NATURAL RADIATION EXPOSURE AND RADON EXHALATION RATE OF BUILDING MATERIALS USED IN TURKEY

by

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Measuring the natural radioactivity levels and radon exhalation rates (surface and mass) in building materials is essential to evaluate the extent of radiation exposure (external and internal) for residents in dwellings. Gamma-ray spectrometry with a high purity germanium detector was used to measure the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in some building materials used in Turkey. Moreover, an active radon gas analyser with an accumulation container was used to measure their radon surface and mass exhalation rates. Results showed that the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K varied from 5.2   0.6 (satin plaster) to 187.0   2.4 (granite) Bqkg^{-1}, 2.6   0.8 (gypsum) to 172.2   7.6 (granite) Bqkg^{-1} and 12.3   17.0 (sand) to 1958.0   83.4 (brick) Bqkg^{-1}, respectively. Radon surface and mass exhalation rates varied from 2.9 (marble) to 2734.6 mBqm^{-2}h^{-1} (granite) and 0.033 (marble) to 53.866 mBqkg^{-1}h^{-1} (granite), respectively. The activity concentration index, indoor absorbed gamma dose rate and corresponding annual effective dose were estimated and compared with the recommended limit values. The results indicated that the building materials sampled presented no significant radiological risk.

Key words: natural radioactivity, radon exhalation rate, AlphaGUARD, annual effective dose, building material

INTRODUCTION

Humans are exposed to ionizing radiation emitted from naturally occurring radionuclides of terrestrial origin such as uranium (^{238}U), thorium (^{232}Th), and potassium (^{40}K) [1]. These radionuclides are present in different quantities in all environments, including the human body [1]. Individuals can be exposed to the emitted ionising radiation in two ways: (1) exposure to gamma rays emitted indoors and outdoors from the radionuclides as whole bodies, known as external irradiation and (2) exposure to alpha and beta rays emitted from the intake of terrestrial radionuclides, especially radon (^{222}Rn) and its short-lived decay products, by inhalation and ingestion, known as internal irradiation.

Building materials used in dwellings, schools and workplaces for structural purposes (brick, cement, concrete, *etc*.) and covering (granite, marble, tuff, stone, *etc*.) contain naturally occurring radionuclides [1]. Therefore, building materials are a secondary source of radon in indoor environments. The ^{222}Rn (3.8 days) is the decay product of ^{226}Ra in the ^{238}U series. It is a natural noble gas and has four short-lived decay products: ^{218}Po (3.05 min), ^{214}Pb (26.8 min), ^{214}Bi (19.9 min), and ^{214}Po (164   s) [1]. Radon and its decay products are the most important sources of human exposure to natural sources of ionising radiation [1]. Epidemiological studies have provided convincing evidence of an association between indoor radon exposure and lung cancer, even at the relatively low radon levels commonly found in residential buildings [2, 3]. Therefore, evaluating the radon concentration and radon exhalation rates (surface and mass) from building materials is important to understand the individual contribution of each material to the total indoor radon exposure. Methods have been developed to measure the radon exhalation rate of building materials such as a passive method using a solid-state nuclear track detector (CR-39 and LR-115), accumulation (E-PERM electret ion chamber) and active methods with radon monitors [4].

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The natural radioactivity levels and radon exhalation rates in building materials have been a matter of concern in recent decades [4-20]. In this study, (1) the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the building materials commonly used in the Western Black Sea region of Turkey were measured using a gamma ray spectrometer equipped with a high purity germanium (HPGe) detector, (2) the ^{222}Rn concentration and ^{222}Rn radon exhalation rates (surface and mass) of these materials were measured by active method using a continuous active radon monitoring system (AlphaGUARD PQ2000 PRO) with an accumulation container, and (3) radiological risks that may arise from the use of building materials were evaluated by estimating their radiological parameters.

MATERIAL AND METHOD

Collection and preparation of samples

A total of 30 commonly used structural and covering building materials were collected from construction sites and markets in the Kastamonu Province, which lies in the West Black Sea region between 41°21'N latitude and 33°46'E longitude. The samples were coded according to the origin and location of the sampling site. The samples were crushed, pulverized and dried in a temperature-controlled furnace at 110 °C for 20 h to remove moisture before being transferred to a 1 L Marinelli container and weighed. Sample containers were hermetically sealed and allowed to stand for at least 4 weeks to ensure short-term equilibrium between ^{226}Ra and its short-lived decay products [21].

Measurement systems

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the samples were measured using a gamma ray spectrometer with a high-resolution coaxial *p*-type horizontal HPGe detector (Canberra GX3018). The resolution of the detector is 1.8 keV for the ^{60}Co gamma ray energy line at 1332.5 keV and has a relative efficiency of 30 %. The detector was shielded to minimize natural background radiation from the environment [21]. A certified multi-nuclide standard distributed in 1.0 gcm^{-3} epoxy (Eckert&Ziegler Isotope Products) in a 1 L Marinelli beaker was used for efficiency calibration of the system in the energy range from 122 to 1836 keV [21]. The counting time for each sample was adjusted to obtain the gamma ray spectrum with good statistics [21]. The activity concentration of ^{226}Ra was measured using the 351.9 keV gamma ray line from ^{214}Pb and the 609.3 keV gamma ray line from ^{214}Bi. The activity concentration of ^{232}Th was measured using the 911.2 keV gamma ray line from ^{228}Ac and the 583.2 keV gamma ray line from ^{208}Tl [21]. The activity concentration of ^{40}K was measured directly by its own gamma ray at 1460.8 keV.

The ^{222}Rn concentration and ^{222}Rn surface and mass exhalation rates from the samples were measured using a radon monitor system (AlphaGUARD PQ2000 PRO). This system is designed for continuously determining the concentrations of ^{222}Rn and its decay products in air, water and soil as well as relevant climatic parameters. It incorporates a pulse-counting ionization chamber with high detection efficiency, wide measuring range, fast response to concentration gradients and permanent, maintenance-free operation [3]. The active volume of the ionization chamber is 0.56 L. The ^{222}Rn concentration measurement range is 2-200.000,000 Bqm^{-3}. To measure the ^{222}Rn concentration and ^{222}Rn surface and mass exhalation rates of the samples, a cubic accumulation container (600 mm × 700 mm × 700 mm) was made from 20 mm of iron. Before measurement, the radon-measuring device was placed in this container and the background ^{222}Rn concentration was measured. Each sample was then placed in this container and measured for at least one month to ensure a balance between ^{222}Rn and its decay products. The background was subtracted from the ^{222}Rn concentration of each sample.

RESULTS AND DISCUSSION

Activity concentration and radon exhalation rate

The activity concentrations and the statistical uncertainty of ^{226}Ra, ^{232}Th, and ^{40}K in the samples are presented in tab. 1. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K ranged from 2.6 0.8 to 172.2 7.6 Bqkg^{-1}, 5.2 0.6 to 187.0 2.4 Bqkg^{-1}, and 123.3 17.0 to 1958.0 83.4 Bqkg^{-1}, respectively. The lowest ^{226}Ra activity concentration was measured in the satin plaster sample, whereas the lowest ^{232}Th and ^{40}K activity concentrations were measured in the sand sample. The highest ^{226}Ra and ^{232}Th activity concentrations were measured in the granite sample, whereas and the highest ^{40}K activity concentration was measured in the pumice brick sample.

The United Nations Scientific Committee on the Effects of Atomic Radiations (UNSCEAR) reported that the average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the Earth's crust was 32, 45, and 412 Bqkg^{-1}, respectively [23]. Figure 1 compares the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the samples with the average of the Earth's crust. This shows that the activity concentration of ^{226}Ra measured in granite, gypsum, pumice brick, sand, clay brick, wall tile, cement, ceramic tile, cellular concrete and lime is greater than that of the average of the Earth's crust. The activity concentration of ^{232}Th measured in granite, pumice brick, wall tile, cement and

Table 1. Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K measured in the samples

Sample (code)	Activity concentration [Bqkg^{-1}]		
	^{226}Ra	^{232}Th	^{40}K
Granite (G)	172.2 7.6	187.0 2.4	1365.0 29.1
Gypsum-1 (GYP-1)	20.8 4.2	17.8 1.8	363.5 46.8
Gypsum-2 (GYP-2)	32.4 5.9	21.2 2.2	467.4 58.4
Pumice brick-1 (PB-1)	91.0 11.9	160.6 6.4	1958.0 83.4
Pumice brick-2 (PB-2)	138.0 9.5	130.0 5.4	1691.0 75.8
Plaster-1 (PL-1)	8.0 2.1	13.9 1.3	315.4 41.0
Plaster-2 (PL-2)	2.9 0.7	12.3 1.2	315.0 42.1
Marble-1 (M-1)	24.0 2.8	6.9 0.5	126.8 14.9
Marble-2 (M-2)	17.6 2.3	6.4 0.5	149.5 16.3
Grout (GRO)	5.7 0.6	14.7 1.2	311.1 33.3
Surface grout-1 (SGRO-1)	21.1 7.6	15.8 0.8	315.5 16.7
Surface grout-2 (SGRO-2)	4.1 0.3	6.6 0.7	172.4 19.6
Surface grout-3 (SGRO-3)	11.2 2.4	7.1 0.7	155.3 18.1
Adhesive mortar-1 (AM-1)	21.4 1.9	7.9 0.6	170.6 15.6
Adhesive mortar-2 (AM-2)	9.4 0.9	9.2 0.6	156.8 16.7
Adhesive mortar-3 (AM-3)	20.4 1.6	10.5 1.5	233.7 17.1
Adhesive mortar-4 (AM-4)	24.8 1.9	23.9 0.8	364.7 17.1
Sand-1 (S-1)	39.7 2.6	5.2 0.6	123.3 17.0
Sand-2 (S-2)	17.3 1.7	15.8 0.7	315.0 15.3
Clay brick (CB)	41.3 3.1	30.0 1.7	597.8 16.7
Wall tile-1 (WT-1)	45.8 3.7	64.3 4.2	906.7 22.2
Wall tile-2 (WT-2)	69.0 2.3	58.0 2.1	658.0 25.1
Cement-1 (CM-1)	86.4 6.2	45.3 2.8	766.1 29.6
Cement-2 (CM-2)	53.3 2.6	21.2 0.7	382.3 20.0
White cement (WCM)	10.9 2.0	9.6 0.5	185.0 17.7
Ceramic tile (CT)	75.8 5.5	41.4 2.3	699.5 21.7
Cellular concrete (CCON)	57.9 2.8	50.9 1.9	1087.0 63.7
Lime (LM)	73.3 4.9	15.4 1.8	402.1 19.6
Satin plaster-1 (SPL-1)	2.6 0.8	13.5 0.6	333.7 41.2
Satin plaster-2 (SPL-2)	10.0 1.7	18.6 0.9	336.2 39.9

lime is greater than that of the average of the Earth's crust. The activity concentration of ^{40}K measured in granite, gypsum, pumice brick, clay brick, wall tile, ceramic tile and cellular concrete is greater than that of the average of the Earth's crust.

The sealed accumulation container was kept until the radioactive equilibrium was reached for 30 days. The ^{222}Rn measurement was carried out after a balance between radon and decay products was averaged every 10 minutes for at least 24 h. The total duration of each measurement varied from 32 to 34 days. The ^{222}Rn activity concentrations measured in the container where ground and walls were covered by granite, marble, wall and floor tile, brick (pumice and clay), and cellular concrete were given in tab. 2. The highest ^{222}Rn concentration is released from the granite sample whereas the lowest ^{222}Rn concentration is released from marble.

The ^{222}Rn exhalation rate is the amount of radon activity concentration emitted per unit surface or mass per unit time. The ^{222}Rn surface exhalation rate (ER_S in Bqm^{-2}h^{-1}) is calculated by the following formula [6]

$$ER_S \quad A_{Rn}\lambda\frac{V}{S} \tag{1}$$

where A_{Rn} is the maximum ^{222}Rn activity concentration in the accumulation container in equilibrium, λ is the decay constant of the ^{222}Rn (7.56 10^{-3} h^{-1}), V [m^3] – the volume of the accumulation container, and S [m^2] – the sample's surface area. The ^{222}Rn mass exhalation rate (ER_M in Bqkg^{-1}h^{-1}) is calculated by the following formula [22]

$$ER_M \quad A_{Rn}\lambda\frac{V}{M} \tag{2}$$

where M [kg] is the mass of the sample. The ^{222}Rn exhalation rates were measured assuming that the accumulation container was leakproof. Therefore, the ^{222}Rn leaking from the container was not taken into consideration.

The ER_S and ER_M values measured for the building materials examined are given in tab. 3. The ER_S and ER_M values measured in granite and marble are compared with the values in the literature in tab. 4.

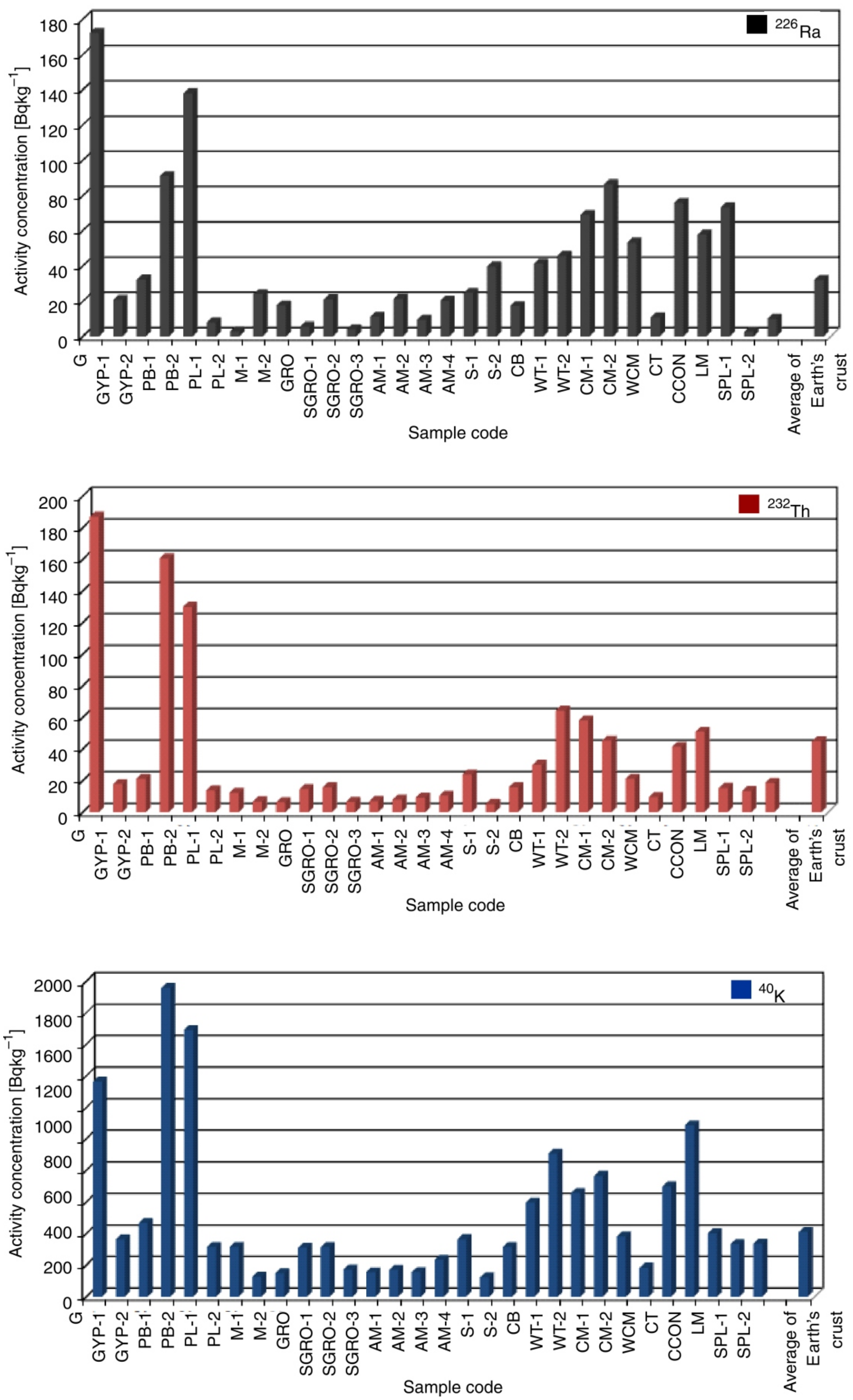

Figure 1. Comparison of the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the samples with the average values of the Earth's crust

Table 2. Activity concentration of ^{222}Rn measured in some building materials

Building materials	Radon activity concentration [Bqm^{-3}]
Granite	1660.0 20.0
Marble	1.0 0.1
Wall tile	6 0.2
Floor tile	3.0 0.1
Clay brick-1	74.0 1.5
Clay brick-2	22.0 0.6
Pumice brick-1	212.0 3.8
Pumice brick-1	10.0 0.3
Cellular concrete	204.0 20.9

Radiological parameter

The activity concentration index ($I\gamma$) proposed by the European Commission (EC) is widely used to evaluate the excess gamma radiation originating from building materials [31]. It is used as a scanning tool for practical monitoring purposes and is estimated as [31]

$$I\gamma \quad \frac{A_{\mathrm{Ra}}}{300} \quad \frac{A_{\mathrm{Th}}}{200} \quad \frac{A_{\mathrm{K}}}{3000} \tag{3}$$

where A_{Ra}, A_{Th}, and A_{K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, in Bqkg^{-1}, respectively. For materials used in bulk amounts such as concrete and bricks, an $I\gamma$ of less than or equal to 1 corresponds to an annual effective dose of 1 mSv, whereas an $I\gamma$ of less than or equal to 0.5 corresponds to an annual effective dose of 0.3 mSv [31]. For superficial and other materials with restricted use such as tiles and boards, an $I\gamma$ of less than or equal to 6 corresponds to an annual effective dose of 1 mSv, whereas an $I\gamma$ of less than or equal to 2 corresponds to an annual effective dose of 0.3 mSv [31]. The $I\gamma$ values estimated for the samples are given in the first columns of tab. 5. The $I\gamma$ values range from 0.1 to 2.0. Only the $I\gamma$ values estimated for pumice bricks are above the recommended upper limit to the dose from building materials to unity.

Table 3. ^{222}Rn exhalation rate from some building materials

Building materials	ER_{S} [mBqm^{-2}h^{-1}]	ER_{M} [mBqkg^{-1}h^{-1}]
Granit	2734.6 32.8	53.866 0.646
Marble	2.9 ± 0.1	0.033 0.001
Wall tile	16.7 0.5	1.140 0.031
Floor tile	7.7 0.2	0.377 0.011
Clay brick-1	79.3 1.5	5.711 0.108
Clay brick-2	22.7 0.7	1.630 0.052
Pumice brick-1	103.0 2.0	13.860 0.278
Pumice brick-2	28.7 0.9	0.872 0.029
Cellular concrete	237.8 0.7	6.891 0.117

The indoor absorbed gamma dose rate (IDR) due to gamma ray emission from ^{226}Ra, ^{232}Th, and ^{40}K in the building material were evaluated as follows [31]

$$IDR \quad DCC_{\mathrm{Ra}} A_{\mathrm{Ra}} \quad DCC_{\mathrm{Th}} A_{\mathrm{Th}} \quad DCC_{\mathrm{K}} A_{\mathrm{K}} \tag{4}$$

where DCC_{Ra}, DCC_{Th}, and DCC_{K} are the dose conversion coefficients estimated for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. In the EC report, DCC were estimated for the center of a standard room (4 × 5 × 2.8 m). The DCC were estimated as 0.92, 1.1, and 0.080 nGyh^{-1} per Bqkg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K, respectively [31].

The corresponding annual effective dose (AED) was estimated using the formula [1]

$$AED \quad IDR\,[\mathrm{nGyh}^{\ 1}] \ 8766[\mathrm{h}] \\ 0.8\,(\text{occupancy factor}) \ 0.7\,[\mathrm{SvGy}^{\ 1}] \ 10^{\ 6} \tag{5}$$

where IDR is given by eq. (4). The indoor occupancy factor implies that 80 % of the time is spent indoors, on average, around the world [1].

The average IDR values estimated for the samples are given in the second column of tab. 5. The IDR values range from 25 to 473 nGyh^{-1}. In the UNSCEAR report, the world average of IDR is given as 84 nGyh^{-1} (range: 40-200 nGyh^{-1}). The IDR values estimated for granite, gypsum, pumice brick, clay brick, wall tile, cement, ceramic tile, cellular concrete and lime are greater

Table 4. Comparison of the ER_{S} and ER_{M} values obtained in this study with those in the literature

Material	Country	E_{RS} [mBqm^{-2}h^{-1}]	E_{RM} [mBqkg^{-1}h^{-1}]	Measurement method	Reference
Granite	Thailand	–	20300	Active (radon monitor)	[24]
	Greece	1240	–	Passive (CR-39)	[25]
	Saudi Arabia	1500	–	Active (radon monitor)	[26]
	Egypt	761-1699	8-19	Passive (LR-115)	[27]
	Serbia	161-576	167-678	Active (radon monitor)	[28]
	Turkey	157	–	Passive (CR-39)	[13]
	Turkey	2735	54	Active (radon monitor)	This study
Marble	Canada	200	–	Active (radon monitor)	[5]
	Serbia	1.4	–	Active (radon monitor)	[29]
	Turkey	130	–	Passive (CR-39)	[13]
	Libya	132	–	Passive (CR-39)	[9]
	Egypt	153-363	2-4	Passive (LR-115)	[27]
	India	26	–	Passive (LR-115)	[30]
	Pakistan	292	–	Passive (CR-39)	[30]
	Turkey	3	0.03	Active (radon monitor)	This study

Table 5. The activity concentration index, indoor absorbed gamma dose rate and corresponding annual effective dose

Sample code	$I\gamma$	*IDR* [nGyh^{-1}]	*AED* [mSv]
G	2.0	473	2.3
GYP-1	0.3	68	0.3
GYP-2	0.4	91	0.4
PB-1	1.8	417	2.0
PB-2	1.7	405	2.0
PL-1	0.2	48	0.2
PL-2	0.2	41	0.2
M-1	0.2	40	0.2
M-2	0.1	35	0.2
GRO	0.2	46	0.2
SGRO-1	0.3	62	0.3
SGRO-2	0.1	25	0.1
SGRO-3	0.1	31	0.1
AM-1	0.2	42	0.2
AM-2	0.1	31	0.2
AM-3	0.2	49	0.2
AM-4	0.3	78	0.4
S-1	0.2	52	0.3
S-2	0.2	58	0.3
CB	0.5	119	0.6
WT-1	0.8	185	0.9
WT-2	0.7	180	0.9
CM-1	0.8	191	0.9
CM-2	0.4	103	0.5
WCM	0.1	35	0.2
CT	0.7	171	0.8
CCON	0.8	196	1.0
LM	0.5	117	0.6
SPL-1	0.2	44	0.2
SPL-2	0.2	57	0.3

than the world average (populated-weighted) indoor absorbed gamma dose rate of 84 nGyh^{-1} [1]. The *AED* values estimated for the samples are given in the last column of tab. 5. The *AED* values ranged from 0.1 to 2.3 mSv.

CONCLUSIONS

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in 9 structural and 21 coating materials commonly used in the Kastamonu Province were measured using gamma ray spectrometry. The ^{222}Rn activity concentration released by some building materials and their ^{222}Rn exhalation rates (surface and mass) were also determined using a continuous active radon-measuring system. For each sample, the activity concentration, indoor absorbed gamma dose rate and corresponding AED were estimated to evaluate the radiological risks. Compared to the average of the Earth's crust, greater activity concentrations were observed for ^{226}Ra, ^{232}Th, and ^{40}K in granite, pumice brick, wall tile, cement and lime, ^{226}Ra and ^{232}Th in gypsum and clay brick, and ^{226}Ra in sand, ceramic tile and cellular concrete. Using the activity concentration index, only the values estimated for pumice bricks were above the upper limit recommended by the EC. The activity concentration results and evaluations showed that the building materials examined presented no significant radiological risk. These findings can be used as guide information for the use and transportation of the building materials sampled.

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AUTHORS' CONTRIBUTIONS

The idea for the research of activity concentration in and radon exhalation rate from building materials was suggested by S. Turhan. Measurements were performed by S. Turhan, A. Kurnaz, and A. T. Temirci and discussion of the results and writing of the article was performed by all authors.

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ИЗЛАГАЊЕ ПРИРОДНИМ РАДИОНУКЛИДИМА И ЈАЧИНА ЕКСХАЛАЦИЈЕ РАДОНА ГРАЂЕВИНСКИХ МАТЕРИЈАЛА КОЈИ СЕ КОРИСТЕ У ТУРСКОЈ

Мерење нивоа природне радиоактивности и површинске и масене јачине ексхалације радона у грађевинском материјалу од кључног је значаја при процени спољашњег и унутрашњег излагања зрачењу становништва у стамбеним објектима. Гама-спектрометријским мерењима на германијуском детектору високе чистоће измерене су концентрације активности ^{226}Ra, ^{232}Th, и ^{40}K у неким грађевинским материјалима који су у употреби у Турској. Додатно, користећи активни анализатор радона са акумулационим резервоаром, измерене су површинска и масена јачина ексхалације радона. Резултати показују да су концентрације активности биле: за ^{226}Ra, у опсегу од 5.2 0.6 Bqkg^{-1} (за сатен гипс) до 187.0 2.4 Bqkg^{-1} (за гранит), за ^{232}Th, од 2.6 0.8 Bqkg^{-1} (за гипс) до 172.2 7.6 Bqkg^{-1} (за гранит) и за ^{40}K, од 12.3 17.0 Bqkg^{-1} (за песак) до 1958.0 83.4 Bqkg^{-1} (за циглу). Површинска и масена јачина ексхалације радона износиле су од 2.9 mBqm^{-2}h^{-1} (за мермер) до 2734.6 mBqm^{-2}h^{-1} (за гранит) и од 0.033 mBqkg^{-1}h^{-1} (за мермер) до 53.866 mBqkg^{-1}h^{-1} (за гранит), респективно. Процењени су индекс концентрације активности, јачина апсорбоване дозе гама зрачења у затвореном простору и одговарајућа годишња ефективна доза и потом упоређене са препорученим граничним вредностима. На основу резултата није уочен значајан радиолошки ризик од узоркованих грађевинских материјала.

Кључне речи: природна радиоактивност, јачина ексхалације радона, годишња ефективна доза, грађевински материјал