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Calculation of radon exhalation rate of Ahlat stone

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Abstract

Natural radioactivity can be found in soil, rocks, plants, water, air, building materials, the homes we live in, and even inside our bodies. Radon (^{222}Rn) gas is the most significant natural source of ionizing radiation. ^{222}Rn decays and emits alpha particle radiation (5.49 MeV). When radon and its decay products are inhaled with normal air, most of the radon (approximately 70%) is exhaled, while the decay products adhere to tissues in the respiratory system, causing permanent damage. Because radon accounts for approximately 60% of total natural background radiation, radon exposure has become a worldwide problem. One of the key parameters for estimating environmental radon levels is the radon exhalation rate from building materials. This study calculated the radon and radium concentrations in the Ahlat Taşı building material, the radon exhalation rate, the radon emission coefficient parameters, and the annual effective dose, which is the radiological effect associated with radon. The study used an LR-115 Type II nuclear track detector. The results were compared with both global limit values and previous studies. The radon concentration value (330.51 Bq/m³) was found to be above the limit value, and the annual effective dose equivalent (8.33 mSv/y) was within the action level range recommended by the ICRP. Surface and mass exhalation rates were 322 mBq/m²h and 12.36 mBq/kg h, respectively. Consequently, some measures are recommended to reduce indoor radon levels in residences when Ahlat Stone is used as a building material.

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1. Introduction

Natural radioactivity can be found in soil, rocks, plants, water, air, building materials, inside buildings, and even inside our bodies. The most important source of ionizing radiation from natural sources is Radon (^{222}Rn) gas. ^{222}Rn is a noble gas produced by the radioactive decay of radium-226, a product of the uranium-238 decay series. With a half-life of 3.82 days, ^{222}Rn decays via alpha emission (5.49 MeV) to produce solid decay products such as ^{218}Po (polonium) and ^{214}Po . Being a gas, radon can move through pore spaces in rocks or soil by recoil (from the alpha decay) and diffusion through solid matrices. When inhaled, radon itself is mostly exhaled, but its solid decay products (especially Po-218 and Po-214) attach to aerosols and stick to lung tissues, delivering damaging alpha radiation doses. This can lead to Lung cancer (most studied and well-established outcome), Skin cancer and kidney disease (from less direct exposure, potentially via bloodstream) [1], [2], [3].

According to the 1993 UNSCEAR report, radon exposure has emerged as a major global health problem because it accounts for approximately 60% of the total natural background radiation to which the population is exposed [1]. In response to the risks associated with radon, the International Commission on Radiation Protection (ICRP) recommends that national regulatory authorities set a reference level for indoor radon concentration in the range of (100-300) Bq/m³ [4]. The World Health Organization (WHO) has stated that indoor radon exposure is the second leading cause of lung cancer after smoking. Consequently, the WHO recommends a lower reference level of 100 Bq/m³ for radon concentrations in buildings to minimize public health risks [5].

Radon exhalation rate is an important factor in environmental radiation exposure and is used to assess the contribution from building construction materials to indoor radon levels. Primary Factors Affecting Radon Exhalation Rate: ^{226}Ra (radium) Concentration (higher Ra-226 content results in more radon production), Radon Diffusion Factor (determines how easily radon passes through the material), Porosity of the Material (more open pore structures facilitate radon movement), Density of the Material (higher density generally inhibits radon diffusion), Radon Diffusion Coefficient (material-specific property that describes radon movement through a solid matrix). Radon exhalation measurement is done by the "Closed Box Method". This is a widely accepted and cost effective method. The sample is sealed in an airtight container with a SSNTD (solid state nuclear track detector). Over time, radon exhaled from the material accumulates and its decay products leave tracks (alpha particles) in the detector. Detectors used are: CR-39, LR-115, CN-85 and all are SSNTD types. The LR-115 Type II detector features: 100 µm polyester base, 12 µm red cellulose nitrate sensitive layer, sensitive to alpha particles in the energy range of 1.7–4.8 MeV. After exposure, the detectors are chemically etched to reveal alpha tracks and are then counted under a microscope or by automated systems [1], [6].

Ahlat Stone is a volcanic stone extracted by traditional methods from the foothills of Mount Nemrut near the Ahlat district of Bitlis province (Figure 1). Ahlat stone has many natural colors such as red, black, gray, white, light yellow and ash color. Ahlat stone is used in architectural works such as houses, mosques, tombs and bridges, as well as in tombstones, inscriptions, fountains and various works [7].



Fig. 1. Ahlat Stone.

This study aimed to calculate the radon and radium concentrations in Ahlat Stone building material, radon exhalation rate, radon emission coefficient parameters and annual effective dose, which is the radiological effect due to radon.

2. Material and method

Ahlat Stone was obtained from the Ahlat district of Bitlis province. It was ground into a fine powder using a mortar and pestle. It was then passed through a 150-micron mesh sieve. The resulting 200-g sample was placed in a cylindrical container measuring 7 cm in diameter and 13 cm in height (Figure 2). Using cotton gloves, the insensitive surface of the LR-115 film detector was cut to $(1.5 \times 1.5) \text{ cm}^2$ and glued to the container lid using patafix. The lid of the cylindrical container was taped to prevent internal gas leaks. Thus, the detector does not record background radiation from the storage environment.



Fig. 2. Cylindrical container in which LR-115 is placed.

The sample was stored in a closed and dry environment for 90 days to minimize contamination and ensure accurate measurements. After exposure, the detector was removed and a chemical etching process was performed by etching with 2.5N NaOH at $60 (\pm 1) ^\circ\text{C}$ constant temperature and 120 rpm in a magnetic heater for 100 minutes in order to count clear traces of alpha particles on the detector film. Then, the film was rinsed with pure water at 120 rpm for 20 minutes and left to dry. Euromex brand optical microscope and ImageFocus Plus V2 image transmission system were used to view the traces of alphas on the detector. The 100x magnification objective of the microscope was selected. Traces were counted and noted for a 1 cm^2 area (Figure 3).

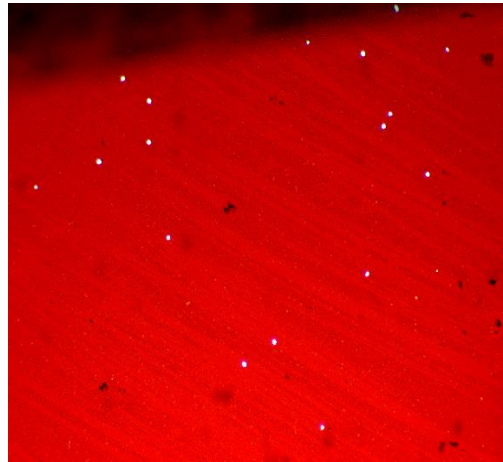


Fig. 3. Radon traces on a 1 mm^2 area.

If it is accepted that the density of the trace formed on the film is proportional to the amount of ^{222}Rn , the radon concentration in that environment is obtained with Equation 1 [8]:

$$C_{Rn} = \frac{\rho}{k_f T_{eff}} \quad (1)$$

Here, C_{Rn} : radon concentration (Bq/m^3), ρ : trace density on the film (trace/cm^2), T_{eff} : effective exposure time (day), k_f : calibration factor ($\text{izm}^3/\text{Bqcm}^2\text{day}$). The calibration factor $k_f = 0.021$ was used in the study [1]. The effective exposure time was obtained from the following equation [3]:

$$T_{eff} = T + \tau(e^{-\lambda T} - 1) \quad (2)$$

Here, T : actual exposure time (day), λ : radon decay constant (day^{-1}) and $\lambda = 0.693/T_{1/2}$, τ : mean lifetime for radon (day) and $\tau = 1/\lambda$. $T_{1/2} = 3.82$ days for radon. The effective radium content of the sample was obtained using the expression below [2]:

$$C_{Ra} = \frac{\rho V}{\eta M T_{eff}} \quad (3)$$

Here, C_{Ra} represents the effective radium content (Bq/kg), M represents the mass of the sample (kg), η represents the detector sensitivity ($\text{trace m}^3/\text{Bqcm}^2\text{d}$), and V represents the void volume of the container. Surface and mass exhalation rates were calculated using the following equations 4 and 5, respectively [2]:

$$E_A = \frac{\rho}{\eta A} \left[\frac{\lambda V}{T_{eff}} \right] \quad (4)$$

$$E_M = \frac{\rho}{\eta M} \left[\frac{\lambda V}{T_{eff}} \right] \quad (5)$$

where, E_A : radon surface exhalation rate ($\text{Bq/m}^2\text{h}$), E_M : radon mass exhalation rate (Bq/kg h), A : area of sample (m^2), M : mass of sample (kg). The annual effective dose equivalent can be calculated from radon concentrations using the conversion factor of $9.10^{-6} \text{ mSv}/(\text{Bqh/m}^3)$ recommended by UNSCEAR. Considering that people spend approximately 80% of their lives indoors (7000 h per year) and assuming a balance factor of 0.4, The annual effective dose from radon exposure may be obtained by equation (6) [9]:

$$D_{eff} (\text{mSv/y}) = \epsilon f T C_{Rn} \quad (6)$$

where, f : conversion factor, T : time spent in the indoor area (7000 h/y), ϵ : balance factor and C_{Rn} : represents radon concentration. The radon emission coefficient (F), an important radiological index, is used to evaluate the amount of ^{222}Rn gas released from building construction materials. The F value is found with the following equation [6]:

$$F (\%) = E_M / C_{Ra} \lambda \quad (7)$$

The contribution of radon released from building materials and soil to indoor space is obtained from the following equation [10]:

$$C_{Rn} = \frac{E_A S_r}{V_r \varphi} \quad (8)$$

where C_{Rn} : radon concentration in construction materials contributing to indoor radon, S_r : area of indoor environment (m^2), V_r : This refers to the total volume of the room or building where radon is potentially accumulating, φ : This represents the rate at which air is exchanged in the room, typically in units of hours (h^{-1}). In this case, it is given as 0.5 h^{-1} . In these calculations, the maximum radon concentration from building materials was assessed by assuming the room to be a cavity with the ratio $S_r/V_r = 2.0 \text{ m}^{-1}$.

3. Results and discussion

Radon concentration, effective radium content, radon surface and mass exhalation rates, annual effective dose due to radon and radon diffusion coefficient values were calculated for Ahlat Taşı and compared with previous studies (Table 1).

Table 1. Radiometric results for Ahlat stone and comparison with previous studies.

Material	C_{Rn} (Bq/m ³)	C_{Ra} (Bq/kg)	E_A (mBq/m ² h)	E_M (mBq/kg h)	D_{etkin} (mSv/y)	F (%)	Referans
Granite	-	49.80	-	81.40	9.71	21.62	[6]
Ceramic	193.71 $\pm$ 45.10	2.59 $\pm$ 0.57	347.42 $\pm$ 79.95	-	4.88 $\pm$ 1.14	-	[2]
Marble	203.8 $\pm$ 7.1	-	154.9 $\pm$ 5.4	27.4	(18.3 $\pm$ 0.6)10 ⁻³	-	[10]
Cement	202 $\pm$ 58	-	379 $\pm$ 51	4.5 $\pm$ 1.29	5.68 $\pm$ 1.63	-	[11]
Sand	108.57	-	70.2	10.8	-	-	[1]
Ahlat Stone	330.51	1.65	322	12.36	8.33	99.8	Present study

Using Equation 7, the contribution of the concentration value originating from Ahlat Stone to indoor radon was calculated as 1.288Bq/m³. The UNSCEAR 2000 report stated that the global average indoor effective dose due to gamma rays emitted from building materials is approximately 0.4 mSv/y [6]. The exhalation rate in the present study sample was calculated as 322 mBq/m²h. The annual effective dose value in case of direct exposure to Ahlat Stone was calculated as 8.33 mSv/y. This is within the action level range recommended by the ICRP 1993 report (3–10) mSv/y. One study reported that radon levels were higher in mud buildings compared to concrete buildings and this could be due to the mud being rich in ²²⁶Ra or ²³⁸U [11]. Another study showed that the highest radon level was in granite (average 506 Bq/m³) [12]. In our study, we obtained the radon concentration value as 330.51Bq/m³ (Table 1). This value is again above the global value (200-300Bq/m³) specified for residential interiors in the ICRP report.

The concentration of radon and its decay products varies depending on a variety of factors, including temperature (Colder temperatures (especially in winter) generally lead to higher indoor radon levels), pressure (Lower indoor pressure compared to the surrounding soil allows soil gas infiltration, which allows radon to enter the home), building materials (Some materials, such as granite, concrete, brick, and even plaster, can emit radon because they contain trace amounts of uranium or thorium), ventilation conditions (Increased air exchange rates can significantly reduce indoor radon concentrations), etc. Assessing radiological hazards in any environment, especially in environments containing human habitation or agriculture, requires careful examination of radium concentration and radon exhalation rate. In a study on building materials, the highest values of surface and mass exhalation rates were reported to be 506.76 mBq/m²h and 15.24 mBq/kg h, respectively [13]. In the present study, the effective radium content was calculated as 1.65 Bq/kg. Surface and mass exhalation rates were obtained as 322 mBq/m²h and 12.36 mBq/kg h, respectively. The histogram comparison between C_{Rn} and E_A is shown in Figure 3. Both C_{Rn} and E_A values are proportional to the track intensity in the LR-115 detector.

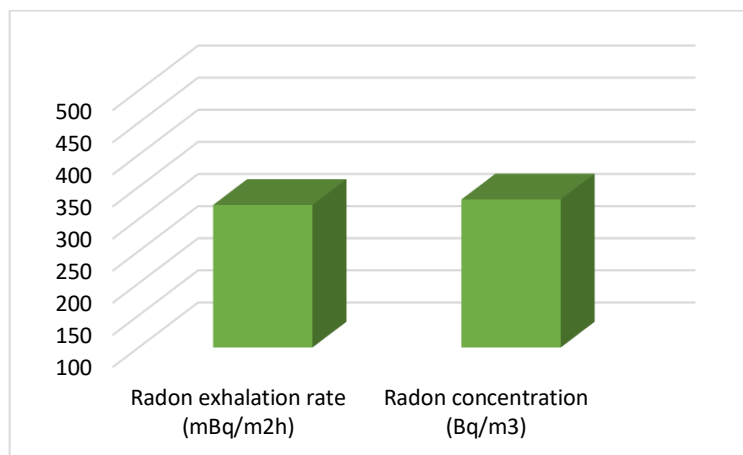


Fig. 4. Comparison of the Radon Concentration (Bq/m³) and Exhalation Rate (mBq/m²h) Histogram of Ahlat Stone.

Since the half-life of radium is 1600 years and the half-life of radon is 3.82 days, radon has enough time to pass through matter and enter the atmosphere. ²²²Rn is transported into the pores of the material and then released into the indoor atmosphere through cracks in the wall. The diffusion fraction of radon determines the diffusion coefficient of that material. In our study, we found the diffusion coefficient for Ahlat Stone to be 99.8%. This may be because Ahlat stone is easy to process when first extracted and the stone has the property of hardening over time. In a study conducted on building materials, it was obtained that the diffusion coefficient values in cement samples varied between 7.13% and 18.10% [6].

4. Conclusion

The radon concentration of Ahlat Stone, effective radium content, surface and mass exhalation rates, diffusion coefficient and the annual effective dose equivalent, which is the radiological effect depending on them, were calculated using the closed box technique and LR-115 film detectors. The radon concentration value is above the global average. D_{eff} is within the action level range determined by the ICRP. However, the contribution of radon emitted from Ahlat Stone to the interior space is high. Ahlat Stone is used in many structures such as houses, mosques, tombs. Therefore, it is recommended that indoor radon reduction measures (such as frequent ventilation of houses, repair of wall cracks) be applied, especially if Ahlat Stone is to be used in housing construction. Calculating E_A and E_M rates in other building materials used in the region will increase awareness about which materials can be used safely and which ones should be restricted.

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There is no conflict of interest.

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