ASSESSMENT OF SUPPORTED RADON IN GROUND WATER FROM HIGHLAND AREA USING PORTABLE CONTINUOUS RADON MONITOR

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ABSTRACT

Ground water contain natural radioactivity associated with uranium and thorium that present naturally in rocks and soils. Humans may be exposed to the emission of energetic alpha particle from supported radon decaying process in this water when it is inhaled or ingested. Assessment of supported radon in ground water was carried out using fourteen ground water samples from Cameron Highlands. The measurement was accomplished by degassing the water samples using pump and then allowing the gas to flow into specially constructed 0.0191 m^3 metal chamber. The activity concentration of supported radon in water sample was measured using continuous radon monitor inside the radon chamber. Measurement was carried out at one hour interval for twenty four hours. The hourly supported radon concentration was found to stabilize after about 8 hours. The stabilized concentration was used to determine supported radon activity concentration in the water samples. Results of the study show that depending on the sampling location, the activity concentrations of supported radon are in the range from 0.09 – 0.48 Bq/L which is lower than the activity concentration of radon in drinking water as proposed by USEPA (11 Bq/L).

ABSTRAK

Air tanih mengandungi radioaktiviti tabii termasuk uranium dan torium yang hadir secara tabii dalam batuan dan tanih. Manusia mungkin terdedah kepada pancaran zarah alfa bertenaga tinggi dari proses penyepaian 'supported radon' dalam air ini apabila dihidu atau diminum. Pengukuran 'supported radon' dalam air tanih dijalankan menggunakan empat belas sampel air tanih dari Cameron Highlands. Pengukuran telah dilakukan dengan menyalurkan udara kepada sampel air menggunakan pam dan membenarkan gas memasuki kebuk logam bersaiz 0.0191 m^3 yang telah diubahsuai. Kepekatan aktiviti 'supported radon' dalam sampel air telah diukur menggunakan alat pengesan radon di dalam kebuk radon. Pengukuran telah dijalankan pada setiap satu jam bagi tempoh 24 jam dan kepekatan 'supported radon' mencapai kestabilan dijumpai selepas 8 jam. Kepekatan yang stabil itu digunakan untuk mengira kepekatan aktiviti 'supported radon' dalam sampel air. Keputusan daripada kajian menunjukkan bahawa bergantung kepada lokasi sampel, kepekatan aktiviti 'supported radon' dalam lingkungan dari 0.09 – 0.48 Bq/L di mana lebih rendah daripada kepekatan aktiviti radon dalam air minuman yang dianjurkan oleh USEPA (11 Bq/L).

Keywords: Ground water, supported radon, radon chamber, natural radioactivity.

INTRODUCTION

Ground water is defined as water present in the saturated zone beneath the water table (Perk, 2006; Baird and Cann, 2005). Since ground water makes up 0.6% of the world's total water supply (Baird and Cann, 2005), it is favored as a source of drinking water in many countries (Skeppström and Olofsson, 2007). The ultimate source of ground water is precipitation that falls onto the surface; a small fraction of it eventually filters down to the saturated zone (Baird and Cann, 2005). This saturated zone composed of porous rocks such as sandstone, or in highly fractured rocks such as gravel, or non-porous but fractured rock such as granite, and water is bounded by a layer of clay or impervious rocks, it constitutes a permanent reservoir and can be called as aquifer (Perk, 2006; Baird and Cann, 2005).

Ground water is subsurface water, often thought to be cleaner and easier to treat as compared to surface water (Skeppström and Olofsson, 2007). However, the precipitation of water that falls onto surface will transfer dissolved pollutants from landfills (waste dumping grounds), agriculture, soil pollution, septic tank and atmosphere deposition, move with the percolating soil water into ground water, while organic liquid pollutants may reach the ground water autonomously. In areas where surface water infiltrates to ground water, surface water pollution is also a potential source of ground water contamination (Perk, 2006).

Since ground water lies above the soils where composed of rocks (sandstone, gravel and granite), it may contains high amounts of natural radionuclide associated with uranium and thorium. These radionuclides are amongst the most incompatible elements and are concentrated in granitic rocks and soils (Örgün *et al.*, 2005) and parent of radon which can lead to health problems if present in high concentrations in ground water (Skeppström and Olofsson, 2007).

Cameron Highlands is one of the major highland areas in Peninsular with agricultural and ecotourism activities and has its own water supply. Its water supply is ground water, coming from high elevation of Mount Pass, Mount Brinchang and Mount Duri whose geological formations made from igneous rocks (intrusive rocks, mainly granite with minor granodiorite) that flows into Terla River, Bertam River and Habu River, respectively. Ground water coming from these mountains is sufficiently large volume to induce an appreciable spread in dissolved supported radon concentration (Abdallah *et al.*, 2007) and this concern specific radionuclide may pose health risk to consumer (Zaini Hamzah *et al.*, 2011). Due to these, the geography, geology and local in nature of Cameron Highlands are ideal and chosen for this study since public water sources used for services and drinking purposes.

Radon is a naturally occurring radioactive inert gas, partially soluble in water colorless, odorless and tasteless (Zaini Hamzah *et al.*, 2011; Ahmad Saat *et al.*, 2007). It is produced continuously by the alpha decay of radium with some atoms escaping to the surrounding fluid phase, such as ground water (Alessandro and Vita, 2003). This gas was found in various concentrations in ground water (Alessandro and Vita, 2003). It is an alpha emitter that decays into a chain of progenies of alpha emitters (Zaini Hamzah *et al.*, 2011; Ahmad Saat *et al.*, 2007) and gamma emitters.

Radon is partially dissolved in water and consumed by human that may pose a health hazard to humans and environment (Zaini Hamzah *et al.*, 2011; Badhan *et al.*, 2010). Study carried out by Department of Environmental Services, New Hampshire (2009) shows that the risk from radon in water is relatively high compared to other drinking water contaminants. The risk on ingesting radon in water is substantially lower than that associated with inhalation (NHDES, 2009). Thus, assessing radon in water, in addition to that in air, is an important step in reducing potential exposure to it.

Normally, radon-in water activity concentrations are measured by means of liquid scintillation counting (LSC). Due to these limitations, several alternative detection methods have been developed (Zaini Hamzah *et al.*, 2011; Zaini Hamzah *et* al., 2011; Misdaq *et al.*, 2007).

In this paper, we described the preliminary results of method developed by using continuous radon in air monitor (CRM that is available in our laboratory) for determination of supported radon activity concentration in ground water samples from Cameron Highlands.

METHODOLOGY

Sampling Site and Samples

Locations of ground water sampling 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 comprise of water source intake, treatment plant and houses in Cameron Highlands, Pahang. Global positioning system (GPS) was used to determine longitude and latitude of each sampling point location. Table 1 shows sampling point location. Samples were collected in 10 liters polyethylene containers from selected locations. The plastic containers was cleaned with 6 M nitric acid (HNO_3) and rinsed with distilled water in order to remove any contaminants, prior to use.

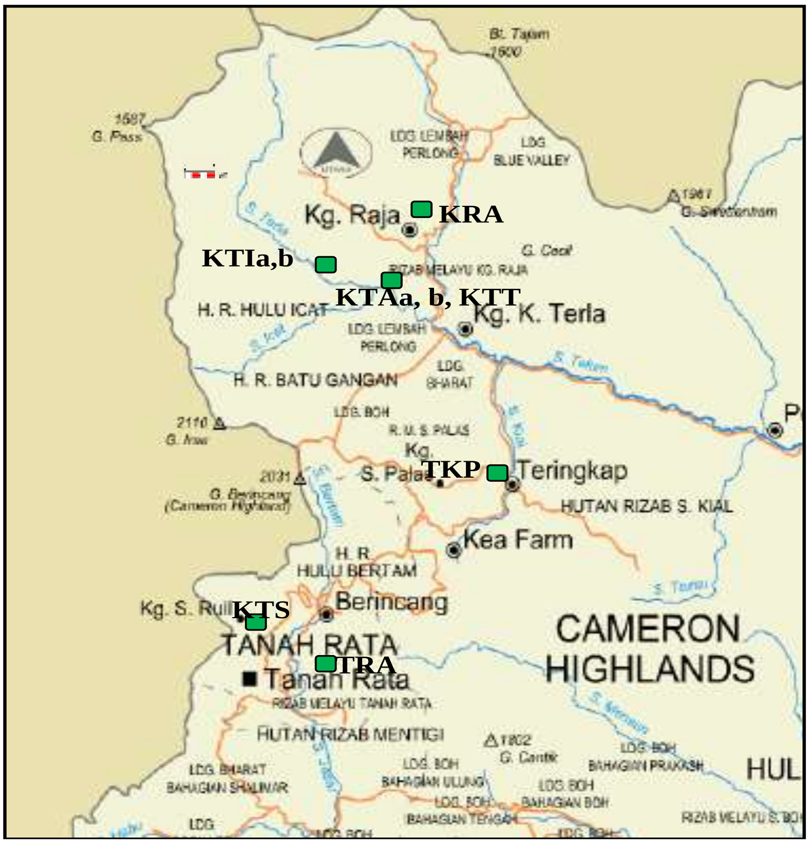


Figure 1: Locations of ground water sampling in Cameron Highlands, Malaysia.

Note: ■ Sampling Location

Water samples from intake and aeration pond at water treatment plant were taken from river surface and below of the river water surface. Samples from treatment plant were collected from the water input, during processes of the treatment and the water output while the residential water were collected from domestic pipes. The containers were filled to capacity in order to avoid air bubbles and closed tightly to prevent radon leakage because it is easily escaped from water to air (Bonotto and Mello, 2006).

Surface doses rate were measured in-situ at water source intake and water treatment plant sampling point using LUDLUM rate meter Model 2241 at the selected point at surface and 1 m height. On the same sampling point, seven water quality parameters were measured in-situ using YSI multi probe water quality parameter. The parameter measured were dissolved oxygen (DO), pH, turbidity, temperature, total dissolves solid (TDS), salinity and oxidation reduction potential (ORP). Total suspended solid (TSS) was measured in the laboratory.

The pH of the water samples were adjusted to below 2 using 6 M nitric acid in UiTM laboratory 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 (Zaini Hamzah *et al.*, 2011).

Table 1: Sampling point location

Location	Sample Code	Type of Water Samples	Latitude	Longitude
Intake Kuala Terla	KTIa	River (surface)	04° 33.500'	101° 23.873'
	KTIb	River (below surface)	04° 33.500'	101° 23.873'
Water Treatment Plant Kuala Terla	KTAa	Aerated river water (surface)	04° 32.796'	101° 24.562'
	KTAb	Aerated river water (below surface)	04° 32.796'	101° 24.562'
Water Treatment Plant Kuala Terla	KTT	Treated water	04° 32.796'	101° 24.562'
Kampung Raja	KRA	Domestic water	04° 34.122'	101° 24.608'
Tringkap	TKP	Domestic water	04° 30.842'	101° 25.646'
Kampung Taman Sedia	KTS	Domestic water	04° 28.620'	101° 22.922'
Tanah Rata	TRA	Domestic water	04° 28.014'	101° 23.083'

EXPERIMENTAL

Two types of water samples were studied which are unfiltered and filtered water samples. For filtered water samples, samples were filtered using cellulose membrane filters (0.45 µm, Whatman) on Buchner funnel attach to pump to eradicate suspended solid and impurities from water samples. Figure 2 shows the schematic diagram of the radon chamber. Water samples were poured into 1 liter polyethylene bottle and properly sealed before placed it outside the specially constructed 0.0191 m^3 metal chamber. A continuous radon monitor Model 1027 of SunNuclear was placed inside the radon chamber. The chamber was completely sealed to prevent the radon leakage. This was important to ensure the gaseous of supported radon was confined in the chamber (Baykara and Doğru, 2006). Initially, the radon chamber was purged by nitrogen gas. Then, supported radon was out gassed from samples by passing a stream of radon free air through the sample where the pumping rate was 1200 cm^3 per minute. The gas was passed through desiccant to absorb water vapor before being released into the tight chamber. By exposing the radon monitor in the chamber for twenty four hours, it automatically monitors radon activity concentration one hour interval for twenty four hours. The CRM was recognized by the USEPA and sensitive to ^{222}Rn isotope. At the pumping rate used, the supported radon concentration in the chamber was expected to reach equilibrium after 8 hours.

Calculation

At equilibrium concentration condition, the CRM will show the delayed radon concentration in the radon chamber, C_o pCi/L (converted to Bq/m^3 by a multiplying factor of 37). If the volume of the radon chamber and water samples used were V_o and V_w respectively, the supported radon concentration in water is calculated by using Equation 1.

$$C_w = C_o\,(V_o/V_w)$$

Equation 1

The value of C_o for the respective sample was obtained by plotting a graph of hourly supported radon concentration recorded by the CRM against elapsed time. The points of the graph were fitted by suitable function (polynomial), and the maximum of the fitted graph was taken as C_o.

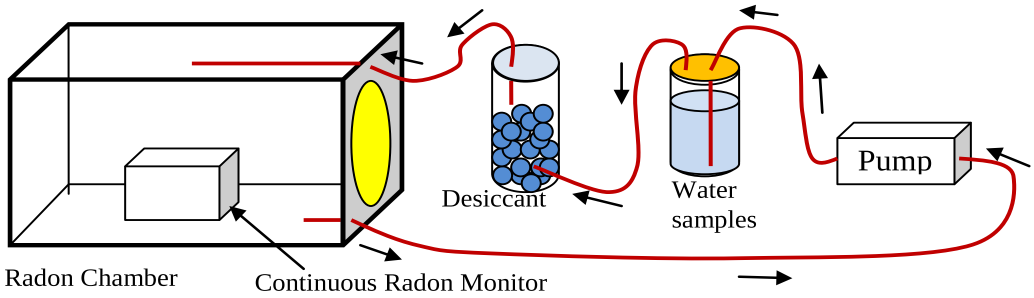


Figure 2: Schematic diagram of the radon chamber

RESULTS AND DISCUSSION

Dose Reading

From Table 2, the surface doses were only measured for KTI, KTA and KTT since the locations are river, aeration pond and treated water tank of the Kuala Terla water treatment plant (LAKT), respectively. The surface dose is ranging from 0.178 to 0.356 µSv/hr for surface, while for 1 meter dose shows value range from 0.206 to 0.342 µSv/hr. These surface dose values indicate higher radiation dose compared to global range value i.e. 0.079 – 0.13 µSv/hr (Zaini Hamzah *et al.*, 2011). The surface dose reading for KRA, TKP, KTS and TRA was not available due to samples taken at domestic pipes.

Table 2: Surface Dose reading

Sample Code	Surface Dose (µSv/hr)	1 Meter Dose (µSv/hr)
KTI	0.178	0.206
KTA	0.259	0.335
KTT	0.356	0.342
KRA	NA	NA
TKP	NA	NA
KTS	NA	NA
TRA	NA	NA

Note: NA – Not available

Water Quality Parameter

Based on Interim National Water Quality Standards for Malaysia (INWQS) (MDOE, 2008), ground water from Cameron Highlands is categorized in Class IIA because it is a water supply II, where conventional treatment is required. Table 3 shows some water quality parameters measured and INWQS limit. Dissolved oxygen (DO) is an essential indicator in natural water due the necessary of it to sustain aquatic life. DO in all water samples is greater than 5 to 7 mg/L which limit given by INWQS (MDOE, 2008). Oxygen is often used as an indicator of water quality because high concentration of oxygen indicates good water quality (Zaini Hamzah *et al.*, 2011). Range of pH in all location is 6.98 to 7.40.The pH still in the range of 6 – 9 in INWQS (MDOE, 2008). According to Chau and Jiang (2002), natural river water is slightly acidic because of its origin of river water and because of tannin and leaves released from forest floor.

Other essential water quality parameter also includes turbidity and total suspended solid (TSS). Turbidity refers to the clearance of the water (Zaini Hamzah *et al.*, 2011). Turbidity value for all samples ranges from 4.75 to 8.73 NTU. KTA contains highest turbidity value (8.73 NTU) due to the untreated sample is at aeration pond at Kuala Terla water treatment plant compared to KTI. All samples turbidity value is lower than INWQS limit (50 NTU) (MDOE, 2008). TSS in all samples is ranges from 3.54 to 8.40 mg/L and still lower than limit of INQWS (50 mg/L) (MDOE, 2008). This suspended solid is caused by the soil erosion with addition to land of the around areas where opening up for agriculture and other activities and extremely significant during rain (Makalahmad et al., 2008).

Table 3: Some of Water Quality Parameters Measured and INWQS Limit

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	0.022	0.010	4.73
KTS	8.87	6.98	4.75	20.04	0.009	0.007	4.91
TRA	9.51	7.13	6.20	20.62	0.031	0.020	5.06
INWQS	5 - 7	6 - 9	50	Normal + 2 °C	1000	10 (1 %)	50

The temperature for all samples ranges from 18.77 °C to 20.62 °C. Being more than 1500 m above sea level, temperature of Cameron Highlands is not higher than 25 °C and rarely falls below 12°C year-around (Eisakhani and Makalahmad, 2009). Since the temperature is low, it slows down the water-rock interaction processes and consequently radon emanation to the ground water (Alessandro and Vita, 2003).

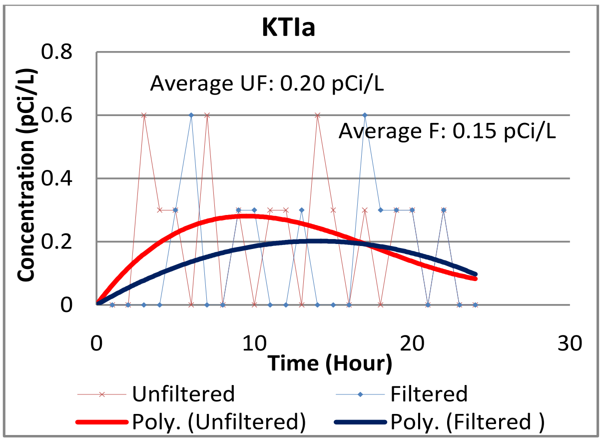
Total dissolved solid (TDS) in all ground water samples are range from 0.009 to 0.031 mg/L which is below than INWQS limit (1000 mg/L). Then, salinity in all water samples is still lower than INWQS limit (1 %).

Radon Activity Concentration

Figure 3 shows the hourly variation and average of supported radon concentration recorded by the continuous radon monitor for each samples studied. Each point on the graph shows the supported radon concentration in air inside the chamber at one hour interval for twenty four hours for every sample measured by continuous radon monitor. These points later were fitted with the polynomial function in order to determine maximum value of the fitted graph. Then, the maximum value of the fitted graph was used in order to determine activity concentration of supported radon in water sample by substituted into Equation 1. Generally, the fitted line in

Figure 3 showed similar pattern of increasing concentration of supported radon for the first few hours and reaching steady value between 8 to 17 hours. Then, all graph showed reduction in concentration. Since this method is closed system, the time started for the measurement of supported radon activity concentration in ground water samples is independent at any hour.

The observation can be explained by the fact that in the earlier hours supported radon-gas building up was taking place in the metal chamber. It will later reach equilibrium or maximum value where the amount of supported radon gas purged by the pump is equal to the amount of formed by the decay of radon parent (radium) that is dissolved in the water sample. In this study, the reasons we choose metal chamber due to less or no electrostatic attraction occur inside surface of the metal chamber compared to plastic chamber. The decaying process of supported radon produce high energetic alpha particle with two neutrons and two protons makes alpha particles positively charged (NHDES, 2007; MDHSS, 2006; Walia *et al.*, 2003). Strong electrostatic attractions occur due to the presence of positively charged alpha particles and negatively charged of the plastic chamber (Serway and Jewett, 2006; USDOEHDBK-1122-99-1.07, 2003). The positively alpha particles attracted to inside surface of plastic chamber (Serway and Jewett, 2006) instead of being detected by continuous radon monitor. Since metal chamber is used, the alpha particles can be detected by continuous radon monitor.



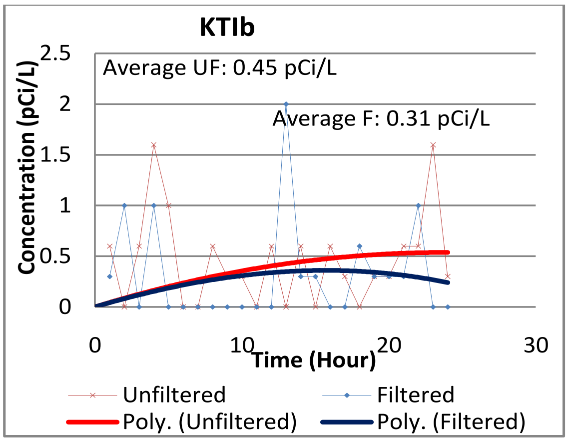
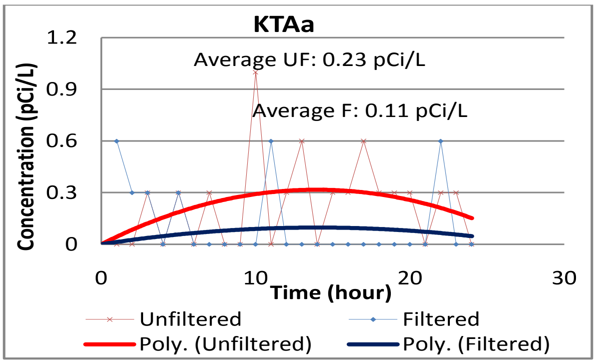


Figure 3. (a)

(b)



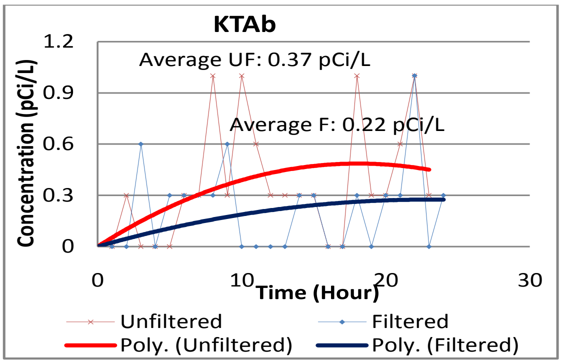
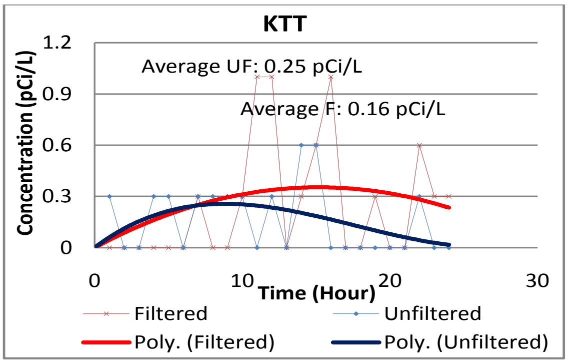


Figure 3 (c)

(d)



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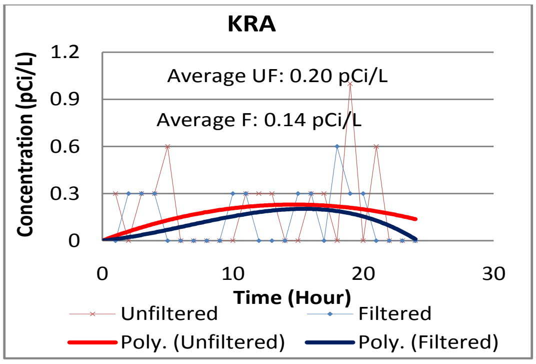

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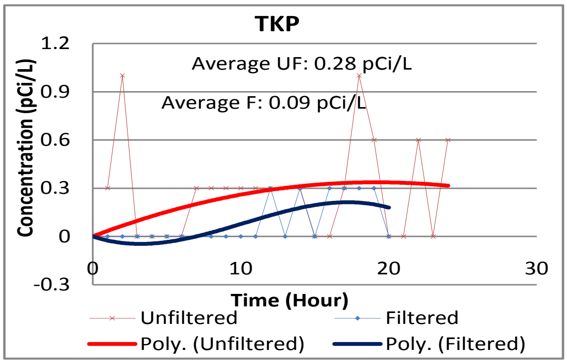

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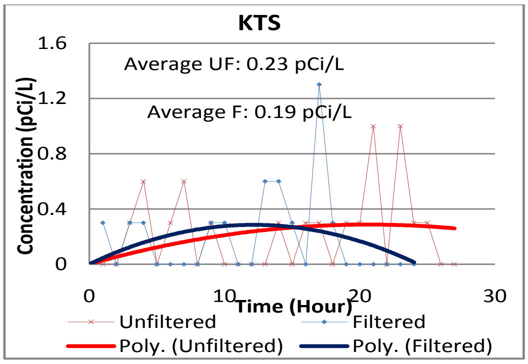

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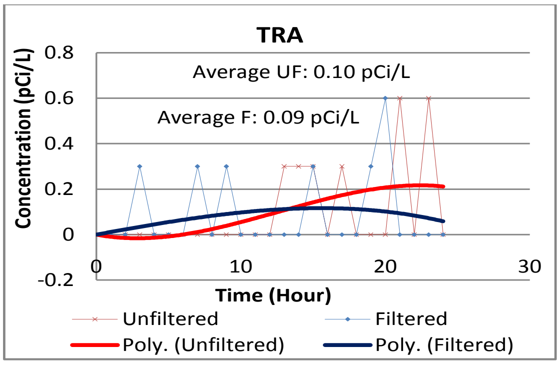

(i)

Figure 3: Hourly variation and average of supported radon concentration recorded by CRM for (a) KTIa, (b) KTIb, (c) KTAa, (d) KTAb, (e) KTT, (f) KRA, (g) TKP, (h) KTS and (i) TRA

The activity concentrations of supported radon in nine samples are presented in Table 4. These activity concentrations of supported radon were ranging from 0.09 to 0.48 Bq/L for unfiltered and filtered samples. The lowest supported radon activity concentration is at KTAa for filtered sample taken at the surface of the river (0.09 Bq/L) and the highest activity concentration of supported radon is at KTIb for unfiltered samples (0.48 Bq/L). Removal of non-dissolve suspended solid that contains a small amount of radionuclides from ground water samples, result in slightly lower of activity concentration of supported radon in filtered water samples than unfiltered samples (Zaini Hamzah *et al.*, 2011). KTI and KTA were divided into surface and below surface where the below surface depth is 0.3 to 0.5 m from the surface of the river. Samples taken from surface of the river contain low activity concentration of supported radon compared to below surface samples because supported radon at surface of the river water is easily escape from water to atmosphere during water movement compared to below surface samples. The geology of the area and bottom sediment also may affect supported radon activity concentration in surface of river water (Zaini Hamzah *et al.*, 2011).

Table 4: Supported radon activity concentrations in ground water samples

Sample Code	Type of Sample	Supported Radon (UF sample) (Bq/L)	Supported Radon (F sample) (Bq/L)
KTIa	Surface	0.25 ± 0.06	0.20 ± 0.05
KTIb	Below surface	0.48 ± 0.12	0.33 ± 0.08
KTAa	Surface	0.33 ± 0.08	0.09 ± 0.02
KTAb	Below Surface	0.46 ± 0.11	0.26 ± 0.07
KTT	Domestic water	0.40 ± 0.10	0.26 ± 0.07
MKR	Domestic water	0.23 ± 0.06	0.20 ± 0.05
TKP	Domestic water	0.34 ± 0.08	0.21 ± 0.05
KTS	Domestic water	0.30 ± 0.08	0.29 ± 0.07
TRA	Domestic water	0.21 ± 0.06	0.11 ± 0.03

Note: UF for unfiltered
F for filtered

Ground water source of LAKT is located at the backbone of Peninsular Malaysia where the rock type is igneous rocks. Igneous rocks contain high amount of uranium and thorium which both are natural source of radon (Ahmad Saat *et al.*, 2007). Based on result tabulated in Table 4, there is a decreasing trend in supported radon activity concentration from KTI to TRA because KRA, TKP, KTS and TRA received its water supply from Kuala Terla water treatment plant (LAKT). The farthest location up to north from LAKT is only KRA while the nearest location to the south from LAKT is TKP followed by KTS and TRA. The activity concentration of supported radon in ground water samples are decreasing from KTI to its distributed area. This is due to some of supported radon may escape to the atmosphere during water distribution and water movement.

Although the source is from same mountain, ground water that comes from different small rivers that enters one main river (Terla River), may results in different in activity concentration of supported radon from the source to the treatment plant. Terla River that carry the ground water sources from Mount Pass to LAKT has its own characteristic that might be different from place to place (Zaini Hamzah *et al.*, 2011).

All values for supported radon activity concentration in all locations are lower than the activity concentration of radon in drinking water as proposed by USEPA (11 Bq/L) (Zaini Hamzah *et al.*, 2011). As the ground water used as services and domestic purposes such as cleaning, cooking and drinking, the supported radon activity concentration still very much lower than limit proposed by USEPA.

The results are compared with those studies on supported radon in water in other location in order to have some idea on acceptability of the present study. This is shown in Table 5. By considering type of water and sampling site is in Malaysia, the comparison of supported radon activity concentration in water samples shows good agreement. Activity concentration of supported radon in ground water samples from Cameron Highlands still in the range of supported radon activity concentration in water samples in Malaysia. However, this method is still preliminary and not validated yet using uranium and thorium standard solutions; even CRM was recognized by the USEPA and sensitive to ^{222}Rn isotope.

Table 5: Comparison of the supported radon activity concentration obtained in the present study with other studied

Place	Type of water	Supported Radon activity concentration (Bq/L)	Method	Reference
Kelantan	River water	0.88 - 44.43	Liquid scintillation counting technique	Zaini Hamzah *et al.* (2010)
Perak	Lake water	0.29 – 1.27	Gamma Spectrometry Technique	Zaini Hamzah *et al.* (2011)
Pahang	River water	0.32 – 1.27	Gamma Spectrometry Technique	Zaini Hamzah *et al.* (2011)
Pahang	Ground water	0.09 – 0.48	Radon continuous monitor	Present study

Table 6: Pearson correlation coefficient, between supported radon activity concentration in ground water samples and water quality parameter

	Supported Radon (UF)	Supported Radon (F)	Temp	TDS	Sal	DO	pH	Turbidity	TSS
Supported Radon (UF)	1								
Supported Radon (F)	0.65	1							
Temp	0.01	-0.07	1						
TDS	0.26	0.11	0.68	1					
Sal	0.08	-0.01	0.52	0.83	1				
DO	-0.26	-0.71	-0.08	-0.23	0.19	1			
pH	0.59	0.58	-0.43	-0.16	0.13	0.13	1		
Turbidity	-0.10	-0.19	-0.63	-0.09	0.05	0.25	0.19	1	
TSS	0.73	0.54	0.48	-0.01	0.16	0.65	-0.59	0.25	1

Table 6 represents Pearson correlation coefficient, between supported radon activity concentration in ground water samples and water quality parameter. There is moderate correlation observed between supported radon activity concentration for unfiltered and filtered samples (r = 0.65) and moderate correlation for supported radon activity concentration for unfiltered and filtered samples with pH measured (r = 59 and r = 0.58). For correlation between water quality parameter measured, there is moderate correlation between temperature and TDS (r = 0.68), salinity (r = 0.52) and TSS (r = 0.48). Strong correlation observed between TDS and salinity (r = 0.83). Dissolved oxygen (DO) has moderate correlation with TSS (r = 0.55).

CONCLUSIONS

As conclusions, the activity concentration of supported radon in ground water samples not exceeded 11 Bq/L as proposed by USEPA but the activity concentration is different at studied site. The differences are possibly due to different depths and pathways of the out flowing water. There is a decreasing trend of supported radon activity concentration from KTI to its distributed area (KRA, TKP, KTS and TRA). The surface dose is range from 0.178 to 0.259 µSv/hr while for 1 meter dose shows value range from 0.178 to 0.335 µSv/h. The surface radiation dose at selected locations are more than the global range for surface radiation dose (0.079 – 0.13 µSv/hr). The correlation between supported radon activity concentration and physicochemical parameters could be different from place to place and depends on geographical locations. To the certain extend this method developed was able to produce acceptable results. To the best of our knowledge, this is the first study of the radioactivity concentration of supported radon in the source of public water supply in Cameron Highlands, Pahang, Malaysia. The data obtained in this study is the baseline for setting standards for water quality in this country.

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