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Distribution of ^{234}U and ^{238}U in Sungai Selangor, Peninsular of Malaysia

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Abstract: In the present study the activities of uranium were measured in water column, suspended particulate matter (SPM) and surface sediment along the Sungai Selangor, which is from river, estuary and near-shore. During high tide, the activities of uranium were found highly in the particulate phase (^{234}U = 350.93±11.69 mBq g^{-1} and ^{238}U = 243.55±9.33 mBq g^{-1}), where the activity of uranium in particulate phases is slightly correlated with the amount of suspended particulate matters. A strong adsorption of uranium occurred onto the suspended particles matter was showed by the distribution coefficient (K_d) values as 2366×10^{-3} L g^{-1} and 1363×10^{-3} L g^{-1} for ^{234}U and ^{238}U, respectively.

Key words: Scavenging, $^{234}U/^{238}U$, suspended particulate matter, distribution coefficient (K_d)

INTRODUCTION

The characteristic and distribution of uranium nuclides in river system is usually influent by the contents of suspended particles either from organic or detrital minerals, where uranium usually efficiently adsorbed onto Fe-oxyhydroxides, clays and other secondary mineral[1,2]. Particulate uranium will release during estuarine mixing process and delivery to the ocean by settling particles[3]. The mechanisms uptake and buried processes of uranium into estuarine particles are not well documented by previous researchers.

Study on uranium isotopes such as ^{234}U and ^{238}U in tropical river systems especially Sungai Selangor of Peninsular of Malaysia is important and relevant to describe the weathering process and transport of chemical constituents occurred in the Southeast Asian river system. In general, Kuala Selangor is a fishing village and famous with fire flies watching along the river side. Sungai Selangor is also one of the major river systems in Selangor and drains into Malacca Straits, and has high fluxes of suspended particulate along the river. The river is 75 km long with the total catchments area 1450 km^2 and 500 m wide at the mouth. Indeed, very little data are available for uranium in water column of Malaysian rivers. Thus, this study has been designed to investigate the effect of suspended particulate matter (SPM) on the scavenging of ^{234}U and ^{238}U occurred along the Sungai Selangor.

MATERIALS AND METHODS

Water and surface sediments samples were collected from eight stations along the Sungai Selangor during high and low tides on September 2002 (Fig. 1 and Table 1) using water sampler and ponar grab, respectively and sediment samples were stored into the plastic bags for further analyses.

Briefly, about 15 L of water sample was filtered as soon as possible through the Millipore filter paper (0.45 μm pore size) for separate aqueous and suspended matters. The aqueous acidify with concentrated HNO_3 (pH<2) and continue adding with 1 mL of $FeCl_3$ (25 mg mL^{-1}) as a carrier and 1 mL of ^{236}U (0.343 Bq mL^{-1}) as chemical yield. Sample was stir for a few minutes and stand for 4 h. After that increase the pH to pH 10 using NH_4OH and Na_2CO_3, then continue stirring for a few minutes and stand for overnight. In the next day,

Table 1: Sampling stations along the Sungai Selangor used for this study

Station	Longitude	Latitude
1	03° 23' 17" N	101° 17' 47" E
2	03° 22' 54" N	101° 16' 47" E
3	03° 21' 41" N	101° 16' 19" E
4	03° 21' 06" N	101° 15' 24" E
5	03° 20' 07" N	101° 15' 17" E
6	03° 21' 16" N	101° 14' 38" E
7	03° 21' 16" N	101° 14' 08" E
8	03° 19' 43" N	101° 13' 32" E

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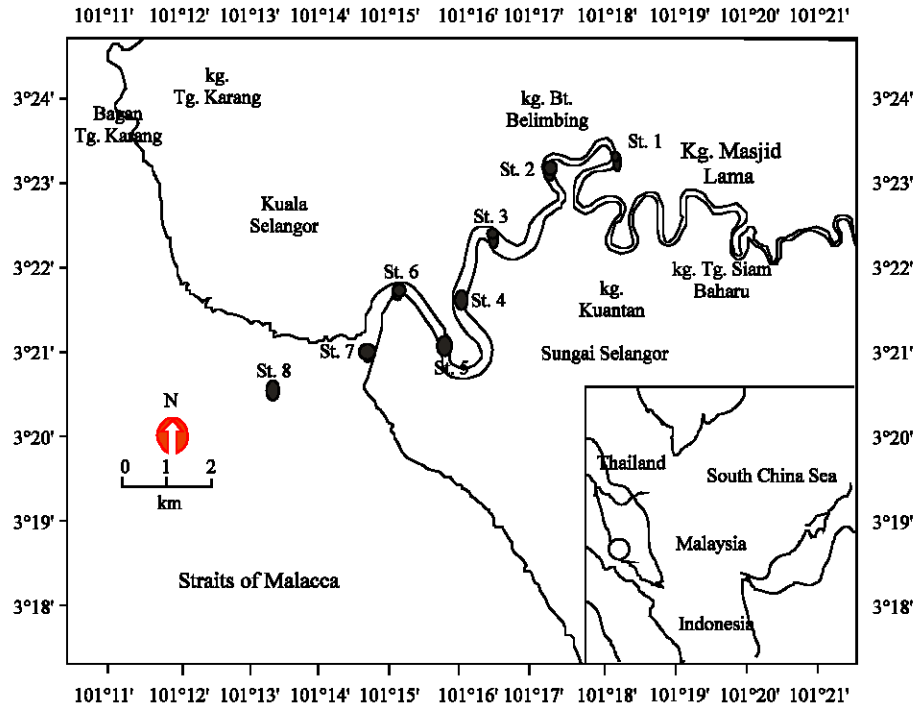

Fig. 1: Sampling stations along Sungai Selangor

siphon off the supernatant and dissolved the precipitate with concentrated HNO_3 and heating on the hot plate for remove carbon dioxide. Dissolved the precipitate with 50 mL of 7 M HCl and purified uranium using an anion exchange column (Dowex AG 1×8, 200 mesh, Cl^-). The purified liquid containing uranium isotopes is plated onto the stainless steel disc at pH 2 using diluted H_2SO_4[3,4] and counting their activities isotope using Alpha Spectrometry[5].

Dried suspended solid samples were digested with diluted HCl as a leaching method digestion procedure[5]. Briefly, the suspended solid sample was weighing, and digest with 50 mL of 8 M HCl and 1 mL of ^{236}U (0.343 Bq mL^{-1}) into glass beaker on the hotplate for 3 h. After cool to the room temperature, the sample was filter using membrane filter of 0.45 μm diameter to collect their aqueous for continue uranium nuclides purification.

Standard reference material of Baltic Sea Sediment (SRM IAEA-300) also analyzed as an analytical quality procedure control and the result obtained in the range of certified values (95% confidence).

RESULTS AND DISCUSSION

Three systems of river environments along the Sungai Selangor were classified base on their Specific conductivity (Spc) values. There are river, estuary and near-shore areas occupied by Station 1 and 2, Station 3 and 4, and Station 5 to 8, respectively (Table 2). The abnormal distribution of Spc value was also found from Station 1 to 4 probably caused by the mixing of seawater with freshwater during the low tide processes (Table 2).

Dissolved ^{234}U and ^{238}U in water column: In general, dissolved concentration of ^{234}U and ^{238}U found in Sungai Selangor slightly high during high tide compared low tide (Table 2). The activities of both uranium isotopes varied widely along the Sungai Selangor, its maybe due to the chemical weathering processes occurred on the riverside. According to the in-situ parameters (i.e., Spc values) which is obtained during sampling (Table 2), it is believed that dissolved uranium enter into the Sungai Selangor occur during the high tide is associated with Fe-organic colloids (Fig. 2). But during the low process as low salinity, they were desorb from the Fe-organic colloids or other colloidal carrier phases into the water column (Fig. 2b and d) as well as described by Anderson *et al.*[4].

During high tide, the activity ratios of $^{234}U/^{238}U$ in dissolved phase ($^{234}U/^{238}U_d$) were slightly fluctuated from 0.86 to 1.26 with mean value of 1.00±0.03. This ratio was also slightly proportional increased with increasing the contents of Fe (r^2>0.19; Fig 2a) but the negative statistical correlation (r^2<0.15; Fig. 2b) was calculated during the low tide. A near-equilibrium activity ratio of

Table 2: Concentration activities of uranium isotopes and *in situ* parameters

Tidal stage	Stn.	Spc (mS cm^{-1})	Sal (psu)	pH	Temp. (°C)	DO (mg L^{-1})	Dissolved phase ^{238}U (mBq L^{-1})	Dissolved phase ^{234}U (mBq L^{-1})	Dissolved phase ^{234}U/^{238}U	Particulate phase ^{238}U (mBq g^{-1})	Particulate phase ^{234}U (mBq g^{-1})	Particulate phase ^{234}U/^{238}U
High	1	20.06	0.00	7.03	28.90	4.22	55.62±3.07	71.10±3.70	1.28±0.10	184.11±29.47	346.77±42.03	1.88±0.38
tide	2	21.65	0.00	7.04	28.11	4.21	95.79±4.67	93.04±4.66	0.97±0.07	211.57±26.66	216.44±27.54	1.02±0.18
	3	33.89	0.00	7.03	28.21	4.18	32.25±2.26	34.28±2.41	1.06±0.11	135.90±4.21	151.64±4.61	1.12±0.05
	4	43.81	0.10	7.04	28.85	4.77	83.70±3.83	82.91±3.87	0.99±0.06	148.18±7.60	251.39±10.42	1.70±0.11
	5	45.17	0.00	7.05	28.90	5.05	33.17±2.29	34.06±2.40	1.03±0.10	75.05±14.60	78.39±15.27	1.04±0.29
	6	41.76	0.00	7.05	29.48	4.46	109.93±6.00	94.01±5.43	0.86±0.07	122.60±10.96	233.82±15.92	1.91±0.21
	7	42.87	2.03	5.66	29.81	4.06	34.75±2.39	33.22±2.38	0.96±0.09	308.20±20.60	403.11±24.76	1.31±0.12
	8	44.29	9.30	6.49	29.93	7.16	69.03±4.37	61.22±4.13	0.89±0.08	762.79±56.20	1125.93±70.65	1.48±0.14
			Mean value				64.28±1.35	62.98±1.34	1.00±0.03	243.55±9.33	350.93±11.69	1.43±0.07
Low	1	19.46	0.13	3.09	28.34	4.41	53.28±3.84	41.51±3.60	0.78±0.09	156.71±33.31	230.16±43.37	1.47±0.42
tide	2	31.15	0.15	2.89	28.26	4.28	37.56±2.40	38.02±2.54	1.01±0.09	78.70±7.76	118.40±10.16	1.50±0.20
	3	40.55	0.16	3.6	28.62	4.25	40.22±2.62	36.92±2.63	0.92±0.09	551.88±24.88	564.85±25.54	1.02±0.07
	4	16.09	0.17	7.15	29.00	6.21	37.84±2.22	39.60±2.34	1.05±0.09	5.61±0.38	10.66±0.55	1.90±0.16
	5	41.85	0.16	7.17	29.31	4.80	45.33±2.64	75.73±3.80	1.67±0.13	10.27±0.43	12.92±0.51	1.26±0.07
	6	40.23	1.51	3.95	29.70	4.65	61.52±3.38	59.23±3.36	0.96±0.08	18.03±4.46	21.50±5.32	1.19±0.42
	7	43.96	4.77	5.14	29.98	4.38	30.32±2.23	26.95±2.24	0.89±0.10	175.32±9.40	191.27±10.17	1.09±0.08
	8	45.35	9.30	6.18	31.06	7.17	30.54±2.24	44.21±2.87	1.45±0.14	999.31±39.98	1074.20±42.77	1.07±0.06
			Mean value				42.08±0.97	45.27±1.05	1.09±0.04	249.48±7.39	277.99±8.48	1.31±0.08

Where, Spc is specific conductivity at 25°C, Sal is salinity, Temp. is temperature and DO is dissolved oxygen

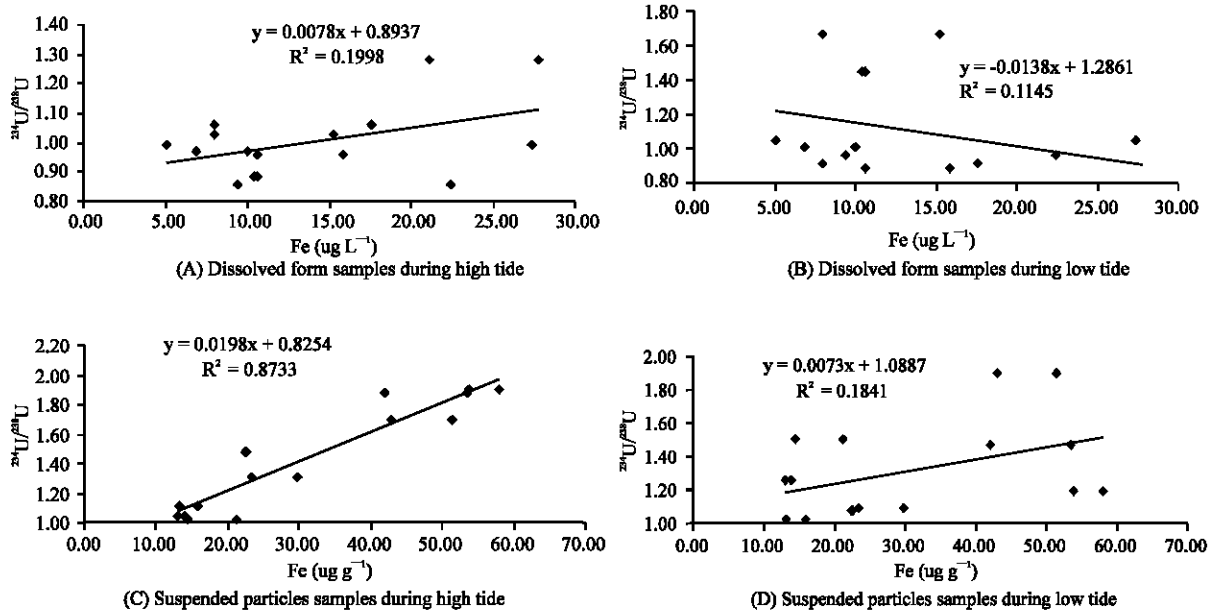

Fig. 2: Statistical correlation in water column calculated between $^{234}U/^{238}U$ ratios with the contents of Fe[10]

$^{234}U/^{238}U_d$ in water column of Sungai Selangor may be implying that uranium isotopes in river water are derived from the weathered materials, where most of the mobile ^{234}U has already been lost or attached with suspended particulate material[6]. This clearly showed that most of uranium in water column at study area was associated with Fe-oxides during high tide and released as the oxides reduction during low tide, which containing high level of ^{238}U[7]. Where as in the Fig. 2c and d were clearly shows that the contents of uranium indicated by ratio of $^{234}U/^{238}U$ was strongly correlated with content of Fe in suspended particles during high tide (r^2>0.8) and slightly weak appear during low tide (r^2<0.2). Then the distribution pattern of $^{234}U/^{238}U_d$ obtained during high tide will be summarized in the order as follows; river (1.13)>estuary (1.03)>near shore (0.94) and inversed shows during the low tide as; near shore (1.24)>estuary (0.99)>river (0.90).

Particulate ^{234}U and ^{238}U: Activity of uranium isotopes in the particulate was slightly higher than dissolved phase, where the mean activity of ^{234}U in particulate phase during the high tide of 350.93±11.69 mBq g^{-1} and about

Table 3: Calculated of K_d values for uranium nuclides during high and low tides

	TSM (g L^{-1})		^{238}U (K_d x 10^3 L g^{-1})		^{234}U (K_d x 10^3 L g^{-1})		$K_d^{234}U/K_d^{238}U$ (ratio)	
Stations	High tide	Low tide	High tide	Low tide	High tide	Low tide	High tide	Low tide
1	0.03	0.17	117.00±10.85	18.94±2.09	195.01±3.91	34.04±6.81	1.67±0.16	1.80±0.41
2	0.05	0.23	42.48±6.03	8.77±0.43	49.44±1.08	13.15±1.48	1.16±0.17	1.50±0.18
3	0.37	0.32	12.66±0.88	42.81±0.92	13.56±3.38	43.59±4.02	1.07±0.28	1.02±0.10
4	0.06	1.74	30.53±1.98	0.14±0.08	50.88±0.01	0.14±0.01	1.67±0.11	1.00±0.58
5	1.61	3.33	1.66±0.29	0.06±0.03	1.66±0.00	0.04±0.00	1.00±0.17	0.67±0.33
6	0.06	3.43	18.72±2.00	0.10±0.01	43.86±0.02	0.10±0.03	2.34±0.25	1.00±0.32
7	0.08	0.72	134.97±11.10	8.23±0.97	174.16±0.73	8.74±0.97	1.29±0.11	1.06±0.17
8	0.01	0.14	1364.00±14	244.38±12.86	2366.00±20	196.11±13.58	1.73±0.02	0.80±0.07

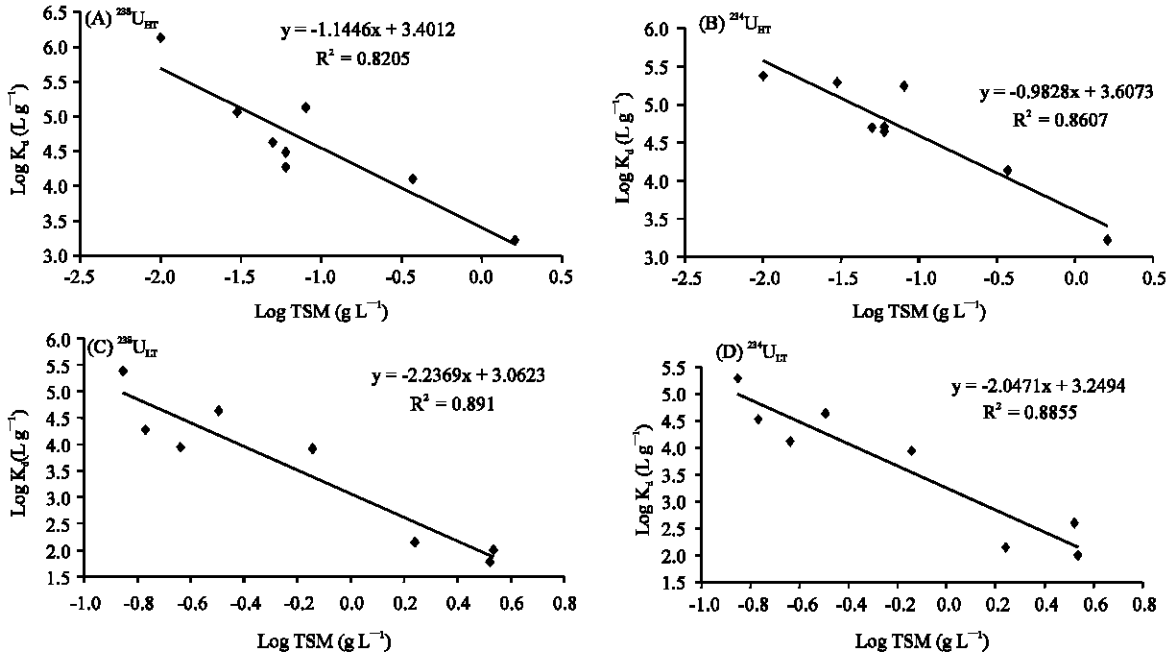

Fig. 3: Correlation regression calculated in water column of Sungai Selangor between K_d values with the contents of TSM for ^{234}U and ^{238}U, where the LT and HT is the low tide and high tide, respectively

277.99±8.48 mBq g^{-1} during the low tide (Table 2). Meanwhile, the mean activity of ^{238}U in particulate phase is slightly same during the high and low tides of 243.55±9.33 mBq g^{-1} and 249.48±7.39 mBq g^{-1}, respectively (Table 2). The activity ratio of $^{234}U/^{238}U$ in particulate ($^{234}U/^{238}U_p$) is also higher than the dissolved phase, with the mean activity ratio in the river stations was to be 1.45 and 1.49 during high and low tides respectively, except near-shore stations during the low tide to be 1.15. High activity ratio of $^{234}U/^{238}U_p$ in river stations especially during low tide might be imply that uranium isotopes are derived from weathered materials, where most of the mobile ^{234}U has already been lost during neap from the dissolved phase[6] and the ^{234}U isotope is more attachable into the particulate phase especially during low tide.

The activity ratio of $^{234}U/^{238}U_p$ is approximately a mirror image of $^{234}U/^{238}U_d$ shows during low tide, which mean uranium at study area is exchangeable between dissolved and particulate phase occur in water column during low tide and the same situation also shows during high tide (Table 2). This phenomenon gives us some idea removal of the uranium from dissolved to particulate phase occurred in the water column as well described by Sarin and Church[6]. The removal of uranium from dissolved to particulate phase at study stations probably related to the sorption or/desorption process of Fe oxyhydroxides[8], where the concentration of Fe in the particulate at study area > 50 μg g^{-1}[9]. But the different of Eh-pH condition in three different environments (i.e., freshwater, brackish water and seawater) could be reduce the Uranyl Ion U(VI) in the dissolved form onto the insoluble uranous ion U(IV)[10].

Distribution coefficient values (K_d) of ^{234}U and ^{238}U: The distribution coefficient (K_d) is widely used as approach for understanding and determinate eventual fate of radionuclides released into the aquatic environment, especially ^{234}U and ^{238}U. Based on interaction some physical and chemical aspects between dissolved and

particulate phases, some mathematical equations have been developed to explain the variability of distribution coefficient in real environments[11]. In this study, the K_d is defined as;

$$K_d = [M_e]_p / ([M_e]_d \times TSM)$$

Where, $[M_e]_p$ is activity of ^{234}U and ^{238}U in particulate phase (Bq g^{-1}); $[M_e]_d$ is activity of ^{234}U and ^{238}U in dissolved phase (Bq L^{-1}) and TSM is concentration of total suspended particulate matter (g L^{-1}).

The calculated K_d values obtained during high tide slightly higher than low tide, with ranged from 1.66×10^3 to 1364×10^3 L g^{-1} and 1.66×10^3 to 2366×10^3 L g^{-1} for ^{238}U and ^{234}U, respectively (Table 3). But for low tide stage their values was ranging from 0.07×10^3 L g^{-1} to 244×10^3 L g^{-1} for ^{238}U and 0.04×10^3 to 196×10^3 L g^{-1} for ^{234}U (Table 3). Highest K_d value was 2366×10^3 L g^{-1} and 1364×10^3 L g^{-1} for ^{234}U and ^{238}U, respectively, obtained at Station 8, indicating there are a strong adoption of both nuclides by suspended particles occur during high tide cycles (Table 3). Moreover, these values may be reflected to the scavenging rate of dissolved in water column and the desorption rate of suspended materials.

A negative statistical correlation ($r^2>0.8$) during high tide and low tide cycles has been showed from the plotted $K_d{}^{234}U$ and $K_d{}^{238}U$ values against the amounts of particle concentration (Fig. 3). This indicating most scavenging process of uranium at study stations was effect by the amounts or chemistry of suspended particulate in water column. Where the distribution coefficients ratio ($K_d{}^{234}U/K_d{}^{238}U$) as the fractionation factor ($F_{234U/238U}$) also calculate during both cycles process was varied from 1.00±0.17 to 2.34±0.25 and 0.67±0.33 to 1.80±0.41 for high tide and low tide stage, respectively (Table 3). High or more than 1.0 of $F_{234U/238U}$ calculated during high tide strongly suggest that the preferential adoption of ^{234}U was much relative than ^{238}U by the particulate materials.

CONCLUSIONS

Activities of uranium isotopes were high in the particulate phase obtained along the Sungai Selangor. High $F_{234U/238U}$ value of uranium was also indicated a strong adoption of ^{234}U onto the suspended particles matter and reflect increased the scavenging rate of dissolved ^{234}U and ^{238}U in water column.

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