Study of Radon and Radium Concentration in Water Samples in Some Regions of Lebanon

دراسة تراكيز الرادون والراديوم في عينات المياه في بعض المناطق في لبنان

Abdalsattar K. Hashim, Burhan R. Al Safaay, Fadhil K. Fulyful
Department of physics, College of science, Karbala University, Iraq.
abdalsattarkareem@yahoo.com

Abstract:

This study describes the results of measurements radon (^{222}Rn) and radium (^{226}Ra) concentrations found in sixteen water samples collected from different locations in Lebanon. Measurements were made, using long-term technique for alpha particles emission with solid state nuclear track detector type CR-39.

The concentrations of ^{222}Rn ranges from 1.08 Bq /L to 9.32 Bq/ L; while the mean value was 2.50 Bq/ L. All results were below the maximum contaminant level (MCL) for ^{222}Rn in drinking water as reported by Environmental Protection Agency (EPA). However, these values were lower than allowed maximum contaminant level (MCL) for ^{222}Rn in drinking water as reported by USA Environmental Protection Agency (EPA) which is about 11.1 Bq/L.

Also, ^{226}Ra alone, in one sample have concentration higher than 0.555 Bq/L as normal level for gross alpha. The maximum concentration of radium was found to be 0.561 Bq/ L and the minimum was 0.065 Bq /L with mean value 0.150 Bq/L. For improvement of the social health level, it is essential that to reduce the radon and radium concentrations in the drinking water before using by people.

Keywords: CR-39 plastic nuclear track detectors, Radon -222 concentration levels, Radium-226, water samples, Lebanon, MCL.

ملخص:

توضح هذه الدراسة نتائج قياسات تراكيز غاز الرادون (^{222}Rn) والراديوم (^{226}Ra) التي وجدت في ستة عشر من عينات المياه التي تم تجميعها من مواقع مختلفة في لبنان. أجريت القياسات باعتماد تقنية القياس طويلة الأمد لانبعاث جسيمات ألفا باستخدام كاشف الأثر النووي الصلب نوع CR-39.

وقد وجدت تراكيز (^{222}Rn) من 1.08 Bq/L إلى 9.32 Bq/L ، في حين أن متوسط قيمة هذه التراكيز بلغت 2.50 Bq/L. لقد كانت جميع تراكيز الرادون في عينات المياه التي تمت دراستها أقل من تركيز الرادون المسموح به في ماء الشرب البالغ 11.1 Bq/L والذي توصي به وكالة الحماية البيئية الأمريكية(EPA).

كذلك الراديوم ^{226}Ra، في عينة واحدة وجد لديه تركيز أعلى من 0.555Bq/L المستوى العادي لألفا الإجمالي .تم العثور على الحد الأقصى لتركيز الراديوم ليكون 0.561Bq/L وكان الحد الأدنى 0.065Bq/L مع متوسط قيمة 0.150Bq/L وهذه القيمه اقل من المستوى العادي لألفا الإجمالي. ولتحسين المستوى الصحي ، فمن الضروري تقليل تراكيز غاز الرادون والراديوم في مياه الشرب قبل استخدامها من قبل الناس.

1. Introduction

Uranium, the heaviest radioactive toxic element is found in almost all types of soils, rocks, sands and water**.** The dominant isotope, uranium-238, forms a long series of decay products that include the key radionuclide's radium-226, and radon-222. Radon is the decay product of radium in the naturally occurring uranium series. As an inert gas, radon can move freely through the soil from its source to a distance which is determined by many factors such as rate of diffusion, effective permeability of the soil and its own half-life [1]. Being a natural alpha emitter, radon can be detected by an alpha sensitive detector. It has been established that radon is a causative agent of lung cancer when present in higher concentrations [2]. Henshaw et al. (1990) has claimed that indoor radon exposure is associated with the risk of leukemia and certain other cancers, such as

melanoma and cancers of the kidney and prostate [3]. If uranium rich material lies close to the surface of earth there can be high radon exposure hazards [4- 6].^{226}Ra is often used as a standard for the evaluation of contamination by transuranic elements. The ^{226}Ra in the environment is widely distributed, being present in various concentrations in waters, soils, sediments and rocks. The quantities of natural radionuclides in water depend upon the quality of the parent rock. The radioactivity of natural radionuclides in granite is somewhat higher than in other rocks **[7].** Rocky, mountainous regions and phosphate rich soil regions and water, all over the world, tend to have varying amounts of Rn-222 [8, 9]. Radon is unstable and breaks down into radon progeny emitting highly ionizing alpha radiation which is very harmful to humans when they are inhaled or swallowed [10]. Radon is a colorless, odorless and tasteless gas. It is a chemically and biologically inert noble gas with a heavily neutron-rich nucleus that makes it a radioactive element [11]. Radon is a natural radioactive noble gas and has three major isotopes, ^{219}Rn (Actinon), ^{220}Rn (Thoron) and ^{222}Rn with half-lives of 3.96 seconds, 55.6 seconds and 3.82 days, respectively [12, 13]. The aim of this study is to measure the radium and radon concentrations in water samples for some regions in Lebanon.

2. Material and Methods

In this work, sixteen water samples were collected from different locations in some areas of Lebanon, shown in figure 1. For each sample about (58 g and 83.154 cm^3) was placed at the bottom of a cylindrical sealed can of 7 cm height and 5.5cm diameter. Solid State Nuclear Track Detector (SSNTD) with sheet thickness 300μm was used in this study, which is usually known as CR-39 plastic detector [14]. Square pieces of detector of size 1 cm × 1 cm were fixed on the top of inner surface of the can, in such a way, that it is sensitive surface always facing the water sample [15]. The detectors were exposed for a period of about 97 days (from 2-10-2013 to 7-1-2014). During exposed period, the sensitive side of the detector always faced the sample and is exposed freely to the emergent radon from the water sample in the can, so that it could record alpha particles resulting from the decay of radon in the remaining volume of the can.

After completion of the exposure, the detectors were collected and chemically etched using 6N KOH at 70 C° for 6 h. after this chemical treatment, these SSNTDs were washed, dried and scanned using an optical microscope with magnification of 400X (40x objective and 10x eyepiece) was used to count the number of tracks per cm^2 in each detector. The determinations of the concentrations of alpha particles from radon gas in samples were performed by using CR-39 from the intercast Europe srl company. The radon gas and radium concentrations in water samples was obtained by using the sealed-cup technique as shown in figure 2.

Fig.(1): The map of Lebanon

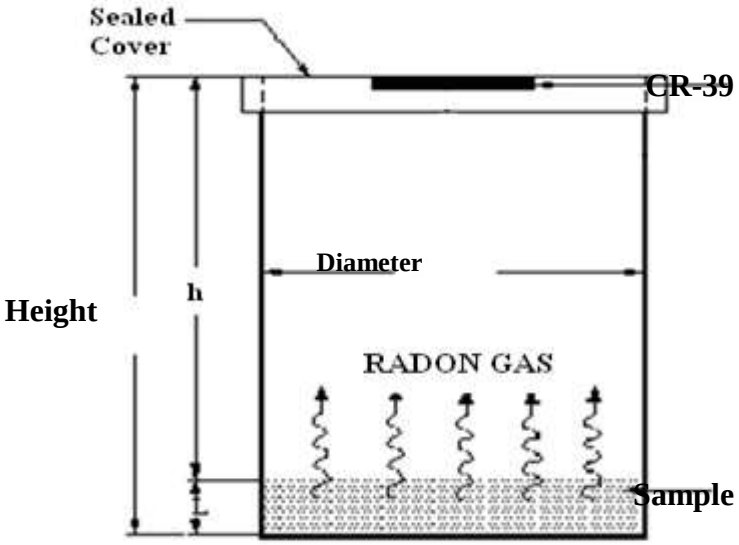

Fig. 2: Experimental set-up for the measurement of radon and radium concentrations in water samples.

3. Results and Discussion

To calculate the concentration of radon in water samples, we will use equation [16]:

$$C_w = C_a\left(\frac{\lambda hT}{L}\right) \qquad (1)$$

where,

C_w : Radon gas concentration in sample water (Bq/m^3) or Bq/L.

C_a: Concentration of radon in the air abovethe water sample Bq/m^3 or Bq/L.

λ: Decay constant of radon gas and is equal to 0.1814 day $^{-1}$.

h: high of detector from the surface of the water (3.5cm).

T: Time exposing the detector to sample the water in days.

L: Height of the water inside the can (3.5cm).

The radon activity density C_a(concentration of radon)in the can air above the water samples was determined by measuring the tracks density on the detector according to the following relation[17, 18]:

$$C_a = \frac{\rho}{K\,T} \qquad (2)$$

where ρ is the measured surface density of tracks on the exposed detectors (Tr/cm^2), T is the exposure time(97 day) and K is the ^{222}Rn gas diffusion constant (calibration factor). The track densities were related to the radon concentration level using calibration factor of 5.7954×10^{-2}Track.cm^{-1}.day^{-1} /Bq.m^{-3}. The radium concentration of the water samples can be calculated by using the relation [19].

$$C_{Ra} = \frac{\rho}{K\,T_e} \qquad (3)$$

T_e: is the effective exposure time which is related with the actual exposure time T and decay constant λ for ^{222}Rn with the relation [20].

$$T_e = [T - \lambda^{-1}(1 - e^{-\lambda T})] \qquad (4)$$

In the presented research, the calculated values of radium and radon (airborne and dissolved) concentration for water samples collected from various places in Lebanon are presented in table 1. The fourth and fifth columns of table 1, present the concentrations of radon in the can air above the water samples by units Bq/m^3 and Bq/L , respectively. While the sixth column of table 1, shows that the dissolved radon concentration for water samples. It is noteworthy from table 1 and also figures 3 that the radon concentration of water samples is least in Riyaq (1.08Bq/L) but highest in North Ba'labak (9.32 Bq/L). In addition to that, the average value of radon concentration was

2.50Bq/L and standard deviation 1.96 Bq/L, as shown in figure 3b. Therefore, the radon levels in water samples are comparatively low since the recommended maximum contaminant level (MCL) of U.S. Environmental Protection Agency is 11.1 Bq/L[21].

It is clear from table 1 that all results are below the maximum contaminate level (MCL) for radon gas in drinking water, as well as , the mean value of radon activity in water sample is about four times lower than the value of (MCL) for radon concentration in drinking water.

The seventh column of table 1, present the concentrations of radium in the water samples. Also, The values of radium concentration are found to be maximum in North Ba'labak (0.561)Bq/L and minimum in Riyaq (0.065 Bq/L). In addition to that, the average value of radium concentration was 0.150Bq/L and standard deviation 0.118Bq/L , as shown in figure 4b .

Specific drinking water standards have not been established for radium- 226 or other alpha emitters, but in one sample (fourth sample) of total samples, ^{226}Ra concentration even higher than MCL for gross alpha is (0.555Bq/L) that determined by U.S Environmental Protection Agency [22]. Also, the mean value of radium concentrations in water sample is lower than the value of (MCL) for gross alpha in drinking water. The measurements indicate different levels of radium and radon concentration in water samples. It can be seen from the results that the radon and radium concentrations vary appreciably from sample to sample. It is due to the fact that the water samples collected from various sites for water samples in some regions of Lebanon may have different Uranium contents.

Table 1. Radon and Radium concentrations of water samples for sixteen different sites in Lebanon.

Number sample	Regions	ρ Trac/cm^2	C_a Bq/m^3	C_a Bq/L	C_w Bq/L	C_{Ra} Bq/L
1	Zahlah	1092.49	194.34	.194	3.42	.206
2	Beirut	372.44	66.25	.066	1.16	.070
3	Juniyah	422.10	75.08	.075	1.32	.079
4	North Ba'labak	2979.52	530.01	.530	9.32	.561
5	Bikfayya	496.58	88.33	.088	1.55	.093
6	Sidon	595.90	106.00	.106	1.86	.112
7	Ba'labak	1117.32	198.75	.199	3.49	.210
8	Alayh	720.05	128.08	.128	2.25	.135
9	Ad Damur	769.71	136.92	.137	2.40	.145
10	Barja	1018.00	181.09	.181	3.18	.192
11	Riyaq	347.61	61.83	.062	1.08	.065
12	Az Zahrani	446.92	79.50	.080	1.39	.084
13	As Sarafand	695.22	123.67	.124	2.17	.131
14	An Nabatiyah	546.24	97.17	.097	1.71	.103
15	Bayt ad Din	645.56	114.83	.115	2.02	.121
16	Jazzin	571.07	101.58	.102	1.78	.107
Maximum value		2979.52	530.01	.530	9.32	.561
Minimum value		347.61	61.83	.062	1.08	.065
Mean value		802.29	142.71	.143	2.50	.150
Standard deviation		647.77	111.86	.112	1.96	.118

Figures 3a and 4a have shown the distribution of radon and radium concentrations in water samples for sixteen different locations in Lebanon, respectively.

Also, Figures 3b and 4b have shown the maximum, minimum, mean and standard deviations of radon and radium concentrations in water samples for sixteen different locations in Lebanon, respectively.

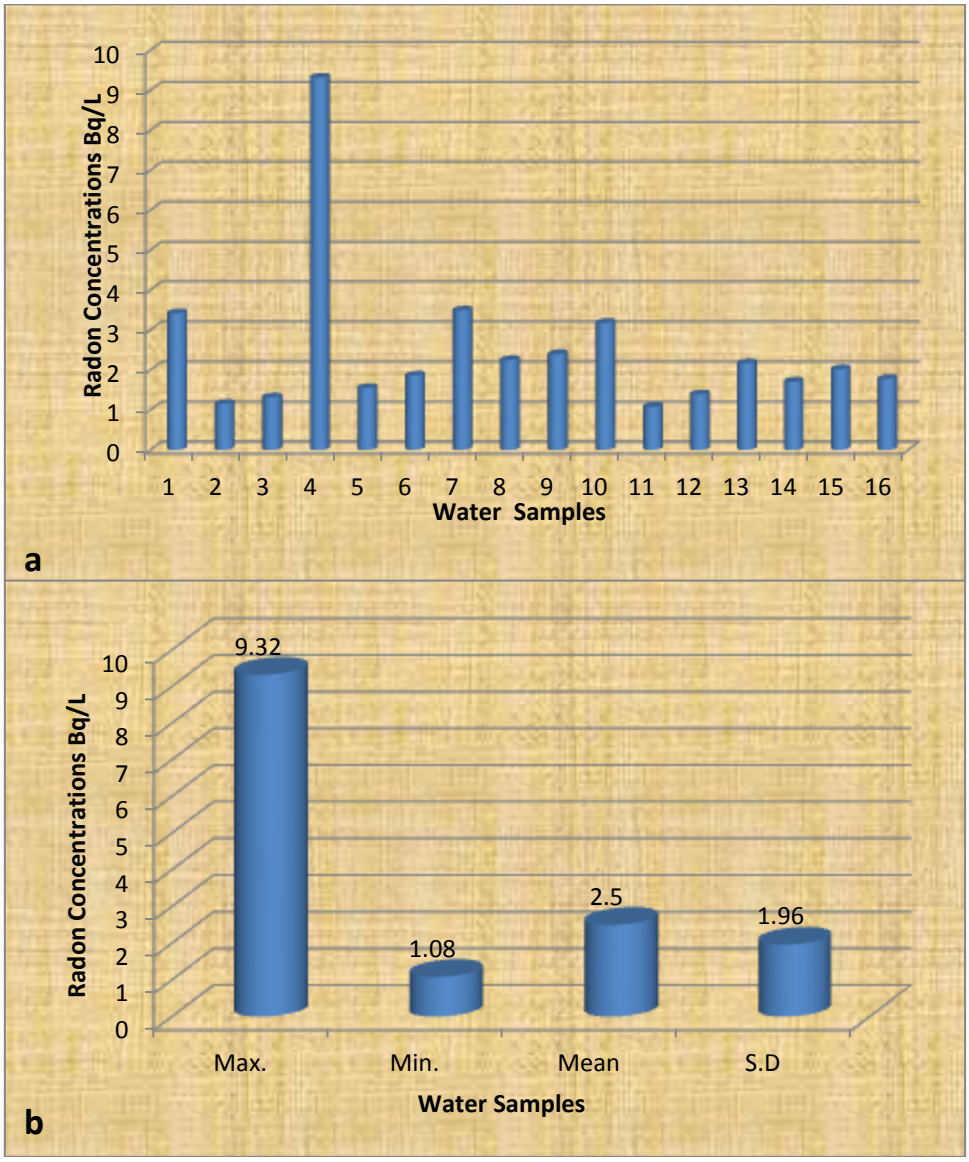

Fig. 3: a) Distributions of radon concentrations for sixteen different water samples. **b)** Maximum, minimum, mean, and standard deviation of radon concentrations for all water samples.

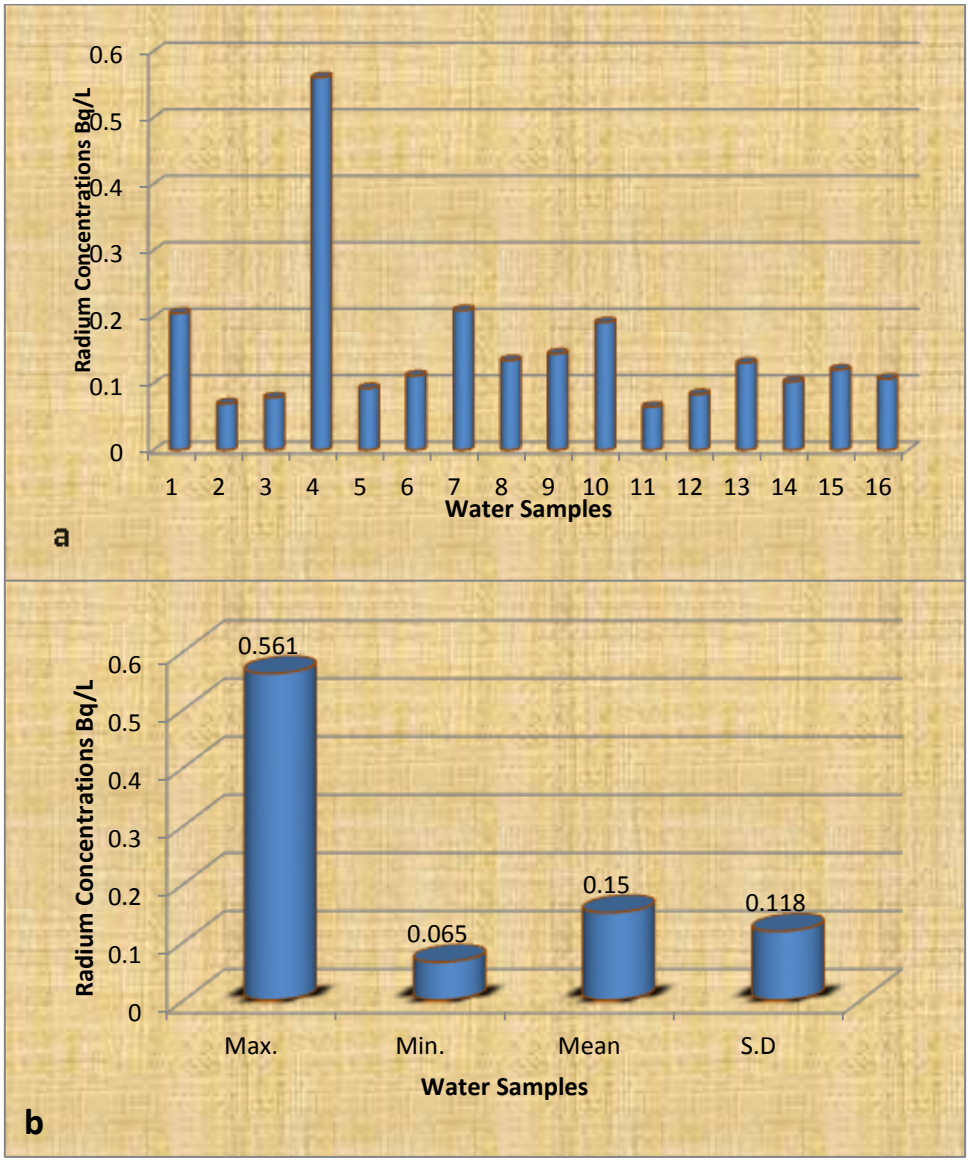

Fig. 4: a) Distributions of radium concentrations for sixteen different water samples. **b)** Maximum, minimum, Mean and standard deviation of radium concentrations for all water samples.

4. Conclusions.

In conclusion, we found that the radon levels in sixteen water samples are within the internationally acceptable values. The results showed that radon concentrations in water samples were ranged between (1.08-9.32) Bq/L and the highest concentrations was found in north B'alabak region, whereas the lowest concentration was found in Riyaq region. These results also show that, the mean value of radon concentrations in water sample is about four times lower than the value of MCL for radon concentration in drinking water.

Clearly the present results shown that the radon concentrations in all water samples were below the allowed limit coined by EPA for ^{222}Rn in drinking water. The results of Radium concentrations of all water samples are small and < 1 Bq/L, but one sample (fourth sample) is upper than 0.555Bq/L.

The mean value of radon and radium concentrations is lower 11.1Bq/L and 0.555Bq/L, respectively. Therefore, radon and radium concentration in the water samples of some regions in Lebanon is not high and this is appropriate.

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