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PHYSICS

Assessment of natural radiation exposure from building materials in Estonia

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Abstract. Radionuclides naturally occurring in building materials may significantly contribute to the annual doses of the public. As information on the radioactivity of such materials is lacking, the study of building materials used in Estonia was carried out in order to estimate the annual dose to the Estonian population due to natural radionuclides in building materials. During the study 53 samples of commonly used raw materials and building products were collected and measured. The activity concentrations were determined by gamma ray spectrometry. Their mean values were in the ranges 7–747 Bq kg^{-1} for ^{40}K, 4.4–69 Bq kg^{-1} for ^{226}Ra, and 0.8–86 Bq kg^{-1} for ^{232}Th. The activity index I in the 53 different building materials varied from 0.02 to 0.74 and the radium equivalent, from 6 to 239. The average annual dose for the people, caused by the building materials of dwellings, was assessed for most commonly used materials. It was estimated to be in the range from 0.16 mSv to 0.44 mSv.

Key words: radionuclides, building materials, dose assessment, gamma spectrometry.

INTRODUCTION

Man is continuously exposed to ionizing radiation from naturally occurring radioactive materials (NORM). The origin of these materials is the Earth's crust, but they find their way into building materials, air, water, food, and the human body itself. Measurement of activity concentrations of radionuclides in building materials is important in the assessment of population exposures, as most individuals spend 80% of their time indoors (Mustonen, 1985). Building materials cause direct radiation exposure, because most of them contain naturally occurring radioactive materials, mainly radionuclides from the ^{226}Ra and ^{232}Th decay chains and ^{40}K. The population-weighted average of indoor absorbed dose rate in air from terrestrial sources of radioactivity is estimated to be 84 nGy h^{-1} (UNSCEAR, 2000), The worldwide average indoor effective dose due to gamma rays from building materials is estimated to be about 0.4 mSv per year (UNSCEAR, 1977, 1993). Elevated indoor external dose rates may arise from high radionuclide content in building materials (Chen and Lin, 1996; Stoulos et al., 2003; Ahmed, 2005; Righi and Bruzzi, 2006; Brígido et al., 2008). Generally, natural building materials reflect the geology of their site of origin. The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the Earth's crust are 35, 30, and 400 Bq kg^{-1}, respectively. However, elevated levels of natural radionuclides causing annual doses of several mSv have been identified in some regions around the world, e.g. in Brazil, France, India, Nigeria, Iran (UNSCEAR, 1977, 1993, 2000). This external radiation exposure, caused by gamma-emitting radionuclides in building materials, can be assessed either by direct exposure measurements in the existing buildings or by radionuclide analyses of building materials with the dose rate modelling. Large-scale surveys of concentrations of radioisotopes in construction materials were summarized by the United Nations Scientific Committee on the Effects of Atomic Radiation, but very little information was found about conditions in Estonia. Consequently, the present study was undertaken with the purpose of determining radioactivity in some Estonian building materials and assessing the annual effective dose to the population due to external gamma ray exposure in dwellings typical of Estonia.

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SAMPLE COLLECTION AND PREPARATION

During the last 10 years 53 samples of the building materials were analysed. Mostly, the sample selection consisted of the commonly available materials, which were obtained from the building material stores. All samples were crushed into grains, dried, homogenized, and put into metallic beakers. For the measurement a low-background ORTEC HPGe gamma spectrometer with a 42% efficiency and 1.7 keV resolution (at 1.3 MeV) was used.

For measurements the samples were placed into gas-tight containers with the capacity of 120 cm^3. So after closure radon and thoron could not escape and during the ingrowth within about three weeks preceding the gamma spectrometric measurement a practical equilibrium between ^{226}Ra and ^{214}Pb/Bi was achieved in the samples. For determination of the ^{226}Ra and ^{232}Th content the photopeaks of their daughters ^{214}Pb, ^{214}Bi and ^{228}Ac, ^{208}Tl, respectively, were used.

For the estimation of the efficiencies a set of high-quality certified reference materials, e.g. IAEA-RG-SET (K, Th, U), with densities similar to the building materials measured after pulverization and calculations were used. The efficiency calibration of the gamma spectrometry systems was performed with the radionuclide specific efficiency method in order to avoid any uncertainty in gamma ray intensities, as well as the influence of coincidence summation and self-absorption effects of the emitting gamma photons.

DOSE RATE ESTIMATION

The method used for estimation of the doses is based on the work presented by Mustonen (1985) and Markkanen (1995). The dose rate is calculated for a rectangular source of uniform density and activity concentration. The following geometry is used: wall w$_1$ 7×2.8 m^2, wall w$_2$ 12×2.8 m^2, floor and ceiling 7×12 m^2. The walls, floor, and ceiling are 0.2 m thick. The room has four windows (2 m × 1.2 m), located symmetrically in the shorter walls at a height of 80 cm from the floor. The gamma dose rate is calculated in the middle of the room presented in Fig. 1 (Markkanen, 1995). Several studies (Mustonen, 1985; Jong and Dijk, 2008; Al-Jundi et al., 2009) have proved that the variation in the dose rate is small (10–15%) and the dose rate in the middle of the room is a good approximation for the average dose rate in the room. It also appears that the dimensions of the room have only a relatively small effect on the dose rate in that room.

Dose rates from the ceiling, floor, and each wall are calculated separately and the total dose rate is calculated as their sum. The influence of windows is taken into account when calculating the dose rate from imaginary wall elements and it is subtracted from the total

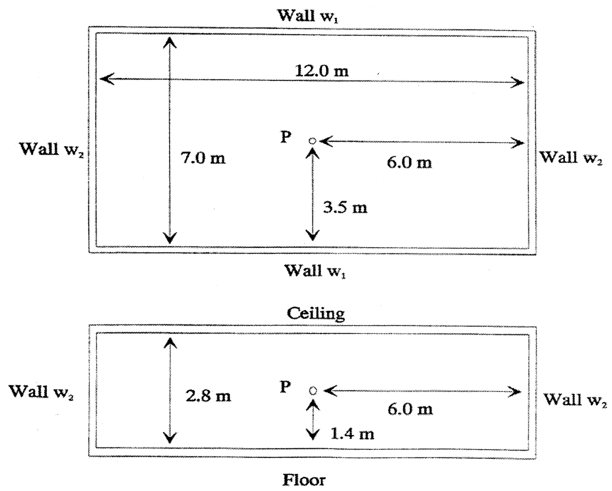

Fig. 1. The geometry used in calculation of the indoor gamma dose rate from building materials.

exposure rate. No external dose rate from the ground surrounding the room is taken into account. It is assumed that the annual exposure/occupancy time in dwellings is 7000 h. The conversion factor from the absorbed dose to effective dose is 0.7 Sv Gy^{-1}. The absorbed dose rate (Gy h^{-1}) in air at point P can be calculated according to the formula by Markkanen (1995):

$$D = 5.77\times10^{-7}\frac{C\rho}{4\pi}\sum\gamma_i\left(\frac{\mu_{\mathrm{en}}}{\rho}\right)_i E_i\int B_i\frac{\exp(-\mu_i s)}{l^2}dV. \quad (1)$$

Two different layers of building materials can be considered in the dose assessment if proper build-up and attenuation factors are used.

The build-up factor according to the Berger model (Berger, 1957; Vrubel, 1973) is

$$B_i = 1 + C(E_i)\mu_i s\exp(D(E_i)\mu_i s), \quad (2)$$

$$s = \left|\frac{z}{z_p - z}\right| l, \quad l = \sqrt{(x_p - x)^2 + (y_p - y)^2 + (z_p - z)^2}.$$

The parameters for equations (1) and (2) are as follows:

C, activity concentration of the layer (Bq kg^{-1});
ρ, bulk density of the layer (kg m^{-3});
γ_i, gamma intensity of gamma line i;
E_i, gamma energy of gamma line i (MeV);
$(\mu_{\mathrm{en}}/\rho)i$, energy absorption coefficient in air for gamma energy E (cm^2 g^{-1});
μ, attenuation coefficient in the layer for gamma energy E (cm^{-1});
$C(E_i)$, coefficient in the Berger model;
$D(E_i)$, coefficient in the Berger model;
l, distance between the point P and the point of integration Q (cm);
s, fraction of 1 within the layer (cm).

Table 1. Averaged gamma energies, attenuation coefficients in concrete, energy absorption coefficients in air, emission probabilities, and coefficients *C* and *D* in the build-up factors

Nuclide	Energy, keV	γ	μ, cm^{-1}	μ_{en}/ρ, 10^{-5} cm^2 g^{-1}	C	D
^{238}U	810	2.12	0.166	0.0285	1.161	0.144
^{232}Th	587	2.05	0.193	0.0295	1.279	0.190
^{232}Th	2615	0.356	0.0927	0.0217	0.734	0.0234
^{40}K	1461	0.107	0.124	0.0257	0.946	0.0755
^{137}Cs	662	0.183	0.183	0.0293	1.237	0.1737

The average parameters used in dose assessments are given in Table 1 (Markkanen, 1995).

RESULTS AND DISCUSSION

The total number of material samples for the activity concentration measurement was 53. The measurement results are presented in Tables 2 and 3. In Table 2 the results of the most common building materials used for dwellings are given. Below these data are used for the estimation of public doses. For these materials at least five different samples were taken. Analysis results for other building materials which do not have a major impact on the annual public doses are given in Table 3.

Table 2. Mean activity concentrations of radionuclides (Bq kg^{-1}) in the most commonly used building materials in Estonia

Material	^{226}Ra	^{238}U	^{232}Th	^{40}K
Aseri I brick	21.1 ± 3.0	33.9 ± 4.3	29.6 ± 3.2	436 ± 20
Aseri II brick	20.3 ± 2.9	32.6 ± 4.2	29.7 ± 3.1	436 ± 20
Aseri III brick	19.6 ± 2.8	31.5 ± 4.0	30.9 ± 3.1	449 ± 21
Misso Light brick	12.6 ± 1.5	19.6 ± 2.0	13.2 ± 1.4	145 ± 7
Misso Dark brick	18.4 ± 2.4	28.8 ± 3.2	19.4 ± 2.2	207 ± 9
Ash-blocks	27.3 ± 2.3	28.5 ± 2.2	14.1 ± 1.7	308 ± 8

Table 3. Mean activity concentrations of radionuclides (Bq kg^{-1}) in some building materials used in Estonia

Material	^{226}Ra	^{238}U	^{232}Th	^{40}K
Finnish building clay	53.6 ± 3.4	56.3 ± 3.3	58.5 ± 3.2	748 ± 17
Siimusti building clay	69.3 ± 3.8	70.6 ± 3.4	82.0 ± 3.5	678 ± 15
German building clay	35.6 ± 2.3	36.9 ± 2.1	55.2 ± 2.4	447 ± 11
Gypsum board	4.4 ± 1.0	4.9 ± 1.1	0.8 ± 0.2	7.0 ± 0.4
Grosso 311 floor tiles	64.1 ± 3.8	48.4 ± 2.4	81.5 ± 2.8	344 ± 9
Pronto 147 floor tiles	48.7 ± 2.5	50.3 ± 2.2	86.3 ± 2.8	284 ± 7
Kunda cement	46.9 ± 2.8	49.0 ± 2.5	21.4 ± 1.6	587 ± 13
Kolumbia stone	25.5 ± 1.8	26.3 ± 1.7	28.4 ± 1.6	480 ± 11
Concrete	35.1 ± 2.1	36.1 ± 1.8	11.3 ± 0.9	207 ± 5

Radionuclides present in the most commonly used building materials are of greater interest. The biggest differences occur in the concentration of ^{40}K, where the lowest mean value is 145 Bq kg^{-1} for Misso Light and highest 449 Bq kg^{-1} for Aseri III bricks. Much smaller variations are found for other radionuclides. From the results it can be seen that the lowest mean value of ^{226}Ra concentration is 12.6 Bq kg^{-1} measured in Misso Light clay brick, while the highest mean value for the same radionuclide is 27.3 Bq kg^{-1} for oil-shale ash block.

The ALARA principle (the dose exposure indoors should be as low as reasonably achievable) for building materials is followed by using the index I. In a general case of a number x of different building materials in a room, the index is calculated as (OECD–NEA, 1979; EC, 1999)

$$I_{ext} = \sum_x \frac{\sum_{i=1}^{n} w_{mi} C_{x_i}}{A_x},$$

where x represents the nuclide of interest, n is the number of different kinds of buildings materials used in a room, C_{x_i} (Bq kg^{-1}) is the measured activity of each nuclide in the building material, w_{mi} is the weight fractional usage of the building material, i, and A_x (Bq kg^{-1}) is the parameter value representing the activity concentration of each nuclide of interest, which promoted an effective dose of 1 mSv per year.

For a specific building material the parameters A_x have the following values: 300 Bq kg^{-1} for ^{226}Ra; 200 Bq kg^{-1} for ^{232}Th, and 3000 Bq kg^{-1} for ^{40}K. Accordingly, this index is defined by international as well as by the Estonian legislation (EC, 1999; Estonian Radiation Act, 2004) as the following sum:

$$I = \frac{C_{\mathrm{Th}}}{200\ \mathrm{Bq/kg^{-1}}} + \frac{C_{\mathrm{Ra}}}{300\ \mathrm{Bq/kg^{-1}}} + \frac{C_{\mathrm{K}}}{3000\ \mathrm{Bq/kg^{-1}}},$$

where C_{Th}, C_{Ra}, and C_{K} represent mean activity concentrations (Bq kg^{-1}) of the radionuclides ^{232}Th (in equilibrium with its daughter nuclides), ^{226}Ra (in equilibrium with its daughters), and ^{40}K.

If the value of the activity index, I, for a building material is 1 or less, the corresponding material can be used, with regard to radioactivity without restriction. If the value exceeds 1, the responsible party (producer or dealer) is required to assess the radiation exposure caused by the material and show specifically that the safety requirement is fulfilled. The guidance for construction materials does not usually include possible radon releases to the indoor air from building materials. The values of the activity concentration index depend on the dose criterion and the mode and the quantity of the material used in a building. According to the recommendations by the European Commission (EC, 1999) for materials used in bulk amounts, the activity index should be less than 0.5. However, for superficial and other materials with restricted use the corresponding activity index should be between 2 and 6.

The radium equivalent concept allows a single index or number to describe the gamma output from different mixtures of uranium (i.e. radium), thorium, and ^{40}K in a material. The radium equivalent Ra(eq) in Bq kg^{-1} can be calculated as follows (OECD–NEA, 1979; El-Hussein, 2005):

$$\mathrm{Ra(eq)} = A(\mathrm{Ra}) + 1.43A(\mathrm{Th}) + 0.077A(\mathrm{K}),$$

where $A(\mathrm{Ra})$ is the activity of ^{226}Ra (which is usually the same as that of ^{238}U) in Bq kg^{-1}, $A(\mathrm{Th})$ is the activity of ^{232}Th in Bq kg^{-1}, and $A(\mathrm{K})$ is the activity of ^{40}K in Bq kg^{-1}.

The index I and radium equivalent values for the most common building materials measured is presented in Table 4.

Clay bricks are widely used for construction of dwellings and their natural radionuclide content affects the population. As there is concern that some of the buildings will cause excessive radiation doses to the residents due to gamma rays emitted, the data obtained from our measurements were used for the estimation of the corresponding doses.

Table 4. Index I and radium equivalent values calculated for the measured building materials

Material	Index I	Radium equivalent Ra(eq)
Aseri I brick	0.47	97
Aseri II brick	0.44	96
Aseri III brick	0.37	98
Misso Light brick	0.16	66
Misso Dark brick	0.23	62
Ash-blocks	0.26	64
Finnish building clay	0.72	196
Siimusti building clay	0.87	239
German building clay	0.54	149
Gypsum board	0.02	6
Grosso 311 floor tiles	0.74	207
Pronto 147 floor tiles	0.69	194
Kunda cement	0.46	123
Kolumbia stone	0.39	103
Concrete	0.24	67

The calculations of annual doses caused by the natural radionuclides present in the building materials of dwellings were performed using the MathCAD software. A rather conservative assumption that all construction elements of the room, e.g. walls, the ceiling and the floor, are made of the same material is accepted in calculations. As a result, the evaluated annual doses represent the maximum values for the corresponding building material. The evaluations are not made for the materials, e.g. the binding materials and tiles, which practically form a relatively small fraction of the construction element. The density of building materials used in calculations is taken equal to 2350 kg m^{-3}. The results are presented in Table 5. From the results it can be concluded that the total effective dose rate is less than 1 mSv per year in dwellings constructed from the building materials measured in the course of the present study.

Table 6 shows the mean values for ^{226}Ra, ^{232}Th, ^{40}K, and radium equivalent activity for all building materials under investigation in other countries. This table also lists the typical world average values for building materials (UNSCEAR, 1993) and soil (UNSCEAR, 2000). We can see that the concentrations of radionuclides in Estonian building materials in the present study are comparable to those suggested by similar studies in other countries. In Estonia, maximum concentration values were recorded in different building clays and cement and minimum ones in gypsum board. Table 7 gives annual effective doses (mSv) for natural radionuclide sources in concrete, calculated from mean activity concentrations for Estonia and for selected countries (UNSCEAR, 2000). The dose values in the table are given for the occupancy factor equal to 1.

Table 5. Total annual dose due to the natural radionuclides in the building materials

Material	Total annual dose, mSv
Aseri I brick	0.42
Aseri II brick	0.42
Aseri III brick	0.44
Misso Light brick	0.16
Misso Dark brick	0.23
Ash-blocks	0.25

Table 6. Range and mean values for ^{226}Ra, ^{232}Th, ^{40}K activity, and Ra(eq) (Bq kg^{-1}) for building materials under investigation in other countries

Country	^{226}Ra		^{232}Th		^{40}K		Ra(eq)	References
	Range	Mean	Range	Mean	Range	Mean		
Ireland	<1–140	32	<1–57	18	4–1977	353	–	Lee et al. (2004)
South Korea	6–27	–	<90	–	17–1081	–	–	Lee et al. (2001)
Sri Lanka	–	35	–	72	–	585	183	Hewamanna et al. (2001)
Pakistan	37–52	–	52–68	–	680–784	–	<370	Khan et al. (2002)
Nigeria	4–67	15.7	12–266	36	86–1073	253	100	Farai and Ademola (2005)
Jordania	10–340	116	3–210	64	2–1500	480	246	Ahmed (2005)
Turkey	18–59	–	7–20	–	66–249	–	–	Mavi and Akkurt (2010)
Malaysia	–	–	13–162	–	328–7541	–	79–1131	Ibrahim (1999)
World average for	50	–	50	–	500	–	–	UNSCEAR (1993)
building materials	35	–	30	–	400	–	–	UNSCEAR (2000)
Present work	4–69	33.5	1–82	37.5	7–678	384	118	

–, no data.

Table 7. Annual effective dose for natural radionuclide sources in concrete, calculated from mean activity concentrations for Estonia and for selected countries (UNSCEAR, 2000)

Country	Total annual effective dose, mSv
Greece	0.39
Egypt	0.41
Algeria	0.44
Switzerland	0.49
Russian Federation	0.49
United States	0.53
India	0.69
Sweden	0.78
Norway	0.86
Estonia	0.27

CONCLUSIONS

To control the indoor exposure caused by the ionizing radiation emitted from building materials, an accurate determination of their natural radioactivity is required. Assuming the ALARA principle, which requires that the indoor doses should be ‘as low as reasonably achievable’, the indices I have been proposed. These indices need to be considered in making decisions about the applicability of a building material in construction. Mean values of the index I of the studied building material samples range from 0.02 to 0.74, which means that all materials pass the accepted regulatory standards. The radium activity equivalent Ra(eq) values of the material samples range from 6 Bq kg^{-1} (gypsum board) to 239 Bq kg^{-1} (Siimusti building clay), both well below the value of 370 Bq kg^{-1}, which corresponds to an annual effective dose of 1 mSv.

The average effective dose annually received by the residents due to building materials varies from 0.16 to 0.44 mSv. Only a few doses are slightly larger than the worldwide average annual external effective dose of 0.41 mSv from natural indoor radiation sources (UNSCEAR, 2010).

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Ehitusmaterjalides leiduvatest looduslikest radionukliididest põhjustatud elanikudoosi hinnang

Merle Lust ja Enn Realo

Ehitusmaterjalides leiduvad looduslikud radionukliidid võivad olla väga olulisteks elanikudoosi tekitajateks. Käesoleva uurimistöö raames määrati looduslike radionukliidide sisaldus 53 erinevas Eestis kasutatava ehitusmaterjali proovis. Analüüsil kasutati kõrge eraldusvõimega HPGe gammaspektromeetrilist analüüsimeetodit. Leiti, et looduslike radionukliidide ^{40}K, ^{226}Ra ja ^{232}Th aktiivsuse kontsentratsioonid varieeruvad uuritud ehitusmaterjalides järgmistes vahemikes: vastavalt 7–747 Bq kg^{-1}, 4,4–69 Bq kg^{-1} ning 0,8–86 Bq kg^{-1}. Aktiivsuse kontsentratsioonide alusel hinnatud ehitusmaterjalide aktiivsusindeksi I väärtused asuvad piirides 0,02 kuni 0,74. Levinumate ehitusmaterjalide jaoks tehti doosihinnangud siseruumides ja selle alusel saadud aastased elanikudoosid jäävad vahemikku 0,16–0,44 mSv.