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ARTICLE

Natural Radioactivity and Associated Radiation Hazards in Local Portland and Pozzolanic Cements Used in Jordan

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Abstract: Activity concentration of the natural gamma-emitting radionuclides (^{40}K, ^{226}Ra and ^{232}Th) in at least forty samples of local Portland and Pozzolanic cement types is measured. The measurements were performed using gamma spectrometric techniques. The range of the mean specific activity (minimum and maximum values) due to all the three radionuclides is found. Radiological hazards of the different samples are estimated using five approaches; the representative level index, the external hazard index, the internal hazard index, the radium equivalent index and the absorbed dose rate. Some of the measured radiological hazard parameters are compared to similar parameters in different countries. The activity concentration of ^{226}Ra for all types of cements varies from 29.1±2.01 to 79.2±3.90 Bq.kg^{-1}. The activity concentration of ^{232}Th for all types of cements varies from 5.8±1.1 to 26.4±1.9 Bq.kg^{-1}. The activity concentration of ^{40}K for all types of cements varies from 231.9±9.8 to 298.0±10.7 Bq.kg^{-1}. The radium equivalent activity concentration, Ra_{eq}, of the total activity of each cement type is obtained. The highest value of Ra_{eq} was seen in white cement (125.49 Bq.kg^{-1}) and the lowest in Portland Pozzolanic cement (5%) with an average value of (78.08 Bq.kg^{-1}). Based on the assessment of potential radiological hazards as inferred from the calculations of Ra_{eq}, representative level index and the dose rate, the investigated cement samples fall within the category of accepted building materials and are safe to use for the construction of inhabited buildings.
Keywords: Radiological hazards, Portland cement, Pozzolanic cement.

Introduction and Objectives

The global demand for cement as a building material is considerable. Cement is an important construction material of houses and buildings built in urban areas in Jordan. Portland cement is the most common type of cement used in construction applications, but it is an expensive binder due to the high cost of production associated with the high energy requirements of the manufacturing process itself [1]. Other cheap inorganic materials with cementitious properties, such as natural pozzolans, e.g. volcanic tuff [2, 3] and clay [4], as well as waste products from industrial plants, e.g. slag [5], fly ash [6, 7] and silica fume [8], can be used as partial replacements for Portland cement; i.e., blended cements [9]. In addition, to reduce the cost of binder, there are potential technological benefits from the use of pozzolanic materials as those blended with Portland cement in concrete applications. These include increased workability, decreased permeability [10], increased resistance to sulphate attack [11], improved resistance to thermal cracking and increased ultimate strength and durability of concrete [12-14].

The first objective of the present work is to measure the naturally occurring radioactive elements in the cement used as a building material in Jordan, since workers are exposed to radiation for a long time, especially in mines and

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at manufacturing sites, in addition the exposure of people, who spend about 80% of their time inside offices and homes [13-15], resulting in exposure to cement or its raw materials as a necessary reality.

The content, of ^{226}Ra, ^{232}Th and ^{40}K in all types of cement can vary considerably, depending on their geological source and geochemical characteristics. The knowledge of radioactivity in these materials is important to estimate the radiological health impact on humans [16]. The radiological effect from natural radioactivity is due to radiation exposure of humans to gamma radiation and irradiation of lungs from inhalation of radon and its progenies. Thus, it is necessary to evaluate the dose limit of public exposure [17]. The external radiation exposure is caused by gamma radiation originating from members of the uranium and thorium decay chains and from potassium ^{40}K. However, the internal radiation exposure, mainly affecting the respiratory tract, is due to the short-lived radon and its daughters' products, which are emitted from construction materials into room air. The second objective of the present work is to calculate the radiological parameters, such as: the representative level index, the external hazard index, the internal hazard index, the radium equivalent concentration, Ra_{eq} and the absorbed dose rate, which are related to the external gamma-dose rate, and their effects on human health. The results of concentration levels and radiation equivalent activities are compared with similar studies carried out in other countries.

Materials and Methods

Preparation of Samples

Twenty five samples of all cement types produced by Cement Jordanian Factory (Portland cement, Portland Pozzolanic cement (5%), Portland Pozzolanic cement (25%), White cement and Sulphate Resistant Cement (S.R.C.)) were collected for this study [18, 19]. The percentage values in parentheses above indicate the percent of Pozzolana in cement. For comparison with products from other factories, 8 samples were taken from the ordinary Portland cement from (Arabia Company, Ashamaliya Company and Al-rajhi Company), 4 samples were taken of Portland Pozzolanic cement (25%) from (Arabia Company and Al-rajhi Company) and 4 samples were taken of (S.R.C.) from (Arabia Company, Ashamaliya Company and Al-rajhi Company).

Each sample, 1kg in weight, was dried in an oven at about 110 ºC to ensure that moisture is completely removed. The samples were crushed, homogenized and sieved through a 200 mesh, which is the optimum size to be enriched in heavy minerals. Weighed samples were placed in a polyethylene beaker of 350-cm^3 volume. The beakers were completely sealed for 4 weeks to reach secular equilibrium, where the rate of decay of the radon daughters becomes equal to that of the parent. This step is necessary to ensure that radon gas is confined within the volume and that the daughters will also remain in the sample.

Instrumentation and Calibration

Measurements were performed using a High Purity Germanium (HPGe) detector supplied by EG&G Ortec. The detector is an n-type gamma-X-ray (GMX) detector, operated at 3500 V, with a useful energy range from 3 keV to 10 MeV, a standard energy resolution of 2.02 keV and a relative efficiency of 56.9% at1.33 MeV of ^{60}Co. The absolute efficiency calibration of the detector was performed using the IAEA standard “soil-6” source within a Petri-dish, 90 mm in diameter and 10 mm thick. Its spectrum was collected for 12 h. Areas under the energy peaks of interest were used for drawing the peak efficiency curve between log of efficiency *versus* log of peak energy. A polynomial was fitted to the curve and the result was stored for further use. Under the assumption that secular equilibrium was reached between ^{226} Ra and its short-lived daughters, gamma ray transitions to measure concentrations of the assigned nuclides in the series are as follows: ^{238}U activities in the samples under investigation were derived from weighted means of the photopeaks of ^{234}Th (63.3, 92.4 and 92.8 keV). ^{226}Ra activity was determined by taking the mean activity of the three separate photopeaks of its daughter nuclides: ^{214}Pb at (295.2 and 352.0 keV) and ^{214}Bi at (609.3 keV). For ^{232}Th determination, the photopeak of ^{228}Ac (at 911.1 keV) and the photopeaks of ^{212}Pb (at 583.1 keV) and ^{208}Tl (at 238.6 keV) were used. ^{40}K was directly determined using the 1460.8 keV photopeak.

The activity concentration (A) in Bq.kg^{-1} in the environmental samples was obtained by the following equation:

$$A = \frac{Np}{e \times E \times m} \quad (1)$$

where N_p is the difference between counts per second of the sample and counts per second of the background, e is the abundance of the γ-peak in a radionuclide, E is the measured efficiency for each gamma-ray peak observed for the same number of channels either for the sample or the calibration source and m is the sample mass in kilograms.

The counting system must have a background as low as attainable with a minimum number of spectral lines originating from natural radionuclides which may be present in the system components and in the surrounding environment of the counting facility. In the present study, measurements of the background count rates for natural radionuclides were carried out at least twice a week, each for a counting time of 80,000 s, and its spectrum was stored in a PC-based multichannel analyzer (MCA). After counting for the specified time, the gamma-ray spectra were automatically calculated and loaded in the PC-based MCA.

Absorbed Gamma Dose Rate, D (nGy/h)

The absorbed dose rates due to gamma radiations in air at 1 m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated based on guidelines provided by *UNSCEAR* (2000). The conversion factors used to compute absorbed gamma dose rate (D) in air per unit activity concentration in Bq.kg^{-1} (dry weight) correspond to 0.462 nGy/h for ^{226}Ra, 0.604 nGy/h for ^{232}Th and 0.042 nGy/h for ^{40}K. Therefore, D can be calculated as follows [2]:

$$D\ (nGy/h) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_k \quad (2)$$

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

Radium Equivalent Index, Ra_{eq}

In comparing the radioactivity of materials that contain ^{226}Ra, ^{232}Th and ^{40}K, a common radium equivalent activity is required to obtain the total activity and is also used to assess the gamma radiation health impact on the public. Since 98% of the radiological effects of the uranium series are produced by radium and its daughter products, the contribution from the ^{238}U and the other ^{226}Ra precursors is usually ignored, so that the Ra_{eq} of a sample can be expressed as [4]:

$$Ra_{eq}\ (Bq.kg^{-1}) = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (3)$$

where C_{Ra}, C_{Th} and C_K are the specific activity values in Bq.kg^{-1}. This equation is based on the assumption that 10 Bq.kg^{-1} of ^{226}Ra, 7 Bq.kg^{-1} of ^{232}Th and 130 Bq.kg^{-1} of ^{40}K produce the same γ-radiation dose rate. The radium equivalent activity of the mean activity of the samples calculated on the basis of the aforementioned relation.

Representative Level Index (Iγr)

Another radiation hazard index was primarily used to estimate the level of γ radiation associated with different concentrations of some specified radionuclides. It is defined as shown in the following formula [16, 20]:

$$RLI(I_{\gamma r})\ (Bq.Kg^{-1}) = (1/150)\ A_{Ra} + (1/100)\ A_{Th} + (1/1500)\ A_K \quad (4)$$

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

External Hazard Index, H_{ex}

In the literature, a number of criterion formulae have been derived over the years to assess the radiation dose rate due to exposure to gamma radiation from the natural radionuclides contained in building materials. The merits of these have been reviewed by the OECD's Nuclear Energy Agency (1979).

Karpov and Krisiuk (1980) have proposed a relation for the activity concentrations that limits the annual gamma dose rate inside a room owing to the building material to about 1 mSv. Also, Krieger (1981) proposed the following conservative model based on infinitely thick walls without windows and doors to serve as a criterion for the calculation of external hazard index, H_{ex}, defined as:

$$H_{ex} = (C_{Ra}/370) + (C_{Th}/260) + (C_K/4810) \leq 1 \quad (5)$$

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

Internal Hazard Index. H_{in}

In addition to the external irradiation, radon and its short-lived products are also hazardous to the respiratory organs. The internal hazard index H_{in} is used to control the internal exposure to

^{222}Rn and its radioactive progeny. The internal exposure to radon and its daughter products is quantified by the internal hazard index H_{in}, which is given by the following equation (Krieger, 1981):

$$H_{in} = (C_{Ra}/185) + (C_{Th}/260) + (C_K/4810) \leq 1 \quad (6)$$

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

Results and discussion

The distribution of natural radionuclides in different brands of cements is presented in Table 1. It can be seen from Table 1 that the activity concentration of ^{226}Ra varies from 29.1±2.01 to 79.2±3.90 $Bq.kg^{-1}$. The activity concentration of ^{232}Th varies from 5.8±1.1 to 26.4±1.9 $Bq.kg^{-1}$. The activity concentration of ^{40}K varies from 231.9±9.8 to 298.0±10.7. The mean ^{226}Ra and ^{232}Th values are slightly higher than the corresponding worldwide average values which are 35 and 30 $Bq.kg^{-1}$, whereas ^{40}K values are lower than the corresponding worldwide average (400 $Bq.kg^{-1}$) as Table 3 shows. Fig. 1 shows the activity concentrations of natural radionuclides for the cement types. Since the distribution of the natural radionuclides in each cement type is not uniform, a common index termed radium equivalent activity (Ra_{eq}) is required to obtain the total activity and is also used to assess the gamma radiation hazards. The radium equivalent of the total activity of each cement type is shown in Table 2. The highest value of Ra_{eq} is seen with white cement (125.49±6.12 $Bq.kg^{-1}$) and the lowest with Portland Pozzolanic cement (5%) with an average value of (78.08±4.03 $Bq.kg^{-1}$).

TABLE 1. The minimum, maximum and mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for Cement Jordanian Factory products.

Types of Cement	Specific γ- ray activity concentrations ($Bq.kg^{-1}$)								
	^{226}Ra			^{232}Th			^{40}K		
	Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.	Mean
Portland Cement	40.3	75.6	66.5	9.7	23.9	18.2	150.5	233.8	227.2
Portland Pozzolanic Cement (5%)	31.2	60.1	43.9	9.1	17.1	11.4	200.7	287.0	232.3
Portland Pozzolanic Cement (25%)	39.9	56.3	49.1	7.9	17.0	12.0	193.3	291.9	221.5
Sulphate Resistant Cement (S.R.C.)	29.1	42.0	42.9	5.8	11.3	10.9	231.9	298.0	265.6
White Cement	56.2	79.2	77.2	17.9	26.4	22.9	178.6	219.0	201.9

TABLE 2. Radium equivalent activity (Ra_{eq}), representative level index ($I_{\gamma r}$), gamma dose rate (D), external hazard index (H_{ex}) and internal hazard index (H_{in}), for different brands of cement.

Types of Cement	D ($nGy.h^{-1}$)	Ra_{eq} ($Bq.kg^{-1}$)	$I_{\gamma r}$ ($Bq.kg^{-1}$)	H_{ex}	H_{in}
Portland Cement	51.19	110.02	0.776	0.296	0.476
Portland Pozzolanic Cement(5%)	36.69	78.08	0.559	0.211	0.329
Portland Pozzolanic Cement (25%)	39.01	83.32	0.595	0.225	0.357
Sulphate Resistance Cement (S.R.C.)	37.29	78.93	0.572	0.213	0.329
White Cement	57.91	125.49	0.878	0.339	0.547

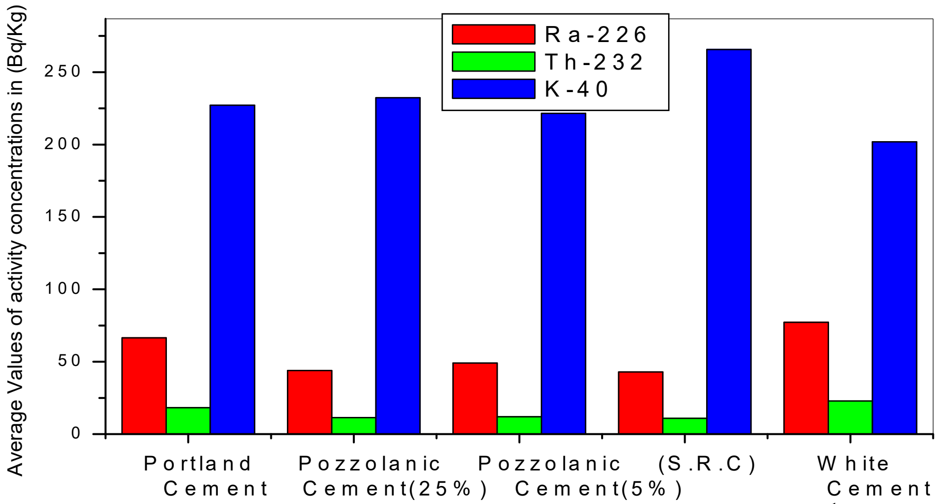

FIG. 1. Different kinds of cements *vs*. activity concentrations ($Bq.kg^{-1}$).

However, all the values obtained in this study for Ra_{eq} fall far below the criterion limit, as the use of materials whose Ra_{eq} concentration exceeds (370 $Bq.kg^{-1}$) is prohibited. It is apparent that the Ra_{eq} of cement samples originating from different types shows considerable variations, which are likely related to the type of raw materials used in cement manufacture. This is important in selecting the suitable cement type for use in building and construction, especially from among those which have large variations in their activities. Fig. 2 shows the variations of radium equivalent activities with the cement types in Jordan.

The radiation hazard indices are primarily used to estimate the level of γ-radiation associated with different concentrations of some specified radionuclides. The representative level index values, the external hazard index values, the internal hazard index values, as estimated using equations (4, 5 and 6) for all types of cements are listed in Table 2. All values are relatively similar compared to each other with the highest value in the representative level index (0.878 $Bq.kg^{-1}$) for white cement as can be seen from Fig. 3. This would tend to confirm that the samples under investigation exhibit a very low gamma radiation level. The obtained results for the products show that the averages of radiation hazard parameters for all products under investigation are lower than the acceptable level, 370 $Bq.kg^{-1}$ for Ra_{eq}, 1 for level index $I_{\gamma r}$ and 59 $nGy.h^{-1}$ for absorbed dose rate, as shown in Table 2.

Table 3 lists the comparison of activity concentrations in Portland cements in different areas of the world. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for all measured samples of Portland cement are comparable with the corresponding values and sometimes less than those of other countries. The radioactivity in Portland cement varies from one country to another because of different materials used in cement manufacture.

Fig. 2 shows a comparison between different kinds of cements used in Jordan in terms of the average values of Ra_{eq} and dose rate. It can be seen that white cement has the greatest values of Ra_{eq} and dose rate among all types of cements, while Pozzolanic cement with higher Pozzolan substitutions has the smallest values.

Another comparison can be noticed from Fig. 3 between different hazard indices among all kinds of cement. The highest values of representative level index $I_{\gamma r}$ and internal hazard index H_{in} are associated with Portland and white cement. The obtained results show that the averages of radiation hazard parameters for all kinds of cements are lower than the acceptable levels.

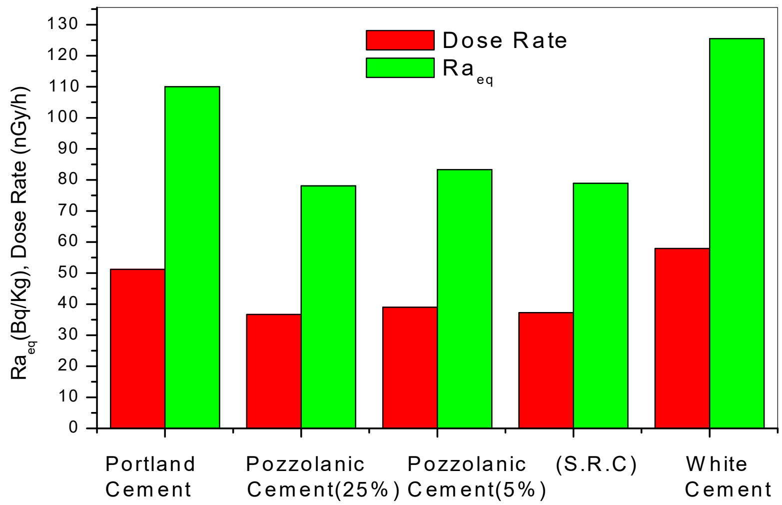

FIG. 2. Comparison between the average values of radium equivalent, Ra_{eq}, and dose rate in different kinds of cement in Jordan.

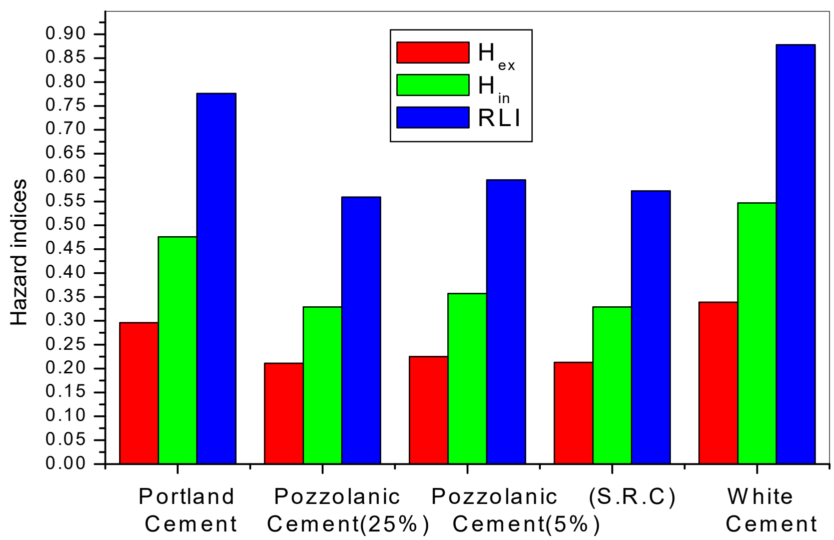

FIG. 3. Comparison between different hazard indices in different kinds of cement in Jordan.

TABLE 3. Comparison between the activity concentrations of Portland cement samples from Cement Jordanian Factory with those of other countries.

Country	Activity Concentration ($Bq.kg^{-1}$)			Reference
	^{226}Ra	^{232}Th	^{40}K	
Finland	44.0	26.0	241.0	NEA-OECD (1979)
Brazil	61.7	58.5	564.0	Malanca et al. (1993)
Pakistan	31.3	26.8	212.0	Tufail et al. (2007)
Cuba	23.0	11.0	467.0	Brigido Flores et al. (2008)
Sweden	96.0	127	962.0	NEA-OECD (1979)
Australia	51.8	48.1	115.0	Beretka and Matthew (1985)
Austria	26.1	14.2	210.0	Sorantin and Steger (1984)
China	69.3	62.0	169.0	Ziqiang et al. (1988)
Germany	26.0	18.0	241.0	NEA-OECD (1979)
United Kingdom	22.0	7.00	141.0	NEA-OECD (1979)
Egypt	31.3	11.1	40.60	Sharaf et al. (1999)
Present work	66.5	18.2	27.20	---
World	35.0	30.0	400.0	UNSCEAR (2000)

Conclusion

Based on the assessment of potential radiological hazards as inferred from the calculations of radium equivalent activity, representative level index and dose rate, the investigated cement samples fall within the category of accepted building materials and are safe to use for the construction of inhabited buildings.

The results may be important from the point of view of selecting suitable materials for use in cement manufacture. Cement products do not pose a significant radiological hazard when used for building construction.

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References

[1] Pakou, A., Assimakopoulos, P. and Prapidis, M., Sci. Total Environ., 144 (1994) 255.

[2] UNSCEAR, Sources and effects of ionizing radiation. Report to General Assembly with Scientific Annexes, United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations, New York (2000).

[3] Ackers, J., Den-Boer, J., de-Jong, P. and Wolschrijn, R., Sci. Total Environ., 45 (1985) 151.

[4] Beretka, J. and Mathew P., Health Phys., 48 (1) (1985) 87.

[5] Khan, K., Khan, H., Tufail, M. and Ahmad, N., J. Environ. Radioac., 38 (1998) 77.

[6] Tufail, M., Ahmad, N., Mirza, S., Mirza, N. and Khan, H., Sci. Total Environ., 121 (1992) 283.

[7] Tzortzis, M., Tsertos, H., Christofides, S. and Christodoulides, G., Radiat. Meas., 37 (2003) 221.

[8] Ziqiang, P., Yin, Y. and Mingqiang, G., Radiat. Prot. Dosim., 24 (1988) 29.

[9] Projin, A., Bourgoignie, R., Marjins, R., Janssens, A. and Jacobs, R., Radiat. Protect. Dosim., 7 (1984) 77.

[10] Sciocchetti, G., Scacco, F. and Baldassini, P., Radiat. Protect. Dosim., 7 (1984) 347.

[11] Malanca, A., Pessina, V. and Dallara, G., Radiat. Protect. Dosim., 24 (1993) 27.

[12] Mollah, A., Ahmed, G., Hussain, S. and Rahman, M., Health Phys., 50 (1986) 849.

[13] Mustonen, R., Sci. Total Environ., 45 (1986) 127.

[14] Paredes, C., Kessler, W., Landolt, R., Ziemer, P. and Panstenbach, D., Health Phys., 53 (1987) 23.

[15] Alam, M., Chowdhury, M., Kamal, M., Ghose, S., and Ismail, M., J. Environ. Radioact., 46 (1999) 243.

[16] Stranden, E. and Berteiz, L., Health Phys., 39 (1980) 275.

[17] Sideris, K., Savva, A. and Papayianni, J., Cem. and Conc. Comp., 28 (2006) 47.

[18] Massazza, F., J. Turkish Cement Manufacturers' Association. Cem. and Conc. World, 3 (1999) 19.

[19] NEA-OECD, Nuclear Energy Agency. Exposure to Radiation from Natural Radioactivity in Building Materials. Report by NEA Group of Experts, OECD, Paris (1979).

[20] Karpov, V. and Krisiuk E., Health Phys., 39 (1980) 819.

[21] Krieger, R., Betonwerk Fertigteil Technol., 47 (1981) 468.