

DETERMINATION OF RADON ACTIVITY CONCENTRATION IN HOT SPRING AND SURFACE WATER USING GAMMA SPECTROMETRY TECHNIQUE

(Penentuan Kepekatan Aktiviti Radon Di dalam Air Panas dan Air Permukaan Dengan Menggunakan Teknik Spektrometri Gama)

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Abstract

Naturally occurring radionuclides in water such as, ^{222}Rn which emit gamma radiation through its decaying process could reach to human. Water samples were chosen to present ground and surface water. The groundwater samples were collected from, Perak, Selangor, Kelantan and Sembilan. The surface water samples were collected from Perak, Kelantan, and Pahang. In this study, the surface doses rate measurements were done in-situ using LUDLUM rate meter, and the radioactivity concentration levels were done by counting the water samples using gamma spectrometer with HPGe detector. The activities are ranged from (0.29-1.41 Bq/l).

Keywords: Radon, Gamma spectrometry, Surface water, Groundwater

Abstrak

Yang berlaku secara semulajadi radionuklid di dalam air seperti, ^{222}Rn yang mengeluarkan radiasi gamma melalui proses mereput dapat sampai kepada manusia. Sampel air telah dipilih untuk membentangkan air bawah tanah dan permukaan. Sampel air bawah tanah telah dikutip dari, Perak, Selangor, Kelantan dan Sembilan. Sampel air permukaan telah dikumpulkan dari Perak, Kelantan, dan Pahang. Dalam kajian ini, pengukuran kadar dos permukaan telah dijalankan di-situ dengan menggunakan meter kadar LUDLUM, dan tahap kepekatan radioaktif telah dilakukan dengan mengira sampel air yang menggunakan spektrometer gamma dengan pengesan HPGe. Aktiviti-aktiviti adalah dari (0,29-1,41 Bq / l).

Kata kunci: Radon, Spektrometri Gamma, Air Permukaan, Air Tanah

Introduction

In the last two decades, there has been a great deal of awareness about the health risks from exposure to radon radioactive gas and its decay products. Large-scale radon surveys have been carried out in Europe and in the United States, more than that have been conducted in any country in the World. However, the study of radon has been increasingly growing throughout the world [1]. The main issue was the monitoring of radon gas in air inside dwellings. Over the last century more emphasis has been placed on measuring ^{222}Rn levels in soils. This is due to the fact that most of the radon in dwellings comes from the underground soil [2]. Nowadays, there are many problems concern the people not only in Malaysia but around the world. One of the problems is radionuclides in water. Radionuclides are isotopes which have the ability to produce radiation. Radiation is produced when elements with an unstable atomic structure undergo decay into another element. Radiation is all around us, every minute of every day. Some radiation is essential to life, such as heat and light from the sun. As used in medicine, radiation helps us to diagnose and treat diseases and to save lives [3]. But other radiation such as radon isotopes, and its decay products of uranium and thorium, can be quite harmful to human beings. The radionuclides in water are member of three natural radioactive series, which are the uranium series, thorium series, and the actinium series [4].

The isotopes in the uranium decay series that may pose a health risk because of their presence in water are ^{226}Ra and ^{222}Rn [5]. The human population typically is exposed to radiation from water, both by ingestion and inhalation. These radiations arise from the uranium and thorium decay series. This paper is focused on radon in water which is a radioactive inert gas produced by the alpha-decay of ^{226}Ra in the ^{238}U series with a half-life of 3.84 days. Since ^{222}Rn occurs naturally in soil and rocks, it is virtually omnipresent on earth. It accounts for more than 50% of the total dose from all sources of ionizing radiation absorbed by the population [6]. Since ^{222}Rn readily dissolves in water under pressure, groundwater is another source of radon. High concentration of ^{222}Rn may cause concern about its effects on health. Either drinking groundwater or breathing can give rise to exposure of humans to its radiation and may result in cancer deaths [7]. The aim of this paper is measuring ^{222}Rn in hot spring and surface water using gamma spectrometry technique.

Experimental

Samples of hot spring water were collected from Perak, Selangor, Kelantan, and Nigeria Sembilan. Because of springs of hot mineral water is very attractive to the external and internal tourism industries, and many people are going there as patients, visitors, and tourists to spend the holidays, and even consumption from the people who live there. However, hot spring water contains many useful elements, such as, calcium, iron, magnesium...etc. also contains natural radioactive isotopes, such as, uranium, radium and radon's progeny (^{214}Bi and ^{214}Pb). Visitors to the bath houses of these springs are unaware that they can breathe ^{222}Rn emanated from the surrounding hot spring water or be exposed to hazardous radiation emitted from radionuclides in the hot mineral water [8]. However, samples of natural water were collected from Kelantan, Perak, and Pahang.

Radiation doses were measured (in-situ) at the selected points at the surface and 1 meter height. The measurements were conducted using LUDLUM rate meter. During collect the water samples, many parameters have been measured to check the quality of the water using (HYDROLAB, model DS5, USA). 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 controlled using nitric acid to stabilize the water, then the samples were divided to two portions (filtered and unfiltered water), to determine dissolve and suspended radon in water. For the filtration, membrane filter 0.45 μm was used because it seems the best filter to remove the suspended materials in the water. Sample preparation was done by filling the water sample (filtered and unfiltered) to the Marinelli beaker 500mL until it is full. The beaker was weighted and sealed using silicon glue [9] to avoid the leakage of radon from water. The beakers were kept for a month, to allow the equilibrium between ^{222}Rn and its progeny to reach. When radon is measured in water sample, usually waits until equilibrium is reached where the disintegration rate of the radionuclides is the same and this depends on the half-life of the radionuclide. However, in this study, equilibrium happens after 6-7 radon decays. Therefore, samples were kept for a month because of the half-life of radon is almost 4 days.

Results and Discussion

The measurement was done by gamma spectrometer with a high-purity germanium detector (HPGe) of high resolution. This advanced spectrometer consists of an HPGe detector of resolution 1.84 keV at 1.33 MeV. The samples were counted for 12 hours (43200s) which found to be enough to get good statistical peaks. However, the calibration of gamma spectrometry was done using standard contained UO_3 to cover energy from 63 until 1001 keV mixed with KCl to present the energy 1460 for ^{40}K , and it is not necessary to have large number of gamma lines above 400 keV to obtain good calibration; two points are enough to have good fit with low uncertainty.

Table 1: Sampling points and Survey meter reading

Location	Latitude	Longitude	Surface Dose	1 Meter Dose
KST	05° 21.253'	102° 14.532'	0.122	0.137
KSM	05° 06.042'	102° 20.959'	0.150	0.135
KKKB	02° 55.814'	101° 51.355'	0.131	0.121
TNKSS	04° 31.121'	102° 30.497'	0.134	0.124
TNKK	04° 31.222'	102° 28.624'	0.230	0.125
TNSTB	04° 26.926'	102° 28.996'	0.080	0.109
KGLL	04° 22.500'	101° 03.200'	0.096	0.088
KGSR	04° 27.170'	101° 04.090'	0.131	0.114
KGSK2L	04° 23.730'	101° 03.960'	0.132	0.122
HSP	02° 37.891'	102° 03.321'	0.232	0.164
HSSSHL1	03° 05.448'	101° 47.670'	0.124	0.103
HSSSHL2	03° 08.353'	101° 50.170'	0.124	0.100
HSSK	03° 59.688'	101° 23.612'	0.138	0.119
HSTB	05° 16.590'	102° 02.595'	NONE	NONE

Table 2: Water Quality Parameters

Code	Temperature (°C)	DO (mg/L)	Conductivity ($\mu\text{S}\cdot\text{cm}^{-1}$)	pH	Salinity (mg/L)	TDS (mg/L)	Turbidity (NTU)
HSP	39.8	5.32	0.2440	7.80	0.12	0.17	8.9
HSSSHL1	42.0	7.60	0.2651	8.67	0.13	0.17	3.3
HSSSHL2	50.0	2.40	0.0509	8.82	0.01	0.03	3.4
HSSK	90.0	2.33	0.1957	8.93	0.09	0.13	0.0
HSTB	N.R	7.40	0.2922	8.93	0.14	0.12	0.0
KST	30.0	3.00	0.0435	7.20	0.01	0.03	802.0
KSM	28.0	2.60	0.0452	7.40	0.01	0.03	842.0
KKKB	25.0	9.20	0.0290	7.10	0.00	0.02	125.0
TNKSS	25.7	2.96	0.0599	6.78	0.02	0.04	21.2
TNKK	27.3	2.12	0.0430	7.16	0.01	0.03	13.9
TNSTB	27.6	1.90	0.0301	7.06	0.00	0.02	33.1
KGLL	28.0	4.35	0.1045	7.04	0.03	0.07	0.0
KGSR	25.3	4.73	0.0233	6.20	0.00	0.02	9.5
KGSK2L	27.6	1.20	0.0589	6.18	0.02	0.09	0.0

Previous studies used the energy 609 keV to obtained ^{222}Rn activity concentration due to no inference with other radionuclides in this energy and the contribution of the intensity to the total intensity of gamma-radiation is almost 50%. However, for other peaks for ^{214}Bi such as, 934 keV, 1120 keV, and 1764 keV lines impose weak intensities. On the other hands, germanium detectors have lower efficiency in high energies which can cause non-detectable counting rates and they give activities with significantly uncertainties. For those difficulties, the calculation of ^{222}Rn activities using 934, 1120, and 1764 keV might result in poor precision and accuracy, therefore, ^{222}Rn activity was calculated from the energy 609 keV only. The peak 352 keV for ^{214}Pb was not considered due to the interference between ^{211}Bi and ^{214}Pb in the same energy.

However, in this study, there is strong inversely correlation between radon activity concentration in hot springs water and conductivity, TDS, and salinity for both unfiltered and filtered samples, and there is inversely correlation between radon activity concentrations in filtered samples only and the temperature. The rest of the parameters, there is very weak correlation with radon activity concentration in hot springs samples which can be ignored in some

cases. Moreover, the Table showed that conductivity has significant correlation with many parameters, which are, temperature, dissolved oxygen, salinity, TDS, and turbidity. On the other hand, TDS has significant inversely correlation with temperature, dissolved oxygen, turbidity and salinity. Finally salinity has significant inversely correlation with turbidity, temperature, and dissolved oxygen.

Salinity is an indicator of the concentration of the amount of dissolved salts, including calcium, magnesium, sodium and potassium) [10]. For the conductivity which can be defined as “the ability of water to carry an electric current” [10], which is proportional to the concentration of ions [10]. In this study, hot springs samples were more salinity than other types of water including rivers and lakes therefore, the correlation between conductivity and salinity in hot springs could be considered normal because specific conductivity is higher in saline systems than in non-saline systems [10]. On the other hand, study was done by [11] showed that radon activity concentration was unrelated to salinity. However, in low salinity environments radium is strongly adsorbed on surfaces, where as radon is dissolved and migrates with the fluid. In groundwater, there is correlation between salinity and radium because of competition between radium and cations for adsorption sites on solids [12], and increases in salinity tend to increase ^{226}Ra activity concentration in groundwater; because of ion-exchange mechanisms a greater partition of the exchangeable radium in pore water than on surface exchanges sites at higher salinities [11].

However, there is strong inversely correlation between radon activity concentration in surface water and salinity only for both unfiltered and filtered samples, for the rest of the parameters, there is no significant correlation with radon activity concentration in surface water. However, conductivity has significant correlation with DO, TDS, and salinity. Salinity has significant inversely correlation with DO.

Table 3: Radon activity concentrations

Sample No.	Location	State	Type of water	^{222}Rn (Bq/L) Unfiltered (U)	^{222}Rn (Bq/L) Filtered (F)
1,2	KST	Kelantan	River	0.99 ± 0.07	0.51 ± 0.05
3,4	KSM	Kelantan	River	1.11 ± 0.07	0.99 ± 0.07
5,6	KKKB	Kelantan	River	1.41 ± 0.08	0.76 ± 0.06
7,8	TNKSS	Pahang	River	0.35 ± 0.04	0.32 ± 0.04
9,10	TNKK	Pahang	River	1.05 ± 0.07	0.92 ± 0.06
11,12	TNSTB	Pahang	River	0.47 ± 0.05	0.38 ± 0.04
13,14	KGLL	Perak	Lake	0.39 ± 0.04	0.29 ± 0.04
15,16	KGSR	Perak	Lake	1.27 ± 0.08	0.66 ± 0.06
17,18	KGSK2L	Perak	Lake	0.40 ± 0.04	0.30 ± 0.04
19,20	HSP	Nigeria Sembilan	Hot Spring	1.12 ± 0.06	1.08 ± 0.05
21,22	HSSSHL1	Selangor	Hot Spring	0.50 ± 0.05	0.41 ± 0.03
23,24	HSSSHL2	Selangor	Hot Spring	0.61 ± 0.05	0.51 ± 0.05
25,26	HSSK	Perak	Hot Spring	0.82 ± 0.08	0.53 ± 0.07
27,28	HSTB	Kelantan	Hot Spring	0.63 ± 0.06	0.50 ± 0.05

Table 4: Pearson correlation coefficient, between radon activity concentration in hot spring samples and water quality parameters

	Rn (U)	Rn (F)	Tem.	DO	Cond.	pH	Sal.	TDS
Temp.	-0.14	-0.74	1					
DO	-0.08	-0.47		1				
Cond.	-0.71	-0.72	-0.79	-0.84	1			
pH	-0.14	-0.05	0.15	-0.15	0.02	1		
Sal.	-0.73	-0.78	-0.58	-0.72	0.98	0.01	1	
TDS	-0.61	-0.63	-0.75	-0.77	0.84	-0.02	0.83	1
Tur.	-0.17	-0.26	-0.71	-0.05	-0.77	-0.01	-0.72	-0.78

Table 5: Pearson correlation coefficient, between radon activity concentration in surface samples and water quality parameters

	Rn (U)	Rn (F)	Tem.	DO	Cond.	pH	Sal.	TDS
Temp.	-0.07	-0.04	1					
DO	0.31	-0.14	-0.10	1				
Cond.	-0.15	-0.16	0.06	-0.66	1			
pH	0.04	0.02	-0.11	-0.13	-0.22	1		
Sal.	-0.74	-0.59	0.04	-0.65	0.76	-0.01	1	
TDS	-0.18	-0.21	0.04	-0.25	0.55	-0.09	0.42	1
Tur.	0.12	0.16	0.05	-0.15	-0.13	0.04	-0.23	-0.22

However, high radon content in hot spring can be due to the leakage of the gas, and alpha-recoil transfer of the radon nuclide, from the uranium [13 and 14], and due to discharge from granite rocks [15]. As the literature indicated, natural radionuclide concentrations in environmental samples can be very different due to geographical, geological factors, the time of sampling as well as on the location, and temperature this may cause different results in different studies [16 and 17]. Radon activity concentration in hot spring samples did not exceed the limit for radon in water, which is 11 Bq/l that proposed by USEPA. However, the results were comparable to the previous study done by [16 and 18].

In surface water radon is not a major concern, due to there is no groundwater contribution to the river water or aeration of river water can decrease radon concentrations very fast because of escape radon from water into atmosphere [19]. On the other hand, once the radionuclides are entered in water, their behavior is very hard to estimate, and every river, and lake it has its own characteristic this might be different from place to place. Radon content in surface water might be affected by several factors, such as the geology of the area, and bottom sediments [18 and 20]. Most of the radionuclides that transport to the surface water can be attach to sediments on the bottom. The process of interaction of dissolved radionuclides in water with suspended solids has been investigated by many researchers. However, the bottom sediments in surfaces water (rivers, and lakes) could be considered as sinks, where as the radionuclides interact with suspended materials and migrate to the sediments [21]. Low ²²²Rn activity concentrations were found in all locations and did not exceed the maximum level for ²²²Rn in water this was proposed by USEPA 1999, and activity concentrations of ²²²Rn in filtered water samples are slightly lower than unfiltered water samples, due to removal of non-dissolve suspended solid from water, which contains a small amount of radionuclides. However, the results obtained from rivers in this study were compared with the reported values from other countries around the world and it was observed that the measured activity concentrations of ²²²Rn water were lower and almost similar than many literatures, [18 and 20].

Conclusion

We have found that the radon levels in hot spring water and surface water are range from (0.29-1.41 Bq/l) which is within acceptable values, but the concentration of radon is different at studied sites. The differences are possibly due to different origins, depths and pathways of the out flowing water. Surface radiation dose measured at 1m above the surface are in range between 0.088 – 0.137 μ Sv/hr for all the locations, and for the surface are range between 0.096-

0.232 $\mu\text{Sv/hr}$. The global range for surface radiation dose is 0.079 – 0.13 $\mu\text{Sv/hr}$. Therefore, it can be concluded that the surface radiation dose are more than the global range at selected sites.

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