Measurement of radon and radium concentrations in different types of water samples in Al-Hindiyah city of Karbala Governorate, Iraq

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Abstract

The largest fraction of the natural radiation exposure we receive comes from a radioactive gas, radon and its decay products (nearly 50%). The radon gas can enter to the body via respiring, drinking and eating and caused damage to respiratory and digestion systems. Also radium-226 is widely distributed in the environment. When a person ingests radium, this element deposits in bones and cause to bone cancer. In this study radon and radium concentrations of the 25 water samples of of Al-Hindiyah city of Karbala governorate in middle of Iraq have been measured by CR-39 detectors. Total samples including 13 samples of tap water(T.W), 6 samples of ground water(G.W) and 6 samples of drill wells water(D.W) has been tested. All samples of tap water have radon concentration lower than 11.1 Bq/L as normal level. Also, ^{226}Ra alone, in all samples have concentration lower than 0.555 Bq/L as normal level for gross alpha.

All samples of drill wells water had radon concentrations higher than 11.1 Bq/L as normal level, while the results showed that radium concentrations were ranging between (0.527-0.644)Bq/L with mean value 0.586 Bq/L, this value higher than 0.555 Bq/L as normal level for gross alpha.

The maximum concentration of radon in ground water samples was found to be 13.573 Bq/L and the minimum was 9.169Bq/L with mean value 11.790Bq/L, this value higher than 11.1 Bq/L as normal level. Also, the results showed that radium concentrations were ranging between (0.340-0.540)Bq/L with mean value 0.437 Bq/L, this value is lower than 0.555 Bq/L as normal level for gross alpha.

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: Radium content, Radium and Radon Concentrations, CR-39 detectors, Can technique, Water.

قياس تراكيز الرادون والراديوم في عينات مختلفة من مياه مدينة الهندية في محافظة كربلاء/ العراق

الخلاصة

الرادون والتحلل الناتج منه يمثل الجزء الاكبر الذي نتعرض له من الاشعاع الذي ياتي من الغازات المشعة، ويمكن ان يدخل الرادون الى الجسم عن طريق التنفس والشرب والاكل ويسبب الضرر الى انظمة التنفس والهضم.

ايضا الراديوم -226 ينتشر على مدى واسع في البيئة. عندما يبتلع الشخص الراديوم، هذا العنصر يستقر في العظام ويسبب سرطان العظام.

تم في هذه الدراسة حساب تراكيز الرادون والراديوم في 25 عينة لانواع مختلفة من مياه مدينة الهندية في محافظة كربلاء وسط العراق باستخدام كاشف الاثر النووي39-CR. وضمت العينات الكلية التي خضعت للاختبار 13 عينة ماء حنفية وستة عينات مياه جوفية وستة عينات اخرى من مياة بئر المثقاب(بئر بريمه) . وكانت تراكيزالرادون لجميع عينات مياه الحنفية اقل من الحد الطبيعي 11.1Bq/L. وكذلك تراكيز الراديوم وجدت اقل من 0.555Bq/L الحد الطبيعي لاجمالي الفا لنفس العينات.

ووجدت تراكيز جميع عينات مياه ابار البريمة اعلى من الحد الطبيعي11.1Bq/L فيما تراوحت تراكيز الراديوم بين(0.527-0.644)Bq/L وبمعدل0.586 Bq/L، وهذه القيمة اعلى من الحد الطبيعي لاجمالي الفا. فيما وجد الحد الاعلى لتراكيز الرادون في عينات المياه الجوفية بقيمة 13.573 Bq/Lوالحد الادنى بقيمة 9.169Bq/L وبمعدل 11.790 Bq/L وهذه القيمة اعلى من الحد الطبيعي. فيما تراوحت تراكيز الراديوم لنفس العينات بين Bq/L (0.340-0.540) وبمعدل 0.437 Bq/L وهذه القيمة اقل من الحد الطبيعي لاجمالي الفا. ولتحسين مستوى الصحة الاجتماعي، من الضروري تخفيض تراكيز الرادون والراديوم في المياه الصالحة للشرب قبل الاستعمال من قبل الناس.

الكلمات المفتاحية : مكونات الراديوم ، تركيز الراديوم ، الرادون ، كاشف CR-39 تقنية can

1. Introduction:

Radium is a naturally occurring radioactive element present in trace amounts throughout the Earth's crust. The decay of radium leads to radon in the environment (indoor and outdoor), soil, ground water, oil and gas deposits. It has been estimated that the radon, largely in homes, constitutes more than 50% of the dose equivalent received by general population from all sources of radiation, both naturally occurring and man-made[1, 2]. The alpha emitted by this radon and other radiation emitted from its decay products increase the absorbed dose in respiratory and digestion systems [3, 4]. Radon in water can enter the human body in two ways. Firstly, radon in drinking water or mineral drinks can enter the human body directly through the gastro-intestinal tract and irradiate whole body which the largest dose being received by the stomach [5]. Secondly, radon can escape from household water and became an indoor radon source, which then enter the human respiratory tract system to deliver radiation dose. Radon is picked up by groundwater passing through rocks and soil containing such radioactive substances; it enters water supplies when this water is pumped up a well [6]. Drinking water is the most important food. Therefore its availability, quality and regulation are delicated and important topics. For this purpose it is fundamental to have regulations about natural radioactivity in drinking water [7]. Exposure to radon occurs when breathing airborne radon while using water: showering, washing dishes, cooking, drinking water that contains radon and other everyday purposes [8].

^{226}Ra in the environment is widely distributed, being present in various concentrations in waters, soils, sediments and rocks [9]. When radium is ingested, the majority of material is rapidly excreted. However, since the chemical behavior of radium is similar to that of calcium, radium absorbed to blood from the GI-tract or lungs follows the behavior of calcium and is primarily deposited in bone [10]. Radium is a common radionuclide in the environment and it is the parent of radon. ^{226}Ra form is the most deadly radionuclides because it produces alpha radiation and has very long a half life (1600 y) [11].

Measurements a natural radioactivity of Radium-226 in drinking water in many countries of the world and that most of the research was to assess the risks resulting from the consumption of such water, these measurements show different levels from one place to another and the measurements varied by a type of natural water sources. Found that levels of radium ranging from1-600 Bq/m^3 in Poland and in France and Slovenia [12], and 21-49 Bq/m^3 in Spain [13] while arrived in the United States to several kBq/m^3 [14] and 2-350 Bq/m^3 in Syria [15] and about 41.1 Bq/m^3 in Iran [16] and the concentration Bq/m^3 was about 21-1040 Bq/m^3 in Turkey [17] and in range 2-15 Bq/m^3 in Argentina [18].

As also measured levels of the radon concentrations in water samples by several researchers have ranged between 2.8–116Bq/L in Jordan (many locations) [19], 0.67–21.25 Bq/L in Tassili, Southeast Algeria [20]. 20–95Bq/L in Eastern Doon Valley, outer Himalaya [21],0.1–576 Bq/L in Northern Venezuela[22], 1.7–376Bq/L; the "medicinal spring water" >74 Bq/L in Sudety Mountains, South-Western Poland[23]; 0.1–5Bq/L (ground water) 0.2–2 Bq/L(tap water) in Cyprus (many locations)[24], Median, 3–50 Max: 16–1220Bq/L in Bavaria, Germany [25], Below DL-161 in Midgonia Basin, Greece [26] and 0.46–49.6Bq/L in Lebanon (many locations) [27].

In 1991, the United States Environmental Protection Agency (EPA) proposed a National Primary Drinking Water Regulation (NPDWR) for ^{222}Rn with a maximum contaminant level (MCL) of 11.1 Bq/L (300 pCi/L) [28]. The National Academy of Sciences (NAS) revised the MLC and established an alternative maximum contamination level (AMCL) [29]. According to Reference [29], the AMCL may be set higher than the MCL such that "the contribution of radon from drinking water to radon levels in indoor air is equivalent to the national average concentration in outdoor air". For the United States, this leads to an AMCL of 4000 pCi/L (146 Bq/L). On the other hand, the European Union (EU) issued a non-binding recommendation in 2001 setting 100 Bq/L as a reference level; a concentration above this level warrants consideration of possible remedial actions. The EU recommendation also sets 1000 Bq/L as the upper bound above which remedial action is definitely required [30].

The aim of this study is to determine radium and radon concentrations were measured in the different types of water samples. Water is very essential in the daily lives of humans.

2. Materials and Methods:

CR-39 solid-state nuclear track detectors (SSNTDs) of thickness $300\mu m$ were used to measure the radium and radon concentration in different type water samples, these samples collected from various locations in Al-Hindiyah city. Which is located in the middle of Iraq on the Euphrates River. This city is surrounded by Karbala from west, Al-Hillah from east, Saddat Al-Hindiyah from north and Al-Haidariyah from south. The map of studied area is shown in Fig. 1.

About 94 gram of sample was placed in plastic can of size 7 cm in height and 7 cm in diameter. While the sample- detector distance is still 4.5cm and volume (96.211 cm^3) of water samples were kept in plastic cans as shown in Fig. 2.

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 [31.32]. The detectors were exposed for a period of about 88 days (from 25-11-2012 to 20-2-2013). 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 88 days 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 was used to count the number of tracks per cm^2 in each detector. To determine the radon and radium concentration from SSNTD readings (which is the track density, or the number of tracks per unit area), one needs to know the detector sensitivity that relates the track density to the total exposure of the detector to radon. Experimental determination of this sensitivity, *i.e.*, calibration of these detectors for radon measurements is carried out by exposing them to known concentrations of radon and/or its progeny in a radon exposure chamber.

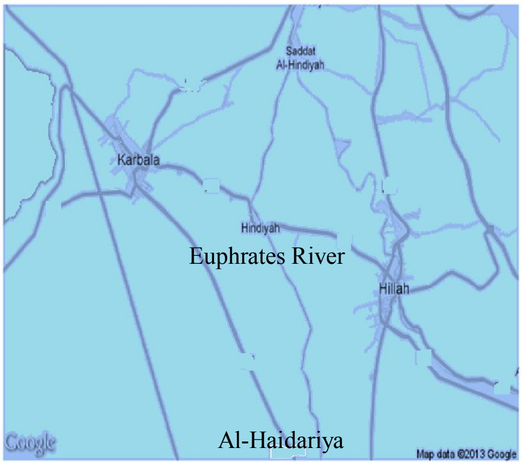

Fig.(1): The map showing Al-Hindiyah city

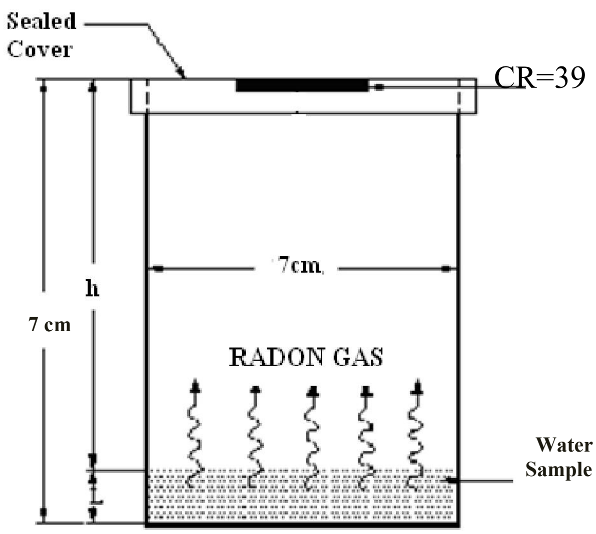

Fig.2 Experimental setup for measurements of radium and radon concentrations in water samples.

3. Theoretical considerations

In order to measure radon concentration levels in water samples, the track density ρ (in track/cm^2) is related to the radon concentration C_{Rn} (in Bq/m^3) and the exposure time T by the formula [31, 32]:

$$\rho = K\, C_{Rn}\, T \qquad (1)$$

Where K is the sensitivity factor of CR-39 plastic track detector, which is equal to 6.0095×10^{-2} Track.cm^{-1}.d^{-1} per Bq.m^{-3}, see Ref.[31].

The dissolved radon concentration in water(C_w) was calculated using equation (2) used by various researchers [1, 32, 33].

$$C_w = \frac{C_{Rn}\lambda h T}{L} \qquad (2)$$

Where C_{Rn} = radon concentration in ambient air (Bq/L), λ = Decay constant for radon (d^{-1}), h = the distance from the surface of water to detector (0.045m), T = Time of exposure (day) and L = the depth of the sample (0.025m)

The can technique [34, 35] was used calculate the radium concentration in water samples. The radium concentration in water was calculated using the relation [31]:

$$C_{Ra}(Bq.\mathrm{m}^{-3}) = \frac{\rho}{KT_e} \qquad (3)$$

The effective radium content of the water samples can be calculated by using the relationb[31,35]:

$$C_{Ra}(Bq.\mathrm{kg}^{-1}) = \left(\frac{\rho}{KT_e}\right)\left(\frac{hA}{M}\right) \qquad (4)$$

WhereC_{Ra} is the effective radium content of water sample (Bq/Kg), M is the mass of the water sample in Kg, A is the area of cross-section of the can in m^2; h is the distance between the detector and top surface of the water samples in meter (0.045m) and T_e is the effective expose time, which is related to the actual expose time T and decay constant λ for ^{222}Rn by the relation [1, 31, 35]:

$$T_e = [T - \lambda_{Rn}^{-1}(1 - e^{-\lambda_{Rn}T}) \qquad (5)$$

4. RESULTS AND DISCUSSION

In the presented research, a total number of 25 water samples including 13 tap water, 6 drill wells of water, 6 ground water samples were collected and analyzed for radon(airborne and dissolved) and radium concentrations. These samples collected randomly from various places in Al-Hindiyah city of Karbala Governorate in the middle of Iraq.

The tables 1, 2 and 3, presents the concentrations of radon and radium concentrations for all different types of water samples.

Also, the mean, maximum, minimum and slandered deviation of radon and radium concentrations for all water samples are shown in figures 3 and 4, respectively.

According to the data from table 1, the radon concentration emanated from tap water samples (air borne) varied from 78.248 Bq/m^3, which is equivalent 0.078 Bq/L to 362.983 Bq/m^3, which is equivalent 0.363 Bq/L with an average of 212.991 Bq/m^3, which is equivalent 0.213Bq/L and standard deviation 90.652Bq/m^3, which is equivalent 0.090 Bq/L.

The dissolved radon concentration for tap water samples varied from 2.248 Bq /L to 10.427 Bq /L with an average of 6.120 Bq/L and standard deviation 2.604 Bq/L. There is no specific national regulation for radioactivity concentrations in drinking water in Iraq. When this is compared to the maximum contaminant level of 11.1Bq/L for radon in public drinking water, suggested by the EPA [5, 28].

The radium concentrations for tap water samples varied from 83.478 Bq/m^3, which is equivalent 0.083 Bq/L to387.145 Bq/m^3, which is equivalent 0.387 Bq/L with an average of 227.223 Bq/m^3,which is equivalent 0.227 Bq/L and standard deviation96,713 Bq/m^3, which is equivalent 0.096Bq/L, while effective radium content for tap water samples varied from 0.154 Bq/Kg to 0.713 Bq/Kg with an average of 0.418 Bq/Kg and standard deviation 0.178 Bq/Kg.

Specific drinking water standards have not been established for radium 226 or other alpha emitters, but all tap water samples, ^{226}Ra concentration lower than MCL for gross alpha is 15 pCi/L(0.555Bq/L) that determined by U.S Environmental Protection Agency [38].

From table 2, the radon concentration emanated from drill wells water samples (air borne) varied from 494.267 Bq/m^3, which is equivalent 0.494 Bq/L to 603.746 Bq/m^3, which is equivalent 0.604 Bq/L with an average of 549.006 Bq/m^3, which is equivalent 0.549 Bq/L and standard deviation 40.963Bq/m^3, which is equivalent 0.041 Bq/L.

As well as, the dissolved radon concentration for drill wells water samples varied from 14.202 Bq /L to 17.384 Bq /L with an average of 15.775 Bq/L and standard deviation 1.177 Bq/L. The results all drill wells water samples had radon concentrations is higher than normal level 11.1 Bq/L.

Too from table 2, the radium concentrations for drill wells water samples varied from 527.300 Bq/m^3, which is equivalent 0,527 Bq/L to 644.095 Bq/m^3, which is equivalent 0.644 Bq/L with an average of 585.699 Bq/m^3, which is equivalent 0.586 Bq/L and standard deviation43.691 Bq/m^3, which is equivalent 0.043Bq/L, while effective radium content for drill wells water samples varied from 0.971 Bq/Kg to 1.187 Bq/Kg with an average of 1.079 Bq/Kg and standard deviation 0.081 Bq/Kg. ^{226}Ra mean concentration for drill wells water samples even higher than MCL for gross alpha is 15 pCi/L(0.555Bq/L) that determined by U.S Environmental Protection Agency [38].

Table 3 shows the concentrations of radium and radon (airborne and dissolved) concentrations in ground water samples.

The radon concentration emanated from ground water samples (air borne) varied from 319.101 Bq/m^3, which is equivalent 0.319 Bq/L to 472.372 Bq/m^3, which is equivalent 0.472 Bq/L with an average of 410.333 Bq/m^3, which is equivalent 0.410 Bq/L and standard deviation 54.373Bq/m^3, which is equivalent 0.054 Bq/L.

Also, the dissolved radon concentration for ground water samples varied from 9.169 Bq /L to 13.573 Bq /L with an average of 11.790 Bq/L and standard deviation 1.562Bq/L.

As well as, the radium concentrations for ground water samples varied from 340.427 Bq/m^3, which is equivalent 0.340 Bq/L to 503,941 Bq/m^3, which is equivalent 0.504 Bq/L with an average of 437.757 Bq/m^3, which is equivalent 0.437 Bq/L and standard deviation57.488 Bq/m^3, which is equivalent 0.058 Bq/L, while effective radium content for ground water samples varied from 0.627 Bq/Kg to 0.928 Bq/Kg with an average of 0.806 Bq/Kg and standard deviation 0.337 Bq/Kg. The mean radon concentration of ground water samples had radon concentrations is higher than normal level 11.1 Bq/L, but the mean ^{226}Ra concentration lower than MCL for gross alpha is 15 pCi/L(0.555Bq/L) .

The measurements indicate different levels of radium and radon concentration in water samples. It can be seen from the results that the radon concentrations vary appreciably from sample to sample. It is due to the fact that the water samples collected randomly from various sites for Al-Hindiyah city may have different Uranium contents.

Figures 3 and 4 indicate the ^{222}Rn and ^{226}Ra concentrations in most of drill wells water samples are high and in tap water are low.

Drill wells water may contain high amounts of natural radioactivity mainly associated with the uranium and thorium-rich soils and rocks, while ground water usually contains lower amounts of ^{222}Rn than drill wells water[38].

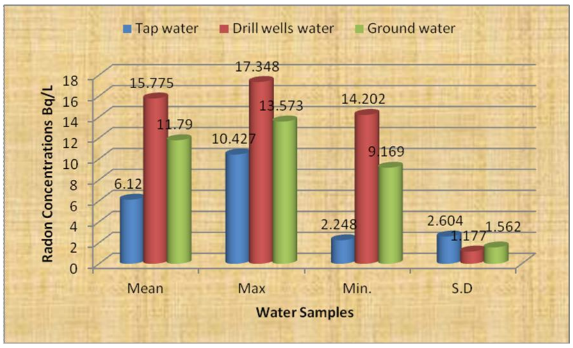

Fig 3: Mean, maximum, minimum and standard deviation of radon concentrations for all types' water samples.

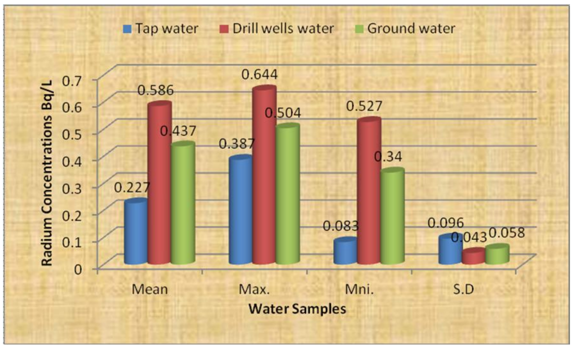

Fig 4: Mean, maximum, minimum and standard deviation of radium concentrations for all types' water samples.

The United Nations Scientific Committee on the Effects of Atomic Radiation has suggested a value of radon concentration in water for human consumption between 4 and 40 Bq/ L.

In this work, the recorded values of radon concentration in water samples are within the safe limit recommended by the United Nations scientific Committee on the Effects of Atomic Radiation [39].

To removal of radon in potable groundwater and drill wells water samples; there exist a number of methods for treating radon-containing water, of which several are reported to have a removal efficiency of over 95%. The methods are essentially based on one (or a combination) of the following:

(i) Aeration: to remove radon from water to the gas phase prior to entry to the household.

(ii) Storage: a storage period which is significant in comparison to radon's short half-life (3.8 days) ensures decay of radon to short-lived isotopes (lead, polonium etc.) which are neither degassed to air nor readily adsorbed during human ingestion.

(iii) Filtration: activated carbon filtration and reverse osmosis have been shown to be effective at removing radon. These methods are expensive, especially for large quantities of water, and require a certain amount of maintenance.

Of these methods, aeration is possibly most suited to treatment of radon from wells in crystalline bedrock. Aeration units are typically based on bubbling or cascading water through a high-surface area aeration medium.

Units are available which are reported to have a 99% radon removal efficiency (although this depends on radon concentration and water flow). After aeration, water should be stored for at least 1 hour, before use, in order to allow daughter products (with short half-lives) to decay [40].

5. Conclusion

Measurement results radon concentration of all tap water samples is lower than normal level 11.1 Bq/L, but the mean of radon concentrations in the drill wells water and ground water samples are higher than the normal level.

Also, the mean value of radium concentrations for all tap water samples is lower than MCL for gross alpha, while the mean value of radium concentrations for all drill wells water is higher than MCL for gross alpha, whereas mean value of radium concentrations for all ground water samples is lower than MCL for gross alpha.

The recorded values of radon concentration in water samples are within the safe limit recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation, so it has no danger on human being life.

There exist several methods for removing radon from water, of which a combination of aeration and subsequent short storage seems to be the most efficient.

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Table 1: Radon (airborne and dissolved) and radium concentrations in tap water samples.

W. S	ρ (Track/cm²)	Radon conc. (airborne) (Bq/m³)	Radon conc. (airborne) (Bq/L)	Radon conc. Dissolved in water (Bq/L)	Radium conc. (Bq/m³)	Radium conc. (Bq/L)	Effective Radium content (Bq/Kg)
1	1571.768	297.206	.297	8.540	317.068	.317	.584
2	529.611	100.144	.100	2.878	106.837	.107	.197
3	1919.154	362.893	.363	10.427	387.145	.387	.713
4	1108.587	209.623	.210	6.023	223.632	.224	.412
5	876.997	165.831	.166	4.765	176.914	.177	.326
6	413.816	78.248	.078	2.248	83.478	.083	.154
7	1455.973	275.310	.275	7.911	293.709	.294	.541
8	761.202	143.936	.144	4.136	153.555	.154	.283
9	1803.359	340.997	.341	9.798	363.786	.364	.670
10	1340.178	253.414	.253	7.282	270.350	.270	.498
11	992.792	187.727	.188	5.394	200.273	.200	.369
12	645.406	122.040	.122	3.507	130.169	.130	.240
13	1224.383	231.518	.232	6.652	246.991	.247	.455
Mean	1126.402	212.991	.213	6.120	227.223	.227	.418
Max.	1919.154	362.893	.363	10.427	387.145	.387	.713
Min.	413.816	78.248	.078	2.248	83.478	.083	.154
SD	400.794	90.652	.090	2.604	96.713	.096	.178

Table 2: Radon (airborne and dissolved) and radium concentrations in drill wells water.

W. S	ρ (Track/cm²)	Radon conc. (airborne) (Bq/m³)	Radon conc. (airborne) (Bq/L)	Radon conc. Dissolved in water (Bq/L)	Radium conc. (Bq/m³)	Radium conc. (Bq/L)	Effective Radium content (Bq/Kg)
1	2613.927	494.267	.494	14.202	527.300	.527	.971
2	3192.903	603.746	.604	17.348	644.095	.644	1.187
3	2845.517	538.059	.538	15.460	574.018	.574	1.058
4	3077.108	581.850	.582	16.719	620.713	.621	1.144
5	2961.313	559.955	.560	16.090	597.377	.597	1.101
6	2729.722	516.163	.516	14.831	550.695	.551	1.015
Mean	2903.415	549.006	.549	15.775	585.699	.586	1.079
Max.	3192.903	603.746	.604	17.348	644.095	.644	1.187
Min.	2613.927	494.267	.494	14.202	527.300	.527	.971
SD	200.166	40.963	.041	1.177	43.691	.043	.081

Table 3: Radon (airborne and dissolved) and radium concentration in Ground water samples.

W. S	ρ (Track/cm²)	Radon conc. (airborne) (Bq/m³)	Radon conc. (airborne) (Bq/L)	Radon conc. Dissolved in water (Bq/L)	Radium conc. (Bq/m³)	Radium conc. (Bq/L)	Effective Radium content (Bq/Kg)
1	2266.540	428.580	.429	12.315	457.222	.457	.842
2	2382.336	450.476	.450	12.944	480.581	.481	.885
3	1687.564	319.101	.319	9.169	340.427	.340	.627
4	2150.745	406.684	.407	11.686	433.863	.434	.799
5	2498.131	472.372	.472	13.573	503.941	.504	.928
6	2034.950	384.789	.385	11.056	410.504	.411	.756
Mean	2170.044	410.333	.410	11.790	437.757	.437	.806
Max.	2498.131	472.372	.472	13.573	503.941	.504	.928
Min.	1087.564	319.101	.319	9.169	340.427	.340	.627
SD	200.875	54.373	.054	1.562	57.488	.058	.337