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An Investigation on the Natural Radioactivity of Building Materials, Raw Materials and Interior Coatings in Central Turkey*

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Aim: The aim of this investigation was to measure the amount of γ-decay of several building materials (brick and cement), raw materials (fly-ash and soil), interior coating materials (foam [used as a thermal insulator], water-based paints, solvent-based paints and phosphogypsum) from central Turkey, in terms of Bq kg^{-1}, and to calculate the biological damage caused by this radioactivity.

Materials and Methods: Gamma-spectrometry technique has been used throughout the research in order to determine the activity of natural radionuclides. Later, annual doses were calculated.

Results: The average radioactivity values were 632.2 Bq $kg^{-1}$for fly-ash, 4.4 Bq kg^{-1} for brick, 73.3 Bq kg^{-1} for soil, 306.6 Bq kg^{-1} for cement, 302.3 Bq kg^{-1} for phosphogypsum and 83.6 Bq kg^{-1} for solvent-based paint in the research region. The activity of water-based paint and foam were below MDA[1]. The annual effective doses ranged between 0.009 and 1.479 mSv y^{-1}.

Conclusions: These results show that annual dose absorbed by inhabitants from construction materials used in central Turkey per kilogram is below 1.0 mSv y^{-1}. However, the dose from fly-ash, a component of cement, is generally over this value. Moreover, H_{in}[2] of phosphogypsum is over the limit, which means it is harmful for the respiratory system. As a result, the issue needs more research, taking into account the density, thickness of walls and percent contribution of fly-ash used in cement and also taking more samples.

Key Words: Gamma-Irradiation, natural radioactivity, building materials, biological damage, annual effective dose

Orta Türkiye'de Kullanılan İnşaat Malzemeleri, Hammaddeler ve İç kaplama Malzemelerindeki Doğal Radyoaktivite Miktarı Üzerine Bir İnceleme

Amaç: Bu çalışmanın amacı orta Türkiye'de kullanılan inşaat malzemeleri (tuğla ve çimento), hammaddeleri (uçucu baca külü ve toprak) ve iç kaplama malzemerindeki (strafor köpük (yalıtım malzemesi olarak),su bazlı boya, solvent bazlı boya ve alçı) gama ışıması miktarının Bq kg^{-1} biriminde ölçülmesi ve bu radyasyon tarafından oluşturulan biyolojik hasarın hesaplanmasıdır.

Yöntem ve Gereç: Çalışma boyunca doğal radyoaktif izotopların aktivitesini belirlemek için gama spektrometrisi tekniği kullanılmıştır. Daha sonra yıllık dozlar hesaplanmıştır.

Bulgular: Çalışılan bölgede ortalama radyoaktivite değerleri uçucu kül için 632.2 Bq kg^{-1}, tuğla için 4.4 Bq kg^{-1}, toprak için 73.3 Bq kg^{-1}, çimento için 306.6 Bq kg^{-1}, alçı için 302.3 Bq kg^{-1} ve solvent bazlı boya için 83.6 Bq kg^{-1} dır. Su bazlı boya ve strafor köpük için aktivite değerleri MDA'nın[3] altındaydı. Yıllık efektif dozlar 0.009 ve 1.479 mSv y^{-1} arasında değişmektedir.

Sonuç: Sonuçlar orta Türkiye'de kullanılan inşaat malzemelerinin yıllık dozunun 1.0 mSv y^{-1} nin altında olduğunu gösteriyor; ancak çimentonun bir bileşeni olan uçucu kül için bu değer genellikle 1.0 mSv y^{-1} nin üzerinde. Ayrıca alçının H_{in}[4] değeri sınırın üzerinde. Bu da alçının solunum sistemi üzerinde zararlı etkileri olduğu anlamına gelmektedir. Sonuç olarak, konunun, duvar yoğunluğu, kalınlığı ve çimentoda kullanılan uçucu külün yüzdelik miktarı dikkate alınarak ve daha çok örnek alarak incelenmesi gerekmektedir.

Anahtar Sözcükler: Gama ışıması, doğal radyoaktivite, inşaat malzemeleri, biyolojik hasar, yıllık efektif doz.

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1 Minimum detectable activity,
2 Internal hazard index
3 Tespit edilebilir en düşük aktivite
4 Dahili hasar indeksi

Introduction

Life on earth has developed under the ubiquitous presence of environmental gamma and charged-particle radiation. Radiation may well be one of the conditions for life and biological development. It is, however, also well established that ionizing radiation may harm life and biological systems (1).

Studies of natural radiation background are of great importance because it is the main source of exposure to humankind (2). Naturally occurring raw building materials and processed products have radionuclides of the three most commonly known radioactive series: Uranium-radium series, thorium series and ^{40}K isotopes (3). High concentrations of natural radionuclides in building materials can result in high-dose rates indoors (4). Gamma-irradiation from naturally occurring radioactive material (NORM) contributes to the whole body dose and in some cases β-irradiation contributes to the skin dose. As β-particles have higher specific ionization (SI) than γ-particles, they lose their energy while they are still in the skin and cannot penetrate further into the body. Apart from β and γ radiations, there are other harmful effects of NORM. One is the toxicity of uranium and radium and the other is the hazardous effects of ^{222}Radon on lungs (4). Seven to nineteen percent of radon originates from building materials (5). Especially the concentration of fly-ash (one of the constituents of cement together with sand and gravel) has the most critical role in the radon release process (6). Moreover, it has been shown that exposure to fly-ash from powder coal combustion implies a moderate genotoxic risk to man (7).

The radiation which people are exposed to may increase if they live in houses or buildings constructed by using materials whose radiation doses are above normal background level in the area. This may be due to fly-ash used in making cement. During the combustion process, the produced ashes are enriched in the above radionuclides (3). This is due to the fact that during combustion, organic material is oxidized and the inorganic material (containing radionuclides) left behind has a smaller volume, so the activity per unit mass is increased.

Therefore, determination of the activity of radionuclides belonging to uranium and thorium series and of ^{40}K radioisotope is important to:

- Evaluate the possible radiological risk and biological damage;
- Make standardizations about building materials.

The aim of this investigation was to calculate the amount of γ-rays emitted by construction materials in unit time and the damage to unit tissue by these γ-rays.

Materials and Methods

Building Materials, Raw Materials and Interior Coatings

A total of 16 samples were collected: 7 fly-ash samples from coal-fired power plants, 2 soil samples from Ankara, 2 cement samples (one containing 8% and the other containing 12% fly-ash), 1 brick sample from Konya, 1 phosphogypsum sample, 1 water-based paint sample, 1 solvent-based paint sample and 1 foam sample (used as insulator).

Preparation of Samples for γ-Spectrometry

Grinding monolithic samples may cause the release of a considerable amount of ^{222}Rn, thus perturbing the equilibrium within the ^{238}U decay chain, so that the activity of ^{214}Bi may be representative of that of its parent isotope only after a period of about 30 days (8). The samples were ground into a fine powder with a particle size less than 1 mm. The samples were then dried in a temperature-controlled furnace at 110ºC 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 and hermetically sealed. The sealed samples were stored for 30 days before counting to allow ^{226}Ra and its short-lived decay products to reach the secular equilibrium.

Measurement of γ-Irradiation

After waiting for one month in containers, γ-irradiation from each sample was measured by γ-spectrometry. Germanium spectrometers (γ-spectrometers with highest resolution) were used throughout the research. 609KeV γ-rays from ^{214}Bi radioisotope (for ^{238}U series), 908KeV γ-rays from ^{228}Ac radioisotope (for ^{228}Th series) and 1460 KeV γ-rays from ^{40}K were measured. Results and errors have been obtained in Bq kg^{-1} (Table 1).

Table 1. Specific activities of radioisotopes to represent each series.

		Activity Rates (Bq kg^{-1})					
		^{214}Bi (for ^{238}U series)		^{228}Ac (for ^{228}Th series)		^{40}K	
	Material	Measurement	Error	Measurement	Error	Measurement	Error
1	Fly-ash	71.88	3.66	28.70	4.43	287.04	19.40
2	Fly-ash	219.57	5.80	138.47	6.78	<MDA[5]	
3	Fly-ash	235.57	5.04	119.79	5.89	<MDA	
4	Fly-ash	501.98	8.78	159.11	4.41	<MDA	
5	Fly-ash	357.78	7.55	123.37	7.06	586.79	16.66
6	Fly-ash	123.10	3.40	71.21	3.20	<MDA	
7	Fly-ash	126.01	3.06	166.25	4.29	670.90	10.39
8	Cement (8% fly-ash)	73.40	2.91	41.48	3.61	<MDA	
9	Cement (12% fly-ash)	86.45	4.19	41.59	6.61	370.21	21.54
10	Soil	22.42	0.74	29.36	1.27	45.18	4.27
11	Soil	18.91	0.83	30.78	1.63	<MDA	
12	Brick	4.40	0.32	<MDA		<MDA	
13	Phosphogypsum	302.28	7.92	<MDA		<MDA	
14	Solvent-based paint	7.07	1.60	<MDA		76.49	12.10
15	Water-based paint	<MDA		<MDA		<MDA	
16	Foam	<MDA		<MDA		<MDA	

[5] MDA: Minimum Detectable Activity

Results

Calculation of Total γ-Irradiation from Each Sample

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in building materials is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq kg^{-1} to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K (2). The radium-equivalent activity was considered (OECD, 1979; UNSCEAR, 1982; Beretka and Mathew, 1985) as:

$$Ra_{eq} = A_{Ra} + 1.34A_{Th} + 0.077A_K$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively. This formula is based on the estimation that 1 Bq kg^{-1} of ^{238}U, 0.7 Bq kg^{-1} of ^{232}Th and 13 Bq kg^{-1} of ^{40}K produce the same γ-ray dose rates (9). The maximum value of Ra_{eq} in building materials must be less than 370 Bq kg^{-1} for safe use (10), which equals an annual dose of 1.5 mSv. However, the European Commission report sets this value as 0.3 - 1.0 mSv y^{-1} for safe use (11). In this case, Ra_{eq} value should be less than 247 Bq kg^{-1}. The radium-equivalent activities of the samples are listed in Table 2.

Calculation of External Annual Dose

The external annual effective dose is calculated for a room with dimensions of 4 m × 5 m × 2.8 m, and it is estimated that the material is used in floor, ceiling and walls. The equation to calculate the annual effective dose is:

$$(0.92 \times C_{Ra} + 1.1 \times C_{Th} + 0.08 \times C_K) \times (10^{-9}\ \text{Gy H}^{-1}) \times (0.7\ \text{Sv Gy}^{-1}) \times (24 \times 365 \times 0.8\ \text{h y}^{-1})$$

where 0.92, 1.1 and 0.08 are the specific dose rates of Ra, Th and K, respectively. C_{Ra}, C_{Th} and C_K are the activity rates of Ra, Th and K. 0.7 is the factor used for conversion from Gray to Sievert for adults and 0.8 is the estimated indoor occupancy factor. But the situation for thick materials is different. They can shield background radiation from outside. Therefore an average activity of 50 nGy h^{-1} is subtracted for the calculation of external effective annual dose of cement (11). Annual effective dose from each sample is listed in Table 3.

Table 2. Radium-equivalent activities of each sample.

	Material	Radium-Equivalent Activity Rates (Bq kg^{-1})
1	Fly-ash	135.023
2	Fly-ash	417.582
3	Fly-ash	406.870
4	Fly-ash	729.507
5	Fly-ash	579.382
6	Fly-ash	224.930
7	Fly-ash	415.407
8	Cement (8% fly-ash)	132.716
9	Cement (12% fly-ash)	174.430
10	Soil	67.884
11	Soil	62.925
12	Brick	4.400
13	Phosphogypsum	302.280
14	Solvent-based paint	12.960
15	Water-based paint	<MDA
16	Foam	<MDA

Table 3. Annual effective dose of each sample.

	Material	Annual Effective Dose (mSv y^{-1})
1	Fly-ash	0.592
2	Fly-ash	1.738
3	Fly-ash	1.710
4	Fly-ash	3.124
5	Fly-ash	2.511
6	Fly-ash	0.940
7	Fly-ash	1.729
8	Cement (8% fly-ash)	0.310
9	Cement (12% fly-ash)	0.515
10	Soil	0.277
11	Soil	0.251
12	Brick	0.020
13	Phosphogypsum	1.364
14	Solvent-based paint	0.062
15	Water-based paint	Cannot be calculated
16	Foam	Cannot be calculated

As mentioned earlier, annual effective dose should be less than 1.0 mSv for safety purposes. Phosphogypsum seems to have a greater value, but since it is used only superficially on the walls there is a different dose criterion index:

$$I = \left(\frac{C_{Ra}}{300}\right) + \left(\frac{C_{Th}}{200}\right) + \left(\frac{C_K}{3000}\right)$$

This external index is derived by using the specific dose rates and 1.0 mSv as the limit. For materials used in bulk amounts, such as concrete or brick, *I* should be less than 1. However, this limit is 6 for superficial and other materials with restricted use, such as tiles or boards (11).

Internal Hazard

In addition to external annual dose, radon and its short-lived products are harmful for the respiratory system. The equation to calculate internal hazard proposed by Krieger is:

$$H_{in} = \left(\frac{C_{Ra}}{185}\right) + \left(\frac{C_{Th}}{259}\right) + \left(\frac{C_K}{4810}\right)$$

H_{in} should be less than 1 for safety (12). Table 4 lists the internal hazard index for each sample.

Table 4. Internal hazard indices of building materials.

	Material	Internal Hazard Index
1	Fly-ash	0.559
2	Fly-ash	1.721
3	Fly-ash	1.736
4	Fly-ash	3.328
5	Fly-ash	2.532
6	Fly-ash	0.940
7	Fly-ash	1.462
8	Cement (8% fly-ash)	0.557
9	Cement (12% fly-ash)	0.705
10	Soil	0.244
11	Soil	0.221
12	Brick	0.024
13	Phosphogypsum	1.634
14	Solvent-based paint	0.054
15	Water-based paint	Cannot be calculated
16	Foam	Cannot be calculated

Discussion

In this study, 16 samples of building materials and raw materials were measured for the natural radionuclides they contain. Later, radium-equivalent activities were calculated using the formula proposed by UNSCEAR (1982) to compare the overall biological damage. This comparison shows that fly-ash samples, averagely, contain high amount of biologically hazardous radioactivity, but the activity of fly-ash samples vary in a wide range. The activity of the most radioactive sample is over 5 times more than the least radioactive one. This difference is due to different geographic characteristics where the coal, which gives fly-ash after burning, is mined. In spite of this averagely high activity of fly-ash, in cement this radioactivity is reduced due to the low percentage contribution of fly-ash. But fly-ash can still be a serious problem for workers in thermal power plants. Phosphogypsum also seems to have a value greater than 247 Bq kg^{-1} (calculated according to 1.0 mSv y^{-1} limit proposed by the European Commission in 1999). However, phosphogypsum has a different condition: Since it is used only superficially, unlike cement used in bulk amounts, it has a limit of 6 for dose criterion index *I* and its dose criterion index is 1.007, which is quite below 6. Yet it contributes not only to the annual effective dose but also the internal hazard. Internal hazard index of phosphogypsum is 1.6 while the limit is 1.0; therefore, the use of phoshogypsum is a critical point for safety in construction. Finally, specific dose rates and wall thickness of the European Commission were used in this study, but these may not be the real values for all Turkish dwellings. For a more accurate result, annual effective doses can be calculated for different values of these parameters. A limited number of samples were used in this study. For a more accurate result that better represents Turkey, similar studies using a higher number of samples can be made. This study can be used as a reference for more extensive studies of the same subject in Turkey.

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