SOIL RADON EXHALATION RATE MEASUREMENT IN DUHOK CITY BY TWO TECHNIQUES

by

Walat A. H. Alhamdi [1] ***and Khairi M-S. Abdullah*** [2]

[1]Department of Physics, College of Science, University of Duhok, Kurdistan Region, Iraq
[2]Department of Water Resources, College of Engineering, University of Duhok, Kurdistan Region, Iraq

Scientific paper
https://doi.org/10.2298/NTRP2203229A

Radon exhalation rate is the rate of radioactive radon gas that escapes from the soil into the atmosphere. In this study exhalation rate of radon was measured for 33 samples of soil, in four districts of Duhok province, using two different techniques: one using radon concentration and other using radium content, by both alpha and gamma spectroscopy. For the radon measurement, alpha-sensitive RAD7 detector was used. While in the second method, radon exhalation rate was expressed as a function of radium concentration, measured by well type NaI (Tl) detector and other soil parameters. Analysis, shows that the average of radon exhalation rate, by the first technique, varies from 14.6 3.2 to 55.7 1.6 $Bqm^{-2}h^{-1}$, while by the second technique, it varies from 13.3 2.4 to 50.6 4.1 $Bqm^{-2}h^{-1}$. Overall, the measured values, for both methods at all the sample points, present a good correlation and less than global mean average recommendation.

Key words: radon gas, radon exhalation rate, alpha spectroscopy, radium, gamma spectroscopy

INTRODUCTION

Radon (^{222}Rn) is a colorless, odorless and tasteless noble gas, with a half-life of 3.8 days. The mechanism through which this gas is produced involves an alpha decay of the parent nuclide which is radium (^{226}Ra) as part of the ^{238}U decay series [1]. Radon, decaying primarily via alpha particle with energy 5.486 MeV, produces two isotopes of polonium (^{218}Po and ^{214}Po) [2]. It is normally found in various amounts in rocks and soil throughout the world [3].

Emanation, advection, diffusion and exhalation of radon from the soil are the main processes of radon release to the atmosphere [4]. Among that, the exhalation rate of radon and radon concentration in soil are the two most important factors used for evaluating and calculating the rate at which radon enters into pore spaces, and subsequently, into the surrounding air [5].

Radon exhalation rate can be described in terms of mass ($Bqkg^{-1}s^{-1}$) and area ($Bqm^{-2}s^{-1}$). Radon exhalation rate depends on many parameters, for example: soil moisture, soil porosity, temperature and the amount of concentration of ^{226}Ra which is a direct source of radon [5]. Exhalation of radon may also be affected by weather conditions [6].

Nowadays radon exhalation measurement from soil, rock, building materials and water becomes an important topic throughout the world [7-9]. Radon poses many health problems because two-thirds of radiation dose, received by living tissue, is caused by radon and its progeny [4]. The present study aims to measure radon exhalation rate in four districts of Duhok region, by two different methods, which are alpha and gamma spectroscopy techniques, making a comparison between both techniques and establishing a data base for Duhok province.

MATERIAL AND METHODS

Study area

Duhok city is the capital of the Duhok Governorate in the Kurdistan Region, located in the north-west of Iraq, as shown in fig. 1. The geological composition of Duhok region is formed of red beds of silt, hard clay and limestone. The soil of the study area is mostly characterized by very low permeability, broad synclines and well-marked folds of asymmetrical anticlines and thick sedimentary cover. [2].

Soil sample collection and preparation

The 33 soil samples have been collected from four districts of Duhok province. Fifteen samples were taken in Duhok district, three in Amedi, four in Zakho

* Corresponding author, e-mail: walat@uod.ac

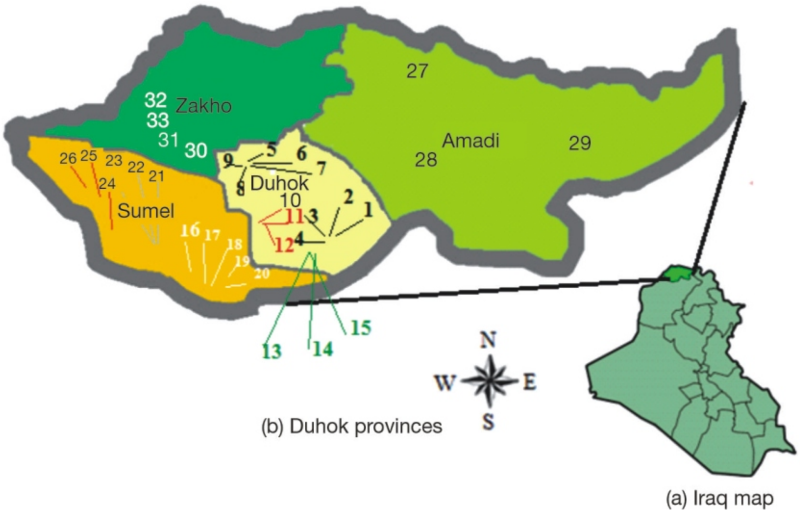

Figure 1. Map of study area

Table 1. study points of geographical locations

Sites	Geographical location		Sites	Geographical location		Sites	Geographical location	
	North	East		North	East		North	East
1	36.84751	43.07947	12	36.86730	42.91770	23	36.85825	42.83727
2	36.84977	43.06578	13	36.58212	43.00692	24	36.85907	42.83617
3	36.84949	43.06690	14	36.91479	43.13966	25	36.86051	42.83522
4	36.84547	43.07595	15	36.91749	43.13580	26	36.85976	42.83428
5	36.88169	42.88883	16	36.86223	42.86183	27	37.09112	43.48454
6	36.88709	42.87949	17	36.86044	42.86114	28	37.09112	43.48776
7	36.86843	42.90092	18	36.86401	42.85436	29	37.09953	43.48613
8	36.86870	42.89994	19	36.85811	42.84647	30	37.13602	42.69203
9	36.84570	42.89994	20	36.85976	42.84046	31	37.13506	42.69118
10	36.86062	42.67685	21	36.86010	42.83865	32	37.13630	42.67658
11	36.85239	42.91805	22	36.85351	42.85616	33	37.13657,42.70525	

and eleven in Somel district. Table 1 gives information about the location of the soil samples. At each point of the study sites, soil was collected from an area of about 50 cm 50 cm, after the soil site was cleared of debris and sediment. The gathered samples of soil were brought to the laboratory. In order to obtain the homogeneous powder, the samples were dried at room temperature for four days approximately, then dried in an oven at 110 °C for one hour to ensure the removal of any remains of significant moisture, then sieved through 250 mm mesh.

Sample storing for radon and radium measurement

In this study accumulation method was used to carry out radon and radium concentration in soil. Thus, cylindrical plastic containers, with two valves in each, were prepared for storing the soil, in order to measure radon concentration. For that reason, prepared dried soil, described previously, of about 125 g of each sample was poured in a plastic container, which vas previously cleaned and dried, as the radon exposure source. At the same time, to estimate radium content, smaller tubes of 13 cm^3 were filled with soil. These plastic containers and tubes were stored for one month to acquire secular equilibrium between radon and its progenies and ^{226}Ra and its progenies. Figure 2 shows the tube used for both radon and radium storage, respectively.

Radon and radium activity measurement

To measure radon concentration in soil, RAD7, which is the solid-state Professional Electronic Radon Detector, is used. The setup of the measurement

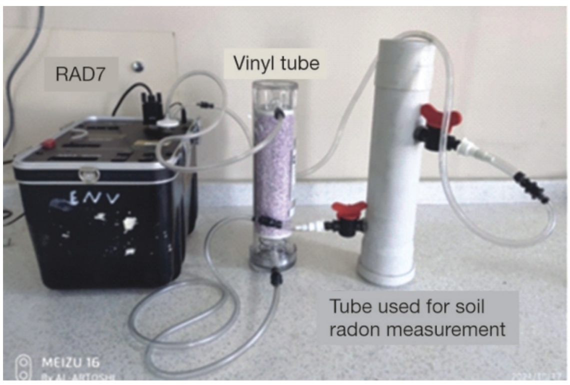

Figure 2. Radon concentration measurement system

shown in fig. 2. It consists of RAD7, a plastic container and a vinyl tube. When plastic container is connected to the closed loop, both valves of the container are opened. The device will pool air from the container to the desiccant tube and after that, to the inlet point of RAD7. The air is then leaving from the outlet of RAD7 to the container again. The RAD7 converts alpha particles, that are emitted from polonium isotopes, to an electrical impulse and is capable to separate the different electrical pulses generated from ^{218}Po and ^{214}Po with energies 7.0 MeV and 6.0 MeV, respectively. In term of test environments, the testing is done in dry conditions (humidity range between 4-8 %) and room temperature between (20-31 °C). Furthermore, to reduce background radiation, radon has been measured for empty tube, following the same manner as for the soil samples in term of starts, stops and exposure time.

Radium content was measured by a gamma-ray spectrometry system consisting of a well-type 3" 3" NaI (TI) scintillation detector. The detector was surrounded by a 4 geometry lead shield with thickness of 6 cm, in order to decrease and limit background radiation.

Energy and efficiency of the detector was determined using gamma standard radioactive sources ^{155}Eu, ^{137}Cs, and ^{22}Na. Based on the spectrum of gamma-ray counts (N) at full width at half maximum of photo-peaks of both energies, 352 keV of ^{214}Pb and 609 keV of ^{214}Bi decay products, the activity per unit mass of radium in the studied samples was measured as the average of both ^{214}Bi and ^{214}Pb radioisotopes [10, 2]

$$Ra \quad \frac{A_{\mathrm{Ra}}}{m}[\mathrm{Bqkg}^{\ 1}] \tag{1}$$

where $A_{\mathrm{Ra}} = N_{\mathrm{Pb}}/[\varepsilon_{E(\mathrm{Pb})} t f_{(\mathrm{Pb})}] + \mathrm{N}_{\mathrm{Bi}}/[\varepsilon_{E(\mathrm{Bi})} t f_{(\mathrm{Bi})}]$ [Bq], t – the detection time, ε_{E} – the detection efficiency at the given energy (351.9 and 609 keV), and f – the gamma fraction.

Assessment of soil parameters

The bulk density ρ_b was calculated according to eq. 2 [11]. Where M is the dry soil sample weight [kg] used in the experiment, and V_{S} is the volume of the dry soil [m^3]. Consequently, it was estimated to be 1100 kgm^{-3}

$$\rho_b \quad \frac{M}{V_s} \tag{2}$$

The density of the soil sample was calculated using the following steps: the samples of soil were dried for 24 hours at 110 °C, as mentioned previously. Then, an empty specific bottle was weighed W_i, then filled with water and weighed W_a. The bottle was then dried again and one-third of the dried bottle was filled with soil and weighed W again. Finally, the remaining two-thirds of the bottle were filled with water. After one week, the bottle was filled with water again and weighed W_b. From each weight, the density of the experiment soil was calculated using eq. (3) [12]. The density of the dry soil was, therefore, calculated as 1700 kgm^{-3}.

$$\rho \quad \frac{W \quad W_i}{(W \quad W_i) \quad (W_a \quad W_b)} \tag{3}$$

Equation 4 was used to estimate the porosity ε of the soil samples. Accordingly, it was estimated to be 0.3. While 3.0 10^{-6} m^{2}s^{-1} is used as the value of effective diffusion coefficient [11]

$$\varepsilon \quad 1 \quad \frac{\rho_b}{\rho} \tag{4}$$

Emanation coefficient f was estimated by the combination of radium activity and radon concentration in soil, the effective volume V of the container and the soil mass M of the sample [12]

$$f \quad \frac{VC_{\mathrm{Rn}}}{MC_{\mathrm{Ra}}} \tag{5}$$

The ^{222}Rn exhalation rate calculated from ^{222}Rn concentration

Radon exhalation rate was calculated by using eq. 6 [12]. Where: (λ = 0.00756 h^{-1}) radon decay constant, V = 0.00104 m^3 which is the volume of the empty container and A = 0.014 m^2 the surface area of the sample and T is the exposure time in hours

$$E_x \quad \frac{\lambda V C_{\mathrm{Rn}}}{A[T \quad (e^{\ \lambda T} \quad 1)/\lambda]} \tag{6}$$

The ^{222}Rn exhalation rate calculated from ^{226}Rn content

Furthermore, ^{222}Rn exhalation rate is mathematically expressed as a function of ^{226}Ra concentrations and the soil parameter such as the soil emanation coefficient f, soil density ρ, porosity ε, and the effective diffusion coefficient D_e [13]

$$E_x \quad \sqrt{D_e \lambda}\, f\rho(1 \quad \varepsilon)\mathrm{Ra} \tag{7}$$

Statistical analysis

The data analysis was based on the different parameters that may affect the concentration of radon mainly: geology of the study area, selecting perfect tubes for soil storing by both techniques, and appropriate for experiment usage. The statistics analyses was used to organize, analyze, summarize, and make predictions regarding the collected data. The main parameters used in measurement of the area are the minimum, maximum, average and standard deviation. Furthermore, the statistical t-test was used to examine if the radon concentrations of these study parameters were significantly different from each other. The P-value of less than 0.05 was always considered significant. Excel software program was used for data entry and analysis.

RESULTS AND DISCUSION

Table 2 shows the minimum, maximum and average of ^{222}Rn, ^{226}Ra, and E_x, by both methods, in four districts of Duhok province. It can be noted from the results that the concentration of ^{222}Rn and ^{226}Ra in soil samples varied appreciably from one point to another. Minimum value of radon concentration was measured at the Point 28, in Amedi district, while the maximum radon concentration was found in Duhok district, which is at the Point 12. Furthermore, the minimum and the maximum values of radium content was measured at sites 28 and 13, respectively. The variation may be due to the geochemical process in soil or, geological condition of the locations. Figure 3 illustrates a positive correlation between radon concentration and radium content with correlation coefficients of 0.73. This means that, radium content in soil is the main source of radon. This trend was observed by many researchers [14, 15].

Figures 4 and 5 illustrate the correlations between radon exhalation rate and concentration of both ^{222}Rn and ^{226}Ra in each sample of soil, respectively. Statistical *t*-test demonstrates that a strong positive correlation has been observed between radon exhalation rate with both radon concentration ($R^2 = 1$) and radium concentration ($R^2 = 0.66$). Also, a positive relation has been found between radon exhalation and the concentrations of both radon and radium in soil. This is because there is a direct relationship between radon and radium concentrations with radon exhalation rate [13]. Findings in this study are consistent with those of many other studies that have been done on this topic [16].

A comparison between radon exhalation rates, measured by radon concentration and radium concentration, is shown in fig. 6. A good correlation has been observed between them. According to *t*-test there is no significant difference between E_x measured by the two methods ($p > 0.05$). According to the previous region studies, the outcomes of this study are broadly in agreement with the research carried out throughout the world [16, 17, 18]. Furthermore, the study has confirmed that ^{222}Rn and ^{226}Ra content at the observed location does not increase the risk to human health, because the total average values of ^{222}Rn, ^{226}Ra, and E_x are below the average global values [3, 17]

Table 2. Average measurements of ^{222}Rn concentration, ^{226}Ra content and radon exhalation rate E_x by both methods in four districts of Duhok province

Duhok districts		Rn [Bqm^{-3}]	Ra (Bqkg^{-1}]	E_x [Bqm^{-2}h^{-1}] by Rn	E_x [Bqm^{-2}h^{-1}] by Ra
Duhok	Minimum	198 2.4	14.7 1.1	28 3.1	25.6 3.5
	Maximum	374 7.8	27 2.6	55.7 1.6	50.6 4.1
	Average	274 4.7	20.8 0.7	40.9 7.1	37.1 0.3
Somel	Minimum	188 2.1	13 1.6	28 1.6	25.4 3.5
	Maximum	341 6.8	26.8 2.5	50.8 2.5	46.1 2.8
	Average	243.7 3.8	18.4 0.1	36.3 1.3	33 1.2
Amedi	Minimum	110 1.1	17 1.8	16.4 1.0	14.9 6.8
	Maximum	289 5.2	21 2.0	43 0.9	39.1 3.6
	Average	169.8 1.5	18.4 0.8	25.3 1.4	23 1.7
Zakho	Minimum	98 0.6	15.8 1.2	14.6 3.2	13.3 2.4
	Maximum	150 1.5	17 2.5	22.4 1.5	20.3 5.1
	Average	119.3 1.0	16.4 1.9	17.8 3.6	16.2 3.3
Average		202 4.6	18.5 2.1	30 1.9	27 3.8

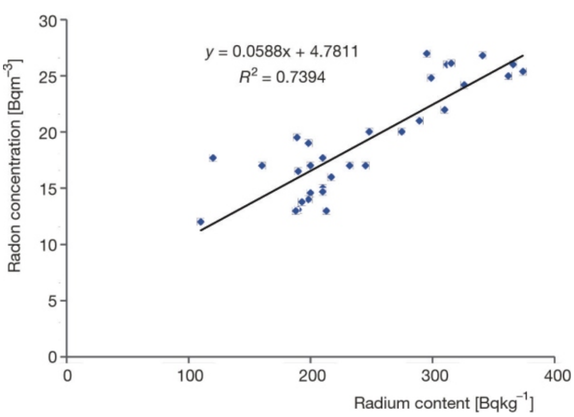

Figure 3. Radon and radium concentration correlation for 33 sites

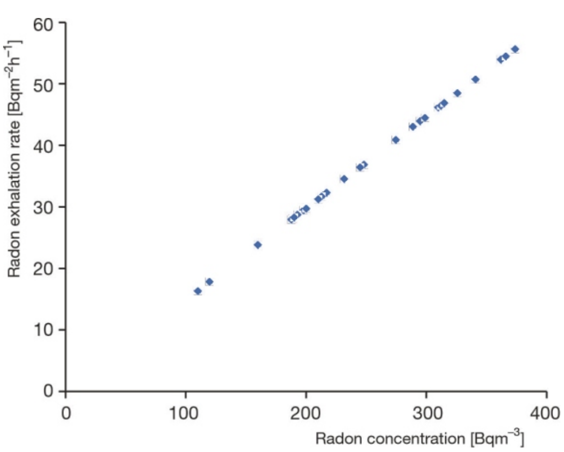

Figure 4. Direct relationship between ^{222}Rn and E_x at all sample points

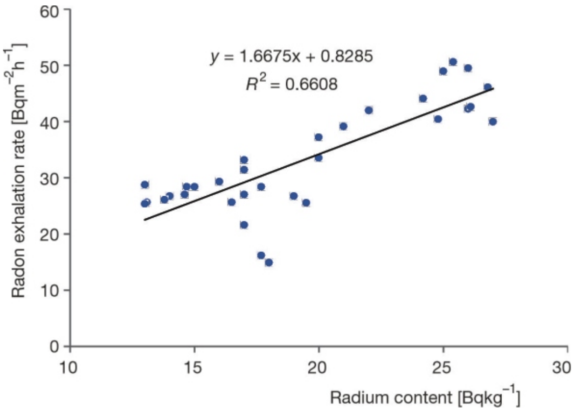

Figure 5. Correlation between ^{226}Ra and E_x at all sample points

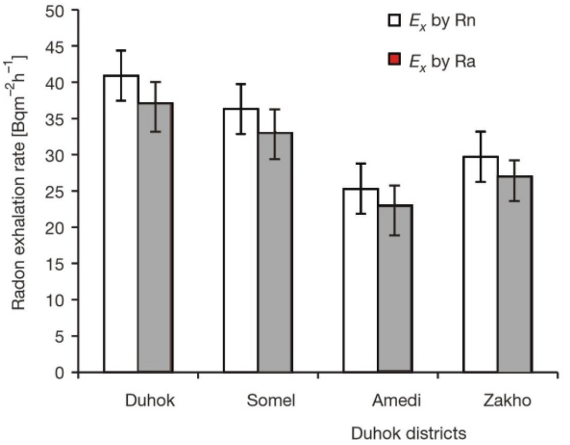

Figure 6. Comparison between the average values of radon exhalation rates estimated by radon concentration and radium content at four districts of Duhok

CONCLUSION

Radon exhalation rate for soil has been calculated in four districts of Duhok province by two different techniques, first by measuring radon concentration and second by estimating radium content with soil parameters. A direct correlation was found between radon exhalation rate measured by both methods and radon concentration. Also, radon exhalation rate obtained by method one is in excellent agreement with radon exhalation rate obtained by method two. Furthermore, positive correlations were observed between radon exhalation with the concentrations of both radon and radium concentrations in soil. The results show that the value of radon concentration, radium content and radon exhalation rates are less than the mean world value limit.

ACKNOLEDGMENT

We acknowledge the support of this work by the University of Duhok, College of science, physics department. Furthermore, a special thanks to the Jomma M. for supporting this work.

AUTHORS' CONTROBUTION

The corresponding author W. A. H. Alhamdi conducted the sample collection, sample storing, experimental procedure, measured the samples, analyzed the data and wrote the paper under the supervision of author K. M-S. Abdullah.

REFFERENCES

[1] Vukanac, I. S., *et al*., Assessment of Natural Radio Activity Levels and Radon Exhalation Rate Potential from Various Buildings Materials, *Nucl Technol Radiat, 35* (2020), 1, pp. 64-73
[2] Alhamdi, W., *et al*., Estimation of Indoor Radon Concentration and Dose Evaluation of Radon and its Progeny in Selected Dwellings in Duhok City, Kurdistan Region, *Iraq. Internation Journal of Radiation Research, 20* (2022), 2, pp.461-466
[3] Mann, N., *et al*., Radon-Thoron Measurements in Air and Soil from Some Districts of Northeren rate of India, *Nucl Technol Radiat, 30* (2015), 4, pp. 294-300
[4] Alhamdi, W., *et al*., Determination of Radium and Radon Exhalation Rate as a Function of Soil Depth of Duhok Province – Iraq. *Journal of Radiation Rsearch and Applied Science, 14* (2021), 1, pp. 486-494
[5] Yanchao, S., *et al*., Study on a New Charcoal Closed Chamber Method for Measuring Radon Exhalation Rate of Building Materials, *Radiation Measurements, 134* (2020), doi: 10.1016/g.radmeas.2020.106308
[6] Azeez, H. H., *et al*., Measurement of Radon Concentrations in Rock Samples from the Iraqi Kurdistan Region Using Passive and Active Methods, *Arabian Journal of Geosciences, 14* (2021), 7
[7] Yang, J., *et al*., Modeling of Radon Exhalation from Soil Influenced by Environmental Parameters, *Science of the Total Environment, 656* (2019), pp. 1304-1311
[8] Dimović, S. D., *et al*., Assessment of Inviromental Radioactivity and Health Hazard at Stara Planing Region, *Nucl Technol Radiat, 35* (2020), 4, pp. 354-360
[9] Kaur, R., *et al*., Environmental Ra don, Its Exhalation Rates and Activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in Northern India, 268 *Nucl Technol Radiat, 35* (2020), 3, pp. 268-282
[10] Hansman, J., Design and Construction of a Shield for the 9" 9" NaI(Tl) Well-Type Detector, *Nucl Technol Radiat, 29* (2014), 2, pp. 165-169
[11] Pisapak, P., *et al*., Correlation Between Radon and Radium Concentrations in Soil and Estimation of Natural Radiation Hazards in Namom District, Songkhla province (Southern Thailand), *Environmental Earth Science, 76* (2017), https://doi.org/10.1007/s12665-017-6439-6
[12] Elzain, A. A., Determination of Radium Concentration and Radon Exhalation Rate in Soil Samples Using CR-39, *Advances in Applied Science Research, 6* (2015), pp. 96-102
[13] Hosoda, M., *et al*., Fukushi, Effect of Soil Mosture Content on Radon and Thoron Exhalation, *Journal of Nuclear Science and Technology, 44* (2007), 4, pp. 664-672
[14] El-Zohry, M., *et al*., The Radiological Impacts of TE-NORM Activity in Upper Egypt, *International Journal of Recent Research and Applied Studies, 33* (2017), pp.7-18

[15] Kaliprasad, C. S., *et al*., Radon Exhalation Rate and Radon Activity in Soils Riverine Environs of South Kaarantaka, *Radiation Protection and Environment. 41* (2018), pp.189-191
[16] Ishimori, Y., *et al*., *Measurement and Calculation of Radon Releases from NORM Residues*, International Atomic Energy Agency Vienna 2013
[17] Tan, Y., *et al*., The Method for Recalibration of Thoron Concentration Reading of RAD7 and Obtaining the Thoron Exhalation Rate from Soil Surface, *Nucl Technol Radiat, 28* (2013), 1, pp. 92-96
[18] Soares, S., *et al*., Comparison of Radon Mass Exhalation Rate Measurement from Building Materials by Two Different Methods, *Radiation Protection Dosimetry, 191* (2020), pp. 255-259

Received on July 7, 2022
Accepted on November 7, 2022

Валат А. Х. АЛХАМДИ, Каири М-С. АБДУЛАХ

МЕРЕЊЕ ЈАЧИНЕ ЕМИСИЈЕ РАДОНА ИЗ ЗЕМЉЕ У ГРАДУ ДУХОКУ ПОМОЋУ ДВЕ ТЕХНИКЕ

Јачина емисије радона представља брзину којом радиоактивни гас радон излази из тла у атмосферу. У овом раду измерена је јачина емисије радона за 33 узорка земљишта у четири округа провинције Духок, двема различитим техникама: једне, коришћењем концентрације радона, и друге, коришћењем садржаја радијума – обе применом алфа и гама спектроскопије. За мерење радона употребљен је алфа осетљив детектор РАД7, док је другом методом јачина емисије радона изражена као функција концентрације радијума, мерена детектором NaI (Tl) облика јаме и другим параметрима земљишта. Анализа показује да просек јачине емисије радона, по првој техници, варира од 14.6 3.2 $Bqm^{-2}h^{-1}$ до 55.7 1.6 $Bqm^{-2}h^{-1}$, док код друге технике варира од 13.3 2.4 $Bqm^{-2}h^{-1}$ до 50.6 4.1 $Bqm^{-2}h^{-1}$. У целини, измерене вредности обема метода на свим тачкама узорка добро су корелисане и мање су од глобалне средње препоручене вредности.

Кључне речи: гас радон, јачина емисије радона, алфа спектроскопија, радијум, гама спектроскопија