MEASUREMENT OF GROSS ALPHA, GROSS BETA, RADON AND RADIUM ACTIVITY CONCENTRATIONS IN AQUEOUS SAMPLES USING LIQUID SCINTILLATION TECHNIQUE

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ABSTRACT

Recently, Malaysia has taken a positive step toward providing a better water quality by introducing more water quality parameters into its Water Quality Standard. With regard to the natural radionuclides that may present in the water, 3 parameters were introduced that is gross alpha, gross beta and radium which need to be measured and cannot exceed 0.1, 1.0 and 1.0 Bq/L respectively. This study was conducted to develop a more practical method in measuring these parameters in aqueous environmental samples. Besides having a lot of former tin mining areas, some part of Malaysia is located on the granitic rock which also contributes to a certain extent the amount of natural radionuclides such as uranium and thorium. For all we know these two radionuclides are the origin of other radionuclides being produced from their decay series. The State of Kelantan was chosen as the study area, where the water samples were collected from various part of the Kelantan River. 25 liters of samples were collected, acidify to pH 2 and filtered before the analysis. Measurement of these parameters was done using liquid scintillation counter (LSC). The LSC was set up to the optimum discriminator level and counting was done using alpha-beta mode. The results show that gross alpha and beta can be measured using scintillation cocktail and radium and radon using extraction method. The results for gross alpha, gross beta, ^{222}Ra and ^{226}Ra are 0.39-6.42, 0.66-16.18, 0.40-4.65 and 0.05-0.56 Bq/L. MDA for gross alpha, gross beta and radium is 0.03, 0.08 and 0.00035 Bq/L respectively.

ABSTRAK

Beberapa tahun kebelakangan ini, Malaysia telah mengambil satu langkah yang positif ke arah menyediakan air yang lebih berkualiti dengan memperkenalkan lebih banyak parameter kualiti air ke dalam Piawai Kualiti Air. Dengan mengambil kira radionuklid semula jadi yang boleh wujud di dalam air, 3 parameter diperkenalkan gros alfa, gros beta dan radium yang perlu diukur dan tidak boleh melebihi 0,1, 1.0 dan 1.0 Bq / L masing-masing. Kajian ini dijalankan untuk membangunkan kaedah yang lebih praktikal dalam mengukur parameter ini dalam sampel larutan alam sekitar. Selain mempunyai banyak kawasan-kawasan bekas lombong bijih timah, ada sebahagian daripada Malaysia terletak di atas batu granit yang juga menyumbang kepada tahap tertentu kadar radionuklid semula jadi seperti uranium dan torium. Seperti yang diketahui, kedua-dua radionuklid ini adalah asal-usul radionuklida lain yang dihasilkan dari siri pereputan mereka. Negeri Kelantan telah dipilih sebagai kawasan kajian, di mana sampel air yang dikumpul daripada pelbagai Sungai Kelantan. 25 liter sampel telah dikumpulkan, diacidkan ke pH 2 dan ditapis sebelum analisis. Pengukuran parameter ini dilakukan dengan menggunakan kaunter scintillation cecair (LSC). LSC telah ditetapkan sehingga ke tahap diskriminasi optimum dan pengiraan telah dilakukan dengan menggunakan mod alpha-beta. Keputusan menunjukkan bahawa alfa dan beta secara kasar boleh diukur menggunakan koktail scintillation, radium dan radon menggunakan kaedah pengekstrakan. Keputusan untuk kepekatan aktiviti bagi gros alpha, gros beta, ^{222}Rn dan ^{226}Ra 0.39-6.42, 0.66-16.18, 0.40-4.65 dan 0.05-0.56 Bq/L masing-masing. MDA untuk gros alfa, gros beta dan radium adalah 0.03, 0.08 dan 0.00035 Bq/L masing-masing.

Keywords: aqueous environmental sample, gross alpha, gross beta, radionuclides, Kelantan River Basin

INTRODUCTION

The important of water in human life is not a question any where in the world. In this study, Kelantan State was chosen as a study area, since river water is still the main source for daily consumption primarily for drinking, cooking and bathing. Thus, an increasing rate of daily water usage due to the increase in population causes an increase in awareness about cleanliness and safety of water used. Moreover, a step to control and monitor the environment and water hygiene is crucial in determining the level of guaranteed health of the people because its ability to transport pollutants. Not only that, most of the Kelantan area is sitting on the granite rocks cause more attention should be given by the scientists because the area not only rich in mineral resources but also radionuclide such as uranium deposits (Geological Map of Kelantan, 2000).

The presence of radioactive material in water is often due to the presence of radioactive elements within earth crust (Damla *et. al.*, 2009). Moreover, mineral mining activities far into the realm of land increase the presence of radioactive material substances in the environment. The risk of drink the water that is having high content of radionuclides may harm the person because of the internal exposure. It may be due to the decay products taken into the body through ingestion or inhalation indirectly obtained from the food chain (Görür *et. al.*, 2011).

The natural radionuclides present in the granite rocks sometimes dissolved in water, predominantly the members of decay series of ^{232}Th, ^{235}U and ^{238}U (Jobbágy *et. al*., 2011). The activity concentrations of radionuclides content depends on the origin of the water, the flow rate and flux of radionuclides and geological characteristics of the area (Jobbágy *et. al*., 2011). Generally, the presences of alpha and beta emitters are natural, but its activity concentration depending on local circumstances and characteristics of existing ground or rocks lies beneath the earth, particularly leaching from minerals (Akyil *et. al*., 2009). According to the UNSCEAR (2000), global average human exposure from natural sources is 2.4 mSv/y (i.e. 0.4, 0.5, 1.2 and 0.3 mSv/y for cosmic ray, terrestrial gamma ray, radon, and food and drinking water, respectively). Since the data on natural radioactivity in environmental samples are not sufficient, effort on more data collection need to be done involving various scientific fields.

Some surveys show that is high in activity concentrations in particular areas in the world (Jobbágy *et. al.*, 2011. Thus, serious attention should be given to the environmental studies because significant doses of radioactive material in water bodies such as river, lake or reservoirs may harm a man (Akyil *et. al*., 2009). According to the WHO (2004), the recommended value for gross alpha and beta is 0.5 and 1 Bq/L respectively, whereas based on the National Water Quality Standards for Malaysia (INWQS) is 0.1 and 1 Bq/L respectively and 1 Bq/L for ^{226}Ra. Gross alpha and beta can be the first steps for the radiological assessment in drinking water and any water that is exceed the recommended value can be screen and only places that have below INWQS is suitable for human consumption. It is important to do radiological assessment especially regarding to the risk and doses that be consumed by the population.

The objective of this study is to measure radioactivity level in water i.e. the gross alpha, gross beta. ^{222}Rn and ^{226}Ra. These results can be used as a baseline for future studies, and to find their relations to the water quality index. This paper also focus on the estimating the annual effective dose in water at the sampling points.

METHODOLOGY

River water samples

25 L of raw sample was collected from Kelantan River tributaries in polyethylene bottle that have been washed with HNO_3 and rinse with distilled water. The samples were taken at the mid depth of the river to ensure that it is representative. Concentrated HNO_3 with the ratio of 1:1 of HNO_3 and water was added to the raw sample as soon as it reached the laboratory to prevent the loss by sorption of the radionuclides in the bottles, to ensure the radionuclides remain in the solution and to reduce biological growth, avoid the collection of organic materials and changes in the state of the ions present in the samples (Reyes & Marques,

2008; Jobbágy *et. al*., 2011; Görür *et. al*., 2011). The samples were filtered using cellulose acetate membrane filter 0.45 µm porosity and 47 mm diameter to eliminate suspended solid in the water samples. Name and location of the sampling area can be found in Table 1 and Figure 1.

Table 1: Name, sampling code and coordinates of the sampling areas

Sampling location	Sampling Code	Coordinates
Jeram Gajah	SK1	N04°44' E101°44'
Kg. Kuala Betis	SK2	N04°55' E101°48'
Sg. Nenggiri	SK3	N05°59' E101°46'
Kg. Pulau Stelu	SK4	N05°06' E101°59'
Kg. Bertam	SK5	N05°09' E102°03'
Kg. Kemubu	SK6	N05°18' E102°01'
Tasik Pergau	SK7	N05°38' E101°42'
Kg. Batu Melintang	SK8	N05°43' E101°45'
Kg. Pasir Dusun	SK9	N05°39' E101°52'
Kuala Terang	SK10	N05°29' E101°54'
Kg. Dalam Balah	SK11	N05°25' E101°55'
Manik Urai	SK12	N05°23' E102°16'
Kg. Chalil	SK13	N05°06' E102°17'
Sg. Temiang	SK14	N05°21' E102°15'
Kuala Krai	SK15	N05°32' E102°12'
Kg. Kerilla	SK16	N05°39' E102°07'
Jambatan Pasir Mas	SK17	N06°02' E102°29'
Jeti Kok Majid	SK18	N06°11' E102°14'
Pulau Haji Nikmat	SK19	N06°11' E102°14'
Kuala Besar	SK20	N06°12' E102°14'

Extraction method for ^{226}Ra and ^{222}Rn

To prepare liquid scintillator, 4 g of 2, 5-diphenyloxazole (PPO) and 0.4 g of 1, 4-bis-(5-phenyl-oxazolyl-2)-benzene (POPOP) were added into one litre scintillation grade toluene (Hamzah *et. al.*, 2011). This solution was stirred for 24 hours to make sure that chemicals dissolved and dispersed evenly in the solution. Then, 100 mL of liquid scintillator was added to the one litre water samples, shook and incubated for three weeks. After three weeks, the extraction mixture was once again being shaken and left for a while until the organic layer and aqueous layer was fully separated. The time of separation was recorded. Finally, the toluene layer was pipette for 20.0 mL and transferred into polyethylene vial and immediately measured in LSC. It is extraction process that trapped radionuclide of ^{222}Rn in the solvent of liquid scintillation.

LSC Measurements

The extracted samples were measured two times for 100 minutes each cycle by using the liquid scintillation counter. ^{226}Ra concentration was calculated from the total alpha peaks of ^{226}Ra and its daughters (^{222}Rn, ^{218}Po and ^{214}Po) in the alpha spectrum region. ^{228}Ra concentration was calculated in the beta spectrum region (Yong, 2001).

Gross alpha and beta sample preparation and counting

Four mL of filtered water was transferred into scintillation vial. Then add 16 mL cocktail containing toluene (with PPO and POPOP) and Triton N101 (emulsifier). After shaking the vial, leave it over night and it is ready for counting. The samples were counted in LSC using alpha beta mode and discriminator level set at 120 minutes (Hamzah *et. al.,* 2011).

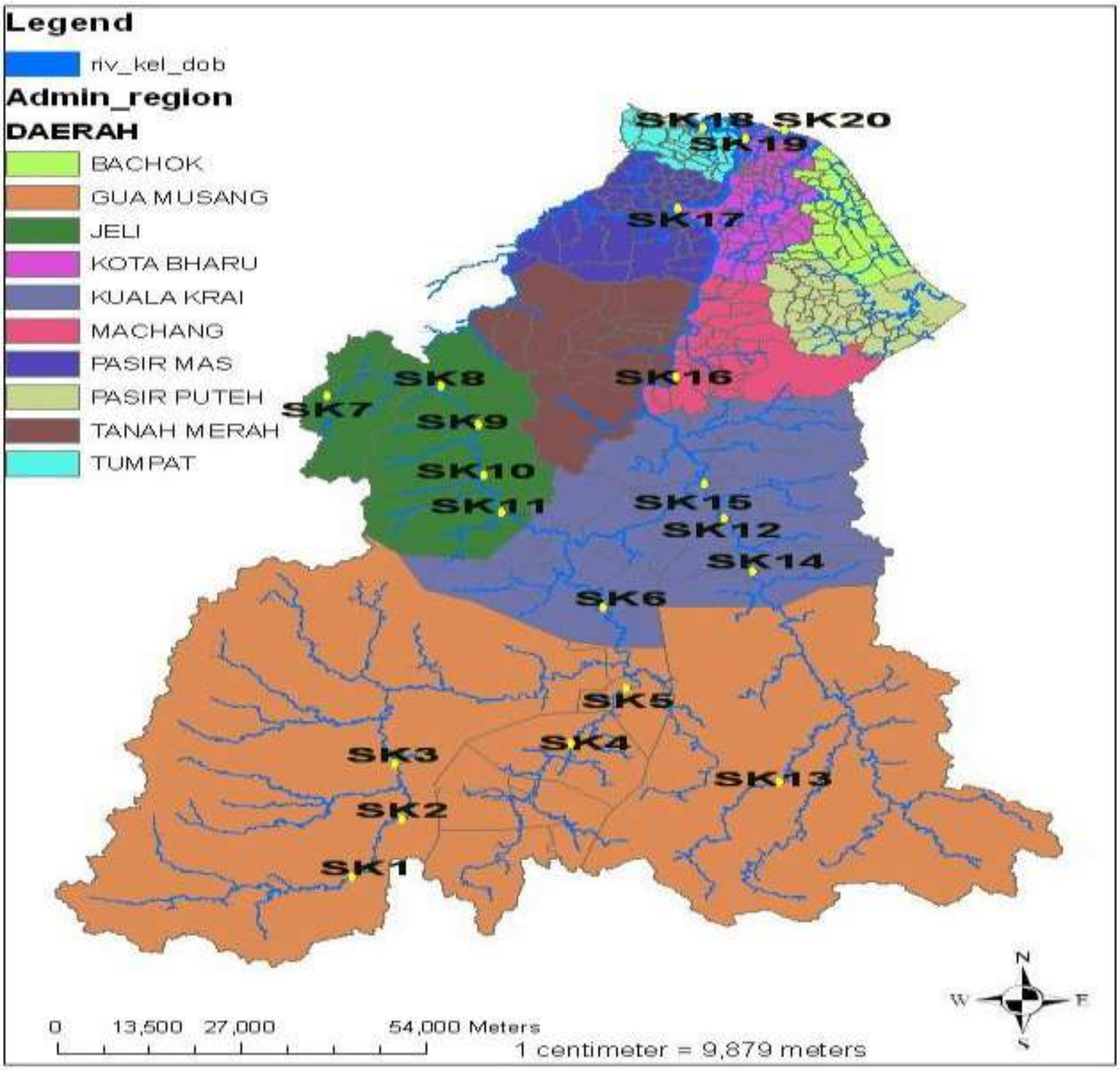

Figure 1: Map of samplings area with sampling codes on the main rivers covering eight districts in Kelantan.

RESULTS AND DISCUSSION

The activity concentration of gross alpha and beta obtained from sampling area of Kelantan are given in Table 2.

Table 2: Gross alpha and beta activity concentrations in water samples collected along Kelantan River

Sampling codes	Gross Alpha (Bq/L)	Gross Beta (Bq/L)
SK1	6.42±0.01	<MDA
SK3	2.19±0.01	<MDA
SK7	1.33±0.01	<MDA
SK10	2.97±0.01	<MDA
SK16	0.39±0.02	16.18±0.13
SK17	3.00±0.02	0.66±0.13
SK18	<MDA	9.63±0.13

Note: MDA Gross alpha = 0.03 MDA Gross beta = 0.08 Bq/L

The measured gross alpha range from 0.39 to 6.42 Bq/L, where as gross beta lies between 0.66 and 16.18 Bq/L. There is only one sample that is below the recommended value gross alpha which is SK18. Gross beta values for two locations which SK16 and SK18 are above the allowable limit and the rest is acceptable for human consumption. These results show that there is certain areas gives high value of gross alpha that is not suitable for daily consumption especially for drinking. For example SK1 and SK16 have the highest activity concentration of gross alpha and beta respectively, both exceeded recommended value by WHO and National Water Quality Standards for Malaysia (INWQS). This is the first detailed study of gross alpha and beta activity concentration in river water of Kelantan state.

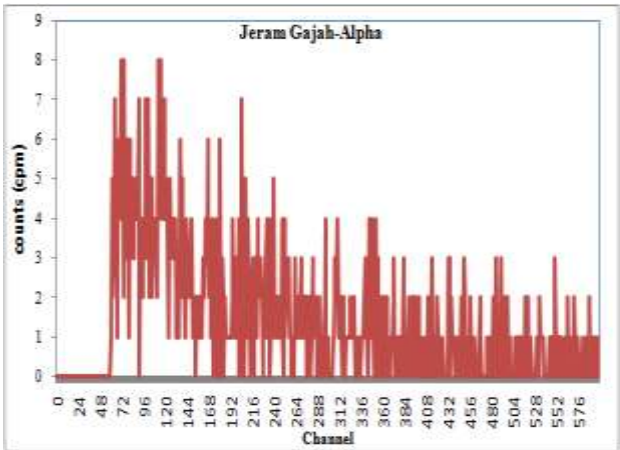

Figure 2: Spectra of gross alpha SK1

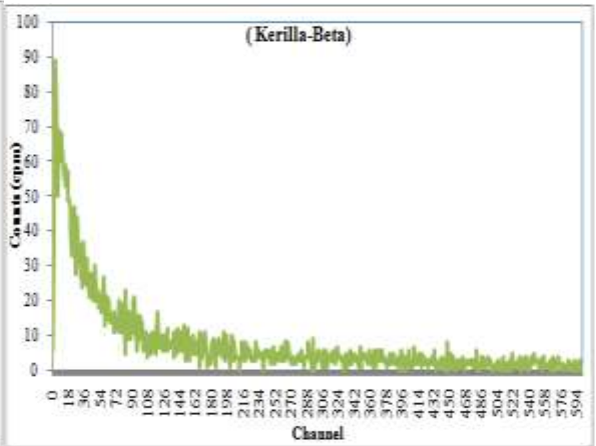

Figure 3: Spectra for gross beta at SK16

Usually, gross alpha in water is originated from uranium isotopes and ^{226}Ra because thorium has low solubility in water (Jobbágy *et. al.*, ; Görür *et. al.*, 2011). It is known that Kelantan state rich in granites and it is possible to have one or more uranium deposit down beneath the earth crust (Geological Map of Kelantan). On the other hand, an activity concentration of beta usually originated from ^{228}Ra, ^{40}K and ^{210}Pb (Jobbágy *et. al.*, 2011).

Figure 2 and 3 are the examples of spectrum obtained from the analysis. From the alpha spectrum, the highest peak shows the counts for the ^{226}Ra, ^{222}Rn and ^{218}Po which originated from ^{238}U. Pre-concentration is necessary to get the good peak for the alpha emitting nuclides. The major beta emitting nuclides in the ^{238}U decays series are ^{214}Bi and ^{214}Po where as ^{40}K decays directly to ^{40}Ca through beta emissions (Bonotto *et. al.*, 2009). Figure 3 shows the beta emitting of ^{40}K at the highest peak from channel 0 to 100. ^{40}K is the major contribution of the gross beta besides ^{214}Bi and ^{214}Po (Bonotto *et. al.*, 2009). The usefulness of gross alpha and beta in screening water samples is very convenient because it is easy and has low cost in terms of laboratory preparations. Furthermore, it is not using longer time as the extraction method thus it is suitable for environmental monitoring that really needs immediate results.

Table 3 shows the activity concentrations of ^{222}Rn and ^{226}Ra in water samples that have collect from Kelantan River Basin. The activity concentration of ^{222}Rn ranging from 0.4 to 4.65 Bq/L and SK13 is at the top. The activity concentrations of ^{226}Ra have the minimum value at SK 16 and the highest is at SK13 with 0.56 Bq/L. it is approved that all location accept two that did not exceed the INWQS and WHO which are SK16 and SK19. From the result of the activity concentrations of ^{226}Ra, it is believed that most of the Kelantan River water is not safe to be used for daily use especially for drinking.

Table 3: ^{222}Rn and ^{226}Ra activity concentrations in water samples collected along Kelantan River

Sampling location	^{222}Rn Activity Concentration (Bq/L)	^{226}Ra Activity Concentration (Bq/L)
SK2	2.29±0.34	0.28±0.04
SK4	2.16±0.34	0.26±0.04
SK5	4.54±0.43	0.55±0.05
SK6	1.94±0.52	0.23±0.06
SK8	3.11±0.46	0.37±0.06
SK9	1.22±0.68	0.15±0.08
SK11	1.67±0.62	0.20±0.07
SK12	4.43±0.27	0.55±0.03
SK13	4.65±0.33	0.56±0.04
SK14	3.89±0.42	0.47±0.05
SK15	2.58±0.33	0.32±0.04
SK16	0.40±0.07	0.05±0.008
SK19	0.52±0.07	0.06±0.008
SK20	3.35±0.27	0.41±0.03

Note: MDA ^{226}Ra = 0.00035 in Bq/L

The minimum detectable activity is expressed using equation below (Görür *et. al.*, 2011):

$$MDA = [4.65\ \sqrt{(B.t)} + 2.71]/(E.V.t) \quad Eq.\ 1$$

E = counting efficiency
V = sample volume (L)
B = background count rate (cpm)
t = counting time (minutes)

The minimum detectable activity at 120 minutes for gross α and β is 0.03 and 0.08 Bq/L respectively. The MDA for ^{226}Ra is 0.00035 Bq/L at 100 minutes.

The following equation is for calculating annual effective dose (Görür *et. al.*, 2011):

$$Annual\ effective\ dose = 730\ L/yr \times 2.8\text{x}10^{-4}\ mSv/Bq \times activity\ concentration\ of\ ^{226}Ra\ (Bq/L) \quad Eq.\ 2$$

Assuming people from these areas using the river water as main water supply, annual effective dose (AED) for consumption of water for an adult were then calculated. The amount of drinking annually in amount is 730 L/yr assuming that adult drinks water for 2L/day and the dose conversation factor for ^{226}Ra are $2.8\text{x}10^{-4}$ mSv/Bq.

Table 4: Annual Effective Dose for activity concentrations of ^{226}Ra

Sampling location	Annual Effective Dose of ^{226}Ra (mSv/year)
SK2	0.06
SK4	0.05
SK5	0.11
SK6	0.05
SK8	0.08
SK9	0.03
SK11	0.04
SK12	0.11
SK13	0.11
SK14	0.10
SK15	0.06
SK16	0.01
SK19	0.01
SK20	0.08
Recommended value	<0.10

From Table 4, there are 3 places that have higher values on AED, above the recommendation value by WHO which are SK5, SK12 and SK13 as well. These are places that need to be monitoring even further so that the risk on health of people would not affect them. Drinking more water from this area could affect the health and harm the people because of the internal exposure.

For comparison of gross alpha and beta activity concentrations from various countries are presented in Table 5. From our study, it is shown that the values of Kelantan River water quite higher than the other places in the world (Jobbágy et. al., 2011; Görür *et. al.*, 2011; Damla *et. al.,* 2009; Akyil *et. al*., 2009; Karahan et. al., 2000; Hancock *et. al.,* 1996). Table 5 shows the summarized data on activity concentration all over the world. Those values are comparable with the present study. This approves the theory that granitic rocks did affect the concentrations of natural radionuclides that presence in the environment.

Table 5: Comparisons on activity concentrations of gross alpha, gross beta, ^{222}Rn and ^{226}Ra activity concentrations in water samples in Bq/L.

Origin	Type	Gross α	Gross β	^{222}Rn	^{226}Ra	References
Kelantan River Basin, Malaysia	River water	0.39-6.42	0.66-16.18	0.66-4.646	0.147-0.558	Present study
Balaton Upland, Hungary	Spring water	0.035-1.749	0.033-2.015	nm	nm	Jobbágy et. al., 2011
Samsun, Turkey	Tap water River water Spring water	0.024-0.116 0.128-0.156 0.061-0.091	0.041-0.191 0.170-0.191 0.121-0.190	nm	nm	Görür *et. al.*, 2011
Batman, Turkey	Tap water Well water River water Spring water	0.011-0.052 0.073 0.047 3.909	0.003-0.347 0.074 0.073-0.083 2.097	nm	nm	Damla *et. al.*, 2009
Izmir,Turkey	Lake	0.03-0.75	1.77-2.62	nm	nm	Akyil *et. al.*, 2009
Istanbul, Turkey	Well water Lake	0.08-0.045 0.02-0.06	0.037-0.11 0.04-3	nm 0.025-0.048	nm 0.015-0.056	Karahan et. al., 2000
Bega River estuary, Autralia	Surface water	nm	nm	nm	0.0006-0.003	Hancock *et. al.*, 1996
Recommended value	All type of water	<0.1	<1	-	<0.1	WHO, 2004

nm=not measured

The water quality parameters of the water samples measured during sampling are shown in Table 5.

Table 5: Water quality parameters along the Kelantan River Basin

Location	Temp.	DO (mg/L)	SPC (mS/cm)	TDS (mg/L)	pH	NH_4 (mg/L)	NO_3 (mg/L)	Cl^- (mg/L)	Turbidity (NTU)
SK1	23.33	9.65	0.04	0.03	7.22	5.36	0.28	47.70	216.8
SK2	25.30	9.20	0.03	0.02	7.10	6.40	6.40	1.02	125
SK3	23.01	9.33	0.04	0.02	7.62	5.47	0.34	47.36	5.64
SK4	26.30	13.00	0.03	0.02	5.60	15.90	3.60	1.02	968
SK5	25.40	9.30	0.03	0.02	5.30	18.20	6.40	1.02	3000
SK6	27.00	11.60	0.04	0.28	5.70	14.80	7.30	1.03	1580
SK7	23.62	10.32	0.02	0.01	6.93	5.12	0.07	47.92	23.80
SK8	24.00	22.20	0.07	0.04	5.66	13.90	11.40	1.03	46.00
SK9	25.00	10.20	0.03	0.02	5.60	16.60	9.10	1.02	110
SK10	23.41	8.95	0.03	0.02	7.06	5.23	0.03	48.05	28.60
SK11	26.00	15.30	0.02	0.01	5.50	15.90	7.60	1.02	12.00
SK12	22.00	11.30	0.00	0.00	6.80	6.80	0.30	1.00	34.70
SK13	26.30	3.70	0.13	0.08	7.60	5.00	0.00	1.05	4.00
SK14	30.00	3.00	0.04	0.03	7.20	5.10	0.00	1.03	8.02
SK15	24.72	9.86	0.05	0.03	6.91	6.25	0.38	1.03	288.62
SK16	23.50	9.56	0.04	0.02	7.27	4.79	0.00	47.82	386.60
SK17	23.50	9.56	0.04	0.02	7.27	4.79	0.00	47.82	386.60
SK18	24.12	6.94	0.02	0.13	6.91	5.62	0.02	51.46	231.40
SK19	23.79	8.46	0.04	0.03	7.07	4.86	0.00	48.19	398.00
SK20	24.87	8.17	0.12	0.08	6.84	8.36	0.45	1.04	285.67
INWQS	-	3-5	-	-	5-9	-	-	-	-

From Table 5, it shows that the water temperature is the range of 22.00-30.00ºC. Dissolves oxygen is in the range of 3.00-22.20mg/L and specific conductivity is in the range of 0.00-0.13 mS/cm, while TDS is from 0.00-0.28 mg/L. Ammonical nitrogen, nitrate and chloride are in the range of 4.79-18.20, 0.00-11.40 and 1.00-51.46 mg/L, respectively. Turbidity ranges from 4-3000 NTU, with SK8 give the highest turbidity. If we relate the activity concentration of ^{226}Ra with turbidity, where SK5 and SK13 are among the highest turbidity reading, it seem that turbidity has no effect on the activity concentration of ^{226}Ra. Thus, turbidity of the river water does not influence the activity concentration of ^{226}Ra in water as if the water is filtered.

Table 6: Pearson Correlation on the Water Quality Parameters with the activity concentration of ^{222}Rn and ^{226}Ra

	^{222}Rn	^{226}Ra	Temp.	DO	SPC	TDS	pH	NH_4	NO_3	Cl^-	Turbidity
^{222}Ra	1.00										
^{226}Ra	1.00	1.00									
Temp.	0.19	0.17	1.00								
DO	0.22	0.23	0.40	1.00							
SPC	0.30	0.29	0.14	0.26	1.00						
TDS	0.03	0.04	0.31	0.03	0.26	1.00					
pH	0.08	0.09	0.05	0.65	0.37	0.12	1.00				
NH_4	0.03	0.04	0.11	0.55	0.28	0.15	0.98	1.00			
NO_3	0.13	0.15	0.03	0.70	0.23	0.15	0.79	0.79	1.00		
Cl^-	0.64	0.64	0.37	0.12	0.09	0.14	0.38	0.43	0.39	1.00	
Turbidity	0.17	0.16	0.11	0.00	0.18	0.30	0.52	0.54	0.21	0.06	1.00

From Table 6, it is known that there are strong correlation between activity concentration of ^{222}Rn and ^{226}Ra which is 1. There are weak correlations between temperature, specific conductivity (SPC), pH and turbidity with the activity concentration of ^{222}Rn and ^{226}Ra. Clearly there is no strong correlation between them that we can determine the water chemistry between them. Further water quality parameters should be investigated to see the chemistry between the activity concentration of ^{222}Rn and ^{226}Ra so that we can easier monitor the river.

CONCLUSIONS

Radiochemical assessments of 20 Kelantan River basins were obtained. The activity concentration of gross alpha, beta, ^{222}Rn and ^{226}Ra higher in certain areas might be due to the geological features and chemical composition of the river basin. The radioisotopes may present in the water caused of geological erosion from the ground water and dissolution of uranium-bearing rocks due to changing of temperature in the earth crust.

This scope of studies must be furthered investigate for the other water bodies such as river basin or reservoirs covering all villages in Kelantan. The data obtained in this study can be used as a baseline to assess any possible changes. Further detailed on radioactivity assessment in environmental should be investigate for estimating of annual dose received by consumers especially for the purposes of agriculture, fishery, tourism/recreation, and water supply.

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