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RESEARCH ARTICLE

RADON EXHALATION RATE AND ANNUAL EFFECTIVE DOSE FOR DIFFERENT ROCK TYPES AND EXCESS LIFETIME CANCER RISK FROM RADON EXPOSURE

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ABSTRACT

Purpose: Radon (^{222}Rn) and its decay products clinging to airborne particles settle in the lungs when inhaled and can lead to lung cancer. The main source of ^{222}Rn is rocks and soil in the Earth's crust and causes indoor radon exposure when local geological material is used as a building material. Accordingly, the primary aim of the study is to determine the radon activity concentrations (C_{Rn}) and exhalation rates (E_A and E_M) from different rock types taken from the Aliağa-İzmir region. The study also estimates the annual effective dose (AED) and the excess lifetime cancer risk (ELCR).
Material and Methods: For the measurement of the C_{Rn}, E_A, and E_M in different rock types, the Can technique with LR-115 detector was utilized. The AED and the ELCR were estimated using the C_{Rn} in the samples.
Results: The results for the examined rock samples were ranged between 66±4 and 1711±13 Bq m^{-3} for C_{Rn}, between 51±3 and 1309±10 mBq m^{-2} h^{-1} for E_A, between 2.68±0.18 and 64.02±0.47 mBq kg^{-1} h^{-1} for E_M, between 1.67 and 43.16 mSv y^{-1} for AED, and between 0.006 and 0.151 for ELCR.
Conclusion: The higher radiological risks in terms of radon exposure were related to the rocks of volcanic origin.

Keywords: Radon, rock types, radon exhalation rates, LR-115 Type II, Sealed Can Technique.

INTRODUCTION

With the development of nuclear technology in the last century and the use of this technology in energy, industry, and research, especially in medicine, human beings have started to be exposed to artificial radiation in addition to natural radiation sources. Despite this, the highest contribution to the radiation dose that people receive from various sources still comes from natural radiation sources. From the radiological viewpoint, the most important natural radionuclides in the terrestrial environment are ^{232}Th, ^{238}U, and its decay product, ^{222}Rn. The source of these radionuclides is the Earth`s crust and their abundance is related to the different rock types. So, the main factors controlling the radiation of terrestrial origin are the geological/geochemical structure and tectonic features of the region. In addition, the factors controlling the ^{222}Rn concentration and ^{222}Rn release in the soil are the structural geological factors such as faults, fractures, shear zones, underground water basins, and physical factors such as soil permeability, moisture content, particle size (1–3).

In terms of public health, it is of great importance to know the main factors that create health problems, to investigate their causes, and to keep them under control. In this respect, one of the most important causes of death due to lung cancer is radon gas.

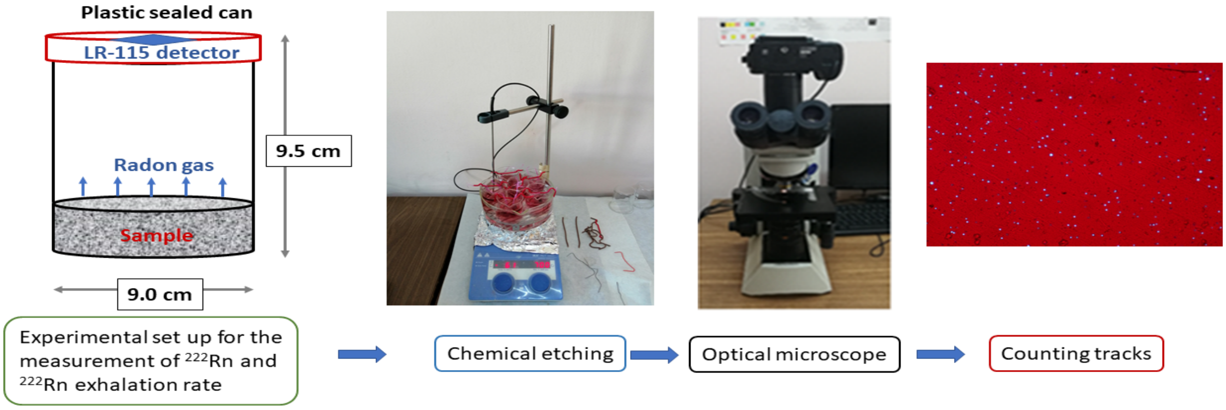

Figure 1. Experimental procedure

Radon, a decay product of radium (^{226}Ra) in the natural radioactive series of uranium, is a tasteless, odourless, colourless, inert gas having a half-life of 3.8 days. While ^{226}Ra acts as a generator in the formation of ^{222}Rn owing to its long half-life, ^{210}Pb in the decay chain effectively protects this decay chain with its long half-life. Radon has poor reactivity, so it does not chemically bound to tissues when inhaled. Also, its solubility in tissues is very low. However, the short half-lived products of radon's radioactive decay are polonium (^{218}Po, ^{214}Po), lead (^{214}Pb), and bismuth (^{214}Bi), and these radioactive products are absorbed by dust and water particles suspended in the air in indoor and outdoor environments, causing the formation of particles that emit active alpha rays. These particles can easily penetrate the upper respiratory tract through respiratory tract, reach the lungs, attach to the walls of the lungs, and act as a permanent local alpha radiation source there. Studies on the effects of these decay products, which can emit alpha-beta rays, on the respiratory system have shown that the biological half-lives of these radionuclides in the lungs can be from a few hours to a day, so they can be very dangerous in terms of the risk of causing lung cancer over time as a result of damage to the lung tissue (2,4-8).

In many countries such as the USA, Finland, England, Sweden, and Norway, a national radon program is carried out under the title of public health program. Within the scope of this radon program, radon maps of the geographical regions that are the riskiest in terms of the population being affected by radon are drawn, and activities are carried out to raise awareness of the public and building material producers. On the other hand, the deficiencies in this area in our country can be listed as the lack of comprehensive regional radon maps in Turkey, the lack of research to establish a connection with radon gas in people who died of lung cancer, the lack of practices for raising awareness of the society on the subject, and the lack of measurements of the radon levels that vary according to the regions where the building products used in building production.

In this context, the study is aimed to establish a regional database by determining the radon activity concentrations and the radon exhalation rates in different rock types taken from the Aliağa-İzmir region and to evaluate these rocks in terms of radiological risk if they are used as building materials.

MATERIAL AND METHODS

The Sealed Can technique in conjunction with Solid-state nuclear track detector (SSNTD) LR-115 Type II (Dosirad, France) was utilized to ascertain the C_{Rn}, E_A, and E_M values in eighteen different rock samples from Aliağa, Izmir, Turkey. The comprehensive geological setting and locations of the sampling area were given by Çam et al 2013 (9). The sample preparation step for radon measurements was carried out at Ege University, Department of Physics, Sample Preparation Laboratory. The rock samples were crushed to fine-grained, and dried in an oven at 105°C for 24 h. After being weighed, the rocks were placed into plastic containers with dimensions of 9.5 cm high and 9.0 cm in diameter (Figure 1).

LR-115 Type II SSNTDs with an area of 1.2x1.2 cm^2 were taped to the inner side of the lid of each container with the alpha sensitive side of the SSNTD facing the rock sample. Then, prepared plastic containers were carefully sealed for 90 days. When the exposure time was over, the samples were transferred to the Dokuz Eylul University, Department of Physics, Radon Measurement Laboratory for chemical processes and radon measurement. The detectors were collected and etched in 2.5 N NaOH solution at 60°C for 90 minutes. After the etching, the LR-115 Type II detectors were rinsed with pure water at room temperature for 20 minutes and left to dry in a clean environment without human touch. The track density in each SSNTD was counted with the help of an optical microscope having 10x10 magnification. The background count of the detector was also determined using an optical microscope and deducted from the count of all SSNTD LR-115 Type II detectors. The error in the trace density (N) measured per unit area fits the Poisson statistics and is given by the square root of the total count ($\sigma = \sqrt{N}$). Presuming the counts of tracks per unit area of detectors are proportional to radon exposure, radon activity concentrations in the rock samples were calculated according to Equation 1 (10,11):

$$C_{Rn} = \frac{D}{KT} \quad (1)$$

where C_{Rn} (Bq m^{-3}) is radon activity concentration, D (tr cm^{-2}) is the net track densities for the alpha particles of the radon, T (d) is exposure time, and K (Bq^{-1} m^3 tr cm^{-2} d^{-1}) is the calibrated detector sensitivity coefficient. The calibration of the detectors was made in the radon exposure chamber at the Ege University, Institute of Nuclear Sciences and the calibration coefficient was determined as 0.131 Bq^{-1} m^3 tr cm^{-2} d^{-1}.

The surface radon exhalation rates (E_A) of rocks were computed from the following expression (12,13):

$$E_A = \frac{CV\lambda}{A\left[T+\frac{1}{\lambda}\{e^{-\lambda T}-1\}\right]} \quad (2)$$

and mass radon exhalation rates (E_M) were modified as:

$$E_M = \frac{CV\lambda}{M\left[T+\frac{1}{\lambda}\{e^{-\lambda T}-1\}\right]} \quad (3)$$

where E_A (Bq m^{-2} h^{-1}) is the expression of the radon exhalation rate in terms of area, E_M (Bq kg^{-1} h^{-1}) is the expression of the exhalation rate in terms of mass, C (Bq m^{-3} h) is the integrated radon exposure, T (hours) is the exposure time, V (m^3) the volume of the container, λ (7.56×10^{-3} h^{-1}) the decay constant for radon, A (m^2) the area of the container, M (kg) is the mass of the sample.

The annual effective dose AED (Sv y^{-1}) was estimated using the radon activity concentrations, exposure times, and the dose conversion factor according to UNSCEAR 2000 report (14):

$$AED = C_{Rn} \times F \times O \times T \times D \quad (4)$$

where C_{Rn} is the radon activity concentration (Bq m^{-3}), F is the equilibrium factor of 0.4 indoors, O is the indoor occupancy factor of 80%, T is the time (8760 hours $year^{-1}$) and D is the dose conversion factor of 9 nSv h^{-1} per Bq m^{-3}.

The excess lifetime cancer risks (ELCR) were computed to assess the increased risk of cancer caused by lifetime exposure to a toxic substance (15). Thus, the excess lifetime cancer risk (ELCR) owing to exposure to indoor radon acquired over the lifetime was calculated using the Equation 5:

$$ELCR = AED \times T \times RF \quad (5)$$

where AED is the annual effective dose (Sv y^{-1}), T is the duration of life (70 years) and RF is the risk factor of 0.05 Sv^{-1}.

RESULTS

The results of the study of the C_{Rn}, E_A, E_M, AED, and ELCR of different rock types from the Aliağa-İzmir region were depicted in Table 1. The C_{Rn} values in the rock samples were ranged from 66 ± 4 to 1711 ± 13 Bq m^{-3} with a mean value of 566 ± 119 Bq m^{-3}. The results disclose that the highest ^{222}Rn activity concentration value was found for the rhyolitic rock sample (R15), while the lowest one was in the volcanoclastic rock sample (R18). Extremely high ^{222}Rn activity concentrations were obtained in rhyolitic (R15), Foca tuff (R11), and volcanic (R17) rocks when compared to other studied rocks.

The surface exhalation rates (E_A) of ^{222}Rn gas from rock samples were in the range of 51 ± 3 and 1309 ± 10 mBq m^{-2} h^{-1}, and the E_M values varied from 2.68 ± 0.18 to 64.02 ± 0.47 mBq kg^{-1} h^{-1} (Table 1). The arithmetic means for E_A and E_M were 433 ± 91 mBq m^{-2} h^{-1} and 19 ± 5 mBq kg^{-1} h^{-1}, respectively.

The correlations between C_{Rn} and E_A and between C_{Rn} and E_M were illustrated in Figure 2 and Figure 3, respectively. High correlations with R^2 values of 1 and 0.98 were obtained between the analysed quantities for the rock samples, respectively.

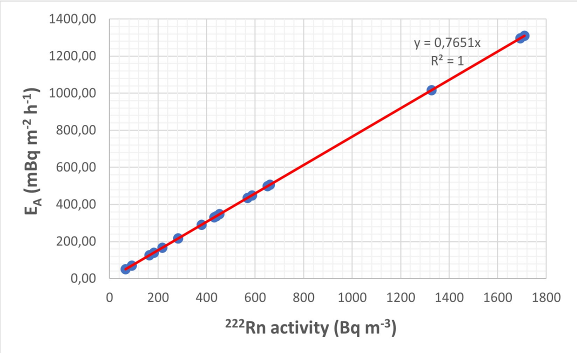

Figure 2. Regression model of the relationship between surface exhalation rates (E_A) and ^{222}Rn activity concentration

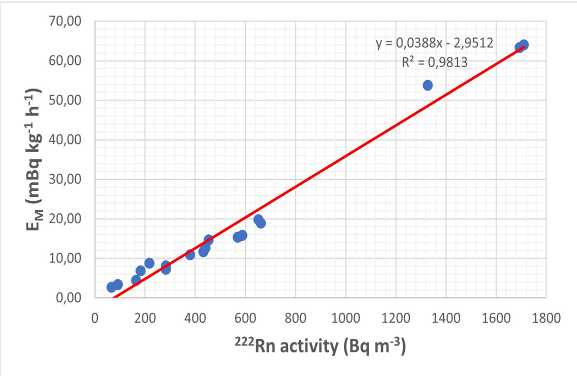

Figure 3. Regression model of the relationship between mass exhalation rates (E_M) and ^{222}Rn activity concentration

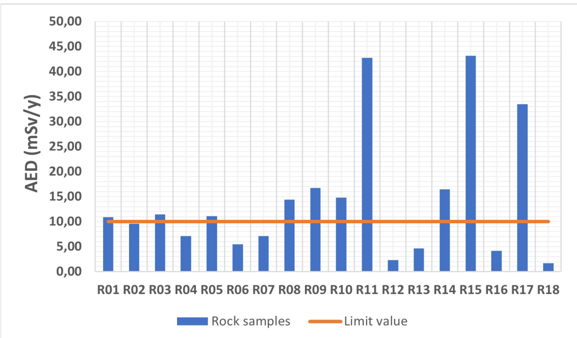

Figure 4. The annual effective doses for rock samples and limit value (14,17)

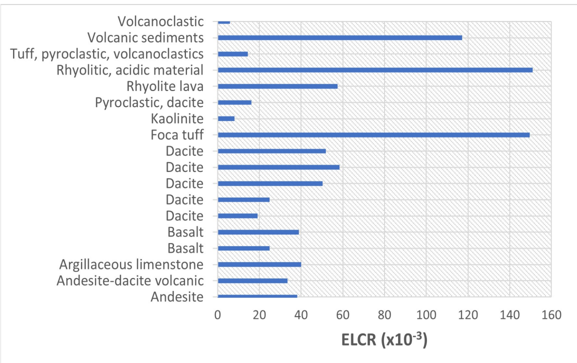

Figure 5. The ELCR values for analysed rock types

The annual effective dose values caused by inhalation of indoor ^{222}Rn gas released from different rock types if used as building materials were found to be varying from 1.67 to 43.16 mSv y^{-1} with a mean value of 14.29 ± 2.99 mSv y^{-1} (Figure 4).

In the case of using the investigated rock samples as building material, the excess lifetime cancer risk (ELCR) values owing to lifetime exposure to indoor radon were found to range between 0.006 to 0.151 with an average of 0.05 ± 0.01. The ELCR results were illustrated according to rock type and demonstrated in Figure 5. Clearly seeing from Figure 4, rhyolitic, acidic material (R15), Foca tuff (R11), and volcanic sediments (R17), are the three rocks with the highest cancer risk in case of indoor radon exposure if they are used as building material.

DISCUSSION

Radon is a natural gas emitted from many substances such as soil, rock, building materials, and artificial fertilizers containing trace amounts of ^{238}U, and is found almost everywhere, even in very small concentrations. Radon in terrestrial materials depends on the ^{226}Ra activity concentrations in the material. Granites, which are plutonic igneous rocks, have high, basalts, which are volcanic igneous rocks, low, and sedimentary and metamorphic rocks have moderate radium content. Even though exceptional values of ^{226}Ra occur in some clastic sedimentary rocks, mainly the results are in agreement with the soil values (16). In the present study, most of the rock samples are of volcanic origin. Besides being of volcanic origin, the higher radon concentrations can be attributable to the high silica content of the rocks as previously stated by Çam et al. (2013). The silica contents of the rocks in decreasing order are in rhyolites, tuffs, dacites, andacites and, basalts (9).

Authorized International Organizations in the Field of Radiation Protection and Safety have published advisory limit values for indoor radon concentration, nonetheless, limits are diverse for each country or region. The International Commission on Radiological Protection (ICRP) recommended a ^{222}Rn concentration of 300 Bq m^{-3} for dwelling (17). United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and World Health Organization (WHO) declared that the limit value of ^{222}Rn activity should not exceed 100 Bq m^{-3} (14,18). In Turkey, ^{222}Rn activity concentrations should not exceed 400 Bq m^{-3} in houses and 1000 Bq m^{-3} in workplaces according to the published in the Official

Table 1. The results of C_{Rn}, E_A, E_M, AED, and ELCR for the analysed rocks

Sample	Rock Type	C_{Rn} (Bq m^{-3})	E_A (mBq m^{-2} h^{-1})	E_M (mBq kg^{-1} h^{-1})	AED (mSv y^{-1})	ELCR x10^{-3}
R01	Andesite	432 ± 7	331 ± 5	11.68 ± 0.19	10.90	38
R02	Andesite-dacite volcanic	380 ± 7	290 ± 5	10.86 ± 0.19	9.58	34
R03	Argillaceous limenstone	454 ± 7	347 ± 6	14.71 ± 0.24	11.44	40
R04	Basalt	282 ± 6	216 ± 5	8.08 ± 0.18	7.13	25
R05	Basalt	441 ± 7	337 ± 5	12.61 ± 0.21	11.12	39
R06	Dacite	217 ± 6	166 ± 4	8.81 ± 0.23	5.48	19
R07	Dacite	282 ± 6	216 ± 5	7.23 ± 0.16	7.13	25
R08	Dacite	569 ± 8	436 ± 6	15.38 ± 0.21	14.36	50
R09	Dacite	662 ± 8	506 ± 6	18.93 ± 0.24	16.69	58
R10	Dacite	587 ± 8	449 ± 6	15.87 ± 0.22	14.82	52
R11	Foca tuff	1694 ± 13	1296 ± 10	63.38 ± 0.47	42.73	150
R12	Kaolinite	91 ± 5	70 ± 4	3.42 ± 0.17	2.30	8
R13	Pyroclastic, dacite	183 ± 5	140 ± 4	6.84 ± 0.20	4.61	16
R14	Rhyolite lava	651 ± 8	498 ± 6	19.81 ± 0.25	16.43	58
R15	Rhyolitic, acidic material	1711 ± 13	1309 ± 10	64.02 ± 0.47	43.16	151
R16	Tuff, pyroclastic, volcanoclastics	164 ± 5	126 ± 4	4.44 ± 0.14	4.15	15
R17	Volcanic sediments	1327 ± 11	1015 ± 9	53.81 ± 0.46	33.48	117
R18	Volcanoclastic	66 ± 4	51 ± 3	2.68 ± 0.18	1.67	6

Table 2. The comparison data for the E_A and E_M with the studies in the literature

Region	Rock Type	E_A (mBq m^{-2} h^{-1})	E_M (mBq kg^{-1} h^{-1})	Reference
Egypt (El-Sebaeya)	Phosphate rocks	520-26160	-	(21)
Egypt (Abu-Tartur)		6200-19070	-	
India (Tusham ring complex, Haryana)	Granitic rocks	230-2110	6.94-63.76	(22)
India	Different types of rocks	-	14.82 - 37.85	(23)
Italy (Campania)	Bauxite	653	33	(6)
	Claystone	696	36	
	Ignimbrites	393-451	21-23	
	Lavic Stones	8900-11847	462-616	
	Limestones	90-622	4-32	
	Marbles	471-1112	24-57	
	Pozzolans	2289-7661	119-398	
	Pumices	11050-15730	574-818	
	Sandstones	645-878	33-45	
	Siltstones	333-727	17-37	
	Tephrites	319-342	16-17	
	Tuffs	372-7730	19-402	
Turkey	Different types of rocks	51 - 1309	2.68 - 64.02	This study

Gazette dated 24/03/2000 and numbered 23999 Radiation Safety Regulation's 37th article which amended by the Regulation on the Amendment of the Radiation Safety Regulation published in the Official Gazette dated 29/09/2004 and numbered 25598 (19). From the radon concentration levels presented in Table 1, 25 percent of obtained results have values below 209 Bq m^{-3}, that of 50% below 436 Bq m^{-3}, and that of 75% less than 654 Bq m^{-3}. Accordingly, the C_{Rn} values measured in 50% of the examined rocks exceed the limit value given for Turkey if they are used as building materials. As mentioned above, the variation in ^{222}Rn activity levels in rock samples can be ascribed to the differences in the activity concentration of ^{226}Ra and silica content in rocks and the geographical and geological conditions.

As seen from Table 1, the surface exhalation rate values are greater than the mass exhalation rates for all rock samples because radon can leak into the interstitial space due to the recoil momentum from radioactive decay. If this event is in the interior of the rock, ^{222}Rn will be constituted in the interior and remain there trapped. Therefore, all radon in the rocks cannot escape the atmosphere. The difference between the exhalation rate values (E_A and E_M) can be ascribable to pore size, grain shape, porosity, and size additionally the ^{226}Ra activity content in the rocks under consideration (20).

The results of radon exhalation rates were compared with the results of the other rock types in literature and were listed in Table 2. According to the data of Table 2, in the current study, the obtained E_A and E_M values were lower than the given results for phosphate rocks by (21), for granitic rocks by (22), and for tuffs, pumices, lavic stones, pozzolans by (6); were higher than the reported values for different types of rocks like granite, gneiss, gabbro, sandstone, limestone, and shales by (23), for marbles, limestones, bauxite, Ignimbrites, tephrites, siltstones, sandstones, and claystone by (6).

The ICRP proposed that the curative action level against indoor radon be 10 mSv, with the action level in the 3–10 mSv y^{-1} range (17). Similarly, UNSCEAR declared the permission level as 10 mSv y^{-1} (14). For the samples studied, the mean annual effective dose owing to the internal exposure to ^{222}Rn gas is slightly above the limit level advised by the ICRP and UNSCEAR. The excess lifetime cancer risks (ELCR) were computed based upon the AED results. Therefore, the limit value for ELCR was obtained as 35×10^{-3} by substituting 10 mSv y^{-1}, which is the limit value for AED, in the ELCR formula. It can be shown from Table 1 and Figure 5 that the ELCR values obtained for 56% of the rock samples were higher than 35×10^{-3}.

CONCLUSION

The study contributes to enriching information about ^{222}Rn activity concentrations and exhalation rates in different rock types in Turkey. Higher ^{222}Rn exhalation rates (E_A and E_M) were observed in rocks of volcanic origin and high silica content. The differences among the results of the examined rock samples showed that the radon activity concentration and the ^{222}Rn exhalation rate were directly related to the lithological origin. These results reveal the importance of local radiological data in terms of using local geological material as a building material.

In addition, considering that the main source of ^{222}Rn is the rock in the crust, and it rises too high concentrations in soils consisting of rocks and building materials having rocks in their origin, the following measures are listed to reduce radon exposure before the building construction process:

- regional monitoring studies for radon and other terrestrial radionuclides (^{238}U and ^{232}Th) should be planned,
- regions with lack or inadequacy of data should be identified and continuously monitored,
- based on the obtained radiometric data, regional natural radiation maps should be drawn,
- construction and even residential areas should be determined in accordance with the created maps of the ^{222}Rn activity concentration or the ^{222}Rn exhalation rate.

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