Materiales de Construcción
Vol. 70, Issue 340, October–December 2020, e233
ISSN-L: 0465-2746
https://doi.org/10.3989/mc.2020.03820

Radioactivity of building materials in Mahallat, Iran – an area exposed to a high level of natural background radiation – attenuation of external radiation doses

A. Shahrokhi[a], M. Adelikhah[a], S. Chalupnik[b], E. Kocsis[a], E. Toth-Bodrogi[a], T. Kovács[a]✉

a. Institute of Radiochemistry and Radioecology, University of Pannonia, (Veszprém, Hungary)
b. Silesian Centre for Environmental Radioactivity, Central Mining Institute, (Katowice, Poland)
✉ kt@almos.uni-pannon.hu

Received 31 March 2020
Accepted 7 July 2020
Available on line 15 October 2020

ABSTRACT: In this study, mass activity of naturally occurring radioactive materials were measured in twenty-three building material samples, use extensively in the area exposed to a high level of natural background radiation (Mahallat, Iran), to determine the radioactivity index and changes to the level of indoor gamma radiation. The mass activity of ^{232}Th, ^{226}Ra and ^{40}K were within the ranges from 18 ± 3 to 44 ± 10 Bq/kg (average of 27 ± 6 Bq/kg), 22 ± 5 to 53 ± 14 Bq/kg (average of 34 ± 6 Bq/kg) and 82 ± 18 to 428 ± 79 Bq/kg (average of 276 ± 58 Bq/kg), respectively. The gamma dose rates for population were estimated between 48 ± 9 and 111 ± 26 nGy/h with exception of radon exhalation from building materials. Since the air kerma rate in the town varies from 0.8 to 4 µGy/h, the attenuation coefficient was calculated for buildings made of the aforementioned materials. Additionally, the annual gamma radiation doses for inhabitants were calculated based on time spent outdoors and indoors.

KEYWORDS: Building Material; HNBRA; Radiation assessment; Dose rate; Shielding effect.

Citation/Citar como: Shahrokhi, A.; Adelikhah, M.; Chalupnik, S.; Kocsis, E.; Toth-Bodrogi, E.; Kovács, T. (2020) Radioactivity of building materials in Mahallat, Iran – an area exposed to a high level of natural background radiation – attenuation of external radiation doses. Mater. Construcc. 70 [340], e233 https://doi.org/10.3989/mc.2020.03820

RESUMEN: *Radiactividad de materiales de construcción en Mahallat, Irán - una zona expuesta a niveles elevados de radiación de fondo natural - atenuación de dosis de radiación externa.* En este trabajo se ha medido la actividad de materiales NORM en veintitrés muestras de materiales de construcción que se emplean ampliamente en una zona expuesta a niveles elevados de radiación de fondo natural (Mahallat, Irán). El objetivo ha sido determinar el índice de radiactividad y cambios en el nivel de radiación gamma en el interior. Las concentraciones medidas de ^{232}Th, ^{226}Ra, ^{40}K fueron respectivamente (Bq/kg) de 18 ± 3–44 ± 10 (27 ± 6), 22 ± 5–53 ± 14 (34 ± 6), 82 ± 18–428 ± 79 (276 ± 58). Se han estimado unas dosis gamma a la población entre 48 ± 9–111 ± 26 nGy/h a excepción de la exhalación de radón de los materiales de construcción. Dado que la tasa kerma en aire en la ciudad varía entre 0.8–4 µGy/h, se calculó el coeficiente de atenuación para los edificios en los materiales mencionados. Además, las dosis de radiación gamma anual para los habitantes se calcularon teniendo en cuenta el tiempo en interiores y exteriores.

PALABRAS CLAVE: Material de construcción; HNBRA; Evaluación de la radiación; Tasa de dosis; Efecto pantalla.

ORCID ID: A. Shahrokhi (https://orcid.org/0000-0003-1272-4869); M. Adelikhah (https://orcid.org/0000-0003-3339-9524); S. Chalupnik (https://orcid.org/0000-0002-2792-4954); E. Kocsis (https://orcid.org/0000-0002-8532-4009); E. Toth-Bodrogi (https://orcid.org/0000-0002-4135-7925); T. Kovács (https://orcid.org/0000-0003-2606-0397)

1. INTRODUCTION

Humans are exposed to background radiation from both natural and artificial sources on a daily basis, however, more than 80% of our entire exposure to ionizing radiation is from natural sources (1). However, the main contributor of human exposure is radon and its decay products, building materials can be also a source of external exposure to ionizing radiation (1, 2). Clay, sands and other raw materials in addition to recycled by-products, used in the production of the majority of building materials, contain trace amounts of naturally occurring radioactive materials, e.g. radionuclides from the uranium (^{238}U) and thorium (^{232}Th) radioactive decay series as well as potassium (^{40}K), another primordial radionuclide (3, 4). The worldwide average mass activity of ^{226}Ra, ^{232} Th and ^{40} K in the soil are 35, 30 and 400 Bq/kg, respectively; but these values can be several times higher in High Natural Background Radiation Areas (HNBRAs), e.g. Ramsar, Mahallat, Yangjiang, Kerala, etc., and people are receiving radiation doses much greater than the worldwide average background dose (1). In accordance with the findings of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the annual effective doses received by inhabitants in HNBRAs are classified as: low (<5 mSv/yr), medium (5–10 mSv/yr), high (20–50 mSv/yr) and very high (>50 mSv/yr) (1, 5). This classification is based on the recommended dose level prescribed by the International Commission on Radiological Protection (ICRP) and the worldwide average dose level for the general public (2.4 mSv/yr) (1, 2, 5–8).

Two well-known HNBRAs in Iran, Ramsar a famous area in radiation point of view, a northern coastal city in Mazandaran province and Mahallat located in central of Iran. In Mahallat, the igneous bedrocks are rich in uranium, which decays into ^{226}Ra as a soluble in groundwater but in the presence of dissolved calcium carbonate ($CaCO_3$) precipitates out as radium carbonate ($RaCO_3$) houses (8). Thus, $RaCO_3$ may ultimately occur as a component of building materials, utilized to construct local. According to the 1, the absorbed dose rate in this area was estimated to fall within the range of 800 to 4000 nGy/h (including cosmic and terrestrial radiation), which is at least fifteen times higher than the worldwide average of 55 nGy/h (1, 9). Therefore, it should be obligatory to monitor building materials as a secondary potential source of emitting indoor gamma radiation, however, the introduction of additional sources of enhanced radioactivity into such an environment would not be preferable. Moreover, according to the new European Union Basic Safety Standards (Council Directive 2013/59/EURATOM), the radioactivity index of building materials was restricted to less than 1 (to correspond to a reference level of external gamma dose -1 mSv/yr - before being released to the market) (1, 2, 10, 11).

Over recent years, many studies have been carried out in High Natural Background Radiation Areas to assess the radiation (5, 8, 12–21). Our study focused on the assessment of gamma-ray emitters in building materials and the estimated exposure to indoor gamma radiation from these materials in the HNBRA of Mahallat, Iran. Attempts were made to evaluate the distribution of naturally occurring radionuclides in different materials, therefore, measurements of the radioactivity of twenty-three of the most common building materials available on the local market were taken. As an objective of the present study and with respect to radiation protection, the radium equivalent activity index, resultant absorbed dose of gamma radiation and annual effective dose of each sample of building material were estimated. An attempt was not made to estimate the excess lifetime cancer risk from indoor gamma radiation based on the life expectancy of the Iranian population because the use of building materials that emit relatively low levels of radioactivity (compared with outdoor values) would be meaningless. On the other hand, the radioactivity index of samples, according to the European Union Basic Safety Standards (EU BSS), was calculated in order to screen the products on the market and avoid the introduction of materials with enhanced radioactivity to the HNBRA. The annual effective dose in relation to time spent outdoors and indoors was estimated to show the shielding effect of building materials from gamma radiation. Additional investigations of the radon levels in dwellings should be undertaken to provide a comprehensive estimation.

2. MATERIALS AND METHODS

2.1. Area of Interest

Mahallat, with a population of approximately 53 thousand and surface area of 37 km^2, is one of the oldest towns in Iran. It is located in Markazi Province in north-western Iran (N 32° 54′, E 50° 27′) and is surrounded by mountains. Mahallat is recognised as a HNBRA (1). It contains several sources of spring water with high levels of radioactivity known as Abegarm-e-Mahallat, a region to the north-east of Mahallat, but the exposure detailed in this study is derived from other sources (8, 13).

2.2. Sampling and Sample Preparation

For the investigations, twenty-three different samples of commonly use building materials in Iran, e.g. gypsum, cement, bricks, sand and gravel, ceramics and tiles, were randomly collected directly from local building material suppliers and construction sites in the year 2015. For every building material investigated, between three and five subsamples were collected, stored and labelled at the time of collection. The samples, after being transferred to the laboratory, were stored at room temperature for several days in order to dry and then pulverised, homogenised and sieved (grain size <3 mm) to achieve the same parameters as the reference material (IAEA-375, Soil standard). Afterwards, the samples were dried in a ventilated oven at 105 °C for 24 hours to reach a constant weight. 500 grams of each prepared and homogenized sample was transferred into a leak-proof Marinelli beaker, weighed and sealed for 30 days in order to allow ^{226}Ra to reach secular equilibrium with its daughters.

2.3. Gamma spectrometry analysis

The concentrations of three naturally occurring radionuclides (^{40}K, ^{226}Ra and ^{232}Th) in the samples of building materials were determined using a High Purity Germanium (HPGe) gamma-ray detector (ORTEC GMX40-76) with a relative efficiency of 40%, and an energy resolution of 1.9 keV at 1332.5 keV. The spectra were recorded by an ORTEC DSPEC LF 8196 MCA (multichannel analyzer) and analysed using Aptec MCA software. The energy calibration was carried out by three sealed sources: ^{137}Cs with a gamma line of 661.6 keV, ^{60}Co with two gamma lines of 1173.2 and 1332.5 keV and ^{241}Am with an X-ray line of 59.5 keV (22).

The mass activity of each radionuclide was determined using its own specific gamma lines or gamma lines of its decay products: for ^{40}K, the line 1461 keV (11%); in the case of ^{226}Ra, the gamma lines of its decay products were used – the lines of ^{214}Pb and ^{214}Bi with energies of 352 keV (35%) and 609 keV (45%), while to measure the decay products of ^{232}Th, that are, ^{228}Ac and ^{208}Tl, gamma lines of 911 keV (28%) and 2614.5 keV (36%) were applied, respectively (22). The concentration of radionuclides in the samples, uncertainty and Minimum Detectable Activity were determined using calculations explained in our recently published papers (23, 24).

The detection efficiencies for particular gamma lines and the detection limits (LID) were determined using a reference material provided by the International Atomic Energy Agency (soil standard IAEA-327) for the system applied in our gamma spectrometric investigations. The detection efficiency was estimated separately for ^{40}K, ^{226}Ra and ^{232}Th as 1.2%, 2.4% and 1.4%, respectively. Meanwhile, the LIDs were calculated as 23.0, 0.5 and 0.7 Bq/kg, respectively.

2.4. Radiation Assessment

The radium equivalent activity from natural radionuclides was estimated for the assessment of radiological hazards of radioactivity in environmental materials (or building materials) for the population health. The radium equivalent was determined according to Equation [1] (25):

$$Ra\left(Eq\right) = a^{226}_{(Ra)} + \left(a^{232}_{(Th)} \times 1.43\right) + \left(a^{40}_{(K)} \times 0.077\right) \quad [1]$$

Where *a* denotes the mas activity of a specific radionuclide (Bq/kg).

The absorbed dose rate, caused by naturally occurring radionuclides in building materials, the radioactivity index and the annual effective dose were calculated using Equations [2, 3, 4], respectively (2, 26–28):

$$D = \left(0.92 \times a^{226}_{(Ra)}\right) + \left(1.1 \times a^{232}_{(Th)}\right) + \left(0.080 \times a^{40}_{(K)}\right) \quad [2]$$

$$I = \left(a^{226}_{(Ra)} / 300\right) + \left(a^{232}_{(Th)} / 200\right) + \left(a^{40}_{(K)} / 3000\right) \quad [3]$$

$$ED_{(indoor)} = D \times T \times 0.8 \times 0.7 \times 10^{-3} \quad [4]$$

where *I* denotes the radioactivity index, *D* represents the gamma dose rate from naturally occurring radionuclides (nGy/h), *ED* stands for the annual effective dose (μSv/yr), 0.7 is the conversion factor for (Sv/Gy), 0.8 is the indoor occupancy factor, and *T* is the number of hours in a year (8760 h/yr).

Additionally, the attenuation coefficient of the outdoor gamma dose rate was estimated – which given the wall thickness of approximately 30–40 cm and the average density of roughly 2 g/cm^3 for the building materials – to be about 500 for gamma rays of 500 keV in energy. As a result, the increased indoor gamma dose rate, due to the high outdoor gamma dose rate, should not exceed 8–10 nGy/h. Therefore, the following estimation was made – the changes in annual effective dose were calculated by taking into consideration different values of indoor occupancy time. As predicted, the highest annual effective doses were estimated for a scenario in which the person spends more time outdoors than indoors. However, this simulation was executed without taking into account possible differences in outdoor and indoor radon levels.

3. RESULTS AND DISCUSSION

Table 1 shows the concentrations of naturally occurring radionuclides recorded in the dry samples of building materials. The average mass activities of ^{226}Ra, ^{232}Th and ^{40}K in the samples measured were

TABLE 1. Measured mass activities and statistical analysis of naturally occurring radionuclides in building materials (Bq/kg).

Building Material	Ra-226	Th-232	K-40	I*
Sand and Gravel	38 ± 8	44 ± 10	204 ± 46	0.4
Sand and Gravel	29 ± 4	25 ± 4	238 ± 38	0.3
Sand and Gravel	33 ± 6	34 ± 6	337 ± 60	0.4
Sand and Gravel	28 ± 5	27 ± 5	378 ± 71	0.4
Mean	**32 ± 6**	**32 ± 6**	**289 ± 54**	**0.4**
Bricks	35 ± 5	23 ± 4	410 ± 78	0.4
Bricks	38 ± 7	32 ± 6	357 ± 66	0.4
Bricks	31 ± 8	28 ± 5	428 ± 79	0.4
Bricks	34 ± 6	27 ± 5	413 ± 80	0.4
Mean	**35 ± 7**	**28 ± 5**	**402 ± 76**	**0.4**
Gypsum	29 ± 4	20 ± 4	82 ± 17	0.2
Gypsum	22 ± 5	18 ± 3	94 ± 16	0.2
Gypsum	25 ± 4	23 ± 5	85 ± 19	0.2
Gypsum	31 ± 4	24 ± 5	107 ± 22	0.3
Mean	**27 ± 4**	**21 ± 4**	**92 ± 19**	**0.2**
Cement	31 ± 4	23 ± 5	224 ± 48	0.3
Cement	42 ± 8	28 ± 6	239 ± 52	0.4
Cement	34 ± 5	23 ± 5	257 ± 56	0.3
Cement	27 ± 4	25 ± 5	198 ± 40	0.3
Mean	**34 ± 5**	**25 ± 5**	**229 ± 49**	**0.3**
Ceramic	35 ± 5	23 ± 5	308 ± 67	0.3
Ceramic	33 ± 5	28 ± 8	258 ± 73	0.3
Ceramic	33 ± 4	33 ± 7	381 ± 81	0.4
Mean	**34 ± 5**	**28 ± 7**	**316 ± 74**	**0.4**
Tile	43 ± 10	29 ± 6	314 ± 71	0.4
Tile	53 ± 14	31 ± 8	292 ± 75	0.4
Tile	41 ± 9	30 ± 8	348 ± 92	0.4
Tile	46 ± 10	33 ± 9	401 ± 82	0.5
Mean	**46 ± 11**	**31 ± 8**	**339 ± 80**	**0.4**
Minimum	**22 ± 5**	**18 ± 3**	**82 ± 18**	**0.3**
Maximum	**53 ± 14**	**44 ± 10**	**428 ± 79**	**0.2**
Average (Bq/kg)	**34 ± 6**	**27 ± 6**	**276 ± 58**	**0.5**
Median (Bq/kg)	**33**	**27**	**292**	**-**
Standard Deviation (Bq/kg)	**7.2**	**5.7**	**112**	**-**
Skewness	**0.8**	**1.0**	**-0.5**	**-**
Kurtosis	**0.9**	**2.2**	**-0.9**	**-**
Geometric Mean (Bq/kg)	**34**	**27**	**250**	**-**

***I**: the radiation index of the material

34 ± 6, 27 ± 6 and 276 ± 58 Bq/kg, respectively. It can be seen that concentrations measured were, in most cases, below worldwide average values (1). The excel program was used for data analysing using built-in commands. Due to the statistical analysis, it was found that the concentrations of ^{40}K and ^{226}Ra were higher than the worldwide average value in 17% and about 26% of the samples, respectively. The highest mass activity of ^{226}Ra, ^{232}Th and ^{40}K were found in various building materials (tiles, sand, gravel and bricks), but did not significantly exceed the worldwide average values (Figures 1 and 2). The activity indexes for all of the samples were calculated and found to be less than 1.0, the value recommended by EU BSS, likewise the statistical analysis of the obtained data with regard to the mass activity of ^{40}K, ^{226}Ra and ^{232}Th in the samples of building materials is presented in as shown in Table 1.

The normal distribution for the specified mean and standard deviation of the measured mass activity of radionuclides in the various building materials is also shown in Figures 1 and 2. The excel Polynomial, 3rd Order, were used to draw the trendlines between the data.

According to Figures 1 and 2, in most samples, the mass activity of ^{40}K fell within the range of ~200 to ~380 Bq/kg, while the corresponding values for ^{226}Ra and ^{232}Th were from ~30 to ~40 and ~25 to ~30 Bq/kg, respectively.

The radium equivalent activities, gamma-ray absorbed dose rates and annual effective doses were estimated for the analysed samples based on the mass activity of the naturally occurring radionuclides. The results of the calculations can be seen in Table 2.

Table 2 shows the results of radium equivalent ($Ra_{(eq)}$), gamma-ray absorbed dose (D) and annual effective dose (ED) for studied building materials. The calculated values of radium equivalent activities were between 55 and 125 Bq/kg with a cumulative average of 95 Bq/kg. Meanwhile, the gamma-ray absorbed dose rates were measured to be between 48 and 111 nGy/h with a cumulative average of 84 nGy/h, which is higher than the worldwide average value of 55 nGy/h (1). Based on the values of estimated gamma-ray absorbed dose rates, the annual effective doses were calculated to be within the range of 234 to 546 μSv/yr with a cumulative average of 412 μSv/yr.

We correlated the values of ^{226}Ra, ^{232}Th and ^{40}K for all samples, and also separately. The correlation study between ^{226}Ra, ^{232}Th and ^{40}K mass activity concentrations of the studied building materials are shown in Figure 3 and Figure 4. Since ^{226}Ra series and ^{232}Th series are usually found together in nature, and good correlation between them is indicative of common sources, which, in general, are associated with a mineralogical component; in the other words, mass ratio between Th and U is dependent on the type of the material but is correlated with activity ratio. Having activity ratio a_{Th}:a_U it can multiply it by a ratio of half-lives of Th (14 billion years) and U (4.5 billion years). That ratio is a constant value roughly 14/4.5 = 3.1(UNSCEAR reports show the average ^{238}U concentration in soil as 35 Bq/kg and similar value for ^{232}Th, therefore, it would give us

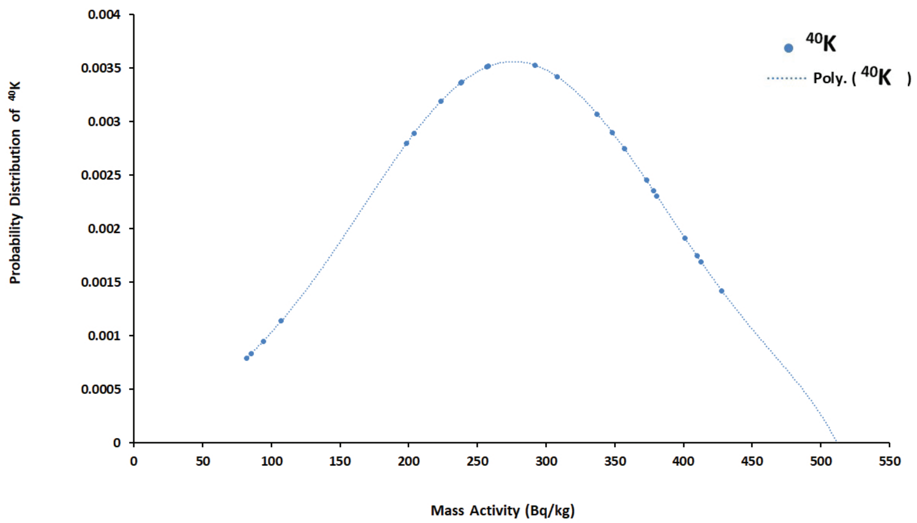

FIGURE 1. Distribution of the ^{40}K mass activity in the various building materials.

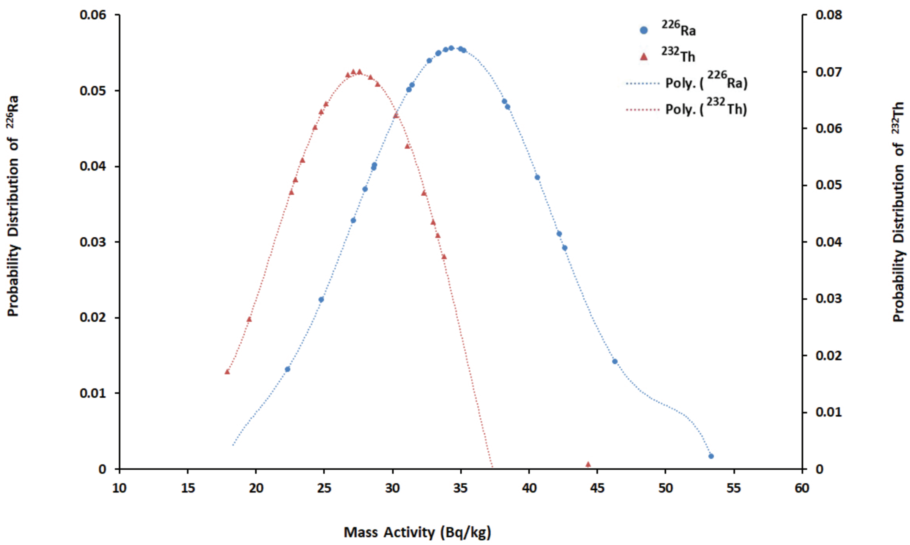

FIGURE 2. Distribution of the ^{226}Ra and ^{232}Th mass activity in the various building materials.

mass ratio of about 3) (1); However, to estimate the dose from the construction materials, activity concentration is needed instead of the mass concentrations or mass ratio.

In most of the studied building materials, there is a positive correlation between ^{226}Ra and ^{232}Th. For instance, as it is revealed that from figures 3 and 4, the strongest linear relationship is observed between ^{226}Ra and ^{232}Th in sand and gravel, and ceramic with $R^2 = 0.98$ and $R^2 = 0.56$, respectively; However, the corresponding correlation coefficient is slightly lower for other building materials. Brick and tile have shown the most positive correlation between ^{232}Th and ^{40}K mass activity concentrations with $R^2 = 0.55$ and $R^2 = 0.48$ than other building materials. The weakest relationships can be also found between ^{232}Th and ^{40}K mass activity concentrations in cement. Moreover, the strongest correlation between ^{226}Ra and ^{40}K with $R^2 = 0.86$ was found in bricks. The weakest corresponding relationships can be also found in ceramic.

To estimate the correlations between the mass activity of ^{40}K, ^{232}Th and ^{226}Ra among all sample as a single component of the building, scatter plots of the mass activity of ^{232}Th vs. ^{226}Ra, ^{40}K vs. ^{226}Ra and ^{40}K vs. ^{232}Th were drawn and are shown in Figures 5 – 7. No strong correlations are visible from the scatter plots shown in Figures 5, 6 and 7.

Table 2. Evaluation of Radium equivalent ($Ra_{(eq)}$), Gamma-ray absorbed dose (D) and Annual effective dose (ED) for studied building materials.

Building Material	**$Ra_{(eq)}$ (Bk/kg)**	**±**	**D (nGy/hr)**	**±**	**ED (μsv/yr)**	**±**
Sand and Gravel	117.4	25.8	100.3	22.0	492	108
Sand and Gravel	82.9	12.6	73.1	11.1	358	55
Sand and Gravel	107.7	19.2	94.9	16.9	466	83
Sand and Gravel	95.3	17.6	85.4	15.8	419	77
Mean	**100.8**	**18.8**	**88.4**	**16.5**	**434**	**81**
Bricks	100.2	16.7	90.9	15.2	446	75
Bricks	111.9	20.7	99.2	18.3	487	90
Bricks	103.6	21.2	93.3	19.2	458	94
Bricks	104.9	19.3	94.4	17.4	463	85
Mean	**105.1**	**19.5**	**94.5**	**17.5**	**463**	**86**
Gypsum	62.8	11.1	54.3	9.5	266	47
Gypsum	55.2	10.5	47.8	9.2	234	45
Gypsum	63.7	12.6	54.5	10.7	267	52
Gypsum	74.2	12.9	64	10.9	314	54
Mean	**64.0**	**11.8**	**55.1**	**10.1**	**271**	**49**
Cement	82.1	14.8	72.5	13.0	356	64
Cement	100.0	20.6	88.3	18.1	433	89
Cement	86.4	16.5	76.9	14.6	377	72
Cement	77.8	14.2	68.1	12.4	334	61
Mean	**86.6**	**16.5**	**76.4**	**14.5**	**375**	**71**
Ceramic	91.5	17.3	82	15.5	402	76
Ceramic	93.2	22.0	82	19.2	402	94
Ceramic	109.8	20.2	97.4	17.8	478	88
Mean	**98.1**	**19.9**	**87.1**	**17.5**	**427**	**86**
Tile	108.1	24.0	96.1	21.5	471	105
Tile	120.3	31.2	106.6	27.7	523	136
Tile	110.6	27.5	98.4	24.5	483	120
Tile	124.8	29.0	111.3	25.5	546	125
Mean	**115.9**	**28.0**	**103.1**	**24.8**	**506**	**122**
Statistical Values of Samples						
Mean	**95.0**	**18.3**	**84**	**16.8**	**412**	**82**
Minimum	**55.2**	**10.5**	**47.8**	**9.2**	**234**	**45**
Maximum	**124.8**	**29**	**111.3**	**25.5**	**546**	**125**

On the other hand, it can be seen that the general trend of these plots is that the mass activity of each particular radionuclide increases as the mass activity of other radionuclides increases.

Since the relative distribution of ^{226}Ra and ^{232}Th is positively correlated with all of the calculated radiological parameters, this result may be due to the rich content of ^{226}Ra and ^{232}Th, which plays an important role in determining the hazardous nature in the building materials; poor correlation was observed between ^{226}Ra and ^{40}K.

The concentrations of radionuclides measured within this study were compared with those from other studies worldwide in Table 3. According to

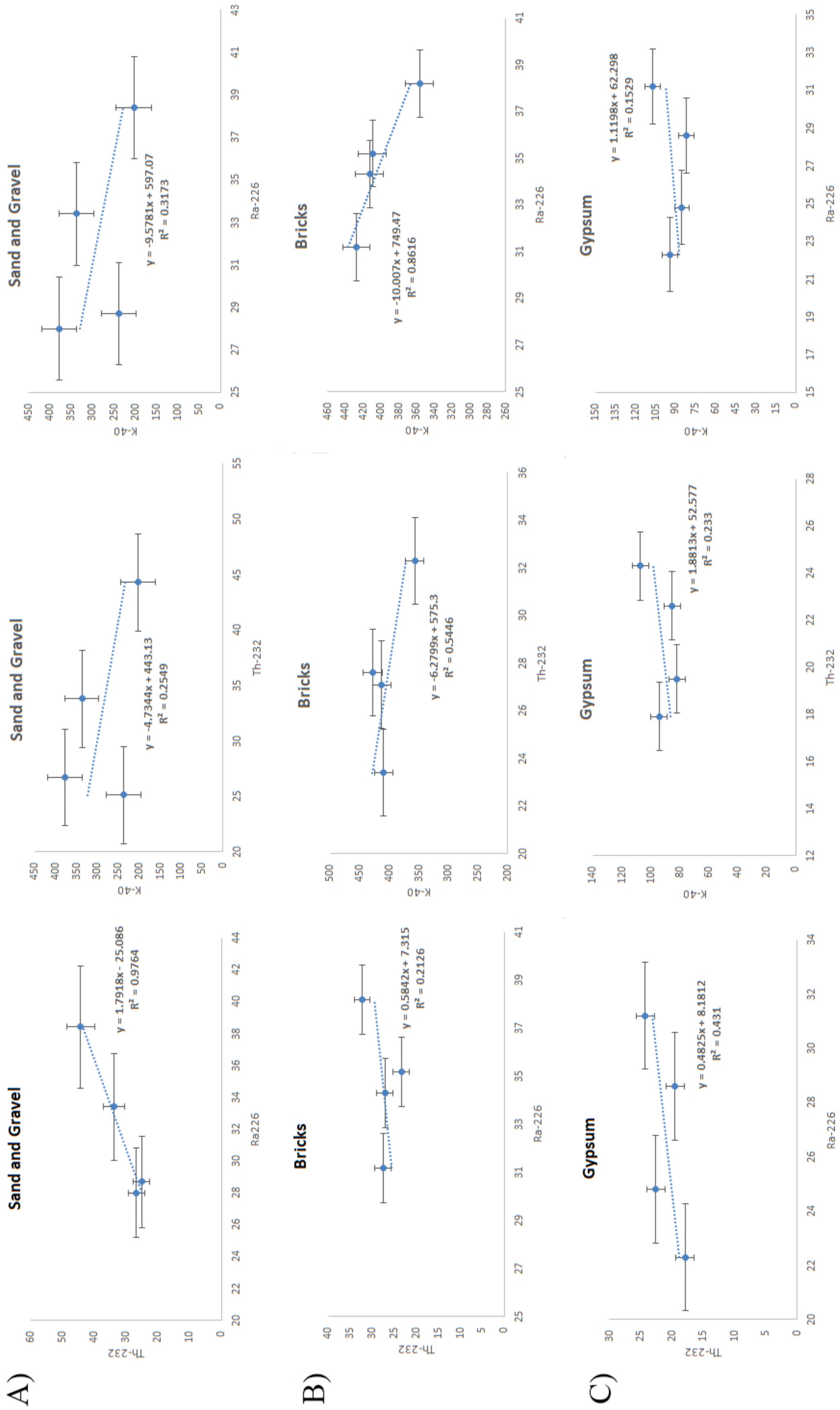

FIGURE 3. Scatter plots of ^{232}Th vs. ^{226}Ra; ^{40}K vs. ^{226}Ra and ^{40}K vs. ^{232}Th for A) Sand and Gravel; B) Brick and C) Gypsum.

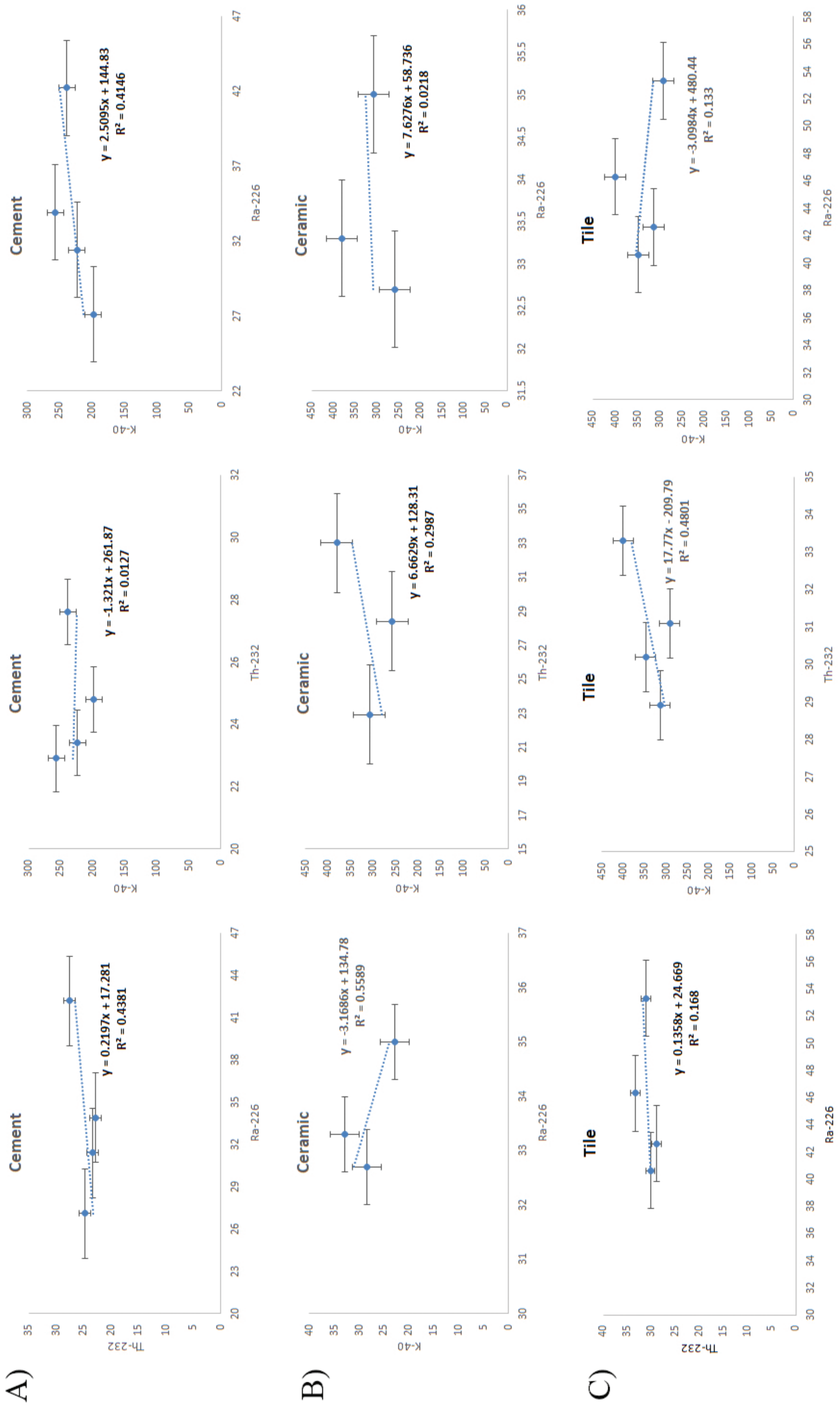

FIGURE 4. Scatter plots of ^{232}Th vs. ^{226}Ra; ^{40}K vs. ^{226}Ra and ^{40}K vs. ^{232}Th for A) Cement; B) Ceramic and C) Tile.

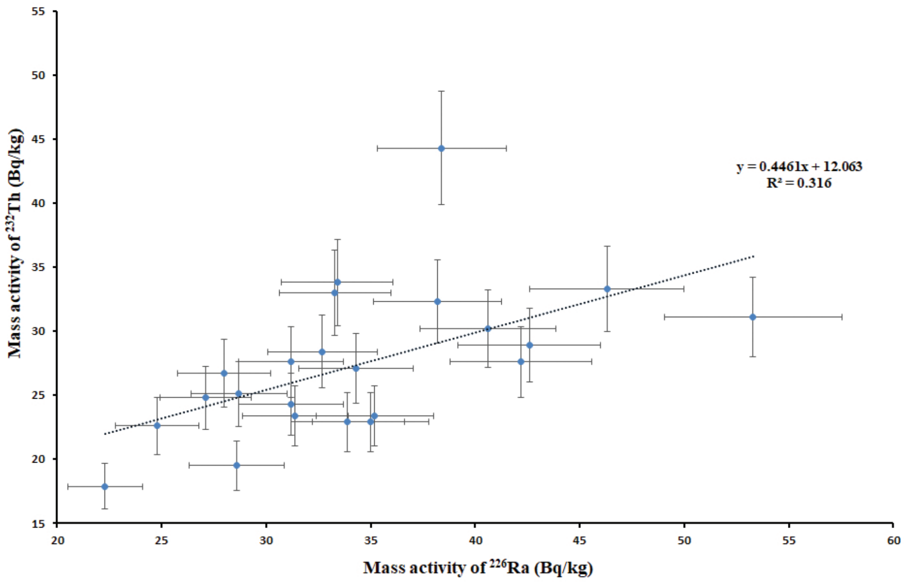

FIGURE 5. Scatter plots of ^{232}Th vs. ^{226}Ra for all samples.

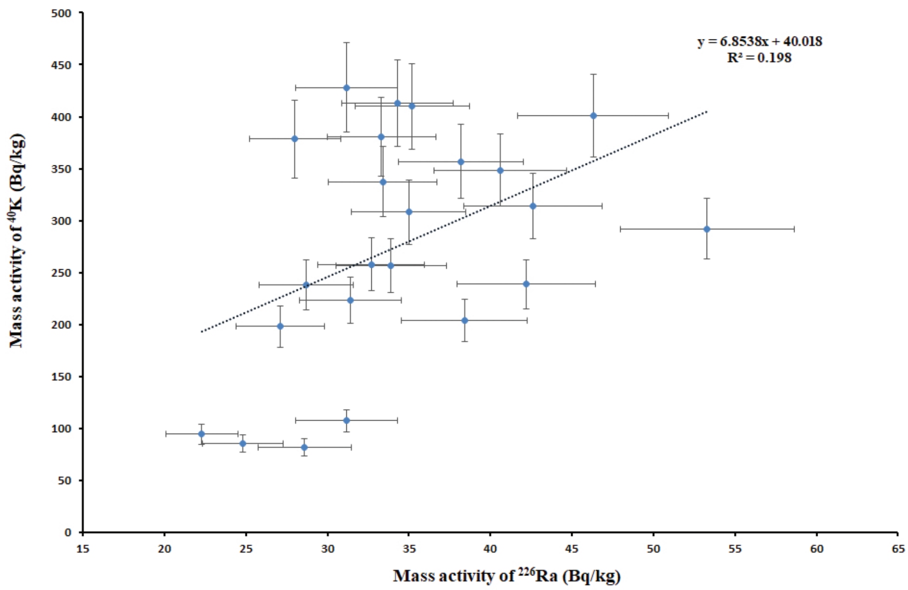

FIGURE 6. Scatter plots of ^{40}K vs. ^{226}Ra for all samples.

Table 3, the results of this study are consistent with other reported results of similar studies in other countries.

Regarding the impact of exposure to gamma radiation on one's health, it was decided not to calculate the excess lifetime cancer risk from gamma dose rates based on the life expectancy of the Iranian population as 74 years based on WHO: Iran national profile in 2012. Such results were published for the studies concerning Jhelum valley (33) or Kirklareli in Turkey (34). The main reason for its absence was that these building materials are used in areas of high natural background radiation with elevated external gamma-ray dose rates (1). Therefore, it is believed that these calculations may yield misleading results. Instead of following the typical approach, the authors decided to calculate the shielding effect of dwellings built from materials with much lower concentrations of radionuclides compared with soils in the area. This approximate

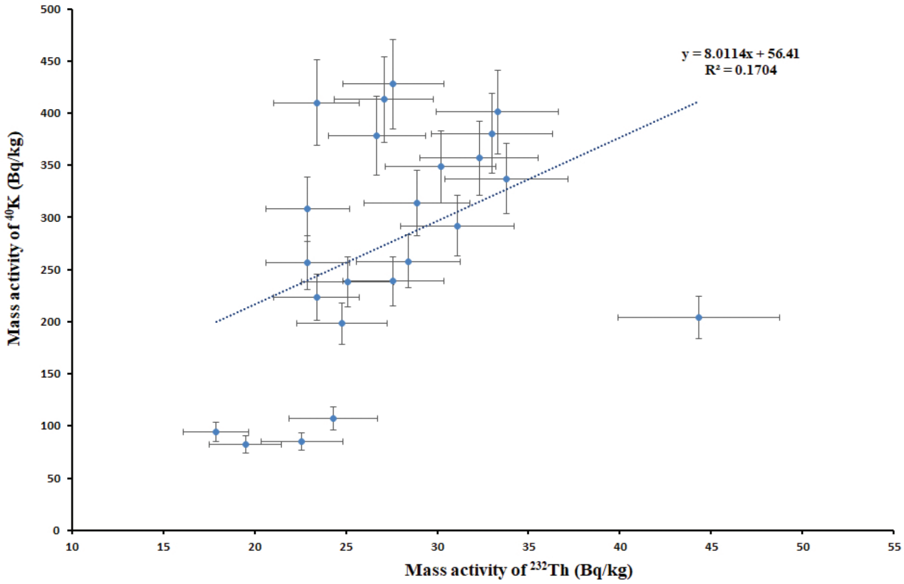

FIGURE 7. Scatter plots of ^{40}K vs. ^{232}Th for all samples.

TABLE 3. Comparison of mass activity of naturally occurring radionuclides in building materials in this study with results from other studies (Bq/kg).

Country	Building Material	a_{Ra-226} (mean)	a_{Th-232} (mean)	a_{K-40} (mean)	References
Austria	Bricks	20-71 (38)	16-112 (45)	520-880 (635)	
	Cement	11-49 (27)	10-26 (14)	89-286 (210)	(29)
	Concrete	7-21 (15)	3-57 (14)	16-382 (164)	
Denmark	Bricks	8-42 (25)	8-34 (21)	280-630 (455)	
	Cement	9-30 (20)	4-21 (12)	20-140 (90)	(29)
	Concrete	15-670 (152)	10-53 (27)	280-1190 (620)	
Hungary	Bricks	8-42(25)	8-34 (21)	280-630 (455)	
	Cement	8-61 (30)	13-53 (22)	95-402 (218)	(29)
	Concrete	13-18 (16)	11-33 (22)	204-437 (356)	
Europe	Brick	2-148 (47)	2-164 (48)	12-1169 (598)	
	Cement	4-422 (45)	3-266 (31)	4-846 (216)	(29)
	Natural gypsum	1-70 (15)	1-100 (9)	5-279 (91)	
Aden, Yemen	Portland cement	33-45 (40)	19-31 (25)	234-502 (428)	
	Sand	14-26 (21)	20-32 (28)	859-1267 (1118)	(26)
	Red bricks	46-60 (55)	30-52 (37)	1209-1343 (1256)	
Egypt	Red-brick	(23)	(23)	(448)	
	Sand	(17)	(13)	(119)	
	Cement	(45)	(10)	(51)	(27)
	Gypsum	(8)	(8)	(85)	
	Ceramic	(51)	(41)	(683)	
Brazil	Sand	(14)	(18)	(807)	
	Cement	(62)	(59)	(564)	
	Gypsum	(6)	-	(18)	(30, 31)
	Granite	(49)	(288)	(1335)	

Continued

Table 3. Continued

Country	Building Material	a_{Ra-226} (mean)	a_{Th-232} (mean)	a_{K-40} (mean)	References
Weinan, China	Sand	97-131 (119)	21-62 (36)	181-274 (250)	(32)
	Red-clay bricks	119-130 (125)	28-30 (29)	377-418 (390)	
	Gravel	91-125 (96)	12-27 (17)	281-398 (325)	
	Ceramic	50-95 (70)	23-52(39)	285-644 (417)	
	Tile	150-266 (395)	35-64 (44)	682-5515 (835)	
	Sand and Gravel	28-38 (32)	25-44 (34)	204-378 (289)	
Mahhalat (current study)	**Bricks**	**31-38 (35)**	**23-32 (28)**	**357-428 (402)**	**current study**
	Gypsum	**22-31 (27)**	**18-24 (21)**	**82-107(92)**	
	Cement	**27-42 (34)**	**23-28 (25)**	**198-257 (229)**	
	Ceramic	**33-35 (33)**	**23-33 (28)**	**258-381 (315)**	
	Tile	**43-53 (46)**	**29-33 (31)**	**292-401 (339)**	

Table 4. Assessment of annual effective dose from external gamma radiation for various occupancy times.

Indoor residence time Percentage (%)	111 nGy/h indoor gamma dose rate (mSv/y)	800 nGy/h outdoor gamma dose rate (mSv/y)	The annual dose (mSv/y)
80	0.55	0.98	1.53
70	0.48	1.47	1.95
60	0.41	1.96	2.37
50	0.34	2.45	2.79
40	0.28	2.94	3.22
30	0.21	3.43	3.64

estimation shows that the attenuation factor calculated for a wall thickness of 30 to 40 cm with an average density of the building material equal to about 2 g/cm^3 may be as high as 500 for a gamma-ray energy of 500 keV. As a result, the additional indoor gamma dose rate would not exceed the previously measured dose in studying area environment as 4 μGy/h (35). If this is compared with the minimum estimated value of gamma-ray dose from building materials in this study (~50 nGy/h), the attenuation effect of the walls of dwellings is confirmed.

Due to the aforementioned factors, the external gamma doses originating from background radiation in the area and building materials were estimated for various indoor occupancy. As we assumed the maximum indoor gamma dose rate to be 111 nGy/h (estimated by this study) and the minimum outdoor gamma dose rate about 800 nGy/h (9), then the annual effective dose without considering the dose from inhaled radon shows in Table 4.

It can be seen that for the maximum an indoor occupancy time 0.8 (80% of one's time is spent in a building), just 37% of the annual effective dose of gamma radiation is caused by radiation produced by building materials. For those who spend more time outdoors (farmers, children), the fraction of the annual effective dose that originates from natural background radiation is greater, while the percentage of the dose that originates from building materials may decrease to 14 % or even less.

Some preliminary measurements of the radon level in dwellings in the investigated area were conducted a few years ago (36). The radon concentration measured was as high as 350 Bq/m^3. By applying the recommendations from ICRP Publication 126, the estimated values of annual effective doses from that area were as high as 12 mSv/yr (the new value based on ICRP-137 were measured by this paper's authors as 13.6 mSv/yr), which is significantly greater than calculated for external gamma radiation. As a result, measurements of radon levels and its decay products in dwellings situated in areas exposed to high levels of background radiation are most important.

Unlike in previous studies, it was decided not to calculate the excess lifetime cancer risk from gamma radiation in dwellings because these building materials are in use in an area exposed to high levels of natural background radiation with elevated values of external gamma radiation dose rates. Therefore, in our opinion, such calculations might yield misleading results.

4. CONCLUSIONS

In the present study, the mass activity of naturally occurring radionuclides in twenty-three samples of building materials available on the market in Mahallat, Iran - a city exposed to a high level of natural background radiation - were determined in order to assess radiation exposure in dwellings. The distribution of results with regard to the measured mass activity of naturally occurring radionuclides found in the samples of building materials was different for ^{40}K compared to ^{226}Ra and ^{232}Th. The average mass activity of ^{226}Ra, ^{232}Th and ^{40}K in all of the samples were determined as 34 ± 6, 27 ± 6 and 276 ± 58 Bq/kg, respectively, and were all below the worldwide average values. The cumulative averages of the gamma absorbed dose rate and annual effective dose rate were estimated to be 84 ± 17 nGy/h and 412 ± 82 µSv/yr, respectively, both of which are higher than the worldwide average value (55 nGy/h) and below the EU BSS recommended annual value (1 mSv/yr). The radioactivity index, calculated according to EU BSS recommendations, was less than 1 for all of the samples. In line with the EU BSS, building materials with a radioactivity index of less than one is exempt from radiological examinations before being placed on the market. However, even for the samples with low radioactivity indexes - due to the absence of national regulations in Iran - to monitor and control the radioactivity of building materials, such measurements are necessary. The legislation of a national standard into the Iranian legal system describing the requirements for the radiological examination of building materials, is necessary before their introduction on the market.

ACKNOWLEDGEMENTS

This work was supported by Hungarian National Research OTKA grant No. K128805 and K128818 and GINOP Grant of the Hungarian Government No. 2016-0016.

REFERENCES

1. United Nations Scientific Committee on the Effects of Atomic Radiation. (2000) Sources and effects of ionizing radiation. UNSCEAR 2000 report to the General Assembly, with scientific annexes. Volume I: Sources. United Nations Publications. New York, (2000).
2. The Council of European Union. 2014. Council directive 2013/59/EURATOM. *OJEU*. L13 [1], 1–73.
3. Kovács, T.; Szeiler, G.; Fábián, F.; Kardos, R.; Gregorič, A.; Vaupotič, J. (2013) Systematic survey of natural radioactivity of soil in Slovenia. *J. Environ. Radioact.* 122, 70–78. https://doi.org/10.1016/j.jenvrad.2013.02.007.
4. Kardos, R.; Sas, Z.; Hegedűs, M.; Shahrokhi, A.; Somlai, J.; Kovács, T. (2015) Radionuclide content of NORM by-products originating from the coal-fired power plant in Oroszlány (Hungary). *Radiat. Prot. Dosim.* 167 [1–3], 266–269. https://doi.org/10.1093/rpd/ncv259.
5. Aliyu, A.S.; Ramli, A.T. (2015) The world's high background natural radiation areas (HBNRAs) revisited: A broad overview of the dosimetric, epidemiological and radiobiological issues. *Radiat. Meas.* 73, 51–59. https://doi.org/10.1016/j.radmeas.2015.01.007.
6. International Commission on Radiological Protection. (2007) The 2007 recommendations of the International Commission on Radiological Protection. *Annals of the ICRP*. 103.
7. Shetty, P.K.; Narayana, Y. (2010) Variation of radiation level and radionuclide enrichment in high background area. *J. Environ. Radioact.* 101 [12], 1043–1047. https://doi.org/10.1016/j.jenvrad.2010.08.003.
8. Mortazavi, S.M.J.; Mozdarani, H. (2012) Is it time to shed some light on the black box of health policies regarding the inhabitants of the high background radiation areas of Ramsar? *Iran J. Radiat. Res*. 10 [3–4], 111–116.
9. Sohrabi, M.M.; Beitollahi, M.M.; Lasemi, Y.; Amin, S.E. (1996) Origin of a new high level natural radiation area in hot spring region of Mahallat, Central Iran. In Proceedings of the 4 th International Conference on High Levels of Natural Radiation. Vienna, (1996).
10. The Council of European Union. (1999) Radiation Protection 112: Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials. Directorate-General, Environment, Nuclear Safety and Civil Protection. 5–16.
11. Nuccetelli, C.; Leonardi, F.; Trevisi, R. (2015) A new accurate and flexible index to assess the contribution of building materials to indoor gamma exposure. *J. Environ. Radioact.* 143, 70–75. https://doi.org/10.1016/j.jenvrad.2015.02.011.
12. Karam, P.A. (2002) The high background radiation area in Ramsar Iran: Geology, Norm, Biology, LNT, and possible regulatory fun. University of Rochester. Rochester, NY, (2002).
13. Ghiassi-Nejad, M.; Mortazavi, S.M.J.; Cameron, J.R.; Niroomand-Rad, A.; Karam, P.A. (2002) Very high background radiation areas of Ramsar, Iran: preliminary biological studies. *Health physics*. 82 [1], 87–93. https://doi.org/10.1097/00004032-200201000-00011.
14. Hendry, J.H.; Simon, S.L.; Wojcik, A.; Sohrabi, M.; Burkart, W.; Cardis, E.; Laurier, D.; Tirmarche, M.; Hayata, I. (2009) Human exposure to high natural background radiation: what can it teach us about radiation risks?. *J. Radiol. Prot.* 29 [2A], A29. https://doi.org/10.1088/0952-4746/29/2A/S03.
15. Jayanthi, D.D.; Maniyan, C.G.; Perumal, S. (2011) Assessment of indoor radiation dose received by the residents of natural high background radiation areas of coastal villages of Kanyakumari district, Tamil Nadu, India. *Radiat. Phys. Chem.* 80 [7], 782–785. https://doi.org/10.1016/j.radphyschem.2011.03.011.
16. Bavarnegin, E.; Fathabadi, N.; Moghaddam, M.V.; Farahani, M.V.; Moradi, M.; Babakhni, A. (2013) Radon exhalation rate and natural radionuclide content in building materials of high background areas of Ramsar, Iran. *J. Environ. Radioact.* 117, 36–40. https://doi.org/10.1016/j.jenvrad.2011.12.022.
17. Sohrabi, M. (2013) World high background natural radiation areas: Need to protect public from radiation exposure. *Radiat. Meas.* 50, 166–171. https://doi.org/10.1016/j.radmeas.2012.03.011.
18. Sahoo, S.K.; Žunić, Z.S.; Kritsananuwat, R.; Zagrodzki, P.; Bossew, P.; Veselinovic, N.; Mishra, S.; Yonehara, H.; Tokonami, S. (2015) Distribution of uranium, thorium and some stable trace and toxic elements in human hair and nails in Niška Banja Town, a high natural background radiation area of Serbia (Balkan Region, South-East Europe). *J. Environ. Radioact.* 145, 66–77. https://doi.org/10.1016/j.jenvrad.2015.03.020.
19. Mubarak, F.; Fayez-Hassan, M.; Mansour, N.A.; Ahmed, T.S.; Ali, A. (2017) Radiological Investigation of High Background Radiation Areas. *Sci. Rep.* 7, 15223. https://doi.org/10.1038/s41598-017-15201-2.
20. Pérez, M.; Chávez, E.; Echeverría, M.; Córdova, R.; Recalde, C. (2018) Assessment of natural background radiation in one of the highest regions of Ecuador. *Radiat. Phys. Chem.* 146, 73–76. https://doi.org/10.1016/j.radphyschem.2018.01.002.

21. Okeyode, I.C.; Oladotun, I.C.; Alatise, O.O.; Bada, B.S.; Makinde, V.; Akinboro, F.G.; Mustapha, A.O.; Al-Azmi, D. (2019) Indoor gamma dose rates in the high background radiation area of Abeokuta, South Western Nigeria. *J. Radiat. Res. Appl. Sci.* 12 [1], 72–77. https://doi.org/10.1080/16878507.2019.1594097.
22. Bé, M.-M.; Chechev, V.P.; Dersch, R.; Helene, O.A.M.; Helmer, R.G.; et al. (2007) Update of x ray and gamma ray decay data standards for detector calibration and other applications, volume 2: data selection assesment and evaluation procdures. International Atomic Energy Agency. Vienna, (2007).
23. Shahrokhi, A. (2018) Applicaton of the European basic safety standards directive in underground mines: a comprehensive radioecology study in a Hungarian manganese mine. *PhD thesis,* Pannon Egyetem. Veszprém, (2019). https://doi.org/10.18136/PE.2018.696.
24. Adelikhah, M.; Shahrokhi, A.; Chalupnik, S.; Tóth-Bodrogi, E.; Kovács, T. (2020) High level of natural ionizing radiation at a thermal bath in Dehloran, Iran. *Heliyon.* 6 [7], e04297. https://doi.org/10.1016/j.heliyon.2020.e04297.
25. Shahrokhi, A.; Szeiler, G.; Rahimi, H.; Kovács, T. (2014) Investigation of natural and anthropogenic radionuclides distribution in arable land soil of south eastern European countries. *Int. J. Sci. Engineer. Res*. 5 [11], 445–449.
26. El-Mageed, A.I.A.; Farid, M.E.A.; Saleh, E.E.; Mansour, M.; Mohammed, A.K. (2014) Natural radioactivity and radiological hazards of some building materials of Aden, Yemen. *J. Geochem. Explor.* 140, 41–45. https://doi.org/10.1016/j.gexplo.2014.01.015.
27. Shoeib, M.Y.; Thabayneh, K.M. (2014) Assessment of natural radiation exposure and radon exhalation rate in various samples of Egyptian building materials. *J. Radiat. Res. Appl. Sci.* 7 [2], 174–181. https://doi.org/10.1016/j.jrras.2014.01.004.
28. Wang, Q.; Song, J.; Li, X.; Yuan, H.; Li, N.; Cao, L. (2015) Environmental radionuclides in a coastal wetland of the Southern Laizhou Bay, China. *Mar. Pollut. Bull.* 97 [1–2], 506–511. https://doi.org/10.1016/j.marpolbul.2015.05.035.
29. Trevisi, R.; Risica, S.; D'Alessandro, M.; Paradiso, D.; Nuccetelli, C. (2012) Natural radioactivity in building materials in the European Union: a database and an estimate of radiological significance. *J. Environ. Radioact.* 105, 11–20. https://doi.org/10.1016/j.jenvrad.2011.10.001.
30. Malanca, A.; Pessina, V.; Dallara, G. (1993) Radionuclide content of building materials and gamma ray dose rates in dwellings of Rio Grande Do Norte, Brazil. *Radiat. Prot. Dosim.* 48 [2], 199–203. https://doi.org/10.1093/oxfordjournals.rpd.a081865.
31. Dabayneh, K.M. (2007). Radioactivity measurement in different types of fabricated building materials used in Palestine. *Arab J. Nuclear Sci. Applic.* 40 [3], 208–219.
32. Lu, X.; Chao, S.; Yang, F. (2014) Determination of natural radioactivity and associated radiation hazard in building materials used in Weinan, China. *Radiat. Phys. Chem.* 99, 62–67. https://doi.org/10.1016/j.radphyschem.2014.02.021.
33. Rafique, M.; Rahman, S.U.; Basharat, M.; Aziz, W.; Ahmad, I.; Lone, K.A.; Ahmad, K.; Matiullah, K. A. (2014) Evaluation of excess life time cancer risk from gamma dose rates in Jhelum valley. *J. Radiat. Res. Appl. Sci.* 7 [1], 29–35. https://doi.org/10.1016/j.jrras.2013.11.005.
34. Taskin, H.; Karavus, M.; Ay, P.; Topuzoglu, A.; Hidiroglu, S.; Karahan, G. (2009) Radionuclide concentrations in soil and lifetime cancer risk due to gamma radioactivity in Kirklareli, Turkey. *J. Environ. Radioact.* 100 [1], 49–53. https://doi.org/10.1016/j.jenvrad.2008.10.012.
35. Sohrabi, M.; Roositalab, J.; Mohammadi, J. (2015) Public effective doses from environmental natural gamma exposures indoors and outdoors in Iran. *Radiat. Prot. Dosim.* 167 [4], 633–641. https://doi.org/10.1093/rpd/ncu372.
36. Shahrokhi, A.; Shokraee, F.; Reza, A.; Rahimi, H. (2015) Health risk assessment of household exposure to indoor radon in association with the dwelling's age. *J. Radiat. Protect. Res.* 40 [3], 155–161. https://doi.org/10.14407/jrp.2015.40.3.155.