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RADIOLOGICAL SIGNIFICANCE OF CEMENT USED IN BUILDING CONSTRUCTION IN TURKEY

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The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in seven cement types from different factories and grinding plants were measured using a gamma ray spectrometry with HPGe detector. The average activity concentrations observed in the studied cement samples (all from 141 samples) were 40.0 ± 27.1, 28.0 ± 20.9 and 248.3 ± 95.0 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The radium equivalent activity (Ra_{eq}), the representative level index, the indoor absorbed dose rate and the corresponding annual effective dose were estimated for the potential radiological hazard of the cement. The Ra_{eq} values were compared with the corresponding values for cement of different countries. The mean indoor absorbed dose rate (87.4 ± 48.5 nGy h^{-1}) is slightly higher than the population-weighted average of 84 nGy h^{-1}, whereas the corresponding effective dose rate (0.4 ± 0.2 mSv y^{-1}) is lower than the dose criterion of 1 mSv y^{-1}. The obtained results indicate no significant radiological hazards arise from using Turkish cement in building construction.

INTRODUCTION

Raw and produced materials used in building sector such as cement, bricks, sand, tile, limestone, gypsum and others derived from rock and soil contain mainly natural radionuclides of the uranium (^{238}U) and thorium (^{232}Th) series, 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. The worldwide average concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the earth's crust are estimated as 50, 50 and 500 Bq kg^{-1}, respectively(1). Radiation exposure due to the building materials can be divided into external and internal exposures. The external exposure is caused by direct gamma radiation whereas the internal exposure is caused by the inhalation of radioactive inert gas radon (^{222}Rn, a daughter product of ^{226}Ra) and its short-lived secondary decay products. In order to assess the possible radiological hazards to human health, it is important to study the radioactivity levels emitted by the building materials. The data obtained from such a study is essential for the development of standards and guidelines for use and the management of these materials.

Cement that is the most important construction material for houses and buildings is considered as one of the basic industries that play an important role in the national economy of Turkey. Turkish Cement Industry is one of the largest of its kind in the world based on the capacity and annual production. Turkish cement is a manufactured product made by blending different raw materials such as lime, silica, clay, limestone, kaolin, marl, iron ore, etc., and firing them at a high temperature in order to achieve precise chemical proportions of lime, silica, alumina and iron in the finished product, known as a cement clinker (or Portland cement clinker). Turkish cement industry mainly produces Portland cement, which is a fine, typically grey powder produced by grinding Portland cement clinker (>90%), containing about the maximum of 5% gypsum, which controls the set time, and up to 5% minor constituents. Portland cement types I–V are the most common and standardised to TS EN 197-1(2) that defines and gives the specification of 27 distinct common cement products and their constituents.

There are few data available about the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Turkish cement. The objective of the present study was to determine the activity concentrations of radionuclides above mentioned in seven different types of cement (total of 141 samples) manufactured in Turkey, to estimate the indoor gamma dose rate and the corresponding the annual effective dose from samples and to assess any radiological hazard. The results were tabulated and compared with the world reported value for cement.

MATERIALS AND METHODS

Sample preparation

A total of 141 cement samples representing seven main types used in building construction in Turkey

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were collected from different cement factories and suppliers for the measurement of radionuclide activity concentrations. The cement samples are already in a powdered form, it could be used without any processing. The samples were dried in a temperature-controlled furnace at 110°C to remove moisture. The samples were packed in the cylindrical plastic containers having a volume of 118 cm^3 (5-cm diameter, 6-cm height), weighted and hermetically sealed. These samples were then stored for 4–5 weeks before counting to reach radioactive equilibrium ^{226}Ra and its short-lived decay products.

Radioactivity measurement

The measurements were made 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. (Meriden, CT, USA). The HPGe detector has a relative efficiency of 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-channels Multi-channel analyzer on advanced digital signal processing techniques.

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[3, 4]. 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 the peak area of samples and references. The accumulating time of both background and sample spectra 25 000 s each. Background measurements were taken and subtracted in order to obtained net counts for the sample. The activity concentrations were calculated based on the weighted mean value of their respective decay products in equilibrium. The gamma ray lines of 295.2, 351.9 keV from ^{214}Pb and the 609.3, 1764.5 keV from ^{214}Bi were used to determine the activity concentration of ^{226}Ra. The gamma ray lines of 338.4, the 911.2 keV from ^{228}Ac, the 727.3 keV from ^{212}Bi 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 minimum detectable activity (MDA) of the present measurement system was calculated as follows[5]:

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

where MDA is in Bq kg^{-1} (confidence level 95%), σ_{N_B} 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 MDA was estimated to be 0.4, 0.3 and 2.7 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively.

RESULTS AND DISCUSSION

The minimum, maximum and average activity concentrations of ^{226}Ra (C_{Ra}), ^{232}Th (C_{Th}) and ^{40}K (C_K) (in Bq kg^{-1}) together with the statistical uncertainty (1σ) and standard deviation (SD) are presented for different types of cement samples in Table 1. The specific radioactivity of ^{226}Ra, ^{232}Th and ^{40}K in the analysed cement samples range from 12.8 ± 0.3 to 165.4 ± 2.5 Bq kg^{-1} with an average of 40.0 ± 27.1 Bq kg^{-1}, 7.2 ± 0.3 to 137.0 ± 2.6 Bq kg^{-1} with an average of 28.0 ± 20.9 Bq kg^{-1} and 54.2 ± 2.9 to 573.1 ± 32.6 Bq kg^{-1} with an average of 248.3 ± 95.0 Bq kg^{-1}, respectively. The average concentration values are lower than the corresponding world mean values that are 50, 50 and 500 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively[1]. As can be seen from Table 1, the average activity concentration of ^{226}Ra and ^{232}Th are to be highest for CEM III (blastfurnace cement) and lowest for SDÇ (sulphate resistant cement). In the case of ^{40}K, the CEM IV (pozzolanic cement) samples showed the highest concentrations, whereas BPÇ (white cement) samples showed the lowest concentrations.

The obtained results indicate that the distribution of natural radionuclides in the cement samples is not uniform. Therefore, to compare the radioactivity concentrations of the cement samples containing Th, Ra and K, a common index is required to obtain the sum of radioactivities. This index is usually called as radium equivalent (Ra_{eq}) activity[6].

$$\mathrm{Ra_{eq}} = C_{Ra} + \left(\frac{10}{7}\right) \cdot C_{Th} + \left(\frac{10}{130}\right) \cdot C_K, \quad (2)$$

where C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}. In the definition of Ra_{eq}, it is assumed that 10 Bq kg^{-1} of ^{226}Ra, 7 Bq kg^{-1} of ^{232}Th and 130 Bq kg^{-1} of ^{40}K produce equal gamma ray dose rate[6–8]. The maximum value of Ra_{eq} in building raw materials

Table 1. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in seven cement types.

Main type[a]	n	Activity concentration (Bq kg^{-1}) ± statistical uncertainty								
		C_{Ra}			C_{Th}			C_K		
		Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD
CEM I	45	14.0 ± 0.4	59.8 ± 0.7	29.5 ± 12.4	7.2 ± 0.3	48.1 ± 1.1	18.9 ± 7.1	92.0 ± 4.9	423.6 ± 19.8	216.4 ± 65.8
CEM II	56	15.7 ± 0.4	139.0 ± 3.6	39.3 ± 22.7	8.3 ± 0.5	87.9 ± 1.3	29.3 ± 16.8	74.7 ± 3.4	469.2 ± 17.4	273.7 ± 95.0
CEM III	5	75.4 ± 1.0	165.4 ± 2.5	121.4 ± 33.6	45.3 ± 0.8	137.0 ± 2.6	90.5 ± 33.6	181.2 ± 6.9	252.5 ± 9.3	222.4 ± 25.9
CEM IV	11	17.9 ± 0.7	89.6 ± 4.1	36.2 ± 19.1	8.8 ± 0.4	112.9 ± 5.7	32.1 ± 28.5	237.6 ± 7.9	573.1 ± 32.6	359.6 ± 97.8
CEM V	10	19.8 ± 0.3	110.8 ± 3.0	73.5 ± 23.9	18.2 ± 0.4	77.6 ± 1.3	43.5 ± 15.3	236.0 ± 8.2	403.2 ± 13.5	298.0 ± 49.4
SDÇ	8	12.8 ± 0.3	37.6 ± 1.6	23.1 ± 9.2	10.5 ± 0.4	18.3 ± 0.9	14.6 ± 2.9	136.9 ± 5.4	233.8 ± 10.7	164.3 ± 34.1
BPÇ	6	26.8 ± 0.4	39.4 ± 0.7	31.8 ± 5.3	8.0 ± 0.3	25.5 ± 0.8	15.9 ± 7.8	54.2 ± 2.9	127.8 ± 6.4	95.8 ± 30.4
Total	141									
Mean ± SD		40.0 ± 27.1			28.0 ± 20.9			248.3 ± 95.0		

[a]CEM I, Portland cement; CEM II, Portland-composite cement; CEM III, blastfurnace cement; CEM IV, pozzolanic cement; CEM V, composite cement; SDÇ, sulphate resistant cement; BPÇ, white cement.

and products must be <370 Bq kg^{-1} for safe use, i.e. to keep to external dose <1.5 mSv y^{-1}[(9)].

The calculated Ra_{eq} values for the cement samples are given in Table 2. In all the cement samples, the Ra_{eq} values vary from 38.3 ± 5.4 to 350.6 ± 10.2 Bq kg^{-1} with an average of 99.1 ± 56.8 Bq kg^{-1}. It is observed that the Ra_{eq} values for all the studied samples are lower than the recommended maximum value 370 Bq kg^{-1}. Thus, these samples are within the recommended safety limit when used as building materials and products.

In Table 3, the average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and of the Ra_{eq} determined in the present study for cement are compared with the corresponding values determined in other different countries. It is observed that the Ra_{eq} values for cement samples originating from different countries show considerable variations, which are likely related to the type of raw materials used in cement manufacture. The comparison also shows that the calculated mean Ra_{eq} in this study is lower than that calculated in Australia, Bangladesh, Egypt, Brasil, China, Malaysia, Algeria, India and Greece and higher than that in Austria, Ireland, Italy, Pakistan, Tunisia and Netherlands.

Another criterion, known as the representative level index ($I_{\gamma r}$), used to estimate the level of gamma radiation hazard associated with the natural radionuclides in specific construction materials, is defined as[(9–11)]

$$I_{\gamma r} = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500}, \qquad (3)$$

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}. For the safe use of materials in the construction of buildings, $I_{\gamma r}$ should be less than unity. The calculated $I_{\gamma r}$ values for the studied cement samples are ranging from 0.3 to 2.4, with an average of 0.7 ± 0.4 (the last column of Table 2). It is clear that the mean $I_{\gamma r}$ values except for CEM III and CEM IV are all below the criterion limit. The mean $I_{\gamma r}$ value for CEM IV is slightly higher than the limit, while the mean $I_{\gamma r}$ value for CEM III is about two times higher than the quoted limit.

The absorbed dose rate in indoor air due to gamma ray emission from the radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in the cement used for a building material is evaluated using data and formula provided by UNSCEAR[(1, 12)] and EC[(13)]. In the UNSCEAR and the European Commissions reports, the dose conversion coefficients were calculated for the standard room centre. Dimensions of the room are 4 m × 5 m × 2.8 m. 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 ^{226}Ra, 1.1 nGy h^{-1}

Table 2. The radium equivalent (Ra_{eq}) activities and the representative level index ($I_{\gamma r}$) of seven cement types.

Main type[a]	N	Ra_{eq} (Bq kg^{-1})		$I_{\gamma r}$	
		Range	Mean ± SD	Range	Mean ± SD
CEM I	45	41.0 ± 5.5–148.3 ± 9.6	73.1 ± 20.6	0.3–1.1	0.5 ± 0.1
CEM II	56	46.9 ± 5.4–269.1 ± 14.8	102.2 ± 47.3	0.3–1.9	0.7 ± 0.3
CEM III	5	211.9 ± 8.0–350.6 ± 10.2	267.8 ± 67.8	1.5–2.4	1.9 ± 0.5
CEM IV	11	53.9 ± 14.5–295.0 ± 33.3	109.7 ± 64.5	0.4–2.1	0.8 ± 0.5
CEM V	10	64.0 ± 8.2–206.7 ± 8.5	158.5 ± 40.2	0.5–1.5	1.1 ± 0.3
SDÇ	8	38.3 ± 5.4–74.0 ± 9.2	56.6 ± 11.7	0.3–0.5	0.4 ± 0.1
BPÇ	6	48.0 ± 4.5–85.0 ± 4.8	61.9 ± 15.3	0.3–0.6	0.4 ± 0.1
Total	141				
Mean ± SD		99.1 ± 56.8		0.7 ± 0.4	

[a]CEM I, Portland cement; CEM II, Portland-composite cement; CEM III, blastfurnace cement; CEM IV, pozzolanic cement; CEM V, composite cement; SDÇ, sulphate resistant cement; BPÇ, white cement.

Table 3. Comparison of the average activity concentrations the radium equivalent activities (Ra_{eq}) of Turkish cement samples with those obtained in other published data.

Country	N	Specific radioactivity (Bq·Kg^{-1})			Ra_{eq} (Bq kg^{-1})	References
		C_{Ra}	C_{Th}	C_K		
Australia	7	51.5	48.1	114.7	129.4	Beretka and Mathew[6]
Austria	18	26.7	14.2	210	63.1	Sorantin and Steger[15]
Algeria	12	41	27	422	112	Amrani and Tahtat[16]
Bangladesh	3	120.2	132.4	505.7	348.3	Mollah *et al.*[17]
	18	62.3	59.4	328.9	172.8	Chowdhury *et al.*[18]
	4	29.7	54.3	523	148	Alam *et al.*[10]
	27	61.1	79.9	1132.6	–	Roy *et al.*[19]
Brasil	1	61.7	58.5	564.0	188.8	Malanca *et al.*[20]
China	–	69.3	62.0	169.0	189.0	Zigiang *et al.*[21]
	46	56.5	36.5	173.2	122.0	Xinwei[22]
Egypt	85	78	33	337	151	El Afifi *et al.*[23]
Finland	11	40.2	19.9	251	–	Mustonen[24]
Greece	22	92	31	310	160	Stoulos *et al.*[4]
	20	62.8	23.8	284.1	118.6	Papastefanou *et al.*[25]
India	1	37.0	24.1	432.2	104.7	Kumar *et al.*[26]
Ireland	–	66	11	130	86	Lee *et al.*[27]
Italy	7	38	22	218	92	Rizzo *et al.*[28]
Japan	16	35.8	20.7	139.4	–	Suzuki *et al.*[29]
Malaysia	–	81.4	59.2	203	181	Chong and Ahmed[30]
Netherlands	6	27	19	230	71.9	Ackers *et al.*[31]
Pakistan	25	26.1	28.6	272.9	87.9	Khan and Khan[32]
Tunisia	2	21.5	10.10	175.5	49.7	Hizem *et al.*[33]
Turkey	145	40.0	28.0	248.3	99.1	Present study

per Bq kg^{-1} for ^{232}Th and 0.080 nGy h^{-1} per Bq kg^{-1} for ^{40}K.

$$D(\text{nGyh}^{-1}) = 0.92 \times C_{Ra} + 1.1 \times C_{Th} + 0.080 \times C_K, \quad (4)$$

where C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}.

To estimate the annual effective dose, one has to take into account the conversion factor from absorbed dose in air to effective dose and the indoor occupancy factor. In the recent UNSCEAR[1, 12] reports, 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 time is spent indoors, on average, around the world. The effective dose rate in units of mSv per year was

Table 4. The indoor-absorbed gamma dose rate and the corresponding effective dose rate from seven cement types.

Main type[a]	*n*	D (nGy h^{-1})		H (mSv y^{-1})	
		Range	Mean ± SD	Range	Mean ± SD
CEM I	45	36.8–129.0	65.2 ± 18.1	0.2–0.6	0.3 ± 0.1
CEM II	56	41.4–237.0	90.3 ± 40.8	0.2–1.2	0.4 ± 0.2
CEM III	5	179.2–295.5	229.0 ± 56.9	0.9–1.5	1.1 ± 0.3
CEM IV	11	49.5–259.5	97.3 ± 54.0	0.2–1.2	0.5 ± 0.3
CEM V	10	57.1–175.9	139.3 ± 34.6	0.3–0.9	0.7 ± 0.2
SDÇ	8	34.3–65.7	50.4 ± 10.4	0.2–0.3	0.2 ± 0.1
BPÇ	6	43.4–73.8	54.4 ± 12.4	0.2–0.4	0.3 ± 0.1
Total	14				
Mean ± SD		87.4 ± 48.5		0.4 ± 0.2	

[a]CEM I, Portland cement; CEM II, Portland-composite cement; CEM III, blastfurnace cement; CEM IV, pozzolanic cement; CEM V, composite cement; SDÇ, sulphate resistant cement; BPÇ, white cement.

estimated using the following formula:

$$H = D \times 8766\text{h} \times 0.8(\text{occupancy factor}) \times 0.7\text{SvGy}^{-1}(\text{conversion factor}) \times 10^{-6}, \quad (5)$$

where D (nGy h^{-1}) is given by equation (4). The estimated results for the indoor absorbed dose rate and the corresponding annual effective dose rate are given in Table 4. The indoor absorbed dose rates ranging from 34.3 to 295.5 nGy h^{-1} with a mean of 87.4 ± 48.5 nGy h^{-1}. The mean indoor absorbed dose rate from all cement samples is slightly higher than the world average (populated-weighted) indoor absorbed gamma dose rate of 84 nGy h^{-1}. From the data in Table 4, the average D values for CEM I, SDÇ and BPÇ are lower than the world average, whereas the average D values for CEM II, CEM III, CEM IV and CEM V exceed the quoted value. The effective dose rates ranging from 0.2 to 1.5 mSv y^{-1} with a mean of 0.4 ± 0.2 mSv y^{-1}. It is noted that the average effective dose rate H is about half of dose criterion of 1 mSv y^{-1}[13, 14]. The mean H values except for CEM III are all below the criterion limit. The mean H value for CEM III is slightly higher than the limit.

CONCLUSIONS

For each sample in this study, the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, the radium equivalent activity, the representative level index, the absorbed gamma dose rate in indoor air and the corresponding effective dose rate were determined to assess the radiological hazards from Turkish cement. It was considered only the external exposure due to gamma radiation emitted by the cement samples. The results indicated that there are considerable variations in the measured values of the cement samples originating from different areas. The results showed that the CEM III cement contains the highest level of the natural radioactivity, particularly ^{226}Ra and ^{232}Th, whereas the SDÇ cement contains the lowest level. The average concentrations of ^{226}Ra and ^{232}Th in CEM I (Portland cement), SDÇ (sulphate resistant cement) and BPÇ (white cement) are more or less similar. The mean Ra_{eq} values are all lower than the recommended maximum level of radium equivalent of 370 Bq kg^{-1} for building raw materials and products. The estimated mean indoor absorbed dose rate is slightly higher than the population-weighted average of 84 nGy h^{-1}. The corresponding effective dose rate from CEM I, CEM II, CEM IV, CEM V, SDÇ and BPÇ are lower than the dose criterion of 1 mSv y^{-1}, whereas CEM III is slightly higher than quoted value. On the basis of the radium equivalent, the representative level index, the indoor absorbed dose rate and the annual effective dose all the cement samples considered do not pose any significant source of radiation hazard and the use of the cement samples in construction of dwellings is considered to be safe for inhabitants.

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