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FIRST CHARACTERISATION OF NATURAL RADIOACTIVITY IN BUILDING MATERIALS MANUFACTURED IN ALBANIA

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This study focuses on the radiological characterisation of building materials manufactured in Albania by using a high-resolution gamma-ray spectrometer. The average activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th were, respectively, 644.1 ± 64.2, 33.4 ± 6.4 and 42.2 ± 7.6 Bq kg^{-1} in the clay brick samples and 179.7 ± 48.9, 55.0 ± 5.8 and 17.0 ± 3.3 Bq kg^{-1} in the cement samples. The calculated activity concentration index (ACI), varied from 0.48 ± 0.02 to 0.63 ± 0.04 in the clay brick samples and from 0.29 ± 0.03 to 0.37 ± 0.02 in the cement samples. Based on the ACI, all of the clay brick and cement samples were categorised as A1 materials. The authors can exclude (at 3σ level) any restriction of their use as bulk materials.

INTRODUCTION

An important contribution to public radioactive exposure is made by external terrestrial radiation, principally due to uranium and thorium decay chains, and by ^{40}K, which is present in the Earth's crust. According to the United Nations Scientific Committee on the Effects of Atomic Radiation[1], this contribution corresponds to ~20 % (ranging between 0.3 and 1.0 mSv y^{-1}) of the average worldwide radiation exposure (2.4 mSv y^{-1}). Considering an indoor occupancy factor of 0.8, the average indoor-to-outdoor ratio of the contribution of external terrestrial radiation is roughly 6.

Building materials contain naturally occurring radionuclides; therefore, they are potential sources contributing to excesses in external and internal radiation exposure due to their final uses in dwellings. According to the European recommendations[2, 3], building materials are classified as suitable for use by regulating an upper external radiation exposure limit of 1.0 mSv y^{-1}.

Scientific interest in studying the radioactivity content of various building materials has increased in the last decade[4–6]. Moreover, industrial waste/by-products (e.g. fly ash, red mud and phosphogypsum) have recently been adapted and used as additives in various building materials[7–9]. This industrial waste and these by-products are themselves subject to regulations[10] because of the potential increase of naturally occurring radioactive materials (NORMs) due to technological processes[11].

Clay bricks and cements are the principal building materials manufactured in Albania, constituting an important proportion of the domestic construction industry, as well as being exported to several foreign countries[12, 13]. However, there is a lack of data about the natural radioactivity content in these building materials.

The aim of this study was to characterise the concentrations of ^{40}K, ^{226}Ra and ^{232}Th in the clay brick and cement samples manufactured in Albania using a high-resolution gamma-ray spectrometer. These data were further used to evaluate the absorbed dose rate of indoor external gamma radiation due to exposure to natural radionuclides. Recent criteria, based on the activity concentration index (ACI)[3], were used to assess the potential radiological hazards associated with the final use of these materials in dwellings. Finally, these data were compared with the values reported in the similar studies performed in those countries that are economic partners of Albania.

EXPERIMENTAL METHODS

Sampling and sample preparation

The Albanian industries of clay brick and cement have become heavily developed over the last few decades, offering quality products for the European market. More than 80 % of the clay bricks and cements commercialised in Albania are produced, respectively, by 9 and 3 manufacturers located across the country (Figure 1). For each of these 12 manufacturers, 5 different samples were collected to obtain a homogeneous data set of 60 samples.

The samples of clay brick were crushed and milled into fine powders with the particle size of <2 mm, while the samples of cement were directly processed because they were already in a powder form. To remove the moisture content, all of the samples were dried in a temperature-controlled furnace at 110°C for at least 24 h (or until constant weight). After cooling in a moisture-free atmosphere, each sample was transferred for measurement to a cylindrical PVC container (with the dimensions of diameter of 7.5 cm × 4.5 cm of height and an effective volume of 180 cm^3) and was then weighted. The hermetically sealed containers were stored for at least 4 weeks prior to measurement to allow ^{226}Ra and its short-lived decay products to reach the secular equilibrium. Each box contained materials coming from the different stocks of clay bricks and cement samples collected in the period from 2010 to 2012.

The same protocol was applied to the three certified reference materials (95% confidence interval)[14], prepared in a powder matrix (240 mesh) containing 4940 ± 30 Bq kg^{-1} of ^{238}U (diluted uranium ore BL-5), 3250 ± 90 Bq kg^{-1} of ^{232}Th (diluted thorium ore OKA-2) in a secular equilibrium and $14\,000 \pm 400$ Bq kg^{-1} of ^{40}K (high-purity K_2SO_4). These certified reference materials were used to calibrate the gamma-ray spectrometer[15, 16].

High-resolution gamma-ray spectrometry calibration and measurements

The natural radioactivity content of the building materials was investigated using a fully automated high-resolution gamma-ray spectrometry system, called the MCA_Rad system[11]. This equipment is composed of two coaxial high-purity germanium p-type detectors with a 60 % relative efficiency, having an energy resolution of 1.9 keV at 1332.5 keV (^{60}Co).

The certified International Atomic Energy Agency reference materials, prepared in a standard geometry, were measured to study the efficiency of the MCA_Rad system. A self-absorption correction, due to variations in the density and the composition between the samples and the reference materials, was performed, considering the approach discussed in detail by Bolivar *et al.*[17] and by Cutshall *et al.*[18]. According to this approach, the self-attenuation correction factor (Eq. 1) is given as a function of the sample density and of the energy of the gamma ray investigated. The ratio of the self-attenuation factor in a reference material (C_{SA}^{ref}) to that of the sample (C_{SA}^{s}) allows for the definition of the self-attenuation correction factor (C_{SA}),

$$C_{SA} = \frac{C_{SA}^{ref}}{C_{SA}^{s}} \tag{1}$$

or

$$C_{SA} = \frac{1 + a_1 E^{-a_2}}{\exp[\rho_s(a_3 + a_4 \ln E + a_5 \ln E^2)]} \tag{2}$$

where E is the gamma-ray energy (in keV), ρ_s is the sample density (in g/cm^3) and a_n are arbitrary parameters. An average mass attenuation coefficient[11] was parameterised in the energy range of 200–3000 keV and used to calculate the best fit of the C_{SA} functions, fixing the following coefficients as $a_1 = 8.8443$, $a_2 = 0.5157$, $a_3 = 1.2609$, $a_4 = -0.2547$ and $a_5 = 0.0134$[11].

The activity concentration of the samples (A_s in Bq kg^{-1}) is then calculated by using the formula:

$$A_s = \frac{[R_s/(m_s \cdot C_\varepsilon)]}{C_{SA}} \tag{3}$$

where R_{ref} and R_s are the count rates (in cpm); A_{ref} and A_s are the activity concentrations (in Bq kg^{-1}) and m_{ref} and m_s are the masses (in kg) for, respectively, in the reference materials and the samples. In Table 1 the values of the conversion coefficient $[C_\varepsilon = R_{ref}/(A_{ref} m_{ref})]$ for the principal gamma-ray energies investigated are reported.

RESULTS AND DISCUSSION

Activity concentrations

Measuring for 1 h the activity concentration of isotopes mentioned in Table 1, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in 60 samples were determined. In appendix A, the results and overall

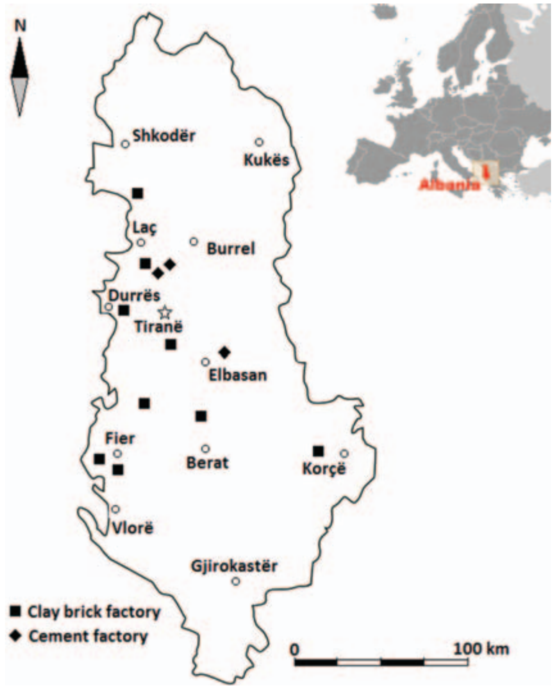

Figure 1. The location of the clay brick (rectangles) and cement (rhombi) factories investigated in this study.

Table 1. The fixed conversion coefficients C_ε (obtained as described in Eq. 3), used for measuring the elements reported in column two, calculated for the principal photopeaks with energy *E* and having the highest gamma yield *I*.

Parent isotope	Daughter isotope	E (keV)	I (%)	$C_\varepsilon \pm \sigma$ (cpm Bq^{-1})
^{238}U	^{234m}Pa	1001	0.84	0.0153 (2)
	^{214}Bi	609	44.8	0.9634 (59)
	^{214}Pb	352	35.8	1.2714 (78)
^{232}Th	^{228}Ac	911	26.6	0.4850 (134)
	^{212}Pb	238	43.3	1.9019 (527)
	^{212}Bi	727	6.58	0.1505 (42)
	^{208}Tl	583	84.5	0.6478 (180)
^{40}K	—	1460	11.0	0.1437 (42)

measurement uncertainties (Supplementary data, Table A1) are reported. The authors note that the radioactivity contents from each manufacturer were homogeneous. Because the standard deviation of the mean for the five samples was comparable to the measurement uncertainty, in Table 2 the mean and combined errors were reported for each manufacturer. The combination of uncertainties was estimated by the Monte Carlo simulation[19]. In particular, to obtain the standard deviation of the mean, matrices were generated with the 10^5 pseudo-random values, using the individual measurement parameters (K, U and Th measures $\pm 1\sigma$). The activity concentrations of K, U and Th in the clay brick samples were found to be lower than or comparable within 1σ to that of the typical activity concentrations of 670, 50 and 50 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th, respectively[2].

Radiological hazard indexation

The radiological hazards of the building materials were calculated by adopting the following ACI, which is widely used at the investigation level for practical monitoring purposes[20]:

$$\mathrm{ACI} = \frac{C_{\mathrm{Ra}}}{300} + \frac{C_{\mathrm{Th}}}{200} + \frac{C_{\mathrm{K}}}{3000}, \tag{4}$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations in Bq kg^{-1} for radium (equivalent to uranium under the secular equilibrium conditions), thorium and potassium, respectively. Following ref. (3), the radiological hazard could be classified into four classes, leading to two categories of materials used in bulk amounts and materials with superficial or restricted uses (Table 3).

Table 2. For each manufacturer, ID, the specific activity (mean $\pm 1\sigma$) calculated for ^{40}K, ^{226}Ra and ^{232}Th (in Bq kg^{-1}), the ACI, the absorbed gamma dose rate indoors (D^{indoor}) and the AEDE are reported.

Manufacturer ID	^{40}K (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	ACI	D^{indoor} (nGy h^{-1})	AEDE (mSv y^{-1})
Clay brick						
A	723.7 ± 65.8	40.5 ± 4.6	50.1 ± 4.7	0.63 ± 0.04	92.4 ± 8.5	0.45 ± 0.04
B	671.1 ± 58.0	42.1 ± 5.8	44.4 ± 3.8	0.59 ± 0.03	83.3 ± 8.2	0.41 ± 0.04
C	644.2 ± 50.4	29.0 ± 2.2	37.4 ± 3.0	0.50 ± 0.02	61.4 ± 5.6	0.30 ± 0.03
D	608.1 ± 75.2	32.6 ± 5.1	38.4 ± 9.4	0.50 ± 0.06	62.8 ± 12.9	0.31 ± 0.06
E	672.3 ± 29.9	33.8 ± 2.2	42.6 ± 3.0	0.57 ± 0.02	79.2 ± 4.5	0.39 ± 0.02
F	566.5 ± 37.2	31.2 ± 2.8	45.4 ± 7.4	0.52 ± 0.04	66.0 ± 9.1	0.32 ± 0.04
G	613.9 ± 21.2	28.7 ± 2.2	35.1 ± 3.4	0.48 ± 0.02	56.2 ± 4.6	0.28 ± 0.02
H	642.8 ± 45.1	28.0 ± 4.7	39.1 ± 5.6	0.50 ± 0.04	62.2 ± 8.4	0.30 ± 0.04
I	654.3 ± 25.0	34.4 ± 5.8	47.6 ± 4.7	0.57 ± 0.03	78.3 ± 7.7	0.38 ± 0.04
Cement						
J	157.0 ± 49.1	49.8 ± 7.4	15.1 ± 3.7	0.29 ± 0.03	17.0 ± 8.8	0.08 ± 0.04
K	227.3 ± 19.7	58.0 ± 3.9	19.8 ± 2.2	0.37 ± 0.02	35.3 ± 4.6	0.17 ± 0.02
L	154.9 ± 12.3	57.2 ± 1.9	16.2 ± 2.0	0.32 ± 0.01	24.8 ± 3.0	0.12 ± 0.01

The uncertainties associated with ACI, D^{indoor} and AEDE were calculated, propagating the errors of specific activities.

Table 3. The scheme for the association of the default dose, according to the ACI criteria defined in European commission recommendation[2,3].

Category (corresponding default dose)	Type 1: materials used in bulk amounts, e.g. concrete, bricks etc.	Type 2: superficial and other materials with restricted use, e.g. tiles, boards, etc.
A (≤ 1 mSv y^{-1})	For ACI ≤ 1 category A1	For ACI ≤ 6 category A2
B (>1 mSv y^{-1})	For ACI >1 category B1	For ACI >6 category B2

In Table 2 the ACIs varied from 0.48 ± 0.02 to 0.63 ± 0.04 in the clay brick samples and from 0.29 ± 0.03 to 0.37 ± 0.02 in the cement samples. An ACI of <1 within 3σ standard deviations categorised these building materials as A1 and therefore suitable for use in bulk amounts without restrictions. Applying the default dose criteria recommended by the European Union (EU) for building materials, the investigated clay brick and cement samples met, within 3σ, the exemption for the annual effective dose criterion of <1 mSv y^{-1} (Table 2).

Estimation of the absorbed gamma dose rate and the annual effective dose equivalent

The total external absorbed dose rates (D^{indoor} in nGy h^{-1}) in the indoor air, due to gamma rays emitted by ^{226}Ra, ^{232}Th and ^{40}K, was estimated according to refs (2, 21), using the absorbed dose conversion coefficients (0.92, 1.10 and 0.08 for ^{226}Ra, ^{232}Th and ^{40}K, respectively and expressed in nGy h^{-1}/Bq kg^{-1}) determined by the Monte Carlo simulation, with a standard room model of 4 m × 5 m × 2.8 m with the thicknesses of the walls, floor and ceiling and the density of the structures of 20 cm and density of 2350 kg m^{-3} (concrete). The resulting external absorbed dose rates in the indoor air, due to gamma rays emitted by the ^{226}Ra, ^{232}Th and ^{40}K, was estimated using the formula:

$$D^{\text{indoor}} = 0.08C_{\text{K}} + 0.92C_{\text{Ra}} + 1.10C_{\text{Th}} \tag{5}$$

where C_{Ra}, C_{Th} and C_{K} are the activity concentrations in Bq kg^{-1} for radium (equivalent to uranium under the secular equilibrium conditions), thorium and potassium, respectively. The excess in the external absorbed dose rates in the indoor air was estimated by subtracting the 'background' outdoor average absorbed dose rate of 58 nGy h^{-1}[1]. In Table 2, the absorbed dose rates in the indoor air are reported, which varied from 56.2 ± 4.6 to 92.4 ± 8.5 nGy h^{-1} in the clay brick samples and from 17.0 ± 8.8 to 35.3 ± 4.6 nGy h^{-1} in the cement samples. The listed values were lower or comparable (within 1σ) to that of the published world average dose rate of 84 nGy h^{-1}[1].

To assess the annual effective dose equivalent (AEDE) rate, the conversion coefficient from absorbed doses in the air to the effective dose received by an adult must be considered as 0.7 Sv Gy^{-1}, published in the UNCREAR[22], and the

Table 4. The mean activity concentrations ($\pm$ 1σ) and, when available, the respective minimum and maximum values (in brackets) for the 45 clay brick samples and 15 cement samples measured in this study and compared with reported bibliographic results.

Author of the study	Country	^{40}K (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	ACI[a]
Clay brick					
This study	Albania	644.1 ± 64.2 (490.6–820.3)	33.4 ± 6.4 (20.8–51.9)	42.2 ± 7.6 (29.2–55.1)	0.54 ± 0.05
Trevisi *et al.*[20]	EU	598 (12–1169)	47 (2–148)	48 (2–164)	0.59
Xinwei [23]	China	713.9 ± 8.2	58.6 ± 4.7	50.4 ± 3.5	0.70 ± 0.04
Zhao *et al.*[24]	China	846 ± 67 (745–961)	46 ± 4 (39–53)	56 ± 7(48–66)	0.70 ± 0.07
Turhan *et al.*[25]	Turkey	775.8 ± 149.6 (587.3–1092.0)	31.2 ± 7.2 (24.7–49.0)	37.2 ± 7.8 (26.6–51.2)	0.54 ± 0.11
Baykara *et al.*[26]	Turkey	201.4 ± 4.4	15.7 ± 1.1	3.8 ± 0.9	0.16 ± 0.01
Krstić *et al.*[27]	Serbia	579 ± 104 (488–700)	34 ± 4 (29–38)	43 ± 8 (35–53)	0.51 ± 0.08
Cement					
This study	Albania	179.7 ± 48.9 (107.3–250.8)	55.0 ± 5.8 (40.9–61.4)	17.0 ± 3.3 (11.5–22.9)	0.33 ± 0.03
Trevisi *et al.*[20]	EU	216 (4–846)	45 (4–422)	31 (3–266)	0.40
Xinwei[23]	China	173.8 ± 8.6	68.3 ± 3.6	51.7 ± 5.4	0.57 ± 0.04
Zhao *et al.*[24]	China	310 ± 76 (219–385)	52 ± 3 (49–55)	103 ± 25 (80–133)	0.71 ± 0.12
Turhan *et al.*[25]	Turkey[b]	316.5 ± 88.1 (196.1–475.7)	39.9 ± 18.0 (17.8–81.6)	26.4 ± 9.8 (7.8–48.8)	0.39 ± 0.15
Baykara *et al.*[26]	Turkey	2493.1 ± 78.9	24.7 ± 1.6	20.7 ± 1.5	1.02 ± 0.04

[a]ACI calculated for the reported mean $\pm\,\sigma$ values.
[b]Reported Pozzolana additive of <3 %.

indoor occupancy factor is ~0.8. The AEDE is given by the following equation:

$$\text{AEDE} = D^{\text{indoor}} \times 10^6 \times 8760 \times 0.8 \times 0.7 \qquad (6)$$

The corresponding average AEDEs are given in Table 2 and include values varying from 0.28 ± 0.02 to 0.45 ± 0.04 mSv y^{-1} in the clay brick samples and from 0.08 ± 0.04 to 0.17 ± 0.02 mSv y^{-1} in the cement samples. Considering the average world AEDE from the indoor terrestrial gamma radiation of 0.41 mSv y^{-1} (corresponding to an indoor absorbed gamma dose rate of 84 nGy h^{-1}), the estimated doses were lower or comparable within 1σ.

According to the INSTAT[13], Albania in 2010 had an export/import trading rates of 'construction materials and metals' of 19.8 and 16.0%, respectively. Albania's main economic partners are EU member states (Italy, Greece, Switzerland, Spain, Germany) and Kosovo, Turkey, China, Serbia and Montenegro, with roughly 85% of exports and 65% of imports. In Table 4 the available bibliographic results found in the similar studies in the EU and in particular in the aforementioned countries are reported to acquire a broader perspective of the building materials used in Albania. It was emphasised that the reported bibliographic values of activity concentrations of building materials in specific studies are only indicative and should not be used to characterise the respective country levels.

These data show that Albanian construction materials have the average concentrations of radioactivity, in the clay brick and cement samples, comparable within 1σ to those reported for EU member states, respectively, for ^{40}K, ^{226}Ra and ^{232}Th. In general, other countries of economic interest report values that are 1σ below the limit value of ACI=1, with the exception of one case in which the cements studied, in Turkey, showed ACIs slightly higher, ranging within 1σ around the limit value. On the basis of this study, it is clear that the clay brick and cement industries in Albania produce building materials of low activity concentrations, as characterised according to the European recommendations.

CONCLUSIONS

The radioactivity content of ^{40}K, ^{226}Ra and ^{232}Th were measured in 45 clay brick and 15 cement samples manufactured in Albania and then were compared with the results from other countries. The average activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th were, respectively, 644.1 ± 64.2, 33.4 ± 6.4 and 42.2 ± 7.6 Bq kg^{-1} in the clay brick samples and 179.7 ± 48.9, 55.0 ± 5.8 and 17.0 ± 3.3 Bq kg^{-1} in the cement samples. These activity concentrations were found to be comparable within 1σ to the measurements from EU member states. These results were useful for the assessment of the radiological hazards due to the final use of these materials as building materials. Adopting the 'new' ACI, these samples can be categorised within a 3σ uncertainty as A1 materials, i.e. suitable for use in bulk amounts without any restrictions. The external absorbed dose rates in the indoor air, due to natural radioactivity, in such building materials was estimated to be in the range from 56.2 ± 4.6 to 92.4 ± 8.5 nGy h^{-1} in the clay brick samples and from 17.0 ± 8.8 to 35.3 ± 4.6 nGy h^{-1} in the cement samples and was comparable, at close to a 1σ level, to the average world dose rate of 84 nGy h^{-1}. This level corresponds to an AEDE varying from 0.28 ± 0.02 to 0.45 ± 0.04 mSv y^{-1} in the clay brick samples and from 0.08 ± 0.04 to 0.17 ± 0.02 mSv y^{-1} in the cement samples. These data are comparable with the average world AEDE from the indoor terrestrial gamma radiation of 0.410 mSv y^{-1}. The clay bricks and cements manufactured in Albania do not pose a significant radiological hazard when used for building construction.

SUPPLEMENTARY DATA

Supplementary data are available at *Radiation Protection Dosimetry.*

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