DETERMINATION OF RADON ACTIVITY CONCENTRATION IN WATER USING LIQUID SCINTILLATION COUNTING TECHNIQUE

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ABSTRACT

Many studies were carried out throughout the world on radon measurement in water especially drinking water for it can cause problem to human health. This study is an attempt to measure the level of radon present in water collect from rivers and lakes. Data gathered from this study provides important information about radiation levels in water at selected sites, because radon gas is the largest contributor to natural radioactive radiation exposure to humans. Exposure to radon gas can cause lung cancer. Liquid scintillation counting (LSC) has been applied to determine the activity concentration of radon (^{222}Rn) in water. Water samples were collected from, ex-mining lake in Perak, Sok River in Kelantan, Tembeling River in Pahang. Water samples were prepared in polyethylene bottles mixed with liquid scintillator and stored for 3 weeks to allow ^{222}Rn and its progeny to reach the equilibrium, and the activity concentrations ranged from 0.24-1.27 Bq/L, and 0.029 – 0.155 Bq/L for radon and radium respectively.

ABSTRAK

Banyak kajian telah dijalankan di seluruh dunia bagi mengukur radon di dalam air terutamanya air minuman kerana ia boleh menyebabkan masalah kepada kesihatan manusia. Kajian ini merupakan percubaan untuk mengukur tahap radon di dalam air yang diambil daripada sungai dan tasik. Data yang dikumpulkan daripada kajian ini memberkani maklumat penting tentang tahap radiasi di dalam air di kawasan yang dipilih, kerana gas radon adalah penyumbang terbesar kepada pendedahan radiasi radioaktif semula jadi kepada manusia. Pendedahan kepada gas radon boleh menyebabkan kanser paru-paru. Pembilang scintillation Cecair (LSC) telah digunakan untuk menentukan kepekatan aktiviti radon (^{222}Rn) di dalam air. Sampel air dari bekas lombong tasik di Perak, Sungai Sok di Kelantan, Sungai Tembeling di Pahang. Sampel air telah disediakan dalam botol polietelena yang bercampur dengan cecair sintilator dan disimpan selama 3 minggu untuk membenarkan ^{222}Rn dan anak-anak untuk mencapai keseimbangan, dan kepekatan aktiviti adalah dari 0.24-1.27 Bq/L dan 0,029 - 0,155 Bq/ untuk radon dan radium.

Keywords: Lake, LSC, radiation level, radon, and river

INTRODUCTION

A number of researchers have done studies on radon concentration in water around the world. The main issue was the monitoring radon gas in air inside dwellings. However, over the last decade more emphasis has been placed on measuring ^{222}Rn in water and soil, due to the fact that most of radon in air comes from water and soil (Durrani, 1999). However, in the recent years, there has been an increasing awareness on the importance of maintaining a good health among Malaysian especially with regard to drinking water, due to the importance of water for human life, for its quality must be maintained (Talha, 2003). For this reason, this study was focusing on determining the radon activity concentration in water in order to make sure that it has a low level of radioactivity. Even though radon is the immediate daughter of ^{226}Ra, and radon's measurements is significant for determining the amount of ^{226}Ra in water as well. Radium is a common radionuclide in the environment and it is the parent of radon. ^{226}Ra form is the most deadly radionuclides because it produces alpha radiation and has very long a half life (1,600 y) (Oyvind S. Bruland, 2008). Radon is a colorless, odorless and radioactive gas, produced from uranium decay series that present everywhere in Earth. Radon is an alpha emitter that decays into a chain of progenies of gamma emitters and alpha emitters, which means that radon atoms in the air can decay to produce other atoms. When the resulting atoms, called radon progeny, are formed, they can attach themselves to a tiny dust particle in indoor air. These dust particles can easily enter to the breathing system and increase the chance of getting lung cancer over long period of time. It has been estimated in the

United State of America that radon is the second cause of lung cancer after smoking (BEIR VI report, 1999). However, the presence of ^{222}Rn in water can pose a health hazard to humans and environment because ^{222}Rn is dissolved in water that means radon can be transferred to the physical and biological materials such as, sediments, vegetation and fish. Therefore, different types of water have been chosen in this study to estimate the radon activity and the health risk. The aim of this study is to determine the radon and radium activity concentrations in water using liquid scintillation counter because no self absorption, high counting efficiency, possible to count two isotopes at the same time and sample preparation and counting procedure are highly developed.

EXPERMENTAL

In this study, three locations were chosen to represent two types of surface water which are, river, and ex-mining lakes. Water samples were collected from the mid-depths to get reprehensive sample. However, during the sampling, many parameters have been measured to check the quality of the water using (HYDROLAB, model DS5, USA), and the sampling points were determined by using a global positioning system (GPS). Then, samples were transferred into plastic containers carefully because radon is a gas and it can be escape from water to air.

In the laboratory, the pH of the water samples was adjust into 2 using nitric acid, to avoid the collection of organic materials and changes in the state of the ions present in the samples (F.K. Gorur, *et al*, 2011), and to avoid loss of radionuclides fractions by adsorption on the wall of the containers, and reduce biological growth (Jobbagy *et al*., 2011, Jobbagy *et al*., 2010, and S. Marques, *et al*., 2008), and the water samples may be preserved for several months (S. Marques, *et al*., 2008), then the samples were divided to two portions (filtered and unfiltered water), to determine dissolve and suspended radon in water. For the filtration, cellulose membrane filter 0.45 µm was used because it seems the best filter to remove the suspended materials in the water.

Sample preparation for LSC is a little bit complex. For determine ^{222}Rn activity. The radon solved in water is extracted using toluene-based liquid scintillator cocktail. 1 L toluene, 4 g PPO (2-5-diphenyloxyole) and 0.4 g POPOP (1,4-bis-2(5-phenyloxaxoly1)-benzene), mix everything together for 24 hours. After that, 1000 mL of water sample (filtered and unfiltered) has transferred into 1 L polyethylene bottle with known actual volume. 100 mL liquid scintillator was added to the polyethylene bottle and the bottle is kept for 3 weeks to allow the equilibrium between ^{222}Rn and its progeny to reach. For the standard, 2 mL of ^{226}Ra is diluted up to 1000 mL with distilled water and 100 mL of liquid scintillator was added and the bottle was kept for 3 weeks.

Table 1: Sampling Points

Code	Location	Longitude	Latitude
KKBM	Kelantan	05° 23.200’	102° 17.063’
KKPD	Kelantan	05° 13.856’	102° 18.795’
KKDB	Kelantan	05° 22.806’	102° 16.277’
TNTRK	Taman Negara	04° 31.121’	102° 30.497’
TNA	Taman Negara	04° 31.287’	102° 30.984’
TNB	Taman Negara	04° 23.043’	102° 24.182’
KGTA	Kg. Gajah	04° 22.500’	101° 03.200’
KGAP1	Kg. Gajah	04° 27.170’	101° 04.090’
KGK3	Kg. Gajah	04° 23.730’	101° 03.960’

RESULTS AND DISSUCSION

Table 2 shows that the basic parameters for Water Quality Index (WQI). First parameter is the temperature, which was high in Kg. Gajah comparing with Taman Negara and Kelantan due to the sampling time because temperature can be change during the day. For the pH, it was ranged in all locations from 6.1-7.5, which could be considered normal for water because pH values between 6.5 and 8.5 usually indicate good water quality. The pH of an aquatic ecosystem is important because it is closely linked to biological productivity (UNEP, 2006). The level of NH_4 in all locations are in the range of 5.9-12.15 µg/L. Nitrogen occurs in water in a diversity of inorganic and organic forms and the concentration of each form is primarily mediated by biological activity. Nitrogen-fixation, performed by cyano-bacteria (blue-green algae) and certain bacteria, converts dissolved molecular N_2 to ammonium (NH_4^+). Aerobic bacteria convert NH_4^+ to nitrate (NO_3^-) and nitrite (NO_2^-) through nitrification, and anaerobic and facultative bacteria convert NO_3^- and NO_2^- to N_2 gas through de-nitrification. Primary producers assimilate inorganic N as NH_4^+ and NO_3^-, and organic N is returned to the inorganic nutrient pool through bacterial decomposition and excretion of NH_4^+ and amino acids by living organisms. Nitrogen in water is usually measured as total nitrogen, ammonium, nitrate, nitrite, or as a combination of these parameters to estimate inorganic or organic nitrogen concentrations (UNEP, 2006). The last parameter is turbidity, it was normal in Kg. Gajah but high in Kelantan and Taman Negara and the range was 3-22. Turbidity refers to water clarity. The greater the amount of suspended solids in the water, the murkier it appears, and the higher the measured turbidity. Turbidity was high in Taman Negara and Kelantan due to the soil erosion, this is let the water mix with high levels of soil making the color of the water brown.

Table 2: Basic Parameters for WQI

Code	Temperature C^o	pH	NH_4 (mg/L)	Turbidity (NTU)
KKBM	28.00	7.40	6.30	21.00
KKPD	28.00	7.30	5.90	11.00
KKDB	27.00	7.50	6.24	4.00
TNTRK	26.00	6.83	8.38	21.20
TNA	25.80	6.83	8.10	47.00
TNB	25.30	7.12	7.52	22.00
KGTA	29.00	7.04	9.73	5.00
KGAP1	30.00	6.88	12.15	5.70
KGK3	28.00	6.18	7.74	3.00

Table 3: Radon and radium activity concentrations (Bq/L)

Code	^{222}Rn Unfiltered	^{222}Rn Filtered	^{226}Ra Unfiltered	^{226}Ra Filtered
KKBM	0.69 ± 0.08	0.56 ± 0.08	0.084 ± 0.004	0.067 ± 0.005
KKPD	1.04 ± 0.08	0.74 ± 0.08	0.125 ± 0.007	0.090 ± 0.004
KKDB	0.40 ± 0.08	0.37 ± 0.08	0.049 ± 0.008	0.044 ± 0.007
TNTRK	0.50 ± 0.07	0.32 ± 0.06	0.063 ± 0.006	0.040 ± 0.009
TNA	0.45 ± 0.07	0.74 ± 0.07	0.090 ± 0.008	0.054 ± 0.005
TNB	1.27 ± 0.07	0.64 ± 0.07	0.153 ± 0.007	0.077 ± 0.006
KGTA	0.40 ± 0.05	0.31 ± 0.05	0.063 ± 0.006	0.036 ±0.005
KGAP1	0.52 ± 0.07	0.30 ± 0.07	0.052 ± 0.008	0.029 ± 0.009
KGK3	0.43 ± 0.07	0.23 ± 0.07	0.155 ± 0.009	0.089 ± 0.010

Table 3 is shown the activities of radon which were in the range from 0.24-1.27 Bq/L, and 0.029 – 0.155 Bq/L for radon and radium respectively. Although there is a variation between radon activity concentrations in the water samples from Kg. Gajah, Kelantan and Taman Negara, Taman Negara has shown the highest. However, Table 3 shows that radon and radium activity concentrations in unfiltered water is slightly higher than in filtered water due to remove the suspended solids from the water during the filtration which contain small amount of radionuclides. On the other hand, radon activities are different due to various parameters, including the physicochemical states of the radionuclides, characteristic of the water and depth of the water. However, radon and radium activity concentrations in this study did not exceed the maximum level of contamination of 11.1 Bq/L and 0.18 Bq/L respectively that proposed by U.S Environment Protection Agency (1999).

The physical and chemical properties of the water might be having an effect on the radioactivity levels in groundwater and surface water (Shuktomova, *et al*., 2011). Some parameters such as pH, temperature, salinity might affect mobilization of radionuclides (USGS, 2006). Positive correlation between water mineralization and activity of ^{40}K, and ^{226}Ra was also observed by Chau, *et al*., (2009). Therefore, the relation among concentrations of radon in water and water quality parameters such as pH, temperature, NH_3, and turbidity were investigated.

Table 4: Pearson correlation coefficient, between radon activity concentration in Kelantan samples and Water Quality Index (WQI)

	^{222}Rn (U)	^{222}Rn (F)	Temp.	pH	NH_3
Temp.	**0.838**	**0.874**			
pH	**-0.999**	**-0.999**	**-0.866**		
NH_3	**-0.820**	**-0.778**	-0.375	**0.788**	
Tur.	0.360	0.424	**0.811**	-0.410	0.239

Table 4 shows strong correlation between radon activity concentration in unfiltered water from Kelantan river with temperature (r^2 = 0.838), and strong inversely correlation between radon activity concentration in unfiltered water with pH (r^2 = -0.999), and NH_3 (r^2 = -0.820). Good correlation found with turbidity (r^2 = 0.360). There is also strong correlation between radon activity concentration in filtered water with temperature (r^2 = 0.874), and strong inversely correlation between radon activity concentration in filtered water with pH (r^2 = -0.999), and NH_3 (r^2 = -0.778). Good correlation found with turbidity (r^2 = 0.424).

Table 5: Pearson correlation coefficient, between radon activity concentration in Taman Negara samples and Water Quality Index (WQI)

	^{222}Rn (U)	^{222}Rn (F)	Temp.	pH	NH_3
Temp.	**-0.944**	**-0.544**			
pH	**0.999**	0.261	**-0.952**		
NH_3	**-0.929**	**-0.580**	**0.999**	**-0.938**	
Tur.	-0.523	**0.704**	0.214	**-0.502**	0.171

Table 5 shows strong correlation between radon activity concentration in unfiltered water from Taman Negara (Tembeling River) with pH (r^2 = 0.999), and strong inversely correlation between radon activity concentration in unfiltered water with temperature (r^2 = -0.944), NH_3 (r^2 = -0.929), and turbidity (r^2 = -0.523). Similarly, there is strong correlation between radon activity concentration in filtered water with turbidity (r^2 = 0.704), and Strong inversely correlation between radon activity concentration in filtered water with temperature (r^2 = -0.544), NH_3 (r^2 = -0.580). Weak correlation was found with pH (r^2 = 0.261).

Table 6: Pearson correlation coefficient, between radon activity concentration in Kg. Gajah samples and Water Quality Index (WQI)

	^{222}Rn (U)	^{222}Rn (F)	Temp.	pH	NH_3
Temp.	**0.721**	**0.803**			
pH	0.105	**0.998**	**0.765**		
NH_3	**0.758**	**0.768**	**0.998**	**0.728**	
Tur.	**0.509**	**0.933**	**0.963**	**0.910**	**0.947**

Table 6 shows strong correlation between radon activity concentration in unfiltered water from Kg Gajah (former tin mining lakes) with temperature (r^2 = 0.721), NH_3 (r^2 = 0.758), and turbidity (r^2 = 0.509). Very weak correlation was noted with pH (r^2 = 0.105). Table 6 also shows strong correlation between radon activity concentration in filtered water with temperature (r^2 = 0.803), pH (r^2 = 0.998), NH_3 (r^2 = 0.768), and turbidity (r^2 = 0.933).

There is a positive correlation between radium concentration and pH of water noted by Lasheen *et al*. (2007), and Baeza *et al*., 1995. High correlation between low pH value and high radium concentration has also been found in study done by (Lauria *et al*., 2002), and low pH values might be from the combination of acidic precipitation, high evaporation rate, and lack of carbonate buffers in soil (Almeida, *et al*., 2004). On the other hand, there is no significant correlation between radon activity concentration with pH values (Choubey, *et al*, 2000 and Badhan, *et al*., 2010). According to Choubey, *et al*, (2000) had shown that no significant correlation was observed between radon concentration and the water temperature, but Oliveira *et al*., (2001) showed that in groundwater, where radon is dissolved in water, and it might be controlled by physical variables, like temperature. Moreover, different correlations have been found due to the behavior of radionuclides in water is very hard to estimate, and every river and lake has its own characteristic which could be different from place to place (Michael, *et al*., 2007). More over the correlation between radon activity concentration and water quality parameters is depends on geographical locations (Paulus, *et al*., 1998), and even no correlation was noted between radon activity concentration and physicochemical parameters in study done by I. Salih, *et al*., (2011).

Table 7: Radon concentration in different types of water

Water type	Radon Activity (Bq/l)	References	Country
Groundwater	0.89-35.44	Alabdula'ali (1999)	Saudi Arabia
Groundwater	24-40	Hopke *et al.* (2000)	USA
Sea Water	0.95-2.67	Ghose et al. (2000)	Bangladesh
Tap water	2.5-4.7	Al-Bataina et al. (1997)	Jordan
River water	0.08-1.17	Al-Masri (1999)	England
River water	0.32-1.27	This work	Malaysia
Ex-mining Lakes	0.24-0.52	This work	Malaysia

CONCLUSION

We found that the radon and radium levels in water from selected sites are within acceptable values, but the activity concentration of radon is different at studied sites due to different origins, depths and pathways of the out flowing water. The correlation between radon activity concentration and physicochemical parameters could be different from place to place and depends on geographical locations.

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