

DETERMINATION OF SUPPORTED ^{222}Rn ACTIVITY CONCENTRATION IN GROUND WATER FROM CAMERON HIGHLANDS AREA USING GAMMA SPECTROMETRY

(Penentuan Kepekatan Aktiviti ^{222}Rn Sokongan di dalam Air Tanah dari Kawasan
Cameron Highlands Menggunakan Spektrometri Gama)

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Abstract

Humans may be exposed to the emission of energetic alpha particles from supported radon decaying process in ground water when it is consumed. Cameron Highland's water supply was chosen as study area since it coming from hilly area of Gunung Pass, Gunung Brinchang and Gunung Cantik whose geological formations made from mainly igneous rocks (intrusive rocks, mainly granite with minor granodiorite) that flows into Sungai Terla, Sungai Ulu Bertam and Sungai Jasar. Determination of supported radon in this ground water was carried out using eighteen ground water samples collected from Cameron Highlands. Water samples (filtered and unfiltered) were poured into Marinelli beaker and closed tightly. These samples were kept at least for three weeks to allow the equilibrium between ^{226}Ra and ^{222}Rn . The measurement was accomplished by measuring water samples using low background gamma spectrometry (ORTEC) using a hyper pure coaxial germanium detector linked to a multi-channel digital analyzer system. ^{222}Rn was measured using its daughter's photo peaks: ^{214}Pb (352 keV) and ^{214}Bi (609 keV). Results of the study show the supported ^{222}Rn activity concentration ranges from 0.19 to 0.66 Bq/L for unfiltered, filtered and untreated samples, 0.32 to 0.39 Bq/L of ^{222}Rn for treated water at water treatment plant and 0.26 to 0.55 Bq/L of ^{222}Rn for domestic water. These values are lower than the activity concentration of radon in drinking water as proposed by USEPA (11 Bq/L).

Keywords: Ground water, supported radon, natural radioactivity, gamma spectrometry

Abstrak

Manusia mungkin terdedah kepada pancaran zarah alfa bertenaga tinggi dari proses penyepaian 'supported radon' dalam air ini apabila digunakan. Sumber air Cameron Highlands dipilih sebagai tempat kajian kerana air tanah mengalir daripada Gunung Pass, Gunung Brinchang dan Gunung Cantik di mana keadaan geologinya adalah kebanyakannya daripada batuan igneous (batuan intrusive (batuan yang terhasil daripada pengkristalan magma di dalam bumi), granit dengan sedikit granodiorite (batuan kecil mengandungi kandungan silika yang tinggi) yang mengalir ke Sungai Terla, Ulu Sungai Bertam dan Sungai Jasar. Pengukuran radon sokongan dalam air tanah dijalankan menggunakan lapan belas sampel air tanah dari Cameron Highlands. Sampel air dimasukkan ke dalam bikar Marinelli dan ditutup dengan ketat. Sampel ini disimpan sekurang-kurangnya selama tiga minggu untuk membenarkan keseimbangan di antara ^{226}Ra dan ^{222}Rn . Pengukuran terlaksana dengan mengukur sampel air menggunakan spektrometri gama berlatar belakang rendah (ORTEC) menggunakan pengesan *coaxial* germanium berketulen tinggi bersambungan kepada sistem digital analisis pelbagai-haluan. ^{222}Rn diukur pada its daughter's photopeaks: ^{214}Pb (352 keV) dan ^{214}Bi (609 keV). Keputusan daripada kajian menunjukkan bahawa kepekatan aktiviti ^{222}Rn sokongan dalam lingkungan dari 0.19 sehingga 0.66 Bq/L bagi sampel air tidak bertapis, bertapis dan air tidak dirawat, 0.32 sehingga 0.39 Bq/L ^{222}Rn bagi air dirawat di loji rawatan air dan 0.26 sehingga 0.55 Bq/L ^{222}Rn bagi lokasi pengagihan air dirawat. Nilai kepekatan aktiviti radon sokongan lebih rendah daripada kepekatan aktiviti radon dalam air minuman yang dianjurkan oleh USEPA (11 Bq/L).

Kata kunci: Air bawah tanah, radon sokongan, radioaktiviti tabii, spektrometri gama

Introduction

In the last decades, radon and its decay product in water has become a great deal of concern to the public since it can pose health hazard to humans and environment [1, 2, 3]. Radon is a naturally occurring radioactive inert gas, partially soluble in water, colorless, odorless with a half-life of 3.84 days which a progeny of uranium and thorium decay series [1, 4, 5, 6]. Radon is an alpha emitter that decays into a chain of progenies of alpha emitters such as ^{218}Po (6.0 MeV), ^{216}Po (6.7 MeV), ^{214}Po (7.7 MeV) and ^{212}Po (8.8 MeV) [1, 4]. It is produced continuously by the alpha decay of radium with some atoms escaping to the surrounding fluid phase, such as ground water when the water moves through granitic rocks containing natural uranium and thorium that released radon to the water [7]. Thus, this gas found in various concentrations in ground water [8].

Cameron Highlands used ground water as its own fully water supply. It was chosen as study area since the ground water is coming from Gunung Pass, Gunung Brinchang and Gunung Duri whose geological formations mainly made from igneous rocks (intrusive rocks, mainly granite with minor granodiorite) that flows into its specific rivers of Sungai Terla, Sungai Habu and Sungai Jasar [9,10]. This ground water is sufficiently large volume to induce an appreciable spread in dissolved supported radon concentration [11]. The ground water was collected from source, river, diverted upstream to the treatment plant and distributed treated water for domestic purposes. The geology and local in nature of Cameron Highlands are ideal for such study since public water source are used for services and drinking purposes.

Normally, radon-in water activity concentrations are measured by means of liquid scintillation counting (LSC) as recommended by USEPA [1,12, 13]. However, alternative method such as gamma spectrometry has been used as alternative method to quantify radon-in-water [1, 12, 13, 14]. The quantification of ^{222}Rn -in-water after at least three weeks to allow the secular equilibrium between ^{226}Ra , ^{222}Rn and its progenies is known as supported ^{222}Rn [15]. The aim of the study is to determine supported ^{222}Rn activity concentration in untreated and treated ground water from Cameron Highlands area using gamma spectrometry technique.

Materials and Methods

Sampling Site and Samples

Ground water sampling location in Cameron Highlands, Malaysia is shown in Figure 1. The sampling was carried out on 25th until 27th of November 2011. Nine ground water samples were collected comprises of water source intake, treated water treatment plant and domestic water from houses in Cameron Highlands, Pahang. The exact sampling point was determined using global positioning system (GPS). Sampling points is shown in Table 1. From each sampling point, 10 liter samples were collected and kept in containers. The containers was cleaned with 6 M nitric acid (HNO_3) and rinsed with distilled water in order to remove any contaminants, prior to use.

As shown in Table 1, there were three types of samples collected which were untreated ground water from Sungai Terla and Kuala Terla water Treatment Plant (LAKT), treated water from LAKT and domestic water from houses. At intake, water samples taken at surface and at 0.5 m below surface. Aerated sample taken at surface and below surface water (0.5 m) at LAKT aeration pond and treated water sample was collected from water output of LAKT. The domestic water was collected from domestic pipes at houses. The altitude for intake sampling point was higher than LAKT results the untreated river water was transferred to the LAKT due to gravity. The treated water was distributed to KRA, TKP, KTS and TRA using buffer pumps since their altitude were higher than LAKT.

On the same sampling point, seven water quality parameters were measured in-situ using YSI multi probe water quality meter. The parameter measured were dissolved oxygen (DO), pH, turbidity, temperature, total dissolved solid (TDS), salinity and oxidation reduction potential (ORP). Total suspended solid (TSS) was determined in the laboratory [16].

Sample Preparation

In laboratory, the pH of the water samples were adjusted to below 2 using 6 M nitric acid in order to stabilize the water, to avoid the loss of radionuclides fractions via adsorption on the wall of the containers and to prevent any biological activities [1, 4]. Each water samples were divided into two portions; one portion represented for unfiltered samples and another portion was for filtered water samples. The filtered samples were filtered using cellulose membrane filters (0.45 μm , Whatman) on Buchner funnel attach to pump to filter away suspended solid and impurities from water samples. This will enable the suspended solids to be determined.

The filtered and unfiltered water samples were then filled into 600 ml Marinelli beaker to brim and closed tightly. The beaker was sealed using silicon glue, to avoid the leakage of radon gas [14]. The beakers were kept for at least three weeks to allow the secular equilibrium between ^{226}Ra , ^{222}Rn and its progenies (^{214}Pb and ^{214}Bi) and ready for measurement using gamma spectrometry.

Measurement of Supported ^{222}Rn

The samples were counted using low background gamma rays spectrometry (ORTEC) using a hyper pure coaxial germanium detector with resolution 1.84 keV, 25% relative efficiency at 1332 KeV ^{60}Co gamma ray linked to a multi-channel digital analyzer system. The efficiency calibration of gamma spectrometry was done using secondary standard made up by mixing known amount of uranium trioxide (UO_3) and KCl in the same container geometry as samples to cover energy from 63 keV to 1001 keV and 1560 keV for ^{40}K [17]. The standard and samples were counted for 8 hours (28800s). Figure 2 shows the efficiency of the gamma rays spectrometer used for calculation of individual radionuclides activity in water samples. Since, the energy of radionuclides interested is higher than 200 keV, Equation (A); Efficiency = $1.143 \times 10^{-6} E^{-0.67}$ were used to calculate the detection efficiency at energy of each radionuclides [18].

Since ^{222}Rn is an alpha emitter, the measurement of ^{222}Rn in water samples was based on ^{222}Rn progenies which were ^{214}Pb and ^{214}Bi with gamma energy peak at 352 keV and 609 keV respectively. This approach is possible for ^{222}Rn that is equilibrium with its progenies [15, 19]. The spectra obtained were analyzed using Gamma Vision-32 version 6.07 software provided by ORTEC®. After analyzing the peak in the spectra, the activity concentration of the individual radionuclide was calculated using Equation 1.

$$A_V = \frac{C_\gamma}{\epsilon_\gamma \times P_\gamma \times M_s} \quad (1)$$

where:

C_γ = Net peak area per counting time (count/sec)
 ϵ_γ = Detection efficiency at the specified energy
 P_γ = Transition probability of gamma radiation
 M_s = Volume of sample (L)

The activity concentrations for both progenies (^{214}Pb and ^{214}Bi) were then averaged to represent the supported ^{222}Rn activity concentration [19]. The gamma spectrometry method for determination was validated earlier and precision of 10 measurements was found to be 2.15% and the minimum detectable activity (MDA) for 609 keV was 0.12 Bq/L [15].

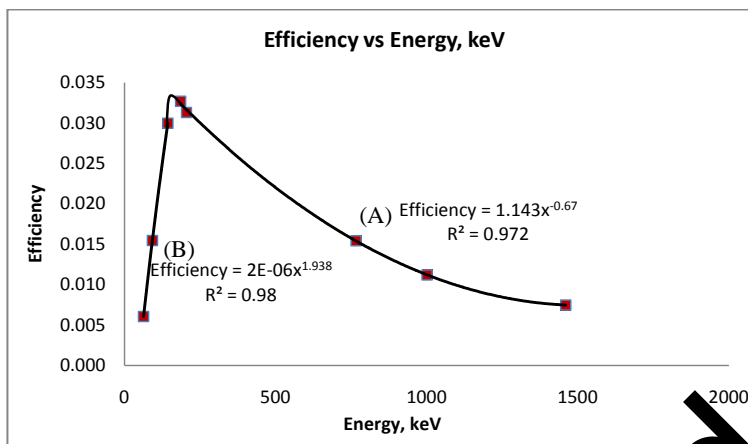


Figure 2: Efficiency Calibration using UO_3 and KCl by Gamma Spectrometry

Results and Discussion

Water Quality Parameter Measurement

Table 2 shows various water quality parameters measured for each sample together with NWQS individual value for Class I and Class IIA Water [20] for comparison purpose. High concentration of DO indicates good water quality [1] because it will reduce the activity of microbial reduction of nitrate to nitrite and sulfate to sulfide and reduced the concentration of ferrous iron [3]. However, DO do not affect ^{222}Rn activity concentration [15]. The pH of KTS is slightly acidic due to the origin of river water, tannin and leaves that released from forest floor [21]. Leaves decay and react with water, forming humic acid and released from forest to the river [21].

KTA has highest turbidity value since the sample was aerated untreated sample at aeration pond. Particulate matter that may be present from water source as a result from inadequate filtration or from re-suspension of sediment and the presence of inorganic particulate matter in some ground waters within the distribution system can cause turbidity [3]. The suspended solid was attributed to soil erosion due to opening up lands around the areas for agriculture and other activities [22]. The effect was extremely significant during rain [22].

The low temperature for all samples due to Cameron Highlands elevation of more than 1500 m above sea level, resulting the temperature to be less than 25°C year-around and rarely falls below 12°C [23]. This low temperature slows down the water-rock interaction processes and consequently radon emanation to the ground water [9]. Since this ground water is used for drinking water purposes, it should be at least in Class IIA or better [20].

Supported ^{222}Rn Activity Concentration

Activity concentrations of supported ^{222}Rn in nine ground water samples (unfiltered and filtered) were presented in Table 3. The uncertainty in the measured activity concentration estimated from the peak area determination and the background [24]. The filtered samples activity concentration of supported ^{222}Rn were lower than unfiltered samples due to removal of non-dissolve suspended solid that contains a small amount of radionuclides from ground water samples [4, 14, 15]. However, the F-value (0.91) shows no significance difference in variance between both types of samples. This was due to small amount of radionuclides that contains in a small amount TSS in samples as shown in Table 2.

Unfiltered samples from Sungai Terla (KT1a and KT1b) contained highest amount of ^{222}Rn due to the raw water was coming from underground. This ground water source of LAKT, is coming from Gunung Pass which is located on the backbone of Peninsular Malaysia that flows into Sungai Terla [10]. Based on the geological map, the geological formation of the area is mainly made from igneous rocks (intrusive rocks, mainly granite with minor granodiorite)

[9]. Igneous rocks normally contain high amount of natural uranium and thorium which natural source of radon [4, 5]. Unfiltered samples of KTAa and KTA b contain lower ^{222}Rn than KTla and KTlb due to samples were aerated at aeration pond. The aeration process enhances the degassing of ^{222}Rn from water samples.

Table 2: Some of Water Quality Parameters Measured and NWQS Individual Value for Class I and Class IIA

Sample Code	DO (mg/L)	pH	Turbidity (NTU)	Temperature (°C)	TDS (mg/L)	Salinity (mg/L)	TSS (mg/L)
KTI	9.25	7.40	6.46	19.47	0.010	0.010	3.83
KTA	9.28	7.16	8.73	18.77	0.011	0.010	8.40
KTT	8.55	7.17	6.04	19.69	0.020	0.010	3.54
KRA	10.88	7.07	7.22	19.84	0.012	0.010	5.94
TKP	7.99	7.00	6.95	20.18	0.021	0.010	4.73
KTS	8.87	6.98	4.75	20.04	0.007	0.007	4.91
TRA	9.51	7.13	6.20	20.62	0.031	0.020	5.06
Class I [19]	≥ 7	6.5 – 8.5	≤ 5		≤ 500	≤ 0.05 (0.5 %)	≤ 25
Class IIA [19]	5 -7	6-9	≤ 50	$(27 \pm 2) ^\circ\text{C}$	≤ 1000	≤ 0.1 (1%)	≤ 50

For KTT, the slightly elevated supported ^{222}Rn in treated water was due to the fact that it was kept in enclosed treated water tank. The increases in supported ^{222}Rn activity concentration possibly due to the accumulation of radon gas in the enclosed tank and back diffused into the water. Domestic water contained 0.26 to 0.55 Bq/L of supported ^{222}Rn . TKP contained higher ^{222}Rn concentration than LAKT. This may be due to the particulate matters such as sand inside the piping system as shown by higher TSS of TKP than KTT in Table 2. For unfiltered samples, the untreated and treated shows appreciable difference of variance with an F-value of 0.37. The same observation prevails for filtered samples where the F-value is 0.11. Since both shows significance differences, the water supply need to be treated before received by the consumer in order to reduce the ^{222}Rn activity concentration.

High activity concentration of supported ^{222}Rn was observed from samples (KTla and KTAa) taken at water surface compared to below surface samples. This possibly due to the ^{222}Rn was easily moves from below to the river water surface during water movement, geology of the area and bottom sediment of the river [4,14].

The farthest location up to north from LAKT was only KRA while the nearest location to the south from LAKT was TKP followed by KTS and TRA. Based on Table 3, there was no trend in supported ^{222}Rn activity concentration from KTI to KTT where the water transportation occurs from intake to LAKT. These mainly due potential changes occurring in the water distribution, the oxidation state of the water and the concentration of suitable complexing agents which can increase the solubility of uranium or thorium [24]. However, there was a decreasing trend of

supported ^{222}Rn activity concentration from TKP to the TRA due to some of supported ^{222}Rn may loss during water distribution to the higher altitude of KTS and TRA [4].

Table 3: Supported radon activity concentrations in ground water samples

Sample	Supported ^{222}Rn Activity Concentration (Bq/L)	
	Unfiltered sample	Filtered sample
KTla	0.66 ± 0.07	0.59 ± 0.07
KTlb	0.43 ± 0.06	0.32 ± 0.05
KTAA	0.36 ± 0.05	0.29 ± 0.05
KTAAb	0.29 ± 0.05	0.19 ± 0.04
KTT	0.39 ± 0.05	0.32 ± 0.05
KRA	0.40 ± 0.06	0.31 ± 0.05
TKP	0.55 ± 0.07	0.43 ± 0.06
KTS	0.36 ± 0.05	0.32 ± 0.05
TRA	0.28 ± 0.05	0.26 ± 0.04

Pearson Correlation of Supported ^{222}Rn and Water Quality Parameter Measurement

Table 4 presents Pearson correlation coefficients between supported ^{222}Rn activity concentration in ground water samples and water quality parameter. A strong correlation observed between supported ^{222}Rn activity concentration for unfiltered and filtered samples ($r = 0.91$) due to some radionuclides may attach to the non-dissolved suspended solids in samples [14, 15]. Moderate correlation observed between ^{222}Rn with temperature. This was due to low temperature slows down the water-rock interaction processes and the releasing of radon to the ground water [8, 15]. Besides that, there was inversely correlation between ^{222}Rn with pH, DO, salinity, and TSS. pH may influence the solubility and mobility of radionuclides in water as the solubility is increasing with decreasing in pH as radium is more soluble in pH less than 7 than at higher pH [15]. DO and salinity does not affect the emanation of ^{222}Rn to the ground water [14, 15]. TSS contained very low amount of ^{222}Rn [15]. In this study, supported ^{222}Rn activity concentration shows inversely very weak correlation with turbidity ($r = -0.08$ and $0-0.38$) and very weak correlation with TSS ($r = 0.15$). However, another study showed good correlation between ^{222}Rn activity concentration with TSS and turbidity [15].

The study was extended to explore the correlation between various water quality parameters. A strong correlation between TDS and salinity ($r = 0.83$) possibly due to the dissolved solids in the samples was the dissolved salts such as calcium, magnesium, sodium and potassium that indicate salinity [14, 15]. Moderate correlation can be observed between turbidity and TSS ($r = 0.74$), temperature with TDS ($r = 0.68$) and salinity ($r = 0.52$). Suspended solids such as soils and particulate matters can affect the turbidity of the water samples [3, 22]. Since TDS and salinity has strong correlation between each other, higher temperature possibly can enhance the dissolving salts and solids in the water samples. The other parameters show no significant correlation between each other.

Table 4: Pearson correlation coefficient, between supported ^{222}Rn activity concentration in ground water samples and water quality parameter

	$^{222}\text{Rn}(\text{UF})$	$^{222}\text{Rn}(\text{F})$	DO	pH	Turbidity	Temp	TDS	Salinity	TSS
^{222}Rn									
(UF)	1								
$^{222}\text{Rn}(\text{F})$	0.94	1							
DO	-0.43	-0.47	1						
pH	-0.23	-0.31	0.13	1					
Turbidity	-0.08	-0.38	0.25	0.19	1				
Temp	0.21	0.48	-0.08	-0.43	-0.63	1			
TDS	0.00	0.15	-0.23	-0.16	-0.09	0.68	1		
Salinity	-0.41	-0.29	0.19	0.13	0.05	0.52	0.83	1	
TSS	-0.43	-0.64	0.36	-0.21	0.74	-0.53	-0.25	-0.01	1

UF – unfiltered samples, F – filtered samples

Comparison with other studies

Table 5 shows comparison of supported ^{222}Rn of the present study with other studies. The measured supported ^{222}Rn activity concentrations in ground water samples were lower than study in India [2] and Korea [25] but similar with studies in Malaysia [14, 15]. Generally, the supported ^{222}Rn activity concentrations in this present study were lower than the activity concentration limit of radon in drinking water as proposed by USEPA (11 Bq/L) [6, 26]. As a consequence from this, by drinking this water does not cause health hazard risk to the user since the dietary intake (D_{ing}) of ^{222}Rn in this water vary from 0.48 to 1.68 $\mu\text{Sv/yr}$. This D_{ing} is lower than $1 \times 10^2 \mu\text{Sv/yr}$ (0.1 mSv/yr) of the world limit [27].

Table 5: Comparison of the supported ^{222}Rn activity concentration obtained in the present study with other studies

Place	Type of water	Supported ^{222}Rn activity concentration (Bq/L)
Punjab, India[2]	Ground water	2.56 - 7.75
Kelantan, Malaysia [14]	River water	0.51 - 1.41
Jeju Island, Korea [25]	Ground water, coastal waters and river water	0.15 to 1.28
Taman Negara Pahang, Malaysia[15]	River water	0.32 – 1.06
Cameron Highlands, Malaysia (present study)	Ground water	0.19 – 0.66

Conclusion

The supported ^{222}Rn activity concentration for unfiltered, filtered and untreated samples ranged from 0.19 to 0.66 Bq/L, the treated and domestic water samples supported ^{222}Rn activity concentration ranged from 0.32 to 0.39 Bq/L and 0.26 to 0.55 Bq/L. All samples contain lower supported ^{222}Rn activity concentration than 11 Bq/L as proposed by USEPA. The variations are possibly or mainly due to the potential changes occurring in the water distribution, the oxidation state of the water and the concentration of suitable complexing agents. The decreasing trend only observed from TKP to KTS and TRA. The correlation between supported ^{222}Rn activity concentration and physicochemical parameters could be different from place to place and depends on geological locations.

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