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NATURAL RADIOACTIVITY OF SAND SAMPLES FROM TRANSYLVANIA AREA AND ASSESSMENT OF THE ENVIRONMENTAL RISKS ON CIVIL AND LIVESTOCK HOUSING

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Abstract

The present paper focuses upon the analysis of ^{226}Ra, ^{232}Th and ^{40}K radionuclides, making use of high-resolution HPGe γ-spectrometry devices. By means of this technique, the radiological risk is investigated in the case of various sand samples. The natural concentrations for ^{226}Ra, ^{232}Th and ^{40}K, from sand samples, ranged from (9.3-20.0 Bqkg^{-1}), (9.1-23.7 Bqkg^{-1}) and (203.5-403.3 Bqkg^{-1}), respectively. The radium equivalent activity of sands under investigation was also analyzed and ranged from 45 to 83 Bqkg^{-1}. The radiation hazard due to the use of sand as a building material on civil and livestock buildings was also estimated by a set of hazard indices such as H_{ext} and I_{yr}, respectively. The geological structure of the sand collection areas is discussed, too. The results of the research show that the samples under analysis are defined by a low natural radioactivity, meaning that they can be used as building materials (mortars, concretes) components, in civil and livestock construction, without enhancing their natural radioactivity.

Key words: environmental radioactivity, γ-spectrometry, hazard indices, sand

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1. Introduction

According to the national standard (SR EN ISO 14688-1, 2004), sands are characterized with respect to the dimension of the particle fractions as follows: coarse sand, medium sand and fine sand with size ranging from 0.063 to 2 mm. Sand size in the Wentworth scale is classified from a very fine sand, with size range between 62.5-125 µm, up to a very coarse sand, with size range between 1-2 mm. The Phi scale (or Krumbein scale) is a logarithmic transformation of the Wentworth scale classifying sands between +4φ (very fine sand) and -1φ (very coarse sand) (Wentworth, 1922). The component parts of sand may vary with geological conditions and origin of rocks, and it is known that more than 93% from sand part are quartz fragments. Sand is used in construction industry in the composition of concrete, mortars and pavements, or as a raw material in other industries.

The main contribution of sand to natural radioactivity is due to gamma radiation, through the presence of natural radionuclides such as ^{226}Ra, ^{232}Th and ^{40}K. Known that exposure to radon and thorium (products of Ra and Th decays) is the second leading cause of lung cancer in the world (Baias et al., 2010;

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Cosma et al., 2013; Todea et al., 2013), the study of natural radionuclides due to gamma-rays allows to understand the implication of the human body, to the gamma radiation exposure and to apply safety measures, whenever the exposure dose is found to exceed the recommended values (Abel-Ghany et al., 2009; Ahmed et al., 2006; Ngachin et al., 2007; Quindos et al., 1987).

Considering the increasing development in the construction area and the overall trend regarding the protection of the environment and population health, the subject of possible radioactivity compounds remains the foundation ground, that is very important to be studied. The laboratory samples were collected from seven construction sites located in Transylvania area (Fig. 1), from drilling studies.

The importance of the present study is also given by the fact that in Romania there is no radioactivity map and the samples collection area is an extended one. Hence, the results obtained in this study will provide important data needed for further radioactivity investigations regarding the Transylvanian area and will also contribute to the protection of natural environment and population health, in terms of natural radioactivity.

In Romania, the content of radionuclides ($Bqkg^{-1}$) in the sands (Table 1) is regulated by the Annex to the Order of the Minister of Health no.51/18.02.1983, which establish the National Norms of Radioprotection of the population and environment (MOH, 1983).

Gamma spectrometry is suitable for both qualitative and quantitative analysis of gamma ray emitting radionuclides in sand. Radionuclides stick to the sand particle in airtight environment and become detectable only by this type of technique.

The aim of the present work lies in determining the concentration of radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in several samples collected from Transylvania Area. Having the results of the radioactivity of the samples analysed, we assessed the hazard caused by the presence of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K in sand, by means of three indices: radium equivalent activity (Ra_{eq}), the gamma radioactivity index (I_{yr}) and the external hazard index (H_{ex}). In addition, the geological structure of the sand collection areas was discussed.

Table 1. Maximum activity concentration of ^{226}Ra, ^{232}Th and ^{40}K and the radioactivity index value (I)

Building material	***Activity concentration ($Bqkg^{-1}$)***			I_{yr}
Sand, gravel	^{226}Ra	^{232}Th	^{40}K	0.5
	29.9	91.3	869.5	

2. Experimental

2.1. Geological description of tested materials

Laboratory studies were carried out on sands collected from the following areas: Aghires (Cluj county), Feleac (Cluj county), Valea Capriorii (Cluj county), Baia-Mare (Maramures county), Valea lui Mihai (Bihor county), Oradea Hills (Bihor county), Arad (Arad county). All the tested samples were collected from different areas from Transylvania. The geologically researches on the studied area have shown that the basis of the Transylvanian Depression is made up in majority of rocks of the same ages, arranged as structures bearing similitude to those in the Eastern and Western Carpathian Mountains.

The Pannonian Block, the massif of the Western Carpathians and the Transylvanian depression have different tectonic evolutions, being defined by radial movements, with differing senses and intensities. While the Western Carpathians are characterised by movements of permanent rising, the Transylvanian Depression underwent alternative modifications of rising, erosion and sinking, so that the general rising of the Carpathian regions finished in the shape of a unitary dry land.

Fig. 1. Areas where the samples were collected from

Table 2. Sample collection area description

Sample sites	***Chronostratigraphical units***	***Geological structure***
Aghires	Priabonian (Pr)	- lower coarse limestone and upper mottled clays
Feleac	Volhinian – Bessarabian (Vh-Bsi) Sarmatian	- marl-sand deposits with tuffs - sandstones parting - in the Western part of the Feleac hill, besides grits with concretions, boulders and gravels can be found
Valea Capriorii	Volhinian – Bessarabian (Vh-Bsi) Sarmatian	- marl-sand deposits with tuffs - sandstones parting
Baia-Mare	Quarternary	- sediment, represented in diluvial-colluvial deposits and slate blocks
Valea lui Mihai	Sarmatian	- dune sand group
Oradea hills	Siltic and Pannonian	- recent alluviums, represented by gravels and sands
Arad	Sarmatian	- recent alluviums from river beds, made up of gravels and sands alike

Geologically analyzing the context, the sands encountered in the North Western part of Romania, which is the Transylvanian Region, are usually Sarmatian sands. In the Western part, especially in Bihor County, Pannonian sands are also encountered (Anastasiu et al., 1999). These types of sands (Pannonian and especially Sarmatian) are very often found in geotechnical drilling in Transylvania area and very often, because of the depth at which they are found, they represent the foundation ground. Regarding the grain size, Sarmatian sands are fine, medium or coarse sands, while Pannonian sands are usually very fine sands (Molnar, 2013; Molnar and Popa, 2013).

2.2. Gamma spectrometry techniques

The activity concentration is detected with gamma spectrometry and hyper-pure germanium detectors. For this purpose, it was used a detector HPGe Ortec of type GMX and resolution 1.92 KeV to 1.33 MeV and a relative efficiency of 34.2%, respectively a GEM – type detector of resolution 1.85 KeV to 1.33 MeV and a relative efficiency of 30%. The gamma spectrometry system was coupled to a multi-channel analyser.

An amount of about 2 kg was collected for each laboratory sample, and after keeping them at normal temperature conditions for about 24 hours, a sample with variable weight (50-200 g) was taken from each amount collected. The activity concentrations was calculated based on the average values from the photopeaks at various energy levels. For ^{226}Ra, the γ – rays of ^{214}Pb (295 keV and 351 keV) and ^{214}Bi (609 keV) were used. For ^{232}Th, γ – rays of ^{228}Ac (338 keV and 911 keV), ^{208}Tl (583 keV) and ^{212}Pb (238 keV). The activity concentration of ^{40}K was computed directly from its γ – rays at 1460 keV. Methods of calculations have been described in detail in earlier publications (Muntean et al., 2012; 2014).

3. Results and discussion

3.1. Specific activity measurements

The analysis of the results given in Table 2 shows that the smallest value of ^{226}Ra concentration is 9.3±0.9 Bqkg^{-1}, measured for sample S4, while the highest value is recorded for sample S5 (20.0±1.4 Bqkg^{-1}). In the case of ^{232}Th radionuclide, the lowest value is recorded for sample S4, while the highest value is found with sample S1. The smallest value for ^{40}K radionuclide is 203.5±13.1 Bqkg^{-1} for sample S7, respectively the highest value is found in sand sample S1. The comparison of the results in Table 2 with the data in Table 1, giving maximum admissible value for Romanian building materials in the case of the three radionuclides, reveals that all the three radionuclides give much lower values in all the analyzed samples.

3.2. Gamma-ray radiation hazard indices

The natural radioactivity in building materials is usually determined from ^{226}Ra, ^{232}Th and ^{40}K contents.

Because radium and its daughter products are responsible for 98.5% of the radiological effects of the uranium series, the contribution from the ^{238}U is replaced with the decay product of ^{226}Ra (Veiga et al., 2006). As the distribution of ^{226}Ra, ^{232}Th and ^{40}K in different types of sand is not uniform, the uniformity, regarding the exposure to radiation, has been analyzed by radium equivalent activity (Ra_{eq}) index. To compare the specific activity of samples containing different concentrations of radionuclides Ra_{eq} index was calculated with the following relationship (Beretka and Mathew, 1985) (Eq. 1):

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (1)$$

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

It was supposed that 10 Bq of ^{226}Ra, 7 Bq of ^{232}Th and 130 Bq of ^{40}K produce the same gamma-ray dose rate (El-Arabi, 2007). Table 3 presents the radium equivalent activity results for the sands coming from Transylvania area. These values vary from 45 Bqkg^{-1} (value found for sample S7) to 83 Bqkg^{-1} (value for sample S5).

All the results are lower than the maximum recommended value of 370 Bqkg^{-1}. As sand is used in the industry of glass and porcelain and building materials, the gamma-ray radiation hazards were assessed by two other indices, gamma radioactivity index (I_{yr}) and the external hazard index, H_{ext}.

Table 3. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in sand samples

Sand sample/location	*Activity concentration (Bq kg^{-1})*		
	^{226}Ra	*^{232}Th*	*^{40}K*
S1/Feleac	16.8±1.2	23.7±2.8	403.3±17.7
S2/Aghires	16.5±1.2	16.4±2.1	396.6±25.7
S3/ Valea Capriorii	13.6±1.2	20.7±2.4	328.0±20.4
S4/ Baia Mare	9.3±0.9	9.1±1.1	292.2±12.9
S5/Arad	20.0±1.4	22.3±2.5	400.8±16.5
S6/ Oradea hills	18.8±1.7	16.6±1.8	357.2±15.5
S7/Valea lui Mihai	11.2±0.9	13.3±1.7	203.5±13.1

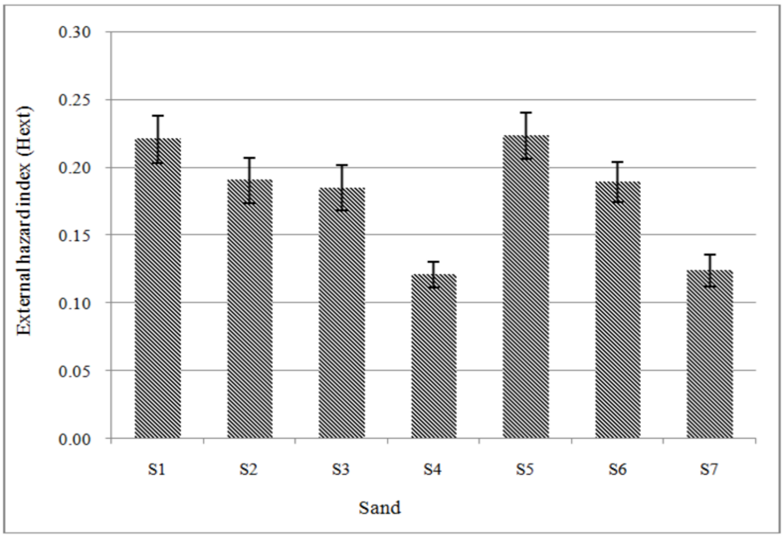

Fig. 2. External hazard index for sand samples

In Romania, the only index defined in standards (MOH, 2004) is the gamma radioactivity index (I_{yr}), calculated with the relationship (Eq. 2):

$$I_{\gamma r} = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \quad (2)$$

The external hazard index H_{ext} (Fig. 2) was calculated based on the model proposed by Beretka and Mathew (1985), to limit the external gamma radiation dose coming from building materials, to the amount of 1.5 mSv. The relationship used is (Eq. 3):

$$H_{ext} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (3)$$

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

The values of the hazard index for sand samples under analysis have a value under 1 (unit) so the hazard risk is negligible. For all building materials, the index must be under 0.5. According to Table 4, one can appreciate that all the sand samples present radioactivity indices under the value of 0.5.

Table 5 presents a comparison between the results found during this investigation and those reported by various countries in Europe. One can notice that sand radioactivity varies from one area to another. The values found for the specific activity of the three radionuclides in the sand samples are in agreement with the values reported in the other countries.

Table 4. Gamma-ray radiation hazard indices for the analyzed sand samples

Sand sample	*Ra$_{eq}$ (Bq kg^{-1})*	*I_{yr}*
S1	82	0.31
S2	71	0.27
S3	68	0.26
S4	45	0.18
S5	83	0.31
S6	70	0.27
S7	46	0.17

4. Conclusions

The results show that the samples analyzed are defined by a low radioactivity compared to the maximum admissible content. The highest activity concentration was found in samples S1 and S5. The evaluation of the hazard indices leads to the conclusion that samples under investigation do not pose any problem regarding natural radioactivity and that they can be used in the construction of dwelling places and livestock housing, having no detrimental effect upon the constructions raised.

As for the Annex of the Ministry of Health Mo. 51/1983, it clearly needs amendments with recent findings and study results about building materials used in practice today.

Table 5. Comparison of activity concentration of sand in Romanian and other European Countries

Country	***Activity concentration (Bq kg^{-1})***		
	^{226}Ra	***^{232}Th***	***^{40}K***
Germany (Tufail, 1992)	15.5	17.8	395.0
Italy (Tufail, 1992)	24.0	27.0	528.0
Netherlands (Tufail, 1992)	8.1	10.6	200.0
Slovakia (Cabanekova, 1996)	8.0 – 21.0	8.0 – 29.0	208.0 – 410.9
Greece (Papaefthymiou and Gouseti, 2008)	11.0 – 27.0	8.0 – 26.0	167.0 – 656.0
Present study	9.3 – 20.0	9.1 – 23.7	203.5 – 403.3

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