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SURVEY ON THE CONCENTRATION OF RADON (^{222}Rn) AND RADIUM (^{226}Ra) IN DOMESTIC WATER IN BAC TU LIEM DISTRICT, HA NOI CITY

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Abstract. Radon (^{222}Rn) and Radium (^{226}Ra) are natural radiation isotopes that greatly affect human health. Based on the present study, we surveyed the ^{222}Rn and ^{226}Ra concentration in domestic water, including: 12 samples of supply water, 18 samples of treated well water and 30 samples of well water in households in the area of Bac Tu Liem District, Ha Noi City. Radon gas concentration was determined by a specialized measuring device, RAD-7 and Radium gas concentration was determined by Radium adsorption method on MnO_2 plate. The results of the study showed that for the treated well water in the survey area there are still 07 (over 18) samples with Radium concentration of ~ 1.03 to ~ 1.27 times higher than the allowed limit (US EPA), i.e. 0.185 Bq.l^{-1}. The Radon concentrations are: from (0.05 ± 0.02) Bq.l^{-1} to (1.05 ± 0.04) Bq.l^{-1}, (0.8 ± 0.03) Bq.l^{-1} to (3.56 ± 0.08) Bq.l^{-1}, and (1.67 ± 0.08) Bq.l^{-1} to (8.87 ± 0.23) Bq.l^{-1}; in supply water, well treated water, and bore well water, respectively, which are within the limits of allowed values by the Environmental Protection Agency (EPA), i.e. 11.1 Bq.l^{-1}. Samples of well water exhibited concentration of ^{222}Rn, ^{226}Ra higher than those of tap water and treated water. In addition, concentrations of ^{222}Rn and ^{226}Ra in the surveyed water did not have a linear relationship.

Keywords: RAD-7, Radon measurement, Radium measurement method, dose.

Classification numbers: 3.4, 3.4.2.

1. INTRODUCTION

^{222}Rn is a direct decay product of ^{226}Ra in the natural uranium radioactive decay series (^{238}U). The ^{238}U isotope is one of the radioactive isotopes that make up the majority of the environment. Due to the widespread distribution on the Earth's crust, ^{238}U is present in most types of rock and minerals. When there are geological tectonic activities or human influence, underground water sources are formed in the ground [1, 2]. The ^{238}U and its descendants then easily diffuse into different sources. The radionuclides ^{226}Ra and ^{222}Rn are highly radioactive isotopes, often present in water with different content. Each isotope has different chemical properties, but both have harmful effects on humans when ingested. Out of all the Radon isotopes, ^{222}Rn is harmful to humans because it is a radioactive noble gas and emits alpha radiation [3, 4]. Domestic water with a lot of ^{222}Rn for a long time has the risk of cancer of

organs in the body, especially the stomach [3]. In addition, ^{222}Rn diffused from water into the air can cause lung cancer when inhaled. Domestic water with isotope ^{226}Ra is at risk of cancer, bone and tooth degeneration.

According to the US EPA surveyed, Radon concentration water causes about 175 deaths each year, of which 87 % death is caused by inhalation of Radon-containing gas and 13 % is due to domestic water with high Radon [5].

Therefore, it is necessary to investigate the concentrations of ^{222}Rn and ^{226}Ra in different water sources because of their dangers to public health and the environment.

In the content of the article, the authors have measured the concentration of ^{222}Rn and ^{226}Ra in supply water including clean water supplied by water plants, water from pre-treated wells and wells taken from wells in households in Bac Tu Liem District, Ha Noi City, Viet Nam.

2. RESEARCH METHOD

2.1. Research area

Bac Tu Liem District was established December 27, 2013, on the basis of separating 9 wards, including: Thuong Cat, Lien Mac, Tay Tuu, Thuy Phuong, Minh Khai, Phu Dien, Dong Ngac, Xuan Dinh and Co Nhue; 9.3 ha of natural area and 596 inhabitants of Xuan Phuong commune; 75.48 ha of natural area and 10,126 people of Cau Dien Town. Bac Tu Liem District has geographical coordinates of 21°03′15″ North latitude 105°40′56″ Eastern longitude. The administrative boundary of Bac Tu Liem District: The East borders Tay Ho district; bordering on Cau Giay District to the Southeast; the West borders Dan Phuong and Hoai Duc districts; the South borders Nam Tu Liem district; Dong Anh district in the North. It is located 16 km west of the center of Ha Noi Capital [6].

Bac Tu Liem District currently has 13 wards: Co Nhue 1, Co Nhue 2, Dong Ngac, Duc Thang, Lien Mac, Minh Khai, Phu Dien, Phuc Dien, Tay Tuu, Thuong Cat, Thuy Phuong, Xuan Dinh and Xuan Tao. Bac Tu Liem District has an area of 4,335.34 ha (43.35 km²), the population in 2019 is 333,675 people [6].

Because Bac Tu Liem District was separated from the former Tu Liem District with the characteristics of agricultural production, many items for people's livelihood have not been invested, including providing clean water for people, some wards. Many households still have to use well water to serve daily activities.

2.2. Water sampling

Radioactive concentrations of ^{222}Rn and ^{226}Ra were determined in some tap water samples, pre-treated well water samples and untreated bore wells in schools and households in Bac Tu Liem District, Hanoi City. The authors took water samples in Co Nhue, Minh Khai, Tay Tuu, Lien Mac and Thuy Phuong Wards. The number of samples is shown in Table 2 and the sampling location diagram is depicted in Figure 1.

There are many different factors affecting the accuracy of the results such as: the amount of water taken in the measured sample, the measurement time, air temperature, the humidity, the background and the environment [7], etc. in which the most important factor is the technique water sampling. Sample water according to the following principles [8, 9]: Water needs to be drained for a while before taking it to stabilize the water flow; the sampling device is rinsed at least twice before it is taken with the water itself.

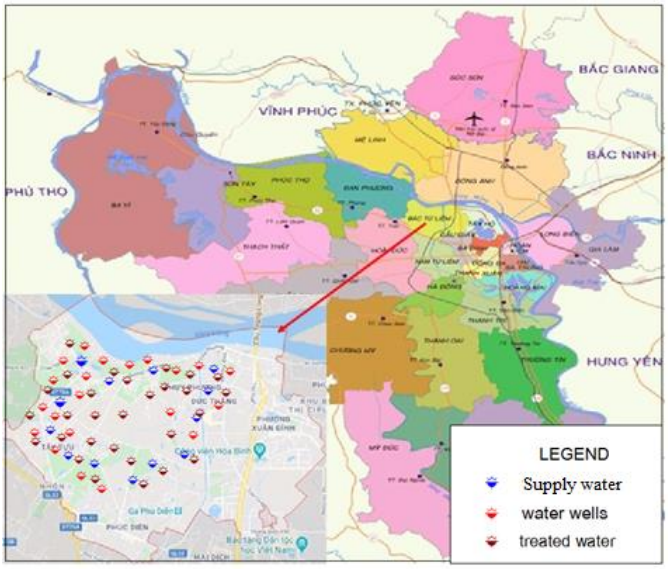


Figure 1. Location of water sampling in the survey area [10].

In addition, the rate of discharge of water also causes a loss of concentration of ^{222}Rn (Table 1). In this study, water samples were taken at a rate of 1-2.5 liters/minute so it is possible to ignore the loss of ^{222}Rn due to the flow rate.

Table 1. The amount of ^{222}Rn lost by the speed of water [9].

No.	Flow rate (liters/minute)	Amount of ^{222}Rn lost (%)
1	~ 4	5.5
2	~ 6	14
3	~ 8	25
4	10-12	32

Table 2. The number of samples collected.

No.	Region (ward/commune)	The number of samples collected			Volume (litre)	Sampling time
		Supply water	Treated well water	Well water		
1	Co Nhue	02	03	05	2	12/1/2020
2	Minh Khai	02	03	05	2	12/1/2020
3	Tay Tuu	02	03	05	2	13/1/2020
4	Lien Mac	02	03	05	2	13/1/2020
5	Thuy Phuong	02	03	05	2	14/1/2020
6	Dong Ngac	02	03	05	2	14/1/2020
	Total	12	18	30		

During sampling, using one end of the plastic tube attached to the drain hose, the other end into the vial, drain the water through the tube until it is overflowing, cover tightly until the sample has not been measured.

2.3. Method of determination of Radon concentration

In the content of the article, the authors determined the Radon concentration through alpha-method, using RAD7 Radon Detector (RAD-7) by DURRIDGE Company. This equipment was calibrated before using. RAD-7 is a device that specializes in measuring Radon concentrations with high sensitivity. RAD-7 consists of a hemispherical chamber of about 700 ml volume. The semiconductor silicon detector is located at the center of the hemisphere. The advantage of the machine is the ability to determine the energy of each alpha particle, thereby distinguishing the isotopes of Radon ^{222}Rn, ^{220}Rn, ^{219}Rn new and old, signal and noise. The device comes with an accessory kit that allows to determine the concentration of Radon in water with a sensitivity from below 4 to over 400,000 Bq.m^{-3} [9]. The RAD-7 device determines the concentration of ^{222}Rn through the isotopes daughters ^{218}Po, ^{214}Po. The relative humidity in the chamber affects the ability of these isotopes to detect. RAD-7 should be dried before each measurement so that the moisture remains below 10 % during measurement [8].

To ensure accurate results, backgrounds were defined. Samples of distilled or reduced ^{222}Rn water were measured for 10 times in the same mode as the analytical sample. Average measured background value was (0.01-0.04) Bq.l^{-1}.

The RAD-H_2O method uses a closed-loop air cycle in which the amount of gas and water are constant. The efficiency of taking ^{222}Rn from water into the air-tight loop reaches up to 99 % for 50 ml vials and 94 % for 250 ml vials. The 250 ml vial is used for water samples with concentrations lower than 100 Bq.l^{-1}, whereas the 50 ml vial is used [9].

The Radon concentration diagram is given in Figure 2.

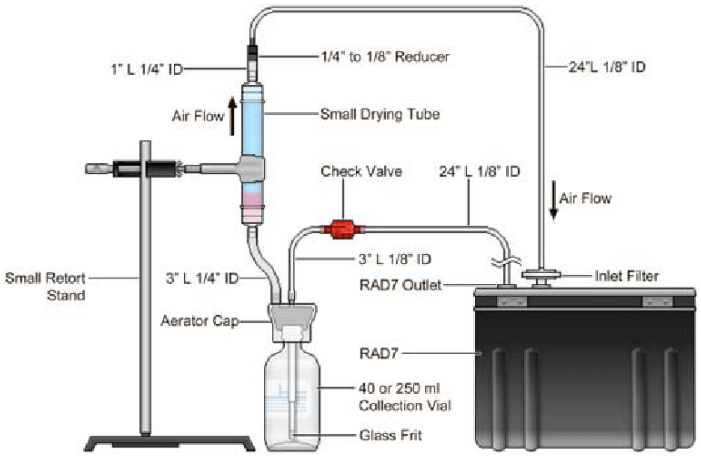


Figure 2. Diagram of Radon concentration in water samples.

2.4. Method of determination of Radium concentration (^{226}Ra)

To determine the concentration of ^{226}Ra in water samples, the authors used Radium adsorption method on MnO_2 plate. The amount of ^{226}Ra is adsorbed on the surface of MnO_2

coated plastic plate in alkaline environment. This is a form of non-electrolytic electrolysis that takes advantage of the different chemical potential of metal ions compared to the surface of MnO_2. This process is highly selective because most of the other ions are present in the chemical solution and cannot cling to the surface of the plastic plate [11].

The manufacturing process of MnO_2 sample was carried out as follows:

** Mixing chemicals*

Mix about 500 ml of $KMnO_4$ in 0.1M solution

** Preparation of an MnO_2 dish*

Used (PA) plastic was cut into small, round discs with a diameter of 2.2 cm and 0.5 mm thick, that were washed with diluted ethanol.

Soak the plastic discs in $KMnO_4$ solution at a concentration of 0.1 M at a temperature of 50 -70 ^oC, in 3 hours, the research group obtained a plastic disc covered with a MnO_2 layer, thick enough to adsorb Radium (^{226}Ra) well. After soaking for 3 hours, remove and rinse the discs with distilled water and dry in room temperature.

** Preparation of sample solution*

We analyzed some water samples and standard ^{226}Ra solution of activity $1Bq.l^{-1}$. For some acidic water samples we neutralized the NH_4OH band to bring the pH to a range of 7-8.5 before the analysis.

** Preparation of measuring sample*

Place the sample cup with a MnO_2 coated plastic dish on the stove, stirring gently for 6 hours. Take care not to stir vigorously the fired sample, causing errors and may damage the MnO_2 layer. After stirring for 6 hours, remove the dish from the solution, rinse with distilled water and allow to dry in the air.

** Measurement of the sample using the alpha spectrometry system*

Place a plastic dish containing the sample on the alpha spectrometer, minus the background value, taking the actual number of samples. The measurement time depends on the radioactivity of ^{226}Ra present in the sample, usually to ensure statistical errors with the environmental sample. The minimum measurement time was about 86,400 seconds (24 hours). It is essential that before the measurements it is to examine the background of the two areas of measurement energy of interest (ROI) of ^{226}Ra and of the isotope of markings.

** Determination of Radium concentration*

The ^{226}Ra isotope was separated from the sample with an efficiency of over 90%, then analyzed by an alpha spectrometer, emitting alpha rays with an energy of 4.7 MeV. Use the standard ^{226}Ra solution to determine the recovery efficiency of all ^{226}Ra transfer processes from the sample to a MnO_2 coated plastic disc.

The activity of the ^{226}R isotope was calculated according to the energy spectral line of 4.7 MeV and is calculated using the following formula [11]:

$$A_{Ra} = \frac{C - C_o}{V \times H_G \times H_T} \times 1000 \quad (1)$$

Inside: A_{Ra}: The activity of ^{226}Ra present in the sample at the time of electrolysis on MnO_2 plastic plates ($Bq.l^{-1}$); C: Counting rate of alpha particles at energy peak of 4.7 MeV; C_0:

Background counting rate; V: Sample volume (liters); H_G: Counting efficiency of alpha system; H_T: Chemical recovery efficiency (> 90 %).

2.5. Calculation of the annual effective dose

The annual effective dose contributed to a person domestic water containing ^{222}Rn or ^{226}Ra as En(Sv) was determined by the following formula:

$$E_n = \varepsilon \times Vn \times C_n \quad (2)$$

in which: ε(Sv.Bq^{-1}) is the effective dose conversion ratio per unit of radioactive concentration, for ^{222}Rn, $\varepsilon = 10^{-8}$ Sv.Bq^{-1} [7]; for ^{226}Ra, ε= 2,8.10^{-}7 Sv.Bq^{-1} [1]; V_n = 730 liters.year^{-1}, which is the volume of water each person drinks every year [12]; C_n (Bq.l^{-1}) is the concentration of ^{222}Rn or ^{226}Ra in water.

In addition, ^{222}Rn in water diffuses into the air. Therefore, the air contains significant amounts of ^{222}Rn released from water. Suppose the water source is placed indoors with the volume of air a person inhales daily is V = 20 m^3 and the volume of water discharged in one hour is W = 0.01 m^3/hour. Then the annual effective dose of ^{222}Rn inhaled from water D (Sv) was calculated by the following formula [13, 14, 15]:

$$D = D = f \times C_n \times (\varepsilon_f + \varepsilon_p F) \quad (3)$$

in which:

$$f = W \times e / V\lambda = 3{,}571 \times 10^{-4} \quad (4)$$

e = 0.5 is the conversion coefficient ^{222}Rn from water into air; λ= 0.7 is the air exchange rate; C_n (Bq.m^{-3}) is the concentration of ^{222}Rn in water; ε_f = 0.33 µSv.m^3/year/Bq is the effective dose conversion coefficient for ^{222}Rn; ε_p = 80 µSv.m^3/year/Bq is the effective dose conversion ratio for ^{218}Po, ^{214}Bi, ^{214}Po.

F = 0.4 is the equilibrium coefficient between 222Rn and descendants.

3. RESULTS AND DISCUSSION

3.1. The concentration of ^{222}Rn in water

3.1.1. Well water from pretreatment

The concentration of ^{222}Rn in well water pre-treated ranged from (0.08 ± 0.03) Bq.l^{-1} to (3.56 ± 0.08) Bq.l^{-1}, lower than the limit concentration from 3.12 to 138.75 times [16]. The average concentration (1.82 ± 0.04) Bq.l^{-1}. Accordingly, the effective annual dose ranged from (0.58 ± 0.22) µSv.year^{-1} to (13.29 ± 0.58) µSv.year^{-1}. The average effective dose was (15.91 ± 0.29) µSv.year^{-1}. If the water source is placed indoors, the annual effective dose for inhalation of ^{222}Rn from the water is approximately (0.96 ± 0.36) µSv.year^{-1} to (42.72 ± 0.96) µSv.year^{-1}. Average effective dose is (21.84 ± 0.48) µSv.year^{-1}.

3.1.2. Clean water supplied by water factory

The concentration of ^{222}Rn in tap water ranged from (0.05 ± 0.02) Bq.l^{-1} to (1.05 ± 0.04) Bq.l^{-1}, lower than the limit of concentration from 10.6 to 222 times [16]. The average concentration (0.38 ± 0.06) Bq.l^{-1}. Thus, the effective annual oral dose ranged from (0.36 ± 0.15)

μSv.year^{-1} to (7.67 ± 0.29) μSv.year^{-1}. Average effective dose was (2.77 ± 0.44) μSv.year^{-1}. If the water source is placed indoors, the annual effective dose for inhalation of ^{222}Rn ranges from (0.60 ± 0.24) μSv.year^{-1} to (12.60 ± 0.48) μSv.year^{-1}. Average effective dose is (4.56 ± 0.72) μSv.year^{-1}.

3.1.3. Well water

The concentration of ^{222}Rn in bore well water ranged from (1.67 ± 0.08) Bq.l^{-1} to (8.87 ± 0.23) Bq.l^{-1}, lower than the limit concentration from 1.25 to 6.65 times [16]. Median concentration (3.34 ± 0.12) Bq.l^{-1}. Accordingly, the effective annual dose ranged from (12.19 ± 0.58) μSv.year^{-1} to (64.75 ± 1.67) μSv.year^{-1}. Average effective dose is (24.38 ± 0.88) μSv.year^{-1}. If the water source is placed indoors, the annual effective dose for inhalation of ^{222}Rn is approximately (20.04 ± 0.96) μSv.year^{-1} to (106.44 ± 2.76) μSv.year^{-1}. Average effective dose is (40.08 ± 1.44) μSv.year^{-1}.

3.2. The concentration of ^{226}Ra in water

3.2.1. Treat well water

Concentrations of ^{226}Ra in well water pre-treated in some houses in Bac Tu Liem District ranged from (0.077 ± 0.024) Bq.l^{-1} to (0.235 ± 0.073) Bq.l^{-1}, whose the medium concentration average was 0.112 ± 0.022 Bq.l^{-1}. In the investigation area, there were 07 samples of treated well water with concentrations higher than the limit concentration from 1.03 to 1.27 times [13]. Accordingly, the effective daily dose for domestic water containing ^{226}Ra is from (15.38 ± 4.91) μSv.years-1 to (48.03 ± 14.92) μSv.year^{-1}. Average effective dose is (25.35 ± 4.50) μSv.year^{-1}.

3.2.2. Supply water

The concentration of ^{226}Ra in tap water in some houses in Bac Tu Liem District ranges from (0.062 ± 0.024) Bq.l^{-1} to (0.171 ± 0.082) Bq.l^{-1}, lower than the limit concentration from ~ 1.08 to ~ 3.24 times [13]. Average concentration is (0.094 ± 0.023) Bq.l^{-1}. At that time, the annual effective dose for domestic water containing 226 rang from (11.71 ± 11.88) μSv.years^{-1} to (35.16 ± 17.01) μSv.year^{-1}. Average effective dose (18.98 ± 3.48) μSv.year^{-1}.

3.2.3. Well water

The concentration of ^{226}Ra in bore water in some houses in Bac Tu Liem District ranges from (0.087 ± 0.043) Bq.l^{-1} to (0.64 ± 0.121) Bq.l^{-1}. In the investigation area, there are 12 well water samples with concentrations higher than the limit concentration from 1.04 to 3.48 times [16]. Average concentration (0.178 ± 0.023) Bq.l^{-1}. Accordingly, the effective annual dose due to domestic water containing 226 ranges from (17.78 ± 8.79) μSv.year^{-1} to (131.63 ± 24.73) μSv.year^{-1}. Average effective dose is (36.39 ± 4.74) μSv.year^{-1}.

Comparison of the average of ^{222}Rn and ^{226}Ra in three types of surveyed water shows that: The average concentration of ^{222}Rn in well water is 1.5 times higher than the average concentration of ^{222}Rn in treated well water and 8.8 times the average ^{222}Rn concentration in tap water; The average ^{226}Ra concentration in well water is about 1.4 times higher than the average ^{226}Ra concentration in treated well water and about 1.9 times the average ^{226}Ra concentration in tap water (Figure 3).

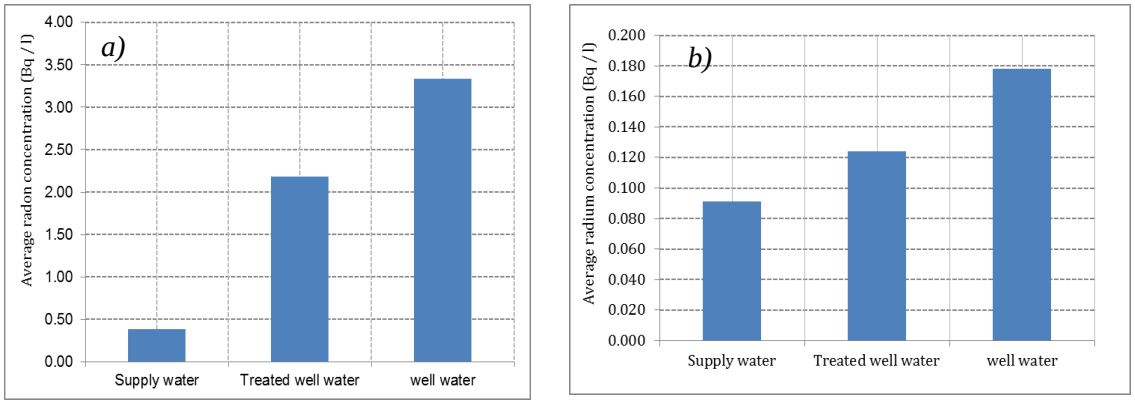
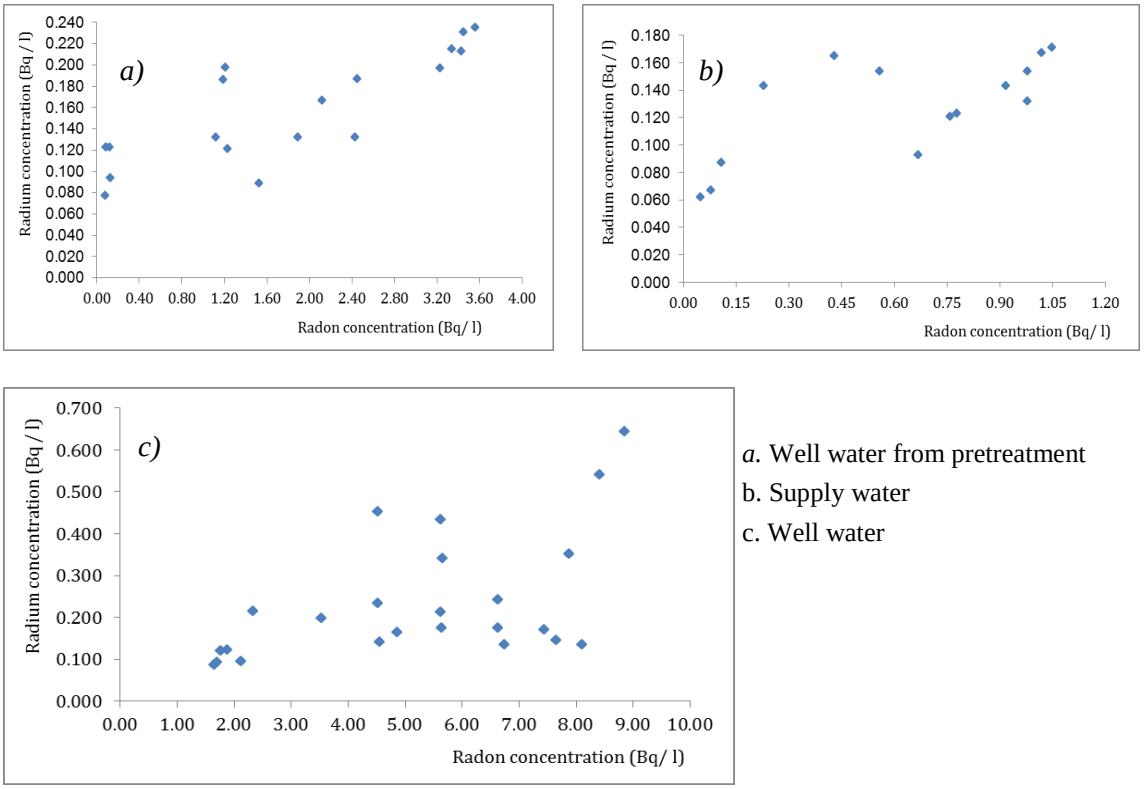


Figure 3. A graph comparing the concentrations of ^{222}Rn (a) and ^{226}Ra (b) average in the surveyed water.

Thus, it can be seen that concentrations of ^{222}Rn and ^{226}Ra depend on the origin of the water. Well water has a higher radiation concentration than the other two. This is due to:

- Mostly treated well water samples have been pre-treated through many stages before being brought to users so the amount of ^{222}Rn, ^{226}Ra has been significantly limited.

- Well water is underground water originated from rock and soil and has not been treated, so the concentrations of ^{222}Rn and ^{226}Ra are still quite high.



a. Well water from pretreatment
b. Supply water
c. Well water

Figure 4. The correlation between Radon and Radium concentration in water samples.

The survey results also showed that the difference of concentration of ^{226}Ra among surveyed water samples was lower than the difference of concentration of ^{222}Rn. This is because ^{222}Rn is in gaseous form, the possibility of loss through the treatment stages will be higher than ^{226}Ra. The difference in radioactive concentration between water samples is due to: characteristics of forming water sources, different geological characteristics in the intake areas, the existence of ^{238}U isotopes in rock or water soil, water depth, temperature, flow rate, treatment, transport and storage, water intake, precipitation, etc.

Because ^{222}Rn is a direct decay product from ^{226}Ra, by the survey results, the authors have built a correlation between them. Figure 4 shows the relationship between the concentration of ^{222}Rn and the concentration of ^{226}Ra in the surveyed water samples. The results showed that, in the water samples surveyed the concentrations of ^{222}Rn and ^{226}Ra did not have a linear relationship (correlation coefficient R^2 = 0.279 - 0.567). For the most parts, the existence of ^{222}Rn in water depends not only on the amount of ^{222}Rn in the water but also on the amount of ^{238}U in the soil and the geological characteristics of each area.

4. CONCLUSIONS

The results showed that the concentration of ^{222}Rn in all surveyed water samples was within the recommended limits of EPA, 11.1 Bq.l^{-1}.

In the survey area, there were 12 samples of well water and 07 samples of pre-treated well water with a concentration of ^{226}Ra exceeding the recommended level of 0.185 Bq.l^{-1}. Therefore, it is necessary to take measures to reduce ^{226}Ra before using for eating purposes.

The concentration of radiation depends on the type of water. In general, samples of well water (groundwater) have ^{222}Rn, ^{226}Ra concentrations higher than samples of tap water, treated water (surface water). The difference of radioactive concentration mainly depends on the origin of the water type, the geological environment of the water source.

Concentrations of ^{222}Rn and ^{226}Ra in the surveyed water sources have non linear relationship. This shows that ^{222}Rn is not only sourced from ^{226}Ra in the surveyed water sources. Most of the ^{222}Rn is generated from the ^{238}U decay chain in the soil. However, its existence in water depends on many influences from the geological environment.

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