MEASUREMENT OF RADON CONCENTRATION IN GROUNDWATER IN PHU HOI COMMUNE, DUC TRONG DISTRICT, LAM DONG PROVINCE AND ASSESSMENT OF ANNUAL EFFECTIVE DOSE FOR THE PUBLIC

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ABSTRACT

Radon is a natural source of radiation that contributes approximately 50% to the public dose. Therefore, it is very important to assess and minimize the impact of radon on human health. In this study, we determined radon concentrations in groundwater samples in Phu Hoi Commune, Duc Trong District, Lam Dong Province by using the liquid scintillation counter of ALOKA LSC 6100. Then, we calculated the annual effective doses for the public when they consume the above-mentioned groundwater on a daily basis. The obtained radon concentrations and annual effective doses for an individual consumer were 2.19 - 8.02 Bq/l and 10.47 - 38.30 μSv/y, respectively. The results show that these values are under the permissible safe limits recommended by the US Environmental Protection Agency (EPA) and World Health Organization (WHO). In conclusion, these sources of groundwater are safe for the public health as far as radon concentration is concerned.

Keywords: *Radon concentration, groundwater, liquid scintillation, annual effective dose*

1. Introduction

In nature, radon has 36 isotopes with the mass numbers from 193 to 228, of which the most common are 3 isotopes: radon (^{222}Rn) with symbol Rn and half-life of 3.82 days, produced from the radioactive decay series of uranium (^{238}U); thoron (^{220}Rn) with the symbol Tn and half-life of 54.5 seconds, belonging to the decay series of thorium (^{232}Th); actinon (^{219}Rn) with the symbol An and half-life of 3.96 seconds, produced from the decay series of uranium (^{235}U). In geological and environmental studies, ^{220}Rn and ^{219}Rn are of little interest due to their very short half-lives; ^{222}Rn is of most interest because it has the longest half-life, enough to penetrate and retain in the human body and cause harm for public health (Passo & Cook, 1994).

Radon (^{222}Rn) is ubiquitous in water, air and soil and contributes about 50% of the public dose (United Nations Scientific Committee on the effects of Atomic Radiation,2000). It is the main lung cancer risk after tobacco (Ahmed et al, 2017). Approximately 20,000 people die from lung cancer related to indoor radon annually in the United States (United States Environmental Protection Agency, 1999). Therefore, the determination of radon concentration in groundwater drinking sources plays a very important role throughout the world in assessing the extent of radon's impact on public health and proposing preventive measures to ensure proper radiation protection when necessary.

Many recent researches on radon in groundwater have been published both inside and outside Vietnam. Radon concentrations in groundwater ranged from 0.11 to 5.61 Bq/l in Kirkuk, Iraq (Ahmed et al, 2017) and were under the safe limit of 11 Bq/l (Ahmed et al, 2017, El-Araby et al, 2019 & Wedad et al, 2015). The radon concentrations in groundwater in Jazan, Saudi Arabia varied from 1.74 to 4.32 Bq/l, lower than the permissible level (El-Araby et al, 2019). However, radon concentrations in drinking ground water in the horst Söderåsen, Sweden were extremely high, ranging from 235 - 358 Bq/l and one of the solutions to these concentrations was to boil water in a coffee machine to release more than 90% of soluble radon (Erlandsson, 2001). In Vietnam, many studies on radon in the groundwater in Cu Chi District, Ho Chi Minh City (0.36 - 5.50 Bq/l) and Long Thanh District, Dong Nai Province (0.45 - 16.83 Bq/l) have been published, in which several concentrations were higher than the safe level (Hanh, 2013 & Ha, 2015).

This study was conducted to investigate radon concentrations in the groundwater in Phu Hoi Commune, Duc Trong District, Lamdong Province – the area without any database on radon up to the present. Then we assessed annual effective doses for the public consuming the groundwater above and make recommendations as well as apply interventions when necessary.

2. Materials and methods

2.1. Sampling and method

- Ten groundwater samples were taken from drilled wells of 10 households located in the area where groundwater is the only source for drinking and cooking in Phu Hoi Commune.

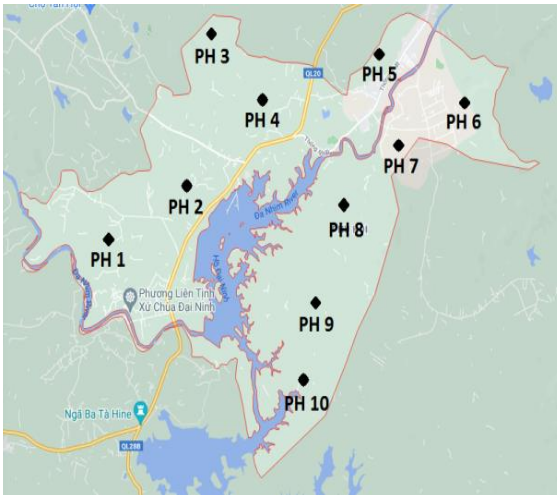

Figure 1: *Map of 10 sampling sites in Phu Hoi Commune*

- Use the analytical procedure for determining radon concentration in groundwater on liquid scintillation counter of ALOKA LSC 6100 validated by the Japan Atomic Energy Agency (JAEA) and applied in both JAEA and the Training Center, Nuclear Research Institute (Tsutomu & Takamitsu, 2015). This is a high-efficiency method that is very well-suited for the environmental radioactivity assessment.

2.2. Analytical procedure

- Use unquenched ^{14}C standard with the activity of 143100 dpm dated on October 3, 2016 and corresponding background standard, both produced by the PerkinElmer.

- Weigh 8 grams of PPO and 0.6 grams of POPOP and place them into a brown glass flask containing 2 liters of toluene solvent and shake until completely dissolved.

- Take 20 ml of toluene scintillation solution, put it into each of 20 vials and tighten the caps.

- Pump and discharge groundwater for 2 minutes and rinse the teflon bottles with that groundwater at each well, then fill up the bottles and tighten the caps. Take 10 groundwater samples shown in Figure 1 and bring them to the laboratory. Note each sampling time.

- Discard part of the groundwater in each bottle until the remaining water volume is 850 ml. Add to each bottle 100 ml of scintillation solution. The remaining air volume was 150 ml.

- Place 10 bottles in a shaker and shake vigorously for 10 mins to extract radon into the organic phase. Then stand the bottles for 10 minutes to separate the aqueous from organic phases.

- Take 20 ml of organic phase from 10 bottles, put it into 10 vials and tighten their caps.

- Stand the vials for 4 hours until radioactive equilibrium is established. Measure each vial for 10 minutes, 3 times, using unquenched ^{14}C standard and number 16 (MY No 16).

Figure 2: *Liquid Scintillation Counter LSC 6100*

3. Results and discussion

3.1. Efficiency Tracing Method (ETM) for radon measurement

Measure the counting rates of ^{14}C standard, background standard, 10 sample vials and 10 background sample vials for 6 windows (KeV): (0.05-2000; 5-2000; 10-2000; 20-2000; 30-2000; 40-2000) and calculate counting efficiencies of ^{14}C standard for these windows.

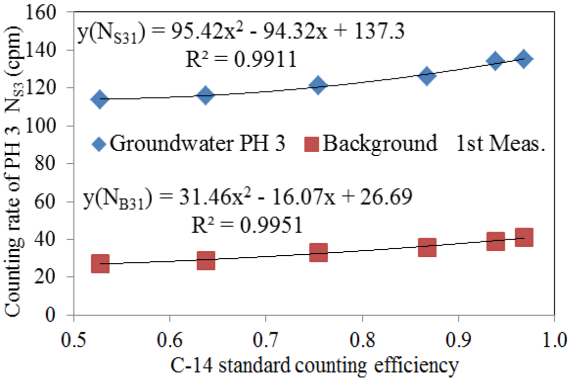

Figure 3: ETM curve for PH 3 and background

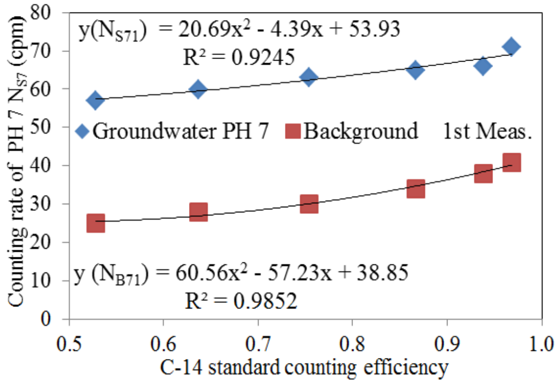

Figure 4: ETM curve for PH 7 and background

Draw the graphs showing the relationships between the counting rates of 10 groundwater samples and 10 background samples with the counting efficiencies of ^{14}C standard for 6 windows and fit with quadratic functions. Functions $y(N_{S31})$, $y(N_{S71})$, $y(N_{B31})$ and $y(N_{B71})$ in Figures 3 and 4, corresponding to samples PH 3 and PH 7, background samples PH 3 and PH 7, are 4 out of 60 fitted quadratic functions obtained, where $y(N_{S31})$, $y(N_{S71})$: total counting rates in 20 ml of organic phase of PH 3 and PH 7(cpm); $y(N_{B31})$ and $y(N_{B71})$: counting rates of background samples of PH 3 and PH 7 (cpm), x(E): counting efficiencies of ^{14}C standard, all for the first measurement.

3.2. Calculation of radon concentrations in groundwater by ETM method

In the decay series of ^{222}Rn, there are 2 alpha emitters and 2 beta emitters as follows:

$$^{222}Rn \xrightarrow[3.82\ days]{\alpha} {}^{218}Po \xrightarrow[3.05\ mins]{\alpha} {}^{214}Pb \xrightarrow[26.8\ mins]{\beta^-} {}^{214}Bi \xrightarrow[19.8\ mins]{\beta^-} {}^{214}Po \xrightarrow[165\ \mu s]{\alpha} {}^{210}Pb$$

$$C_T = (N_S - N_B) \times (1/60) \times (1/5) \times (1/f_e) \times (1/20) \times \exp(0.693t/3.825) \quad (1)$$

This equation (Tsutomu & Takamitsu, 2015) is used for calculating ^{222}Rn concentrations in the organic phase C_T (Bq/ml).

where N_S: total counting rates of groundwater samples extrapolated at 100 % efficiency (dpm); N_B: counting rates of background samples extrapolated at 100 % efficiency (dpm); 1/5: when equilibrium is reached, the radioactivity of radon and the other 4 isotopes in the decay chain above are the same. The ETM method measures 5 isotopes simultaneously. Therefore, radon activity is 1/5 of the total activity measured; f_e: radon correction factor of 0.75 for a 20-ml vial; t: time interval from sampling to measurement (second).

According to the law of radioactivity conservation, ^{222}Rn concentration in groundwater C_0 (Bq/ml) is calculated as follows (Tsutomu & Takamitsu, 2015):

$$C_O V_W = C_W V_W + C_A V_A + C_T V_T \quad (2)$$

$$C_O = \left\{\frac{D_W}{D_T} + \frac{1}{D_T}\left(\frac{V_A}{V_W}\right) + \left(\frac{V_T}{V_W}\right)\right\} C_T \quad (3)$$

where $D_W = 9.12/(17.0 + T)$ (4)
$D_T = 18.2 \times \exp(-T/46.5)$ (5)

where C_0: radon concentration in groundwater sample (Bq/ml); C_W: radon concentration in aqueous phase (Bq/ml); C_A: radon concentration in air phase (Bq/ml); C_T: radon concentration in organic phase (Bq/ml); V_W: volume of aqueous phase, V_W = 850 ml; V_A: volume of air phase, V_A = 150 ml; V_T: volume of organic phase, V_T = 100 ml; D_T: ratio between radon concentration in toluene and in air C_T/C_A; D_W: ratio between the concentration of radon in groundwater and in air C_W/C_A; T: laboratory temperature, T = 20°C.

Extrapolate the radioactivities N_{S31}, N_{S71}, N_{B31}, N_{B71} (dpm) at 100 % efficiency (E=1) from 4 quadratic functions in Figures 3 and 4. Then calculate the radon concentrations in groundwater based on the formulas 1 and 3. The calculation is similar for the 2nd and 3rd measurements. Finally calculate the average radon concentrations for 3 measurements. The same calculation is conducted for the rest of the samples.

3.3. Calculation of the annual effective doses of radon in groundwater for the public

Radon in groundwater enters the human body through two pathways of inhalation and ingestion. The total effective dose for the public includes the dose for both inhalation and ingestion.

Annual effective dose for inhalation of radon gas E_{INH} (μSv/y)

Radon in groundwater enters the indoor air through water-disturbing activities such as showering, washing dishes, boiling... According to UNSCEAR, the formula for calculating the annual effective dose for inhalation of radon gas from domestic water (United Nations Scientific Committee on the effects of Atomic Radiation, 2000).

$$7000\ (\text{hrs/y}) \times 0.4 \times 9 \left(\frac{\mu Sv}{Bq/l.hrs}\right) \quad (6)$$

where E_{INH}: annual effective dose for inhalation of radon released from domestic water (μSv/y); C_0: concentration of radon in groundwater (Bq/l); 10^{-4}: ratio between the amount of radon released from water into the air and radon in the water; 7000 (hrs/y): the total time the public breathe indoor air annually; $9 \left(\frac{\mu Sv}{Bq/l.hrs}\right)$: dose conversion factor for radon dose for inhalation; 0.4: equilibrium coefficient between radon and its isotope (equilibrium factor) F.

$$F = \frac{C_{eq}}{C_{Rn}} = 0.4 \quad (7)$$

Annual effective doses for ingestion of radon gas E_{ING} (μSv/y)

$$E_{ING}\ (\mu Sv/y) = C_0\ (Bq/l) \times 730\ (l/y) \times 0.0035 \left(\frac{\mu Sv}{Bq}\right) \quad (8)$$

where E_{ING}: annual effective dose for ingestion of radon in groundwater (μSv/y); C_0: radon concentration in groundwater (Bq/l); 730 (l/y): average annual amount of groundwater consumed by the public (equivalent to 2 l/day). This is the only source of drinking and cooking water for those households; 0.0035 (µSv/Bq): dose conversion factor for ingestion of radon gas (United Nations Scientific Committee on the effects of Atomic Radiation, 2000).

The values of radon concentrations and annual effective doses are given in Table 1.

Table 1: *Radon concentrations in groundwater (C_0) and total annual effective doses (Total E)*

No	Samples	Coordinates		Average C_o (Bq/l)	E_{INH} (µSv/y)	E_{ING} (µSv/y)	Total E (µSv/y)
		Latitude	Longtitude				
1	PH 1	11.66837	108.29726	3.60 ± 0.27	9.06 ± 0.68	8.11 ± 0.61	17.18 ± 0.91
2	PH 2	11.67974	108.31625	2.95 ± 0.22	7.43 ± 0.57	6.65 ± 0.51	14.07± 0.76
3	PH 3	11.70954	108.32241	8.02 ± 0.59	20.21 ± 1.48	18.09 ± 1.32	38.30 ± 1.98
4	PH 4	11.69702	108.33563	4.47 ± 0.40	11.27 ± 1.01	10.09 ± 0.91	21.36 ± 1.36
5	PH 5	11.70583	108.36443	5.23 ± 0.40	13.19 ± 1.00	11.80 ± 0.89	24.99 ± 1.34
6	PH 6	11.69542	108.38551	6.98 ± 0.53	17.59 ± 1.34	15.74 ± 1.20	33.33 ± 1.80
7	PH 7	11.68823	108.36848	2.46 ± 0.25	6.20 ± 0.62	5.55 ± 0.56	11.75 ±0.83
8	PH 8	11.67636	108.35473	4.42 ± 0.35	11.13 ± 0.88	9.96 ± 0.79	21.09 ± 1.18
9	PH 9	11.65573	108.34864	3.36 ± 0.32	8.46 ± 0.82	7.57 ± 0.73	16.03 ± 1.10
10	PH 10	11.64145	108.34578	2.19 ± 0.15	5.53 ± 0.38	4.95 ± 0.34	10.47 ± 0.50

Radon concentrations in 10 groundwater samples ranged from 2.19 to 8.02 Bq/l, less than the EPA's maximum contamination level (MCL) of 11.1 Bq/l (Ahmed et al, 2017, El-Araby et al, 2019 & Wedad et al, 2015). The total annual effective doses of radon for the public varied from 10.47 µSv/y to 38.30 µSv/y. These doses are lower than the permissible dose of 0.1 mSv/y recommended by WHO (Ahmed et al, 2017, El-Araby et al, 2019 & Wedad et al, 2015). However, the consumers in the study area often boil water before drinking and eating and approximately 50% of the houses there are very naturally well-ventilated, so the true total effective doses for the public were less than the calculated values shown in Table 1 because boiling releases from 10% to 90% of water-soluble radon (Erlandsson, 2001) and reduces the doses for ingestion while the highly well-ventilated houses such as outdoor kitchens decrease the doses for inhalation.

Our research team discovered that almost all consumers in the study area were unaware of radon. Therefore, it is necessary to disseminate the basic knowledge about radon, its effects on human health and the measures to reduce radon levels to enhance health protection, even when the radon concentrations are under the permissible level. Nonetheless, in order to conclude that the radon concentrations in the groundwater sources in the entire Phu Hoi Commune as well as the total annual effective doses for the whole public there are under the permissible limits, our team will have to monitor radon for a longer time and take more samples based on the differences in geographic, topographic, geological, seasonal, climatic and weather conditions, depths of water sources, geo-hydrological processes... as well as assess the true annual effective doses more accurately so that the obtained results are guaranteed to be representative of the whole commune.

4. Conclusion

Radon concentrations in groundwater in 10 households in Phu Hoi Commune, Duc Trong District, Lam Dong Province are all lower than the EPA's permissible limit of 11.1 Bq/l. The total annual effective doses of radon for the public are also under the WHO's recommended limit of 0.1 mSv/year. Therefore, those households can use these groundwater sources for daily domestic consumption without any restriction or intervention. However, our research team still recommends that those households improve their basic knowledge about radon and be aware of minimizing the doses as low as reasonably achievable (ALARA) in order to limit the stochastic effects that radon gas can cause for them by consistently boiling water before eating and drinking and further enhancing the ventilation of indoor air with outdoor air.

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XÁC ĐỊNH NỒNG ĐỘ RADON TRONG NƯỚC NGẦM TẠI XÃ PHÚ HỘI, HUYỆN ĐỨC TRỌNG, TỈNH LÂM ĐỒNG VÀ ĐÁNH GIÁ LIỀU HIỆU DỤNG HẰNG NĂM ĐỐI VỚI DÂN CHÚNG

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TÓM TẮT

Radon là nguồn bức xạ tự nhiên đóng góp khoảng 50% vào liều dân chúng. Do đó, việc đánh giá và giảm thiểu mức độ ảnh hưởng radon đối với sức khỏe con người là một vấn đề rất quan trọng. Trong nghiên cứu này, chúng tôi khảo sát nồng độ radon trong nước ngầm tại xã Phú Hội, huyện Đức Trọng, tỉnh Lâm Đồng bằng hệ đo nhấp nháy lỏng ALOKA LSC 6100. Từ đó, tính toán liều hiệu dụng hằng năm mà dân chúng nhận được khi sử dụng các nguồn nước ngầm nói trên. Hàm lượng radon và liều hiệu dụng thu được đối với dân chúng tương ứng là 2,19 - 8,02 Bq/l và 10,47 - 38,30 µSv/năm. Kết quả cho thấy các giá trị này nằm trong giới hạn an toàn cho phép so với tiêu chuẩn của Cơ quan Bảo vệ Môi trường Hoa Kỳ (EPA) và Tổ chức Y tế Thế giới (WHO) đề ra. Do vậy, đây là nguồn nước có mức radon đảm bảo an toàn và không gây hại đến sức khỏe khi dân chúng sử dụng ăn uống hằng ngày.

Từ khóa: *Nồng độ radon, nước ngầm, nhấp nháy lỏng, liều hiệu dụng hằng năm*