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ASSESSMENT OF NATURAL RADIOACTIVITY AND ITS SIGNIFICANT HAZARDS OF DIFFERENT KINDS OF MARBLES IN ELAZIG (TURKEY)

ABSTRACT

The environmental radioactivity is very important to Humans and it increases by the building materials. Radon takes a very important part of the environmental radioactivity. Since the widespread use of marble as a building/construction or decoration material, the presented work has been aimed to determine the natural radioactivity (^{238}U (^{226}Ra), ^{232}Th and ^{40}K) and radon concentrations of fourteen different marble samples by using a NaI(TI) gamma spectroscopy and track detector (CR-39), respectively. The gamma analysis has been carried out by using gamma spectroscopy including NaI(TI) detector and the radon exhalation rate was calculated by using passive methods of CR-39 passive nuclear track detectors. The radium equivalent activity (Ra_{eq}), the absorbed dose (D), the effective dose rate (EDR), the external (H_{ex}) and internal (H_{in}) hazard index were also calculated and compared to the international standards values.

Keywords: Natural radioactivity, Radon, Marble, Hazard

ELAZIĞ'DAKİ (TÜRKİYE) FARKLI TÜRDEKİ MERMERLERİN DOĞAL RADYOAKTİVİTESİ VE KAYDEDEĞER RİSKİNİN DEĞERLENDİRLİMESİ

ÖZET

Çevresel radyoaktivite insanlar için oldukça önemlidir ve bu bina yapım materyalleri ile artmaktadır. Radon, çevresel radyoaktivitenin çok önemli bir bölümünü teşkil etmektedir. İnşaat/yapı veya dekorasyon malzemesi olarak mermerlerin kullanılmasının artmasından dolayı, bu çalışmada iz detektörü (CR-39) ve NaI(TI)gama spektroskopisi kullanılarak 14 farklı mermer örneğinde doğal radyoaktivite (^{238}U (^{226}Ra), ^{232}Th ve ^{40}K) ve radon konsantrasyonlarının belirlenmesi amaçlanmıştır. Gama analizleri NaI(TI) detektörü içeren gama spektroskopisi kullanılarak yapılmış olup,radon yayılım miktarı ise CR-39 pasif nükleer iz detektörlü pasif metot kullanılarak hesaplanmıştır. Bunların yanı sıra, radyum eşlenik aktivitesi (Ra_{eq}), absorblanmış doz (D), etkin doz oranı (EDR), dış (H_{ex}) ve (H_{in}) iç risk indeksleri de hesaplandı ve uluslararası standart değerler ile karşılaştırıldı.

Anahtar Kelimeler: Doğal radyoaktivite, Radon, Mermer, Risk

1. INTRODUCTION (GİRİŞ)

Because of polsihed surface and availability in variety of attractive colors of marbles are widely used as a building/construction or decoration materials. It is mostly used lining on walls and floors in dwellings [1]. Therefore, persons are more contact than concentrate or other construction materials in their daily life. Natural radioactivity is composed of primordial and cosmogenic radionuclides. Natural occurring primary radionuclides (Nat-U, Nat-Th and ^{40}K) and their progenies (especially ^{222}Rn) play an important role in public health. These radionuclides are the main sources of the radiation derived from soil, rocks and building materials (cement, roof asbestos, sand, granite, marble, etc.) in living area. Monitoring of any releases radioactivity to the environment is important for environmental protection of radiation exposure [2 and 3]. Especially, ^{238}U, ^{232}Th and ^{40}K radionuclides have externally and internally exposure risks due to their gamma radiation and alpha particles inhalation, respectively. The environmental radioactivity depends on geological features (kind of rock and soil) and it varies from place to place [4]. Since the public health risks related to the indoor radiation exposure, many societies such as European Commission (EC) have proposed to accept the exposure dose limits [5]. The indoor radiation (especially radon) concentration levels change with type of the building structures, air infiltration rates, wall materials and the traditional living. Emanation of ^{222}Rn is associated with the presence of radium which is a progeny of the uranium. The inhalation of short-lived products of uranium is a major contributor to the total radiation dose. In this way, the high radon concentration in indoor can be caused of lung cancer [6]. Therefore, the limit have been set on the concentrations of radionuclides in various building material and also, radionuclides concentrations in these materials and indoor radiation levels should be continuously measured.

One of the materials of building construction is different kinds of marbles which are used in building as having a purpose decorative material for interior and exterior surface and its exposure to persons a long time. Due to these materials are soft stones, it can be easily cut and polished compared to granites. Many of kind of marbles is extensively used as decorative materials in building around the world. Marbles is basically metamorphic rock composed essentially of a crystalline aggregate of calcite or dolomite. The calc-silicate marble, also known as calciphyre, isa rock with conspicuous calcium and magnesium silicate minerals. Nevertheless, any calcareous and/or dolomite rock with a polishable surface is called a marble. Metamorphic rocks are formed from igneous and/or sedimentary materials; the primary U, Th and K contents will be redistributed according to the degree of the metamorphic transformation. Therefore marbles may contain a large amount of radioactive materials leached out from porous uranium-rich rocks. The presence of other minerals in marble gives it a variety of attractive colors [1].

In the present work, we have carried out determination of natural radioactivity (^{238}U, ^{232}Th. ^{40}K) concentration of fourteen types of marble samples which extracted from Elazığ district in Turkey. This marbles are exported to many countries (e.g., Italy, France, USA, etc.) around the world. While the determination of primal radionuclides was used gamma spectroscopic technique based on gamma counting with NaI(TI) detector; measurements of radon, effective radium and radon exhalation rate were obtained by using passive nuclear track detectors (CR-39). Nevertheless, the gamma internally and externally dose levels are evaluated with radionuclides concentrations in different marbles samples are also compared to the proposed activity levels by the United Nation Scientific Committee on Effects of Atomic Radiation (UNSCEAR) [6].

2. RESEARCH SIGNIFICANCE (ARAŞTIRMANIN ÖNEMİ)

Especially, the marbles are used decorative purposeful in dwellings, so the active bone marrow and bone surface cells can be affected by radiation emitted from this materials. Therefore, study of radioactive materials contains of used materials in dwellings are more important.

3. EXPERIMENTAL PROCEDURE (DENEYSEL İŞLEM)

3.1. Sampling and Sample Preparation for Gamma Counting (Örnekleme ve Gama Sayımı İçin Örnek Hazırlama)

The studied marbles are extracted from Elazığ district in Turkey. After extraction, they are transported to the different marble factories in the district. In the presented work, fourteen types of marble samples of about 1 kg were collected from different factories located in the district. The samples were dried at 100 ^{0}C for 2-3 h to ensure that the moisture is completely removed. Each sample were sealed in the plastic container (5 cm height x 5 cm dia) which is limited the possible escape of radon and stored for a period of about 1 month before counting to allow the ^{238}U to reach equilibrium with its progeny such as ^{214}Bi (609 keV) and ^{214}Pb (352 keV).

3.2. Experimental Method for Gamma Spectroscopy (Gama Spektroskopisi İçin Deneysel Metot)

Gamma spectrometric system consist of a 2″x2″ NaI(TI) (sodium iodide gamma scintillator which is thallium-activated optically coupled to a photomultiplier tube) well-type detector which was housed in a cylindrical lead shield of about 13.7 cm and 15.5 cm in diameter and length, respectively [7]. The lead shield thickness was about 3.5 cm and this is suitable for limiting the gamma background. The detectors entrance window consists of 0.50 mm thick aluminum. The energy calibration was performed by using ^{60}Co (1 μCi) and ^{226}Ra (10 μCi) point sources. The photopeak efficiency of NaI(TI) detector has been found to be 24% at 186 keV. The determination of ^{238}U activity concentrations in soil sample is based upon the detection of 609.3 keV gamma rays emitted by ^{214}Bi. The counting time for each sample was 12,000 s. The background activities from ^{238}U (^{214}Bi) energies at 609.3 keV were 0.0023 counts/s. The activity concentrations were calculated by using the following equation;

$$A_U(Bqkg^{-1}) = \frac{C}{\varepsilon x P_\gamma x M_S} \tag{1}$$

Where, C is the counting rate of gamma rays (counts per second). ε is the detector efficiency of the specific γ-ray. P_γ is the transition probability of γ- decay and M_S is the mass of the sample (kg) [8].

3.3. Experimental Method for Radon and Radon Exhalation Rate (Radon Yayılım Oranı ve Radon İçin Deneysel Metot)

The radon concentration, radon exhalation rate and radium concentration were carried out by using passive track detectors (CR-39) which is capability to register tracks at the different levels of registration sensitivity. The 0.5 kg samples were ground homogenized and sieved to about 150 meshes by a crushing machine. The samples were dried at 100^{0}C for 48 h to ensure that moisture is completely removed. Weighed samples were placed in polyethylene bottles, 500 cm^3 volumes, each. The bottles were completely sealed for more than one month to allow radioactive equilibrium to be reached. This step was necessary to ensure that the radon gas is confined within the volume and that the progenies will also remain in the sample. Each sample was placed in a plastic bottles chamber of radius 4.5 cm and length 9 cm. Square pieces (2 cm x 2 cm) of solid-state nuclear track detectors (SSNTDs) CR-39 were mounted on the inner side of the lid of cylindrical plastic cans. These cans were sealed and stored for 30 days. Exposed detectors were collected and chemically etched using 6 M NaOH at 60^{o}C for 12h. Etched detectors were washed thoroughly in running water for 5 min and then in distilled water for another 5 min. The track density (track.cm^{-2}) on CR-39 detector samples was counted using the optical microscope at a magnification of 100X.

For the purpose of calculating ^{222}Rn concentration levels in marble sample was determined by measuring the tracks density on the passive detector according to the Equation 2.

$$C_{Rn} = \frac{\rho}{\eta T} \tag{2}$$

Where, ρ is the measured track density of the background corrected tracks on the exposed detectors (track.cm^{-2}), T is the exposure time of the samples and η is the detection efficiency (0.096 tracks cm^{-2} d^{-1}recorded per Bq m^{-3}of radon).

The radon exhalation rate, E, in soil sample was calculated by using Eq. 3.

$$E = \frac{\rho h \lambda}{\eta T_{eff}} \tag{3}$$

The effective radium content is calculated from following equation.

$$C_{Ra} = \frac{\rho V}{\eta M T_{eff}} \tag{4}$$

Where, h is height of sample cup, V is effective volume of the cylindrical container (m^3), λ is the decay constant of ^{222}Rn. T is the total exposure time, T_{eff} is the effective exposure time $[T_{eff} = T + 1/\lambda(e^{-\lambda T} - 1)]$, M is mass of the sample (kg) and η is the detector efficiency.

3.4. Estimation of Dose Rate (Doz Oranının Tahmini)

Distributions of ^{238}U, ^{232}Th and ^{40}K in the environmental samples is not likely uniform. Generally, exposure of radiation spread from these radionuclides has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq.kg^{-1} and this definition is calculated through the Eq. 5 and also used conversion factors in absorbed dose rates (in nGy.h^{-1}) are calculated by using the Eq. 6.

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_K \tag{5}$$

$$D = 0.430C_U + 0.666C_{Th} + 0.042C_K \tag{6}$$

In order to estimate the annual effective dose rates, the conversion factors from absorbed dose in air to the effective dose (0.7 Sv.Gy^{-1}) and indoor occupancy factor (0.8) proposed by UNSEAR were used [6]. The effective dose rate was calculated from Eq. 7.

$$EDR\ (mSv/y) = D(nGy/h)x8760(h/y)x0.8x0.7(Sv/Gy)x10^{-6} \tag{7}$$

Annual external hazard index (H_{ex}) due to emitted gamma-ray of each samples are given by following equation.

$$H_{ex} = C_U/370 + C_{Th}/259 + C_K/4810 \leq 1 \quad (8)$$

and the internal exposure to ^{222}Rn and its radioactive progeny is controlled by the internal hazard index which is given by

$$H_{in} = C_U/185 + C_{Th}/259 + C_K/4810 \leq 1 \quad (9)$$

Where, C_U, C_{Th} and C_K are activity concentrations of U, Th and K in Bq.kg^{-1}, respectively [9].

4. FINDINGS AND DISCUSSION (BULGULAR VE TARTIŞMA)

The measured ^{238}U, ^{232}T, ^{40}K concentrations and calculated radium equivalent activity for fourteen different marble samples are reported in Table 1. It can be seen from Table 1 that high uranium and potassium concentrations in the some of the marble samples have been determined. Since the many of studied marbles have formed by calcite which is generally formed in sedimentary rocks, the samples of marble did not show significant thorium concentrations. It is known that sedimentary rocks have low radioactivity than igneous rocks. Consequently, the obtained thorium concentrations ranged from 5.63±0.60 to 46.43±1.74 Bq.kg^{-1} and average thorium of studied marbles samples are 20.40±1.22 Bq.kg^{-1}. However, average uranium and potassium concentrations in marbles samples are 29.55±1.23 and 1260±36.78 Bq.kg^{-1}, respectively. Generally, potassium concentration in the some type of the sedimentary rocks is very high and also potassium concentration of marble samples was found higher than other building materials [4 and 10]. The highest values of radium equivalent in marble samples are 255.15±4.42 Bq.kg^{-1} which is lower than the recommended maximum value 370 Bq.kg^{-1} [11]. The lowest dose rate was 48.08 nGy.h^{-1} in sample 3, while the highest dose rate was 123.49 nGy.h^{-1} in sample 1 (see Table 1). The mean value of dose rate was 82 nGy.h^{-1} such that this value was higher than the worldwide average value 55 nGy.h^{-1} [12].

Table 1. Natural radioactivity and dose values of marble samples
(Tablo 1. Mermer örneklerinin doğal radyoaktivite ve doz değerleri)

Number of Sample	^{238}U (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})	^{40}K (Bq.kg^{-1})	Ra_{eq} (Bq.kg^{-1})	Dose rate (nGy.h^{-1})	Eff. Dose Rate (mSv.y^{-1})	H_{ex}	H_{in}
1	114.16±2.45	22.32±1.32	1416.54±40.71	255.15±4.42	123.44	0.60	0.68	1.02
2	33.89±1.27	41.23±1.65	435.97±22.53	126.41±3.50	60.34	0.30	0.35	0.43
3	20.39±1.12	9.30±0.73	787.81±31.90	94.35±2.85	48.04	0.23	0.25	0.30
4	21.22±1.20	18.93±1.19	1601.83±45.82	171.63±3.82	89.00	0.43	0.46	0.52
5	13.04±0.90	5.95±0.60	1762.09±49.48	157.22±3.75	83.57	0.41	0.42	0.46
6	12.75±0.82	46.43±1.74	613.74±29.40	126.40±3.50	62.18	0.30	0.34	0.37
7	28.52±1.22	6.50±0.69	1651.87±46.49	165.00±3.80	85.97	0.42	0.44	0.52
8	39.35±1.53	45.58±1.70	481.94±22.33	141.63±3.65	67.51	0.33	0.38	0.49
9	30.16±1.25	6.88±0.73	1600.89±44.43	163.26±3.70	84.78	0.41	0.44	0.53
10	81.52±1.94	6.20±0.65	1442.38±41.52	201.44±4.10	99.76	0.48	0.54	0.76
11	12.33±0.80	5.63±0.60	2381.40±53.75	203.76±4.13	109.07	0.53	0.55	0.58
12	38.50±1.30	41.00±1.55	1163.53±35.43	186.72±3.95	92.72	0.45	0.50	0.60
13	13.31±0.82	12.15±1.05	1670.81±47.62	159.33±3.78	83.98	0.41	0.43	0.46
14	39.20±1.65	17.59±1.17	641.29±29.12	113.73±3.45	55.50	0.27	0.30	0.41

The annual effective dose rates in air ranged from 0.23 to 0.60 mSv.y^{-1} with an average value of 0.39 mSv.y^{-1}. These values do not exceed the average worldwide exposure of 2.4 mSv.y^{-1} due to natural sources [13]. However, the mean specific activities of the three radionuclides under study and the corresponding Ra_{eq} concentrations for marble samples reported from different countries are given for comparison in Table 2 [14]. In order to evaluate the hazard of natural radiation from marbles external (H_{ex}) and internal (H_{in}) index was calculated. The average calculated values of H_{ex} and H_{in} index are found to be 0.43 and 0.53, respectively. Since the high uranium concentration was found in sample 1, the internal index in this sample was found to be 1.02.

Radon concentrations, radon exhalation rate and effective radium concentration in fourteen different marble samples are determined and illustrated in Table 3. The mean measured radon concentration and radon exhalation rate in samples was found to be about 1017 $Bq.m^{-3}$ and 22.46 $Bqm^{-2}d^{-1}$, respectively. The relationship between radon exhalation rates and uranium concentrations for marble samples are shown in Fig.1. The relationship between the uranium and radon exhalation rate is not found to be in a strong correlation. For this reason, porosity and density of this marble samples are very variable. But, there is a positive correlation between them (0.60). The effective radium content varied from 3.02 to 9.56 $Bq.kg^{-1}$. The fluctuations in these values may be attributed to the variation of uranium concentrations in different kind of marbles. The analyses of the results indicate that there is a positive correlation between the effective radium content and uranium concentration in samples.

5. CONCLUSION (SONUÇ)

A very important part of the radioactivity exposion is originating by environment. For this reason, the importances of monitoring of radioactivity levels in the environmental samples and also in building materials are getting very important for the public health. In the presented work, the levels of natural radioactivity in different marbles were determined using gamma-ray spectroscopy and solid state nuclear track detectors. Within the measured marble samples; uranium concentration, effective radium and radon concentration in sample 1 is higher than others. The highest level for thorium was found in sample 6, while highest level for potassium was found in sample 11. According to obtained results, the radioactivity levels of marbles are within recommended values [6]. Since uranium values in samples 1 are high, the calculated external and internal indexes in those samples are higher than unity. Based on the average values of dose rate; effective dose rate, external and internal index of the marble samples are acceptable for use as a building materials and decoration.

Table 2. Comparison of activity concentrations and radium equivalent in marbles in different areas of the world
(Tablo 2. Dünya'nın farklı bölgelerindeki mermerlerde radyum eşleniği ve aktiviye konsantrasyonlarının karşılaştırılması)

Country	Activity ($Bqkg^{-1}$)			Ra_{eq} ($Bqkg^{-1}$)	Reference
	^{238}U	^{232}Th	^{40}K		
Algeria	23±2	18±2	310±2	73±4.1	Ngachin et al. (2007)
Jordan	20.1	11.4	85	42.9	
Kuwait	3.9±0.5	0.22±0.08	3.7±0.5	4.2	
Cameroon	8±2	0.35	19±2	10.15	
Pakistan	33	32	57	62	Iqbal et al.(2000)
Elazig (Turkey)	29.55±1.25	20.40±1.22	1407.78±39.63	161.85±3.65	Present study

Table 3. Radon concentrations, radon exhalation rate and effective radium concentration in different marble samples.
(Tablo 3. Farklı mermer örneklerindeki radon konsantrasyonu, radon yayılım oranı ve etkin radium konsantrasyonu)

Number of Sample	Radon Concentration ($Bq.m^{-3}$)	Radon Exhalation Rate ($Bqm^{-2}d^{-1}$)	Eff. Radium Content ($Bq.kg^{-1}$)
1	1567.70	34.32	9.56
2	1194.15	26.13	7.27
3	949.19	20.77	5.78
4	1154.42	24.93	6.34
5	979.81	21.44	5.96
6	754.45	16.51	4.59
7	918.57	18.89	5.25
8	1102.29	24.12	6.71
9	1077.79	23.59	6.56
10	1113.31	24.36	6.78
11	499.70	10.93	3.02
12	1163.53	25.46	7.08
13	745.88	16.32	4.54
14	1224.76	26.80	7.46
Average	1016.98	22.46	6.20

We hope the measurements of concentration of natural radioisotopes ^{238}U, ^{232}Th and ^{40}K performed in the this study will help to cerate a primary data base on radionuclides content in using building materials and their components available in different region of Turkey. The present study will make it possible to develop the national standard on the radioactivity of building materials.

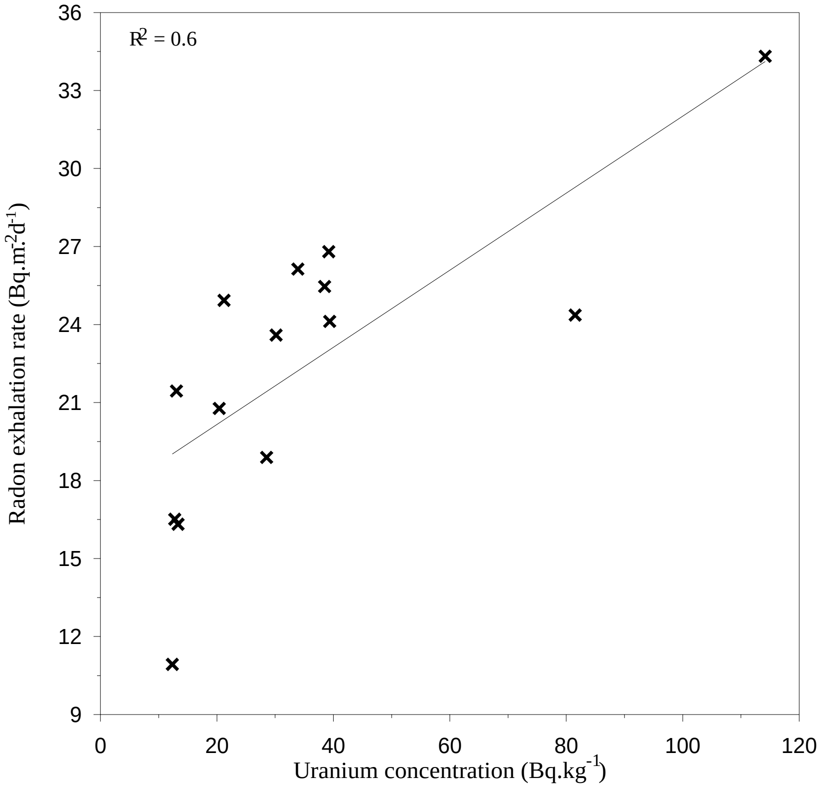

Figure 2. Correlations between radon exhalation rate and uranium concentrations.
(Şekil 2. Uranyum konsantrasyonu ve radon yayılım oranı arasındaki ilişki)

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