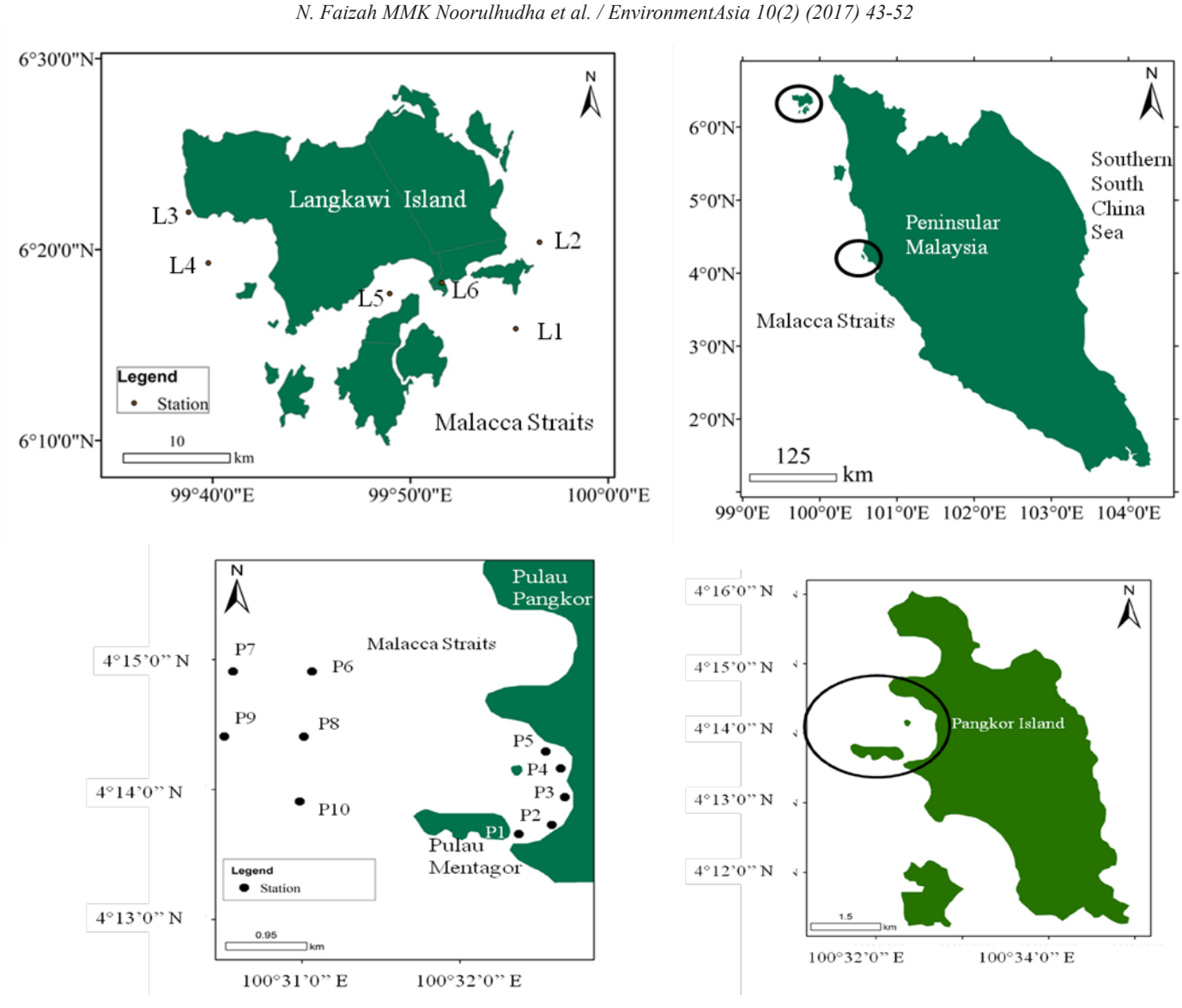

Figure 1. Sampling stations at Langkawi Island and Pangkor Island

Table 2. Sampling dates, coordinates, surface water salinity, dissolved oxygen, pH and conductivity

Location	Sampling date	Station	Coordinate		Salinity (psu)	Dissolved oxygen (mg/L)	pH	Conductivity (µs/cm)
			Latitude °N	Longitude °E				
Langkawi Island, Kedah	12-May-14	L1	06°15.849'	99°55.329'	32.67	9.47	7.70	56418
		L2	06°20.383'	99°56.530'	32.61	12.38	7.51	56352
		L3	06°21.953'	99°38.790'	32.31	12.50	7.87	55428
		L4	06°19.291'	99°39.788'	32.51	9.61	7.58	55995
		L5	06°17.688'	99°48.955'	32.42	14.55	7.17	56541
		L6	06°18.256'	99°51.597'	32.63	11.37	7.33	50146
Pangkor Island, Perak	11-Oct-14	P1	04°13.655'	100°32.373'	32.14	5.20	8.03	49375
		P2	04°13.727'	100°32.583'	32.28	5.31	8.08	49571
		P3	04°13.940'	100°32.665'	32.17	5.65	8.09	49417
		P4	04°14.161'	100°32.639'	32.25	5.49	8.09	49559
		P5	04°14.292'	100°32.544'	32.23	5.59	8.10	49524
		P6	04°14.907'	100°31.066'	32.12	5.14	8.11	49358
		P7	04°14.907'	100°30.566'	32.01	5.71	8.15	49219
		P8	04°14.405'	100°31.015'	32.08	5.24	8.13	49299
		P9	04°14.905'	100°30.511'	31.98	5.65	8.12	49021
		P10	04°13.907'	100°30.989'	30.05	5.13	8.11	49264

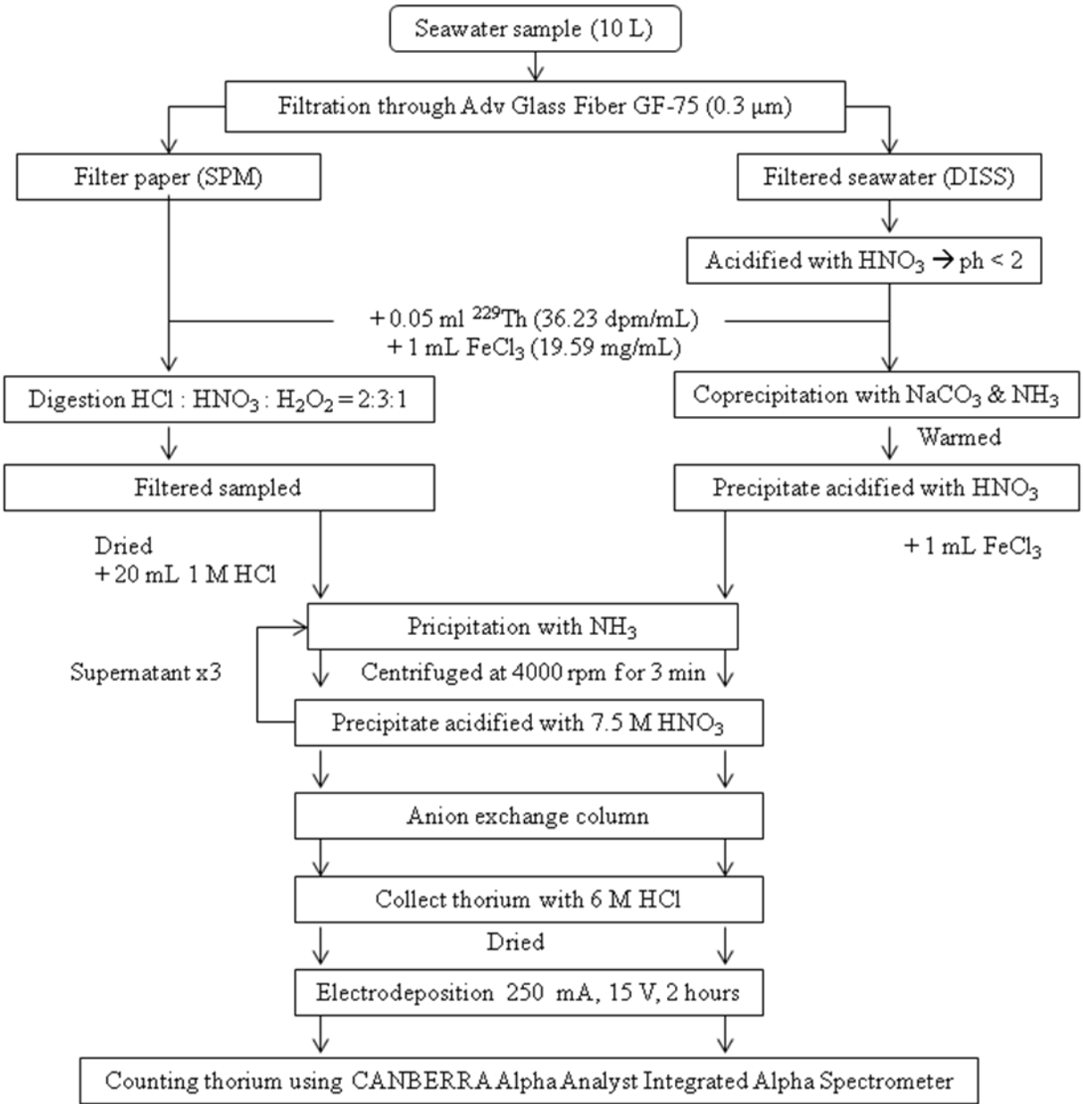

Figure 2. The summarized flow chart for the procedure done in this study

Filter papers were then digested using concentrated hydrochloric acid (HCl), nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) in the ratio of 2:3:1. The digested samples were filter to remove unwanted residue. The filtered solution was dried and about 20 mL of 1 M HCl was added to the solution.

Meanwhile, the filtered seawater samples were coprecipitated using sodium carbonate and ammonia and left to stand for one night for the formation of white precipitate. Supernatant were removed using the siphon method, while the precipitate was collected and warmed using a hotplate to remove carbon dioxide in the sample (Lally, 1982). The precipitate was than acidified with concentrated HNO_3 until clear. Another 1 mL of $FeCl_3$ was added to each sample. The samples were then left overnight for the complete reaction between radionuclides and Fe.

Both SPM and DISS samples were purified with an anion exchange resin (AG 1-X8 Bio-RAD) as decsribed by previous researchers (Mohamed *et al*., 1996; Mohamed *et al*., 2001). Thorium was eluted from the column by using 50 mL of 6 M HCl and electro-deposited onto thin silver disks at 250 mA, 15 V for 2 hours. It was then ready for the counting of thorium activity using CANBERRA Alpha Analyst Integrated Alpha Spectrometer model S570 that was equipped with Genie-2000.

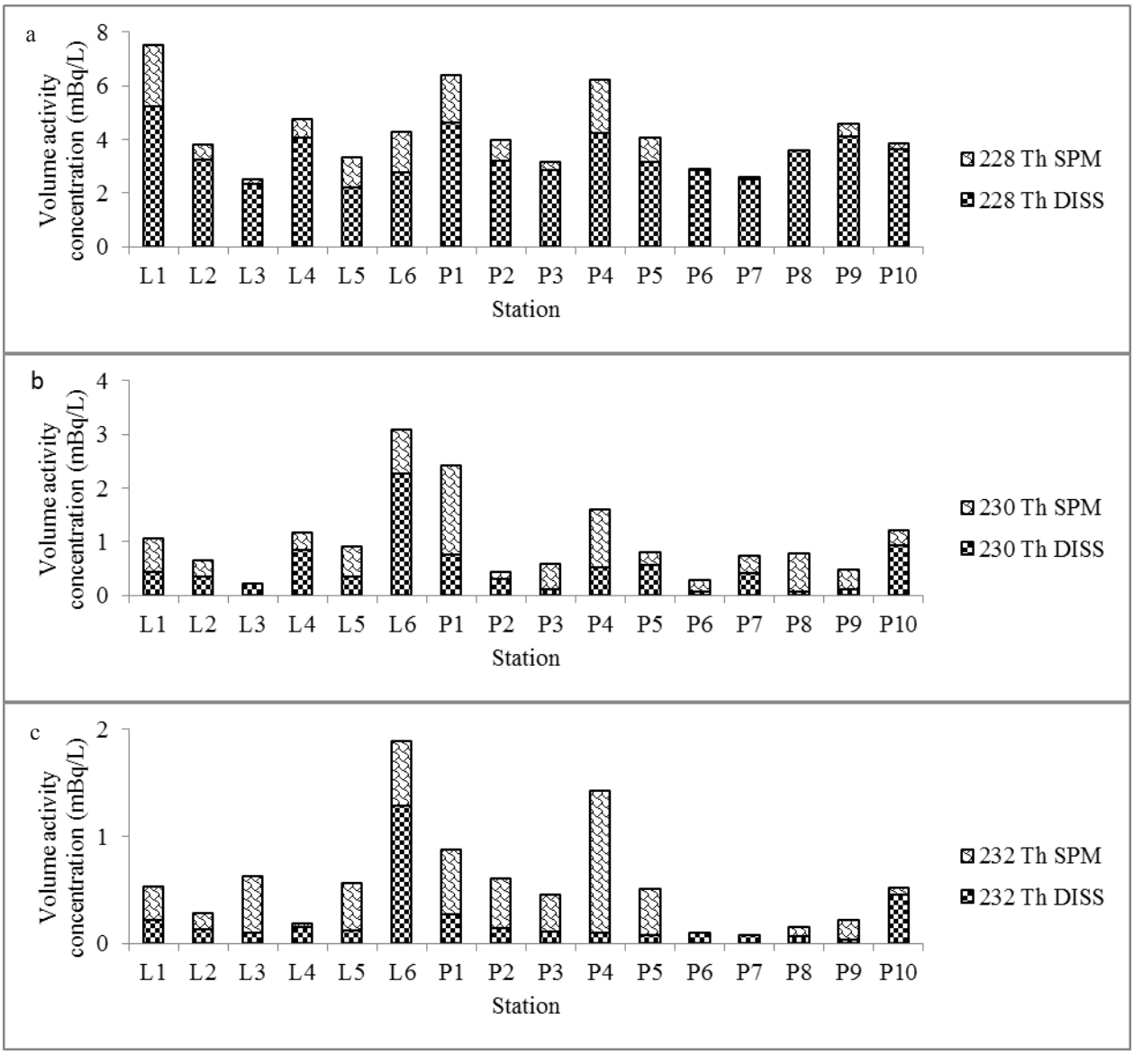

Figure 3. Volume activity concentrations of ^{228}Th (a), ^{230}Th (b) and ^{232}Th (c) both in DISS and SPM phases at Langkawi Island and Pangkor Island stations.

3. Results and Discussion

3.1 Volume activity concentration of ^{228}Th, ^{230}Th and ^{232}Th

Volume activity concentration (mBq/L) of ^{228}Th, ^{230}Th and ^{232}Th in dissolved and suspended particle matters phases fluctuated slightly between stations (Fig. 3).

In general, the activity of $^{228}Th_{DISS}$ is higher compared to $^{228}Th_{SPM}$, $^{232}Th_{DISS}$, $^{232}Th_{SPM}$, $^{230}Th_{DISS}$, and $^{230}Th_{SPM}$ at all stations which is possibly due to high concentrations of its parent, ^{228}Ra and the high turnover of ^{228}Th due to its short half-lives (1.913 years) (Zhang *et al*., 2004). The high concentration of ^{228}Ra was possibly from its desorption behaviour which may be related to salinity, sediment concentration and/or tidal effects (Webster *et al.*, 1995). Another explanation for the high ^{228}Th activity is from the weathering of rock (Latham and Schwarcz, 1987). Latham and Schwarcz (1987) further suggest that ^{228}Th and ^{228}Ra are rapidly removed from rock and thus have high mobility. It is well known that weathering is high in tropical countries. Given that Malaysia is a tropical country, it is thus likely that the high ^{228}Th (^{228}Ra) activity is a result of the weathering of rock.

Table 3. The approximate distance of the sampling stations with nearby coasts and their SPM concentrations

Station	Estimate distance from coastal (km)	SPM concentration (mg/L)
L1	3.33	4.57
L2	3.27	2.38
L3	0.46	8.92
L4	4.58	9.79
L5	2.1	5.26
L6	< 0.1	6.61
P1	0.1	11.63
P2	0.1	13.88
P3	0.12	16.17
P4	0.11	13.92
P5	0.33	14.89
P6	2.1	10.23
P7	3.16	7.66
P8	3.14	8.19
P9	4.14	8.51
P10	1.38	8.45

For ^{232}Th, stations L4, L6, P7, P6 and P10 show a high volume of activity concentration in dissolved phase compared to in SPM phase. Previous study suggested that, at open ocean, the level of $^{232}Th_{DISS}$ at the ocean surface is used to monitor the input of atmopheric dust (Andersson *et al*., 1995; Hsieh *et al*., 2012). Hayes *et al*. (2013) has hypothesized that $^{232}Th_{DISS}$ from dust is present as colloids in seawater. These stations (L4, P7, P6, and P10) are far from the coast (Table 3), thus it is possible that a high volume of activity concentration of $^{232}Th_{DISS}$ comes from atmospheric input especially during the southwest monsoon.

In contradiction to the above however, station L6, which is near the coast and recorded high SPM concentrations (6.61 mg/L), showed low volume activity concentrations of $^{232}Th_{SPM}$ (0.60 ± 0.17 mBq/L) compared to $^{232}Th_{DISS}$ (1.29 ± 0.17 mBq/L). It is known that an area with organic compound pollution such as oil tends to show low conductivity since oil does not conduct electricity. Station L6 is near the ferry terminal, therefore it is possible that this station receives organic compound pollution since conductivity is low (50146 µs/cm) compared to other stations. Thus it is possible that the addition of anthropogenic organic matter from the nearby ferry terminal can increase the solubility of radionuclides and/or lessen the possibility of radionuclides being absorbed into the mineral surface (Evans and Heath, 2004).

In the case of ^{230}Th at Langkawi Island, the high total activity recorded at stations L1, L2 and L6 is possibly the result of nearby limestone weathering as suggested by Von Gunten *et al*. (1996) in which the concentrations of ^{230}Th and ^{226}Ra are high at the limestone Karst region.This will be further discuss in section 3.2. As for stations L2 and L6 together with station L3, L4, P2, P5, P7 and P10 show high activities of $^{230}Th_{DISS}$ compared to $^{230}Th_{SPM}$. According to Osmond and Ivanovich (1992), the presence of ^{230}Th in solution may come from the desorption of particles and colloids, α-recoil or mineral dissolution and decay of its parent, ^{234}U.

3.2 Activity ratio of (^{232}Th/^{230}Th and ^{228}Th/^{232}Th)

The activity ratio of thorium is important to study the behaviour of its isotopes in seawater and the input of SPM into the water column (Zhang *et al*., 2004).

At the Langkawi Island sampling stations, the activity ratio of $(^{232}Th/^{230}Th)_{SPM}$ and $(^{232}Th/^{230}Th)_{DISS}$ generally ranges from 0.12 to 0.81 and 0.18 to 0.56, respectively (Table 4). A higher range of $(^{232}Th/^{230}Th)_{DISS}$ and $(^{232}Th/^{230}Th)_{SPM}$ was observed at Pangkor Island with a minimum ratio of 0.13 up to 1.39 and 3.16, respectively. Meanwhile for $(^{228}Th/^{232}Th)_{SPM}$ and $(^{228}Th/^{232}Th)_{DISS}$, generally higher ratios were obtained in the dissolved phase except for stations L6 and P10.

Table 4. The activity ratio for thorium for both dissolved and SPM phases

Location	Station	SPM		DISS	
		$^{232}Th/^{230}Th$	$^{228}Th/^{232}Th$	$^{232}Th/^{230}Th$	$^{228}Th/^{232}Th$
Pulau Langkawi, Kedah	L1	0.50	7.36	0.50	23.77
	L2	0.48	3.68	0.38	23.93
	L3	ND	0.31	0.45	24.30
	L4	0.12	17.19	0.18	26.98
	L5	0.81	2.51	0.34	18.42
	L6	0.75	2.46	0.56	2.17
Pulau Pangkor, Perak	P1	0.36	2.94	0.37	16.63
	P2	3.16	1.63	0.47	22.69
	P3	0.70	0.92	1.07	24.28
	P4	1.22	1.53	0.20	41.28
	P5	1.82	2.01	0.13	42.20
	P6	ND	ND	1.39	27.02
	P7	ND	ND	0.18	32.94
	P8	0.13	ND	0.84	55.31
	P9	0.50	2.64	0.32	113.89
	P10	0.23	3.94	0.49	7.94

*ND Not detected

In explaining the presence of thorium radioisotopes (^{232}Th, ^{230}Th, and ^{228}Th) in the water column at Langkawi Island, we try to describe it in terms of the distance from the coast and the input from rock weathering since the sampling location is situated in the GEOPARK.

In this study, the use of $^{232}Th_{SPM}$ as a proxy to detect input from the coast is clear in Langkawi Island. In terms of distance, the stations that are far from the coast (stations L1, L2, and L4) show low activity ratios of $(^{232}Th/^{230}Th)_{SPM}$, while stations that are near the coast (stations L6 and L5) show high activity ratios. This pattern is opposite to ratios of $(^{228}Th/^{232}Th)_{SPM}$ and $(^{228}Th/^{232}Th)_{DISS}$ in which the station that is far from the coast shows higher ratios compared to stations which are near the coast.

In terms of input from rock weathering, stations L5 and L6 are close to granite sources. Thus it is possible that the high ratio of $(^{232}Th/^{230}Th)_{SPM}$ at stations L5 and L6 was caused by the weathering of granite which further enriches ^{232}Th at these stations. On the other hand, stations L1 and L2 which are near limestone sources show low activity ratios of $(^{232}Th/^{230}Th)$ in DISS and SPM phases, thus supporting the hypothesis that these stations receive ^{230}Th input from limestone weathering.

In addition, it is interesting to notice that the ratio for $(^{232}Th/^{230}Th)_{DISS}$ at L6 is the highest. This further supports the possibility that there is possible anthropogenic organic input from the nearby jetty as discussed previously.

As was the case in Langkawi Island, stations far from the coast of Pangkor Island (P8, P9 and P10) show low activity ratios of $(^{232}Th/^{230}Th)_{SPM}$ and stations that are near coastal areas (P2, P3,P4 and P5) show high activity ratios of $(^{232}Th/^{230}Th)_{SPM}$ (Table 4). Nevertheless, station P1 which is located near the coastal have considerably low activity ratios of $(^{232}Th/^{230}Th)_{SPM}$ compared to other nearer coastal stations. It should be noted that station P1 recorded considerably high activities of $^{232}Th_{SPM}$ (0.59 ± 0.21 mBq/L) and the highest activity of $^{230}Th_{SPM}$ (1.67 ± 0.36 mBq/L). According to Andersen *et al*. (2009), ^{234}U is easily removed from granite that has actively weathered. Thus it is possible that the high activity of ^{230}Th is due to the enrichment of its parent radionuclide ^{234}U from the weathering of nearby granite since station P1 is located in between Pulau Pangkor and Pulau Mentagor which can contribute more of weathered materials.

Table 5. Partition coefficient values for ^{228}Th, ^{230} and ^{232}Th at Pulau Langkawi Island and Pulau Pangkor Island

Location	Station	Suspended particulate matters (SPM) (mg/L)	Partition coefficient [k_d x 10^5 L/g]		
			^{228}Th	^{230} Th	^{232}Th
Pulau Langkawi, Kedah	L1	4.57	211.99	684.60	684.60
	L2	2.38	284.83	1470.37	1851.58
	L3	8.92	8.67	ND	685.90
	L4	9.79	17.70	39.38	27.77
	L5	5.26	185.83	574.40	1361.34
	L6	6.61	121.72	80.58	107.71
Pulau Pangkor, Perak	P1	11.63	27.89	163.83	157.74
	P2	13.88	12.16	25.26	168.84
	P3	16.17	4.23	168.47	111.16
	P4	13.92	24.71	107.78	665.68
	P5	14.89	12.55	19.31	263.60
	P6	10.23	1.79	251.81	ND
	P7	7.66	5.07	132.10	ND
	P8	8.19	ND	1370.45	207.62
	P9	8.51	16.02	435.61	689.88
	P10	8.45	9.46	40.14	19.08

*ND Not detected

As mentioned above in 3.1, stations that are far from the coast (P6, P8 and P10) may receive inputs of ^{232}Th from the atmosphere since sampling was conducted during the southwest monsoon. This hypothesis is strengthened by the high ratio of $(^{232}Th/^{230}Th)_{DISS}$

3.3 Partition coefficient of thorium (^{232}Th, ^{230}Th, ^{228}Th)

Partition coefficient (k_d) explains the equilibrium partitioning of an element between the particulate and dissolved phases (Chase *et al*., 2002). In this study, both locations generally show a wide range of k_d for all the three thorium isotopes (Table 6). Even though the study by Periáñez and Martínez-Aguirre (1997) was conducted in an estuary, the result is almost parallel, in which there were large variations of ^{232}Th and ^{230}Th k_d between sampling points. Periáñez and Martínez-Aguirre (1997) further suggest that this is due to (i) unsteady input from the sources, (ii) adsorption and desorption of radionuclides to bottom sediment and SPM and (iii) tidal effect.

In addition, ^{228}Th showed lower k_d values compared to ^{230}Th and ^{232}Th except for L6. Statistical analysis shows that there is a significant difference between k_d values for ^{228}Th with ^{232}Th but not with ^{230}Th. Moreover, k_d values for ^{232}Th are not significantly different from ^{230}Th.

Based on the earlier discussion of the distribution of thorium activities and its ratios, we grouped L1, L2, L4, P6, P7, P8, P9 and P10 as sampling stations far from the coast, while station L3, L5, L6, P1, P2, P3, P4 and P5 are stations near the coast. By differentiating according to distance, it is hoped that the behaviour of thorium in northwest of Peninsular Malaysia can be revealed.

Guo *et al*. (1995) mentioned that if the distribution between dissolved and SPM is only affected by equilibrium surface adsorption, the k_d value should be independent of particle concentration. In this study, as SPM concentrations increased, the k_d value decreased for both groups but only ^{228}Th showed strong negative correlations with SPM (Table 6). According to Guo *et al*. (1995) the negative correlation is due to

Table 6. The equation for log SPM vs k_d values for thorium isotopes

Location	Log SPM vs	Equation	r^2
Near to coastal	^{232}Th	y = -0.986x + 0.518	0.181
	^{230}Th	y = -1.516x - 0.997	0.324
	^{228}Th	y = -2.631x - 3.820	0.701
Far from coastal	^{232}Th	y = -2.511x - 3.196	0.539
	^{230}Th	y = -1.743x -1.334	0.357
	^{228}Th	y = -3.076x - 5.380	0.752

the particle-concentration effect in which the dissolved fraction contains colloidal thorium isotopes. It should be noted that the relationship between SPM and ^{232}Th (far from the coast) even though not significant, is higher compared to ^{232}Th ratios (near the coast).

4. Conclusions

In general, volume activity concentrations of $^{228}Th_{DISS}$ are higher compared to $^{228}Th_{SPM}$ at all stations. For stations L4, P7, P6 and P10 which are far from the coast, $^{232}Th_{DISS}$ is higher compared to $^{232}Th_{SPM}$ which may come from atmospheric input. It is interesting to note that the unusual partitioning of thorium between the dissolved and SPM phases can be used to determine anthropogenic organic pollution, as demonstrated by samples taken at station L6, provided that physical parameters (conductivity) is known. In addition, the use of activity ratios, especially $(^{232}Th/^{230}Th)_{SPM}$ was used to determine lithogenic input. Higher ratios of $(^{232}Th/^{230}Th)_{SPM}$ were observed near the coastal. Moreover the k_d value at Langkawi Island and Pangkor Island did not show any clear pattern for the ^{232}Th, ^{230}Th and ^{228}Th. This is most probably due to the adsorption and desorption of radionuclides in bottom sediment and SPM, as well as the tidal effect. Indeed, the correlation between k_d and SPM shows that there is a possible presence of colloidal thorium that passes through the filter paper especially for ^{228}Th and ^{232}Th (far from the coast).

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