Determination of Uranium concentration, Radium content and Radon Exhalations Rates in soil samples for Some Regions in Lebanon

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Abstract:

In this study provides measurement of uranium concentration, radium content and radon exhalations rates in the sixteen soil samples collected from surface soil for some regions in Lebanon. Measurements were made, using long-term technique for alpha particles emission with solid state nuclear track detector type CR-39. The uranium concentration varies from 0.131 ppm to 6.772 ppm with mean values of 1.467 ppm. While, the values of effective radium content (^{226}Ra) are found to vary from 0.096 Bq /kg to 4.982 Bq /kg with mean values of 1.079 Bq/kg.

Also, the values of mass exhalation rates of radon vary from 0.68×10^{-8} Bq /kg. sec to 34.97×10^{-8} Bq /kg.sec with a mean value of 7.58×10^{-8}Bq/ kg.sec, while the surface exhalation rates vary from 1.40×10^{-7} Bq/m^2.sec to 72.69$\times 10^{-7}$ Bq/m^2.sec with a mean value of 15.74$\times 10^{-7}$ Bq/m^2.sec. The values of radon exhalation rates and radium content are found less than the world average value; also the values of uranium concentrations in soil samples of these areas are close than the values reported by many researchers.

Keywords: Uranium concentration, Radium content, Radon exhalation rates, Soil, CR-39 detectors, Can technique, Lebanon.

تحديد تراكيز اليورانيوم ومحتوى الراديوم ومعدل انبعاث الرادون في عينات تربة بعض المناطق من لبنان

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الخلاصة:

في هذه الدراسة تم قياس تراكيز اليورانيوم و محتوى الراديوم ومعدل انبعاث الرادون في ستة عشرة عينة جمعت من سطح تربة بعض المناطق في لبنان. وتمت القياسات باعتماد تقنية القياس طويلة الأمد لانبعاث جسيمات ألفا باستخدام كاشف الأثر النووي الصلب نوع CR-39. لقد أظهرت نتائج هذه الدراسة ان تراكيز اليورانيوم في عينات التربة تتراوح بين ppm (0.131-6.772) وبمعدل 1.467ppm. بينما كان محتوى الراديوم- ٢٢٦ في عينات التربة يتراوح بين Bq/kg(0.096- 4.982)وبمعدل 1.079Bq/kg. فيما كانت قيم انبعاث الرادون لوحدة الكتلة من عينات التربة تتراوح

بين 0.68×10^{-8}Bq/kg.se و 34.97×10^{-8} Bq/kg.sec وبمعدل 7.58×10^{-8} Bq/kg.sec فيما كانت قيم انبعاث الرادون لوحدة السطح من عينات التربة تتراوح بين 1.40×10^{-7} Bq/m^2.sec و 72.69×10^{-7} Bq/m^2.sec وبمعدل 15.74×10^{-7} Bq/m^2.sec.وجدت قيم معدل انبعاث الرادون ومحتوى الراديوم اقل من معدل القيمة العالمية، وكذلك تراكيز اليورانيوم في عينات تربة هذه المناطق قريبة من القيم التي ذكرت من قبل العديد من الباحثين.

الكلمات المفتاحية: : تركيز اليورانيوم ، محتوى الراديوم، معدل انبعاث الرادون،التربة، كاشف CR-39 ، تقنية كان، لبنان.

1. Introduction

Exhalation of ^{222}Rn (α-radioactive inert gas) is associated with the presence of ^{226}Ra and its ultimate precursor uranium in the earth crust. Although these elements occur in virtually all types of rocks and soils, their concentration varies with specific sites and geological materials. Radon can diffuse through rocks and soil, can move from one place to the other and can leak out in the atmosphere from the soil [1, 2].

^{226}Ra in the environment is widely distributed, being present in various concentrations in waters, soils, sediments and rocks [3, 4]. 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 [5].

The immediate radon precursor is ^{226}Ra. Radium, spread widely, particularly in materials which are made from mineral products. The forerunner of radium is ^{238}U; which has a half-life of 4.47×10^9 years. It is present in all types of rocks and soil and therefore in most of the raw materials from which we process final products. Uranium concentrations are commonly expressed in parts per million by weight (ppm) or in terms of the "specific activity" expressed in Becquerel per kilogram (Bq/kg) [6]. These units are related by the conversion factor for ^{238}U, 37 Bq/kg =3.0 ppm [6]. To consider the uranium concentrations in some typical rocks, granite, which is relatively rich in uranium, has an average concentration of about 59.2 Bq/kg (4.8 ppm) and basalt, which is relatively uranium-poor, has an average of about 11.1 Bq/kg [7]. The average value for rock in earth's crust is about 37 Bq/kg. Soils average is about 25.9 Bq/kg.

The alpha particles given off by radium and radon bombard the bone marrow and destroy tissues that produce red blood cells. It may cause bone cancer. The radium content of a sample also contributes to the level of environmental radon as radon is produced from ^{226}Ra through α-decay. Higher values of ^{226}Ra in soil contribute significantly to the enhancement of indoor radon. Uranium is the proximate source of radium and radon in the soil and rocks. Uranium prospection through the analysis of soil, rocks, plants and water has been reported by many workers [8, 9]. Uranium, present in the earth, is transferred to water, plants, food supplements and then to human beings. Uranium accumulated in humans may have a dual effect due to its chemical and radioactive properties. High intake of uranium and its decay products may lead to harmful effects in human beings. [10]

The aim of this study is to measure the uranium concentrations, radium content and radon exhalations in sixteen soil samples for some areas in Lebanon. The map of studied area is shown in Fig.1.

2. Area of Study

Lebanon is a country with an area of 10452 km^2 and an estimated population of 3.5 million people of which 88% live in urban areas. The country lies around the conjuncture of three tectonic plates: the Arabian, Anatolian and African plates. A major geological feature in Lebanon is the northern extension of the great African/Red Sea/Dead Sea rift valley. This is manifested in a major fault line with many branched sub-faults.

Currently, and in the recent past, the area has not been very active geologically, although mild earthquakes are not uncommon. From north to south, the coastline in Lebanon extends over 217 km, while from east to west it measures 80 km at its widest point. Lebanon’s landforms fall into four parallel belts that run from northeast to southwest: a narrow coastal plain along the Mediterranean shore; the massive Mount Lebanon rising steeply from the coastal plain to 3083m to dominate the entire country before dropping eastward; a fertile between-mountains basin, with an approximately uniform altitude of 800–1000 m, called the Beqaa Valley; and the ridges of the Anti-Lebanon Mountains dominated in the south by the volcanic Mount Hermon at 2814 m.

3. Experimental part:

Surface soil samples were collected from sixteen sites in the some regions of the Lebanon. The sites, Zahlah, Beirut, Juniyah, Az Zahrani, Bikfayya, Sidon, Alayh, Ba'labak, Ad Damur, Barja, Riyaq, North Ba'labak, As Sarafand, An Nabatiyah, Bayt ad Din, and Jazzin, see Fig. 1.

Fig.1. The map showing Lebanon

Passive integrating methods of radon measurement by Solid-State Nuclear Track Detectors (SSNTDs): A passive method (closed-can technique) using CR-39 plastic detector with sheet thickness 300µm from the intercast Europe srl company, as a solid state nuclear-track detector was developed for measurements of radon exhalation rate of different samples materials, in which the samples of interest is enclosed in a sealed can [11-14]. The CR-39 is a very useful detector for the direct registration of alpha particles, the sensitive surface of the detector faced to the samples, the experimental arrangement is shown in Fig. 2. The tracks detected by these plastic detectors are not directly visible and have to be enlarged by adequate chemical processing. The sixteen different samples of soil were collected by the grab sampling method (surface soil samples) from different places for some areas in Lebanon.

Soil samples were dried at 100 °C for 3 h in an oven to ensure complete moisture removal. All samples were crushed to a fine powder form, the crushed samples were then sieved through a small mesh size to remove the larger grains size and render them more homogenous. Dried samples (20g) were placed at the bottom of the plastic Cans of size 6.5cm height and 3.5 cm in diameter. A piece of detector of size 1cm ×1cm was fixed on the top of inner surface of the Can, in such a way, that it is sensitive surface always facing the soil sample. The Can is sealed air tight with adhesive tape and kept for exposure of about 100 days (from 29-9-2013 to 7-1-2014).

After that the CR-39 detectors were collected and chemically treated by etching using 6N solution of NaOH at temperature of 70°C, for 6 hours. At the end of etching process, the detectors were washed thoroughly with distilled water and then dried. Each detector was counted visually using optical microscope(A.KRÜSS-OPTRONIC, Germany) with power of 400X (40x objective and 10x eyepiece), then the average number of track per cm^2, track density of soil, were obtained by taking the average of twenty views in the microscope.

The uranium concentration, radium content and radon exhalations rates in soil samples was obtained by using the sealed-cup technique as shown in Fig.(2).

The radon track density ρ_{Rn} (in track/cm^2) is related to the radon activity concentration C_{Rn} (in Bq/m^3) and the exposure time T by formula [15].

$$C_{Rn} = \frac{\rho_{Rn}}{K_{Rn}.T} \qquad (1)$$

where, K_{Rn} is the calibration factor of CR-39 plastic track detector.

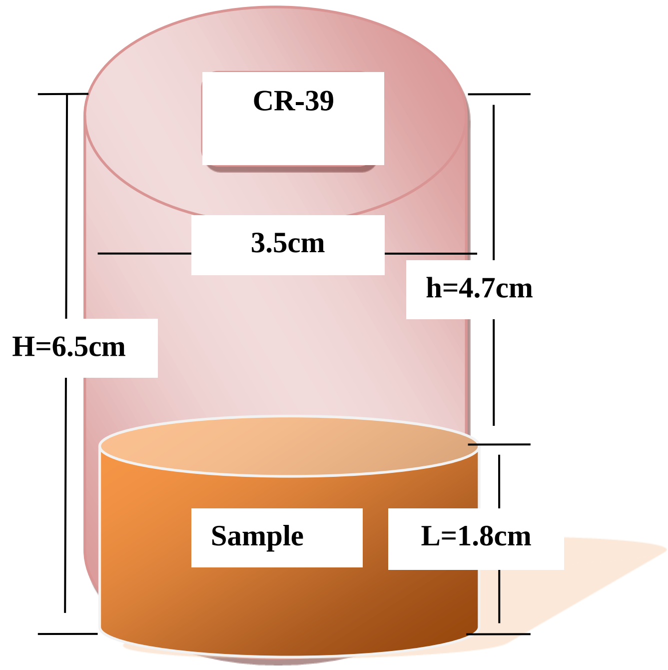

Fig2: Test tube technique used in the study.

The value of K_{Rn} will depend on the height and radius of the measuring Can [16]. Such that

$$\boldsymbol{K = \frac{1}{4}\, r \left[2 \cos \theta_c - \frac{r}{R_\alpha}\right]} \qquad (2)$$

where,r is the test tube radius (1.75cm), θc is the critical angle for CR-39 (35 degree) and R_α is the range of ^{222}Rn alpha particle in air (4.15cm). By substituting , $\boldsymbol{\theta_c}$ and $\boldsymbol{R_\alpha}$ in equation (2) we get:-

$$K = 4.5987 \times 10^{-2} \left(\frac{Trac / cm^2 .day}{Bq / m^3} \right)$$

Since the half-life of ^{226}Ra is1600 years and that of ^{222}Rn is 3.82 days, it is reasonable to assume that an effective equilibrium (about 98%) for radium-radon members of the decay series is reached in about three weeks. Once the radioactive equilibrium is established, one may use the radon alpha analysis for the determination of steady state activity concentration of radium. The activity concentration of radon begins to increase with time T, after the closing of the can, according to the relation [4]:

$$\mathrm{CRn} = \mathrm{CRa}\left(1 - \mathrm{e}^{-\lambda \mathrm{RnT}}\right) \qquad (3)$$

The "Can technique" was used to calculate the radium content in soil samples. The Effective radium content in soil was calculated using the relation [17, 18]:

$$C_{Ra}(Bq\ kg^{-1}) = \left(\frac{\rho_{Rn}}{K_{Rn}.T_e}\right)\left(\frac{hA}{M}\right) \qquad (4)$$

where C_{Ra} is the effective radium content of soil sample (Bq/kg), M is the mass of soil sample (0.020 kg), A is the area of cross section of the cylindrical can in m^2 and h is the distance between the detector and top of the solid sample (4.7cm).

T_e Is the effective exposure time, which is related to the actual exposure time T and decay constant λ_{Rn} for ^{222}Rn by the relation [٤, 17].

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

The mass exhalation rate $E_x(M)$ of the sample for release of the radon can be calculated by using the expression [19, 20]:

$$E_M(Bq.\mathrm{kg}^{-1}.\mathrm{d}^{-1}) = C_{Ra}\left(\frac{\lambda_{Ra}}{\lambda_{Rn}}\right)\frac{1}{T_e} \qquad (6)$$

The surface exhalation rate $E_x(S)$ of the sample for release of radon can be calculated by using the expression [19, 20]:

$$E_S(Bq.\mathrm{m}^{-2}.\mathrm{d}^{-1}) = \left[C_{Ra}\left(\frac{\lambda_{Ra}}{\lambda_{Rn}}\right)\frac{1}{T_e}\right]\frac{M}{A} \qquad (7)$$

$$E_S(Bq.\mathrm{m}^{-2}.\mathrm{d}^{-1}) = E_M\left(\frac{M}{A}\right) \qquad (8)$$

where λ_{Ra} is the decay constant of ^{226}Ra.

To find uranium concentrations (C_U)in units of part per million (ppm) using the following equation [21, 22]:

$$C_U\ (ppm) = \frac{W_U}{W_s} \qquad (9)$$

where W_s *is* the weight of sample (20gram).

W_U is uranium weight in sample can calculate from the following equation [23]:

$$W_U\ (gm) = \frac{N_U\ W_{mol.}}{N_{AV.}} \qquad (10)$$

where $W_{mol.}$ is weight molecular uranium 238.07gm/mol, N_{AV} is number of Avogadro 6.023×10^{23}atom/mol, N_U is numbers of uranium atoms.

4. Results and Discussion

Table 1 show that collected data for all values of uranium concentrations, radium content and radon exhalation rates in the sixteen soil samples for some areas in Lebanon.

Fig. 3 (a and b) have shown the distribution of radium contents and uranium concentrations in soil samples for sixteen different locations in Lebanon, respectively.

While, Fig. 4(a and b) have shown the distribution of mass exhalation rates and surface exhalation rates of radon for same samples, respectively.

Uranium content in soil samples of the study area was found to range from 0.131 ppm to 6.772 ppm with a mean value of 1.467 ppm . Also, the radium content varies from the lowest values 0.096 Bq/kg to highest value is 4.982 Bq/kg with mean value 1.079 Bq/kg. See Fig. 3.

The values of exhalation rates of soil samples vary from one sample to another. This variation is due to the content of uranium and radium and to the porosity of the soil samples.

The radon exhalation values were calculated in terms of (surface) area (E_S) and mass (E_M). The values of E_M ranged from 0.68×10^{-8}Bq/kg.seto 34.97×10^{-8} Bq/kg.sec with mean value 7.58×10^{-8} Bq/kg.sec. While the values of E_S range from 1.40×10^{-7} Bq/m^2.sec to 72.69×10^{-7} Bq/m^2.sec with mean value 15.74×10^{-7}Bq/m^2.sec.

Radon exhalation rates observed in the present study are well below the world average of 57600 mBq/m^2.h(0.016 Bq/m^2.sec) and hence do not pose any health hazards to the residents[24].

From Table 1, it has been observed that there are variations in the values of radon exhalation rates among the samples. This variation may be arisen due to the difference in the nature of the samples, and radium content of the samples because radium is present in varying levels all over the world. The values of effective radium content are less than the permissible value of 370 Bq/kg as recommended by Organization for Economic Cooperation and Development [25]. Hence, the result shows that this study area is safe as for as the health hazards of radium are concerned.

Also, we found the uranium content in the soil samples was low and not significant from a health hazard point of view. The values of uranium concentrations in soil samples of these areas are close than the values reported by many researchers [26- 27].

5. Conclusion

Uranium concentrations, Radium content and radon exhalation rates (both the mass and surface exhalation rates) have been measured successfully using CR-39 plastic track detectors by the sealed can technique.

The values of radon exhalation rates and radium content are found to be below the world average value of 57.600Bq/m^2.h (0.016 Bq/m^2.sec) [24] and 370Bq/kg recommended by OECD, 1979[25], respectively**.** Hence it can be concluded that the study area is safe from the health hazard of radon and radium point of view.

The mean uranium concentration in the soil samples was low and not significant from a health hazard point of view**.** The values of uranium concentrations in soil samples of these areas are close than the values reported by many researchers [26 - 28].

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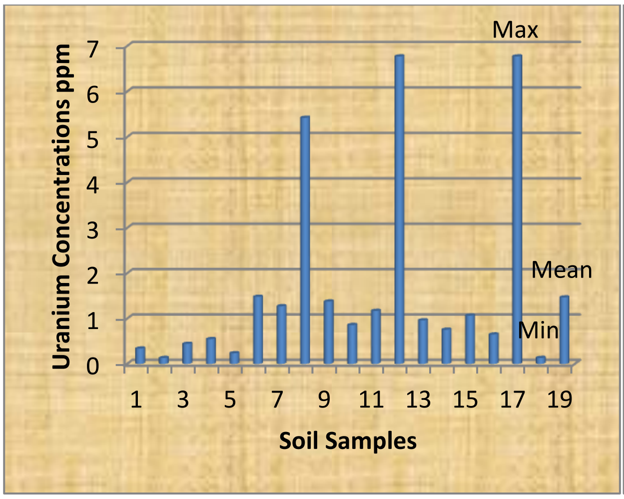

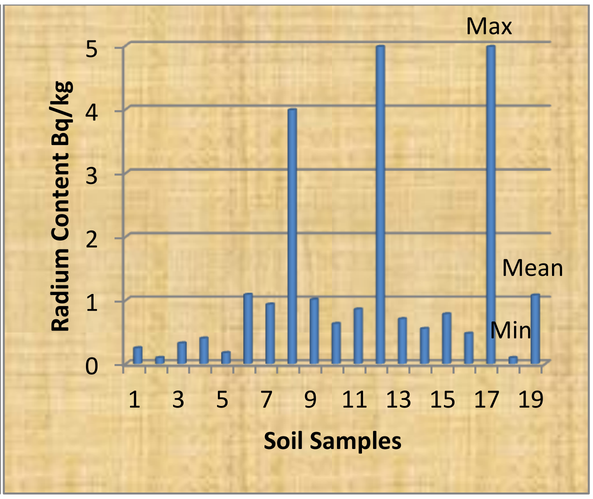

Fig.3 : (a) Uranium concentrations by unit ppm, and (b)) Radium content by unit Bq/kg for all soil samples

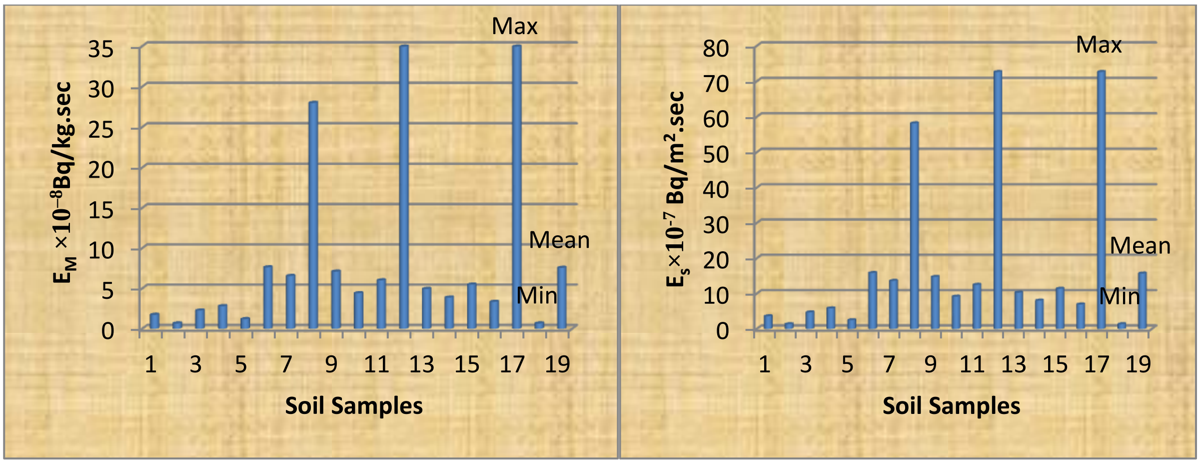

Fig. 4: (a) The mass exhalation rate (E_M)of radon by unit Bq/kg.Sec, and b) The surface exhalation rate (E_S) of radon by unit Bq/m^2.sec. for all soil samples

Table 1: Uranium concentrations, Effective radium content, mass exhalation(E_M) and surface exhalation (E_S)rates of radon in different soil samples for sixteen different sites.

Number sample	Regions	ρ Trac/cm^2	C_{Rn} Bq/m^3	C_{Ra} Bq/kg	Radon Exhalations Rates		CU ppm
					$E_M \times 10^{-8}$ Bq/kg.sec	$E_S \times 10^{-7}$ Bq/m^2.sec	
1	Zahlah	478.53	104.05	.249	1.75	3.63	.338
2	Beirut	185.09	40.24	.096	.68	1.40	.13
3	Juniyah	625.24	135.96	.325	2.28	4.74	.442
4	Az Zahrani	771.96	167.86	.402	2.82	5.86	.546
5	Bikfayya	331.81	72.15	.173	1.21	2.51	.235
6	Sidon	2092.44	455.00	1.089	7.64	15.88	1.480
7	Alayh	1799.00	391.19	.936	6.57	13.65	1.272
8	Ba'labak	7667.76	1667.37	3.990	28.01	58.21	5.423
9	Ad Damur	1945.72	423.10	1.012	7.11	14.77	1.376
10	Barja	1212.12	263.58	.631	4.43	9.20	.857
11	Riyaq	1652.28	359.29	.860	6.03	12.54	1.169
12	North Ba'labak	9575.11	2082.13	4.982	34.97	72.69	6.772
13	As Sarafand	1358.84	295.48	.707	4.96	10.31	.961
14	An Nabatiyah	1065.40	231.67	.554	3.89	8.08	.754
15	Bayt ad Din	1505.56	327.38	.783	5.50	11.43	1.065
16	Jazzin	918.68	199.77	.478	3.36	6.97	.650
Maximum value		9575.11	2082.13	4.982	34.97	72.69	6.772
Minimum value		185.09	40.24	0.096	0.68	1.40	0.131
Mean value		2074.09	451.01	1.079	7.58	15.74	1.467