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TECHNICAL NOTE

RADIOACTIVITY LEVELS OF LIMESTONE AND GYPSUM USED AS BUILDING RAW MATERIALS IN TURKEY AND ESTIMATION OF EXPOSURE DOSES

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The natural radionuclide contents of limestone and gypsum samples used as building raw materials in Turkey were measured by means of gamma spectrometer system with HPGe detector. The measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclide were compared with the reported data from other countries. Radium equivalent activity and internal hazard index, absorbed gamma dose rate in indoor due to the external exposure and corresponding annual effective dose were determined to estimate the exposure risk arising due to the use of these raw materials. This study has shown that examined gypsum and limestone raw materials do not pose any excess radiological health problem and are safe for use in the construction of buildings.

INTRODUCTION

Natural radioactivity in raw materials used for the construction of dwellings result in external and internal exposure to the occupants. All building raw materials derived from rock and soil such as, limestone, gypsum, pumice, etc. contain varying amounts of natural radionuclides, including uranium (^{238}U) and thorium (^{232}Th) and their decay products and the radioactive isotope of potassium (^{40}K). In the ^{238}U series, the decay chain segment starting from radium (^{226}Ra) is radiologically the most important and, therefore, reference is often made to ^{226}Ra instead of ^{238}U. External exposure is caused by direct gamma radiation emitting these radionuclides while internal exposure is caused by the inhalation of the radioactive inert gas radon (^{222}Rn, a daughter product of ^{226}Ra) and its short-lived decay products. Knowledge of the level of natural radioactivity in building materials is not only needed to assess the possible radiological hazards to human health but also to develop standards and guidelines for the use and management of these materials.

Limestone is sedimentary rock consisting of calcium carbonate ($CaCO_3$) at least 90 %. Limestone is an important raw material for the cement and concrete industries. Especially limestone is the chief raw material for manufacturing of Portland cement. It is also an important raw material in iron and steel making, glass manufacture and sugar refining. Turkey is a major producer of limestone. In 2004, Turkey produced 60 million tons of limestone and aggregate[1].

Gypsum is a mineral consisting of water-containing calcium sulphate ($CaSO_4.2H_2O$). Gypsum is one of the most widely used minerals in the manufacture of building products such as cement, plaster and plasterboard in the construction sector. Gypsum is used mainly in Portland cement, which is used in concrete for highways, bridges, buildings and many other structures. Turkey produced about 4.4 million tons of gypsum in 2006[2]. The total reserves including proven, probable and possible reserves are about 39.5 billion tons for limestone and 3.9 billion tons for gypsum[3].

The levels of natural radioactivity in limestone and gypsum used as building raw materials have been assessed by many authors[4–13]. Up to now several studies related to the hydrogeological properties and tectonic significance of gypsum and limestone have been reported[14–19]. However, the detailed information of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in limestone or gypsum raw materials in Turkey is not available in the literature. In the present study, a total of 104 limestone and gypsum samples were analysed using gamma-ray spectrometry to determine the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K. The results are used to assess the potential radiological hazards associated with the use of these materials for buildings by estimating indices of radium equivalent activity and internal hazard index, indoor absorbed gamma dose rate and corresponding annual effective dose.

MATERIALS AND METHODS

Sampling and sample preparation

Sixty-six limestone samples and 38 gypsum samples were collected from different quarries as shown in

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Figure 1. Map showing the sampling locations.

Figure 1. The samples weighing from 0.5 to 1 kg were catalogued, coded properly and ground into a fine powder with a particle size <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 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 those 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 more than 30 d before counting to allow ^{226}Ra and its short-lived decay products to reach the secular equilibrium.

Radiometric analysis

The radiometric analyses were performed with a gamma-ray spectrometry system equipped with a coaxial p-type HPGe detector (GC11021) with an relative efficiency of 110 % relative to a 7.62 cm (diam.) × 7.62 cm cylindrical NaI(Tl) detector, an energy resolution of 2.1 keV at 1332.5 keV of ^{60}Co and of 1.3 keV at 122 keV of ^{57}C, 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 the background. It features 10-cm lead thickness, which is jacketed by a 9.5-mm steel outer housing. The graded liner comprises 1-mm-thick tin layer and 1.5-mm-thick copper layer to prevent interference by lead X-rays. To minimise the scattered radiation from the shield, the detector was centred in it. The detector was interfaced to the digital spectrum analyser (DSA-1000), which was a full-featured 16 K channel multichannel analyser on advanced digital signal processing (DSP) techniques. DSA-1000 operates through Genie-2000 gamma spectroscopy software including peak searching, peak evaluation, energy/efficiency calculation mode, nuclide identification, etc.

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 systems was carried out using the radionuclide specific efficiency method in which the efficiency values of gamma-ray lines belonging to the specific radionuclide existing in both the reference material and sample were only used. Thus, the uncertainty in gamma-ray intensities, the influence of coincidence summation and self-absorption effects of the emitting gamma photons were avoided for the close geometry conditions[20,21]. 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 same geometry was used to determine the peak

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area of samples and references. The counting time for each measurement was ranged between 30 000 and 40 000 s to obtain good counting statistics. Background measurements were taken under the same conditions of sample measurements and subtracted in order to get net counts for the sample.

The activity concentrations were averaged from gamma-ray photopeaks at several energies assuming the secular equilibrium between ^{226}Ra and ^{222}Rn and ^{232}Th and ^{228}Ra. The activity concentration of ^{226}Ra was derived from the weighted average of the activities of the gamma-ray line of 609.3 keV from ^{214}Bi and of two gamma-ray lines of 295.2 keV, 351.9 keV from ^{214}Pb, while the gamma-ray lines of 338.4 keV, the 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 obtained using its 1461 keV gamma-ray line.

The minimum detectable activity (MDA) of the gamma-ray measurements was calculated as follows[22]:

$$\mathrm{MDA}\ (\mathrm{Bq\,kg^{-1}}) = \frac{F_c \cdot \sigma_{NB}}{\varepsilon \times P \times t \times w}, \qquad (1)$$

where F_c is the statistical coverage factor equal to 1.64 (confidence level 95 %), σ_{NB} the standard deviation of the background in the region of interest and equals square root of the number of counts for the background spectrum, ε the absolute efficiency of the detector, P the absolute emission probability of the gamma decay, t the measurement time in seconds and w the weight of the dried sample expressed in kilograms. The mean values of MDA for ^{226}Ra, ^{232}Th and ^{40}K were estimated to be 0.8, 1.3 and 5.2 Bq kg^{-1} for gypsum and 0.7, 1.2 and 4.9 Bq kg^{-1} for limestone, respectively.

The total statistical uncertainty of the radioactivity measurements was calculated by taking into consideration the systematic uncertainties in the efficiency calibration and counting statistical uncertainty.

RESULTS AND DISCUSSION

Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K

The statistical data for the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the studied samples are shown in Table 1. It can be seen from Table 1 that the mean values of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the gypsum and limestone samples are significantly lower than the median values in the earth's crust (35 Bq kg^{-1} for ^{226}Ra, 30 Bq kg^{-1} for ^{232}Th and 400 Bq kg^{-1} for ^{40}K[23]) except the mean value of the activity concentration of ^{226}Ra measured in the limestone samples. This value is about half of the quoted median value. From Table 1, the values of skewness and kurtosis coefficients, which indicate whether the distributions are normal or log-normal for the activity concentrations of ^{226}Ra, ^{232}Th and

Table 1. The statistical data for the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}) measured in the samples examined.

Sample	n		^{226}Ra	^{232}Th	^{40}K
Gypsum	38	Minimum	0.8[a]	1.3[a]	9.0
		Maximum	36.8	18.2	188.0
		Mean	7.2	3.4	40.7
		Median	2.8	2.2	29.4
		Standard deviation	10	3.3	36.9
		SE	1.6	0.5	6.0
		Skewness	1.9	3.1	2.0
		Kurtosis	2.6	11.0	5.7
		Frequency distribution	Log-normal	Log-normal	Log-normal
Limestone	66	Minimum	0.7[a]	1.2[a]	10.1
		Maximum	55.1	20.9	258.4
		Mean	19	4.3	55
		Median	18.5	2.9	24.7
		Standard deviation	12.7	4.3	68.5
		SE	1.6	0.5	8.4
		Skewness	0.7	2.1	1.9
		Kurtosis	0.3	3.9	2.3
		Frequency distribution	Log-normal	Log-normal	Log-normal

SE, Standard error.
[a]MDA taken as the minimum value

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Table 2. Comparison of the mean values of activity concentrations measured in the present study for the samples examined with those obtained in other countries.

Raw materials	Country	Activity concentration (Bq kg^{-1})			References
		^{226}Ra	^{232}Th	^{40}K	
Gypsum	Bulgaria	13.3	5.3	39.5	Krstić *et al.*[7]
	EU	10.0	10.0	80.0	European Commission Report[26]
	India	8.3	—	26.7	Xinwei and Xiaolan[10]
	Macedonia	32.4	70.3	143.8	Krstić *et al.*[7]
	Pakistan	6.2	13.3	173.7	Khan and Khan[5]
	Syria	14.0	2.0	31.0	Othman and Mahrouka[8]
	Tanzania	5.0	—	—	Msaki and Banzi[6]
	Turkey	9.4	3.9	40.7	Present study
Limestone	Bangladesh	60.2	60.8	928.0	Alam *et al.*[4]
	Egypt	20.4	4.4	19.3	Sharaf *et al.*[27]
	China (Baoji)	24.2	20.2	113.2	Xinwei and Xiaolan[10]
	Greece	16.0	1.6	—	Papaefthymiou and Gouseti[9]
	India	73.9	—	64.6	Kumar *et al.*[13]
	Italy	13.1	6.0	13.5	Righi and Bruzzi[12]
	Pakistan	21.9	8.6	73.8	Khan and Khan[5]
	Syria	29.0	2.0	22.0	Othman and Mahrouka[8]
	Turkey	20.2	5.0	55.0	Present study

^{40}K are positive values indicating the non-existence of normal distribution. In Table 2, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the present study for gypsum and limestone are compared with the corresponding values measured in other countries. As can be seen in Table 2, the mean value of the activity concentration of ^{226}Ra obtained for the gypsum sample is comparable to those obtained from other studies except Macedonia while the mean value of the activity concentration of ^{226}Ra obtained for the limestone sample is comparable to those obtained from other studies except Bangladesh and India. The mean values of activity concentration of ^{232}Th and ^{40}K obtained for the gypsum samples are comparable to those obtained for the gypsum samples in Bulgaria and Syria. The mean value of activity concentration of ^{232}Th obtained for the limestone sample is comparable to those obtained for the limestone samples in Egypt and Italy while the mean value of activity concentration of ^{40}K obtained for the limestone sample is comparable to that obtained for the limestone samples in India.

Estimation of exposure risk

In this study, the radiological parameters such as indices of radium equivalent activity and internal hazard index, indoor absorbed gamma dose rate and annual effective dose were determined to estimate the exposure risk from usage of gypsum and limestone samples as raw materials in construction.

Equivalent activity index

A common radiological index has been used to compare the activity concentrations of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K and evaluate the radiation hazards associated with these radionuclides. This index is usually known as radium equivalent (Ra_{eq}) activity[24].

$$Ra_{eq} = 370\,\mathrm{Bq\,kg^{-1}} \cdot \left(\frac{A_{Ra}}{370\,\mathrm{Bq\,kg^{-1}}} + \frac{A_{Th}}{259\,\mathrm{Bq\,kg^{-1}}} + \frac{A_K}{4810\,\mathrm{Bq\,kg^{-1}}}\right), \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}. The results of the Ra_{eq} index calculated for all gypsum and limestone samples are summarised in the fourth column of Table 3. The mean Ra_{eq} index values are significantly lower than the recommended upper value of 370 Bq kg^{-1} for building materials, which is equivalent to an annual effective dose of 1.5 mSv y^{-1}[25].

Internal hazard 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[12]. In the present study, to address radiation hazard respiratory organs due to radioactive inert gas radon (^{222}Rn) and its short-lived secondary

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Table 3. The values of the indices of the radium equivalent activity (Ra_{eq}) and the internal hazard (H_{in}), the indoor absorbed gamma dose rate (D_R) and the corresponding annual effective dose (E_{eff}) determined for the samples examined.

Sample	n		Ra_{eq} (Bq kg^{-1})	H_{in}	D_R (nGy h^{-1})	E_{eff} (mSv)
Gypsum	38	Range	3.3–56.5	0.01–0.24	2.9–50.4	0.01–0.25
		Mean ± SE	15.1 ± 2.4	0.06 ± 0.01	13.6 ± 2.1	0.07 ± 0.01
Limestone	66	Range	3.2–81.6	0.01–0.35	2.8–73.5	0.01–0.36
		Mean ± SE	29.4 ± 2.1	0.13 ± 0.01	26.7 ± 1.9	0.13 ± 0.01

products the internal health index (H_{in}) was determined by the next formula[24]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}, \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}. For the safe use of materials in the construction of buildings, H_{in} should be less than unity. It is observed in the fifth column of Table 3 that the H_{in} values for all the studied samples are below the recommended upper level of unity.

Indoor absorbed gamma dose rate and annual effective dose

The absorbed dose rate in indoor air due to gamma-ray emission from the radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in the gypsum and limestone used as raw materials in the manufacture of building products such as cement, plaster and plasterboard was evaluated using data and formula provided by UNSCEAR Report[23] and European Commission Report[26]. The specific dose rates (0.92 nGy h^{-1} per Bq kg^{-1} for ^{226}Ra, 1.1 nGy h^{-1} per Bq kg^{-1} for ^{232}Th and 0.08 nGy h^{-1} per Bq kg^{-1} for ^{40}K) were calculated in a standard room with dimension of $4 \times 5 \times 2.8$ m^3, wall thickness of 20 cm and density of 2350 kg m^{-3}.

$$D_R(\text{nGy h}^{-1}) = 0.92 \times A_{Ra} + 1.10 \times A_{Th} + 0.08 \times A_K, \quad (4)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}.

In estimating the annual effective dose, 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. The annual exposure time used is 7012.7 h y^{-1}, implying that 80 % of time is spent indoors. The annual effective dose (in terms of mSv) was estimated using the following formula[28–29]:

$$E_{eff} = D_R \times 0.7\,\text{Sv Gy}^{-1}(\text{conversion factor}) \times 7012.7\,\text{h y}^{-1} \times 10^{-6}, \quad (5)$$

where D_R (nGy h^{-1}) is given by Eq. (4). The estimated results for the absorbed gamma dose rate (D_R) in indoor and the annual effective dose (E_{eff}) are given in the sixth and seventh column of Table 3 for the studied gypsum and limestone samples, respectively. According to the UNSCEAR Report[23], the corresponding worldwide average values for the indoor absorbed dose rate range from 20 to 200 nGy h^{-1} with a population-weighted average of 84 nGy h^{-1} for the absorbed gamma dose rate. The mean value of the D_R estimated for gypsum samples is about six times lower than the quoted population-weighted average value while the mean value of the D_R estimated for limestone samples is about three times lower than the quoted value. The mean values of E_{eff} estimated for all samples except that one limestone sample (0.36 mSv) are lower than the exemption dose criterion of 0.3 mSv.

CONCLUSION

For each sample in this study, the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, the indices of the radium equivalent activity (Ra_{eq}) index and internal health (H_{in}), the indoor absorbed gamma dose rate and the corresponding annual effective dose have been determined to assess the radiological hazards from the samples of the gypsum and limestone commonly used as building raw materials in Turkey. These indices have been used only as screening tools for ready-to-use building materials. The calculated values of Ra_{eq} and H_{in} for all the studied samples are significantly lower than the recommended upper levels. Turkey applies a strict criterion based on a dose in the range of 0.3–1 mSv y^{-1} for building materials used in bulk amounts, e.g. concrete. It is concluded that the values of the annual effective dose estimated for all the studied gypsum and limestone samples are below the criterion limit of the exemption corresponding to a dose rate of 0.3 mSv y^{-1}.

In regard to the results obtained in this study, the gypsum and limestone samples commonly used as raw materials in Turkey are within the recommended safety limit and do not pose any significant source of radiation hazard.

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