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NATURAL RADIONUCLIDE CONTENT AND RADIOLOGICAL HAZARD ASSOCIATED WITH USAGE OF QUARTZITE SAND SAMPLES FROM OVACIK–SILIFKE–MERSIN OPEN PIT AS BUILDING MATERIAL IN TURKEY

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Activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in quartzite sand samples collected from the Ovacık–Silifke–Mersin open pit located in the East Mediterranean region of Turkey were determined by using a gamma-ray spectrometry technique. The mean activity concentrations of the ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in quartzite sand samples were found as 81.7 ± 22.9 Bq kg^{-1}, 6.3 ± 2.8 Bq kg^{-1}, 77.5 ± 24.3 Bq kg^{-1}and 140.0 ± 124.1 Bq kg^{-1}, respectively. The gamma index (I_γ), the internal exposure index (I_α), the indoor absorbed dose rate (D_{in}) and the corresponding annual effective dose (H_{in}) were evaluated for the public exposure to radiological hazard arising due to the use of quartzite sand samples as building material. The values of I_γ, I_α, D_{in} and H_{in} ranged from 0.20 to 0.75, with a mean of 0.34 ± 0.11, 0.23 to 0.77 with a mean of 0.39 ± 0.12, 58.27 to 201.51 nGy h^{-1} with a mean of 93.33 ± 27.63 nGy h^{-1} and 0.29 to 0.99 mSv with a mean of 0.46 ± 0.14 mSv, respectively.

INTRODUCTION

Naturally occurring radionuclides of terrestrial origin are present in various degrees in all materials derived from rock and soils. Irradiation of the human body from external sources is mainly by gamma radiation from naturally occurring radionuclides in the uranium-radium (^{238}U–^{226}Ra) and thorium (^{232}Th) series and from the radioactive isotope of potassium (^{40}K). The specific levels of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K are related to the types of rock from which the soils originate. The median values of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in the earth's crust are 35, 30, 35 and 400 Bq kg^{-1}, respectively[1]. In order to assess the possible radiological hazards to human health from the external gamma radiation, it is important to study the radioactivity levels emitted by the materials such as rock, soil, sand, granite, cement, pumice, marble, limestone etc.[2–10]

Quartzite sands in the Ovacık–Silifke–Mersin open pit are formed in sedimentary and metamorphic formations[11]. The Ovacık–Silifke–Mersin open pit mine (Figure 1) is one of the biggest quartzite mines in Turkey. The length of economical quartzite deposit in Ovacık pit mine is about 370 m and the average thickness is nearly 45 m. Also the economical producible depth is about 40 m[12]. Quartzite sands obtained from Ovacık open pit are the most commonly used in ferrochromites, ferrosilisium, iron-steel, light gas concrete, silica bricks, ceramic, chemistry, plastic and colour industries. Nowadays a few studies relate to the Ovacık open pit[12,13]. However, detailed information of the activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in quartzite sand samples from collected Ovacık open pit in Turkey is not available in literature.

The aim of the present study is to determine the activity concentration of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in quartzite sand samples collected from the Ovacık open pit using gamma-ray spectrometry and to assess any radiation hazard for exposure of the public from external gamma radiation arising due to the use of quartzite sand samples as building material by estimating the gamma index, the internal exposure index, the indoor gamma dose rate and the corresponding the annual effective dose.

MATERIALS AND METHODS

Sample preparation

Thirty quartzite sand samples weighing from 1 to 1.5 kg were collected from the Ovacık–Silifke–Mersin open pit. Fifteen sampling sites were chosen to cover the whole open pit as much as possible. At every sampling site, quartzite sand samples were collected from the different profile depths (0–100 cm) and lengths (20 m approximately) of walls on each location. The distance between two sample locations at each sampling site is 50 m. These sampling sites

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were numbered according to the frequency and appearance of the quartzite ore in the open pit (Figure 2).

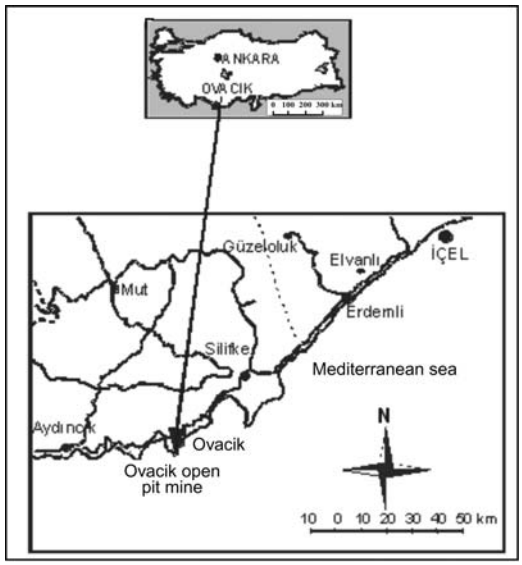

Figure 1. Location map of the study area.

The samples were properly coded according to the location of the sampling site. The samples were ground into a fine powder with a particle size of <1 mm. The samples were then dried in a temperature-controlled furnace at 110°C for 20–24 h to remove moisture. After moisture removal, these samples were cooled in a moisture-free atmosphere. Each sample was then filled into cylindrical plastic containers ($\phi = 5$ cm, h = 6 cm), weighed and hermetically sealed. The geometrical dimensions of the samples were kept identical to that of the reference materials which were used for the efficiency calibration of the gamma spectrometry system. The sealed samples and the reference materials were stored for a few months before counting to allow the secular equilibrium in the ^{238}U (i.e. ^{238}U–^{234}Th equilibrium and ^{226}Ra–^{222}Rn equilibrium) and ^{232}Th decay series even if ^{238}U and ^{234}Th are not in equilibrium.

Experimental set-up and gamma-ray measurements

The activity concentration of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in the quartzite sand samples were measured with a high resolution HPGe gamma-ray spectrometry system. The system was equipped with a coaxial p-type HPGe detector (GC11021) with an active volume of 451 cm^3 manufactured by Canberra Inc. The HPGe detector has a relative efficiency of

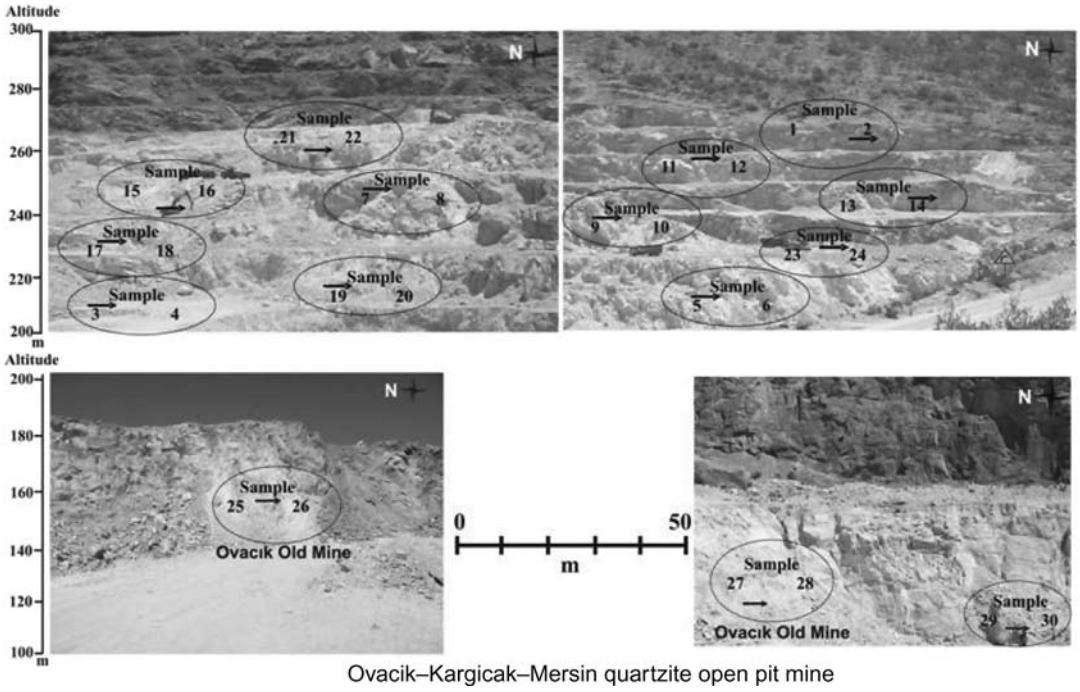

Figure 2. Sampling sites.

110%, an energy resolution of 2.1 keV at 1332.5 keV of ^{60}Co and of 1.3 keV at 122 keV of ^{57}Co, and a peak-to-Compton ratio of 85 : 1. For gamma ray shielding, a front opening split-top shield (Canberra Model 767) was used to reduce background. The detector was interfaced to the Digital Spectrum Analyzer (DSA-1000), which was a full-featured 16 K channel multichannel analyzer on advanced digital signal processing techniques.

Energy calibration of the detector was performed using point sources (^{60}Co, ^{137}Cs and ^{241}Am). The absolute efficiency calibration of the gamma spectrometry system was carried out using the radionuclide specific efficiency method in order to reduce the uncertainty in gamma ray intensities, as well as the influence of coincidence summation and self-absorption effects of the emitting gamma photons[14,15]. The IAEA reference materials RGU-1 (U-ore), RGTh-1 (Th-ore) and RGK-1 (K_2SO_4), with densities similar to the samples to be measured after pulverisation, were employed for the efficiency calibration of the system.

The sample containers were placed on top of the detector for counting. The same geometry was used to determine peak area of samples and references. The accumulating time of the sample spectra was 30 000 s in order to obtain a gamma spectrum with good statistics. Background measurements were taken and subtracted in order to get net counts for the sample.

The activity concentrations of ^{238}U and ^{226}Ra were calculated taking into account the secular equilibrium between ^{238}U and ^{234}Th, ^{226}Ra–^{222}Rn and averaged from gamma-ray photopeaks at several energies. The gamma ray lines of 63.3 and 92.8 keV from ^{234}Th were used to determine the activity concentration of ^{238}U, while the gamma ray lines of 351.9 keV from ^{214}Pb and 609.3 keV from ^{214}Bi were used to determine the activity concentration of ^{226}Ra. In the case of thorium series, the existence of the equilibrium can be assumed throughout the series if the emanation is prevented. The gamma ray lines of 911.2 keV from ^{228}Ac and 583.2 keV from ^{208}Tl were used to determine the activity concentration of ^{232}Th. The activity concentration of ^{40}K was measured directly by its own gamma ray at 1460.8 keV. The statistical uncertainty of the radioactivity measurements was calculated by taking into consideration the systematic errors in the efficiency calibration (0.5–5 %) and counting statistical error (up to 10 %).

The minimum detectable activity (MDA) of the present measurement system was calculated as follows[16]:

$$\mathrm{MDA} = \frac{1.64\ \sigma_{\mathrm{NB}}}{\varepsilon \cdot P \cdot t \cdot w} \quad (1)$$

where MDA is in Bq kg^{-1}(confidence level 95 %), σ_{N_B} is the standard deviation (SD) of the background in the region of interest and equals the square root of the number of counts for the background spectrum, ε is the absolute efficiency of the detector, P is the absolute emission probability of the gamma decay, t is the measurement time in seconds and w is the weight of the dried sample expressed in kg. The mean value of the MDA was 0.6 Bq kg^{-1} for ^{238}U, 0.7 Bq kg^{-1} for ^{232}Th, 1.2 Bq kg^{-1} for ^{226}Ra and 3.7 Bq kg^{-1} for ^{40}K.

RESULTS AND DISCUSSION

Radionuclide content

The activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K measured in the studied quartzite sand samples in Bq kg^{-1}, together with the statistical uncertainty (1σ) and SD are shown in Table 1. The activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in the studied sand sample ranged from 52.7 ± 4.8 to 159.4 ± 15.4 Bq kg^{-1} with a mean of 81.7 ± 22.9 Bq kg^{-1}, 2.1 ± 0.2 to 15.8 ± 1.2 Bq kg^{-1} with a mean of 6.3 ± 2.8 Bq kg^{-1}, 46.8 ± 3.3 to 153.6 ± 11.2 Bq kg^{-1} with a mean of 77.5 ± 24.3 Bq kg^{-1}and 40.1 ± 4.9 to 484.5 ± 40.1 Bq kg^{-1} with a mean of 140.0 ± 124.1 Bq kg^{-1}, respectively. ^{226}Ra/^{238}U ratios in all analysed sand samples are in the range 0.86–1.06 (with a mean of 0.94), suggesting that the actual specific activities of ^{226}Ra in the sand samples could be on the average 5.1 % lower than the mean value of ^{238}U. This assumes that ^{226}Ra and ^{238}U are in the secular equilibrium. As can be seen Table 1, the highest values for the activity concentration of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K are measured in the sand sample collected from the sample site 13 in the Ovacık old open pit. The mean values of the activity concentrations of ^{232}Th and ^{40}K are lower than the median values in the earth's crust (30 Bq kg^{-1} for ^{232}Th and 400 Bq kg^{-1} for ^{40}K) while the mean values of the activity concentrations of ^{238}U and ^{226}Ra are significantly (two times) higher than the median value (35 Bq kg^{-1} for ^{238}U and ^{226}Ra).

Gamma index

Gamma index was calculated as proposed by the European Commission (EC) report[17]. The Commission suggests that building materials should be exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from them increases the annual effective dose of a member of the public by 0.3 mSv at most. Controls should be based on dose criterion, which was established considering the overall nations circumstances. It is therefore recommended that controls should be based on an annual effective dose in the range 0.3–1 mSv. Gamma index (I_γ) was calculated using the following formula proposed by the

Table 1. The activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K measured in the studied quartzite sand samples.

Location	Activity concentration (in Bq kg^{-1} ± 1σ)			
	^{238}U	^{232}Th	^{226}Ra	^{40}K
Sample site 1	78.0 ± 6.2	5.4 ± 0.7	68.4 ± 6.2	50.5 ± 6.3
	74.9 ± 6.1	3.2 ± 0.4	64.2 ± 5.6	40.1 ± 4.9
Sample site 2	77.2 ± 5.6	5.2 ± 0.8	72.9 ± 7.7	106.2 ± 14.4
	123.3 ± 8.5	4.3 ± 0.6	116.4 ± 10.4	85.1 ± 11.5
Sample site 3	82.7 ± 7.2	2.8 ± 0.4	77.9 ± 6.7	380.9 ± 41.0
	57.4 ± 4.9	4.5 ± 0.6	50.9 ± 4.4	373.7 ± 39.7
Sample site 4	73.6 ± 7.1	6.9 ± 0.8	69.5 ± 7.1	84.0 ± 9.7
	67.9 ± 6.3	7.8 ± 0.9	64.5 ± 6.4	70.1 ± 7.9
Sample site 5	75.6 ± 7.4	5.1 ± 0.4	74.7 ± 5.2	87.3 ± 7.8
	68.9 ± 6.8	4.1 ± 0.3	65.7 ± 4.6	76.3 ± 6.8
Sample site 6	89.3 ± 8.2	9.4 ± 1.2	81.4 ± 5.5	116.8 ± 14.5
	68.9 ± 6.4	6.6 ± 0.8	62.2 ± 4.3	112.7 ± 13.9
Sample site 7	65.0 ± 7.2	6.5 ± 0.6	58.7 ± 4.7	242.3 ± 29.1
	74.6 ± 8.1	4.7 ± 0.4	66.2 ± 5.4	222.2 ± 28.6
Sample site 8	81.4 ± 8.5	6.7 ± 0.5	77.1 ± 6.5	98.6 ± 10.3
	72.3 ± 7.7	8.2 ± 0.6	65.7 ± 5.8	85.6 ± 9.1
Sample site 9	70.8 ± 5.7	2.1 ± 0.2	73.9 ± 6.0	81.9 ± 7.5
	121.8 ± 10.5	4.0 ± 0.4	126.5 ± 9.9	65.3 ± 6.3
Sample site 10	84.2 ± 8.9	6.7 ± 0.9	75.8 ± 5.0	91.9 ± 12.5
	67.5 ± 7.3	5.7 ± 0.8	62.0 ± 4.2	78.1 ± 10.8
Sample site 11	72.5 ± 6.8	6.5 ± 0.7	66.5 ± 4.1	70.7 ± 7.3
	52.7 ± 4.8	4.8 ± 0.5	46.8 ± 3.3	56.3 ± 5.7
Sample site 12	85.7 ± 7.5	7.9 ± 0.9	81.5 ± 6.4	150.6 ± 16.3
	60.6 ± 5.2	7.5 ± 0.9	56.7 ± 4.0	140.4 ± 14.7
Sample site 13	96.6 ± 9.3	11.2 ± 0.8	94.1 ± 6.8	484.5 ± 30.0
	159.4 ± 15.4	15.8 ± 1.2	153.6 ± 11.2	468.5 ± 30.2
Sample site 14	123.9 ± 10.8	8.2 ± 1.1	131.8 ± 12.8	65.0 ± 6.3
	64.6 ± 5.5	10.2 ± 1.3	67.1 ± 6.6	54.3 ± 5.2
Sample site 15	85.4 ± 9.7	3.8 ± 0.5	82.8 ± 6.9	82.6 ± 11.0
	74.1 ± 8.7	4.3 ± 0.6	70.3 ± 6.1	76.1 ± 10.4
Mean ± SD	81.7 ± 22.9	6.3 ± 2.8	77.5 ± 24.3	140.0 ± 124.1

EC report:

$$I_\gamma = \frac{C_{Ra}}{300\ \text{Bq kg}^{-1}} + \frac{C_{Th}}{200\ \text{Bq kg}^{-1}} + \frac{C_K}{3000\ \text{Bq kg}^{-1}} \quad (2)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}. The activity index should be used only as a screening tool for identifying materials which might be of concern. For materials used in bulk amounts, e.g. concrete $I_\gamma \leq 1$ corresponds to an annual effective dose less than or equal 1 mSv, while $I_\gamma \leq 0.5$ corresponds to an annual effective dose less than or equal 0.3 mSv. The values of the calculated I_γ for quartzite sand samples are shown in the first column of Table 2. It is observed in Table 2 that the mean value of I_γ for sand samples (0.34 ± 0.11) is about 1.5 times lower than the criterion of 0.5 corresponding to an effective dose of 0.3 mSv.

Internal exposure index (alpha index)

Up to now, several indexes dealing with the assessment of the excess alpha radiation due to inhalation originating from building materials such as alpha indexes and internal health indexes were developed[18]. In the present study, the alpha index was determined by the next formula:

$$I_\alpha = \frac{C_{Ra}}{200\ \text{Bq kg}^{-1}} \quad (3)$$

where C_{Ra} is the activity concentration of ^{226}Ra in Bq kg^{-1}. The recommended upper level indoor radon exposure in buildings set by the Turkish Standard TS 12614[19] is 200 Bq m^{-3} while the recommended value for remediation of indoor radon exposure in existing building set by Turkish Radiation Safety Regulation[20] is 400 Bq m^{-3}. $I_\alpha \leq 1$ corresponds to the ^{226}Ra activity concentration less than or equal 200 Bq kg^{-1}. When the activity concentration of ^{226}Ra in a building material

Table 2. The values of the gamma index, the alpha index, the indoor absorbed gamma dose rate and the corresponding annual effective dose.

Location	Gamma index I_γ	Alpha index I_α	Absorbed gamma dose rate (nGy h^{-1}) D_{in}	Annual effective dose (mSv) H_{in}
Sample site 1	0.27	0.34	81.78	0.40
	0.24	0.32	75.64	0.37
Sample site 2	0.30	0.36	85.25	0.42
	0.44	0.58	124.97	0.61
Sample site 3	0.40	0.39	109.66	0.54
	0.32	0.25	87.65	0.43
Sample site 4	0.29	0.35	81.93	0.40
	0.28	0.32	76.66	0.38
Sample site 5	0.30	0.37	82.16	0.40
	0.26	0.33	74.00	0.36
Sample site 6	0.36	0.41	101.78	0.50
	0.28	0.31	79.66	0.39
Sample site 7	0.31	0.29	86.36	0.42
	0.32	0.33	91.58	0.45
Sample site 8	0.32	0.39	90.15	0.44
	0.29	0.33	82.38	0.40
Sample site 9	0.28	0.37	74.01	0.36
	0.46	0.63	121.68	0.60
Sample site 10	0.32	0.38	92.26	0.45
	0.26	0.31	74.62	0.37
Sample site 11	0.28	0.33	79.54	0.39
	0.20	0.23	58.27	0.29
Sample site 12	0.36	0.41	99.59	0.49
	0.27	0.28	75.23	0.37
Sample site 13	0.53	0.47	140.00	0.69
	0.75	0.77	201.51	0.99
Sample site 14	0.50	0.66	128.29	0.63
	0.29	0.34	75.00	0.37
Sample site 15	0.32	0.41	89.35	0.44
	0.28	0.35	79.09	0.39
Mean $\pm$ SD	0.34 ± 0.11	0.39 ± 0.12	93.33 ± 27.63	0.46 ± 0.14

exceeds the value of 200 Bq kg^{-1}, it is possible that the radon exhalation from this material could cause indoor radon concentration exceeding 200 Bq m^{-3}.

The values of the calculated I_α for sand samples are shown in the second column of Table 2. The calculated I_α varied from 0.23 to 0.77 with a mean 0.39 ± 0.12 for sand samples. It is observed in Table 2 that the calculated I_α values for all the studied samples are below the recommended upper level.

Evaluation of the indoor absorbed dose rate and the annual effective dose

It is possible to evaluate the absorbed dose rate in indoor air and the corresponding annual effective dose from the activity concentrations of ^{238}U, ^{232}Th and ^{40}K measured for each sample. These results are of great interest in the radiological protection study for exposure to the public from the external gamma radiation, since quartzite sand samples are widely used to make lightweight building materials such as gas concrete and concrete blocks and bricks.

The absorbed dose rate in indoor air (D_{in}) due to the external gamma radiation from the ^{238}U, ^{232}Th and ^{40}K radionuclides in the building material, were evaluated using data and formulae provided by the EC report[17]. In the EC report, the dose conversion coefficients were calculated for the centre of the standard room. The dimensions of the room are 4 m $\times$ 5 m $\times$ 2.8 m. The thickness of walls, floor and ceiling and density of the structures are 20 cm and 2350 kg m^{-3} (concrete), respectively. These coefficients correspond to 0.92 nGy h^{-1} per Bq kg^{-1} for ^{238}U, 1.1 nGy h^{-1} per Bq kg^{-1} for ^{232}Th and 0.080 nGy h^{-1} per Bq kg^{-1} for ^{40}K:

$$D_{in}(\text{nGy h}^{-1}) = 0.92 \times C_U + 1.10 \times C_{Th} + 0.08 \times C_K \quad (4)$$

where C_U, C_{Th} and C_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}.

In estimating the indoor annual effective dose, the conversion factor from absorbed dose in air to effective dose and the occupancy factor are important. In the UNSCEAR report[1], a value of 0.7 Sv Gy^{-1} was used for the conversion factor from absorbed dose in air to effective dose received by adults, and 0.8 for the indoor occupancy factor, implying that 80 % of the time is spent indoors, on average, around the world. The annual effective dose in units of mSv per year was estimated using the following formula:

$$H_{in} = D_{in} \times 8760\,h \times 0.8 \times 0.7\,Sv\ Gy^{-1} \times 10^{-6} \quad (5)$$

where D_{in}(nGy h^{-1}) is given by Equation (4).

The results of the D_{in} and H_{in} evaluated for quartzite sand samples from the Ovacık open pit are given in the second and the third columns of Table 2, respectively. The values of the D_{in} and H_{in} ranged from 58.27 to 201.51 nGy h^{-1} with a mean of 93.33 ± 27.63 nGy h^{-1} and 0.29 to 0.99 mSv with a mean of 0.46 ± 0.14 mSv, respectively.

CONCLUSIONS

The natural radioactivity due to the presence of ^{238}U, ^{232}Th and ^{40}K radionuclides in sand samples collected from the Ovacık–Silifke–Mersin open pit was measured using a gamma-ray spectrometer with an HPGe detector. The results show that the mean values of activity concentration of ^{232}Th and ^{40}K are lower than the corresponding world median values while the mean activity concentration value of ^{238}U is about two times higher than the corresponding world median value. For each sample in this study, the gamma index, the internal exposure index, the absorbed gamma dose rate and the corresponding annual effective dose have been determined to assess the radiological hazard of exposure of the public resulting from the use of the quartzite sand samples as building materials in Turkey. The calculated values of the gamma index and the internal exposure index for all the studied sand samples are below the recommended upper level of unity. The evaluated mean value of the indoor absorbed gamma dose rate is about 10 % higher than the population-weighted average value of 84 nGy h^{-1} for the indoor absorbed dose rate. The EC suggests that building materials should be exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from them increases the annual effective dose of a member of the public by 0.3 mSv at most[17]. Controls should be based on the dose criterion, which was established considering the overall nations circumstances. The annual excess gamma doses incurred by inhabitants of dwellings due to natural radionuclides in building materials exceeding 1 mSv y^{-1} are very exceptional and can hardly be disregarded from the radiation protection point of view. It is therefore recommended that controls should be based on an annual effective dose in the range 0.3–1 mSv. The evaluated values of the indoor annual effective dose for all the studied sand samples are below the recommended upper level of 1 mSv. The mean value of the indoor annual effective dose is about 50 % lower than upper level of 1 mSv.

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