Chapter 4

Levels of Radon and Granite Building Materials

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Additional information is available at the end of the chapter

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Abstract

Natural radioactivities in granite building materials that are commonly used in the world have been surveyed two type methods: a passive measurement method and an active measurement method. In the passive measurement method, after sample preparation, the concentration of ^{226}Ra is measured in the samples. The measuring procedure carried out by using a gamma-ray spectrometry system with a high purity germanium detector. Also, in the active measurement method, an alpha GURD model PQ 2000 radon meter is used directly. In this method, a standard room is considered ((4.0 m × 5.0 m area × 2.8 height) that ground and walls have been covered with granite stones, we calculated radon concentration and radon exhalation rate. Moreover, the results of the exhalation rates measured by passive and active methods are compared and the results are the same, within errors better than 22%.

Keywords: building materials, natural radioactivity, granite, radon, exhalation

1. Introduction

Radioactive radon-222 (^{222}Rn) gas has a half-life of 3.8 days, which emanates from rocks and soils and tends to concentrate in enclosed spaces like underground mines or houses. Radon is a major contributor to the ionizing radiation dose received by the general population [1].

Most terrestrial materials contain ^{238}U and radon gas emitters from these materials, since ^{222}Rn is a decay product of Ra-226, which in turn is taken from the longer lived prior U-238. Concentrations of ^{238}U and ^{226}Ra in some terrestrial materials such as alum shale and black shale are high. Certain granites are typical of radioactivity-bearing natural materials, but it is always feasible to find natural radioactivity-rich bedrocks of different kinds such as construction materials. Construction materials that used as building materials are sources of indoor airborne radioactivity and external radiation from the decay series of uranium in buildings [2].

Short-lived radioelement outcomes from radon are the most important contributors to human exposure to ionizing radiation from natural sources. This contribution represents 50% of the total annual human dose. The parameters such as relative humidity, airflow, type of building materials, indoor-outdoor temperatures and ventilation rate as well as geological formations affect indoor radon concentrations. It is important to estimate the impress and contribution of the different building materials that can deed as Rn sources or Rn absorber inside of buildings for residence and work.

According some reports, the granite stones have a higher radon exhalation rate than other terrestrial materials on average [3–6].

2. Granite building materials

The most abundant of plutonic rock in mountain belts and continental shield areas were made by granite stones. Types of batholith stones that may in dwell thousands of (km^2) are usually intimately associated with granodiorite, quartz, gabbro and diorite. They are scratch resistant and extremely durable; their hardness lends themselves for the stone to be mechanically polished to a high gloss finish. They mainly contain of large grains of potassium, sodium feldspars and quartz. Granite is a plutonic rock in which quartz makes up between 10 and 50% of the felsic components and alkali feldspar accounts for 65–90% of the total feldspar content. Applying this definition requires the mineral identification and quantification abilities of a competent geologist. Other components of granites stones are hornblende and mica. A typical granite rock, in chemical view, is composed of 5% soda ($NaHCO_3$), 5% potassium oxide (KO_2), 12% aluminium (Al), 75% silica (SiO_2), as well as lime (CaO), iron (Fe), magnesia (Mg $(OH)_2$) and titanium (TiO_2) in minor quantities.

In general, it was widely believed that granite stones were formed mostly from magmatic differentiation of basaltic magma. The evidence of this exegesis was considered to indicate a metamorphic origin [7].

Figure 1 illustrates the generalized mineral composition of igneous rocks. Granites and rhyolites (compositionally equivalent to granite but of a fine grain size) are composed mainly of orthoclase feldspar, quartz, plagioclase feldspar, mica and amphibole.

2.1. Radon-222 in granite building materials

In nature, two types of radioactivity sources including technologically enhanced naturally occurring radioactive materials (TENORMs) and naturally occurring radioactive materials (NORMs) consist of materials, usually industrial wastes or by-products enriched with radioactive elements found in the environment, such as U, Th and K and any of their decay products, such as Ra and Rn. All natural products with terrestrial origin, especially granite stones, sand and minerals, include trace amounts of some NORM radioactive elements that can produce measurable amounts of radiation and sometimes Rn-222. This includes all concrete products, clay bricks, most non-plastic plates and dishes, coal and the fly ash produced in coal-fired power plants, natural gas (contains Rn), phosphate fertilizers used in your arable

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land (all contain K and small amounts of U and Th) and the vegetables grown using those fertilizers. All glasses made by using silica (even wine glasses, eye glasses, windows, mirrors, etc.) and granite stone too. **Figure 2** shows the percentage of radon source in residential place. As seen in figure, only 2.5% of radon gas is released from building materials, whereas granite building material has highest contribution to radon exhalation rate among other materials.

A fraction of the radon activity produced by decay of ^{226}Ra in building materials enters buildings by diffusion. The area exhalation rate can be expressed as

$$R = \lambda_{Rn} \rho_{Build} C_{Build,\,Ra}\, F_r L_{Rn} tagh\left(\frac{L_h}{L_{Rn}}\right) \tag{1}$$

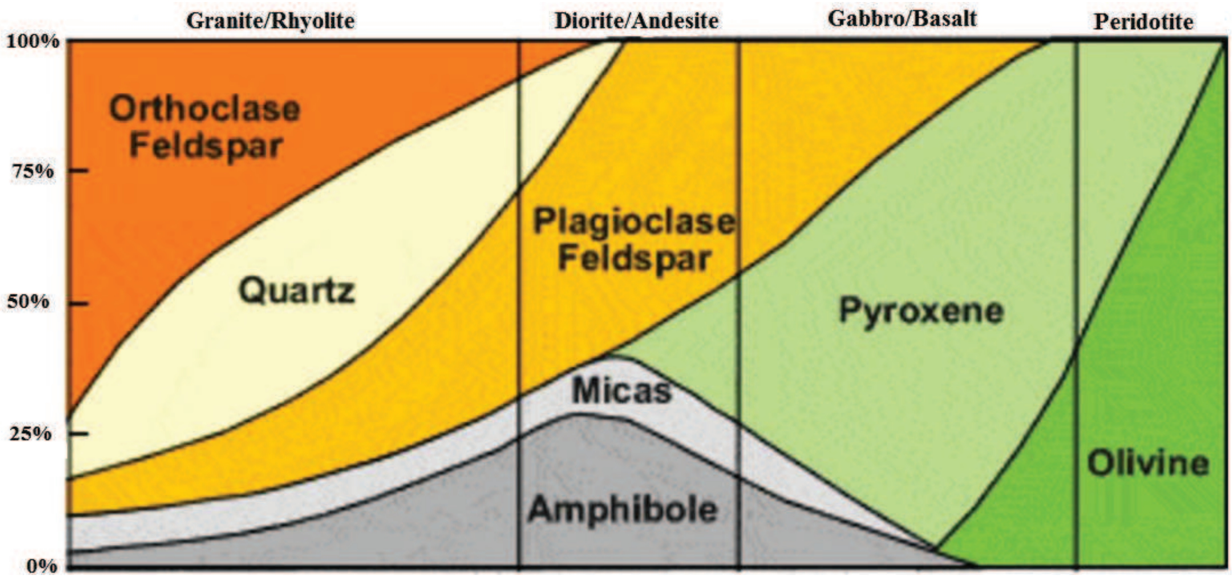

Figure 1. Granite composition diagram.

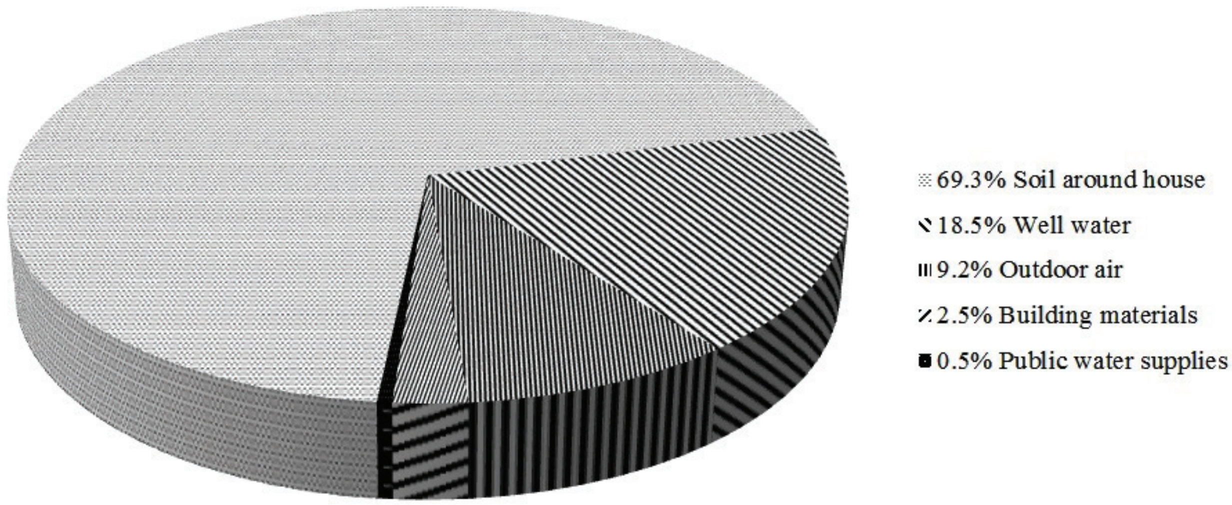

Figure 2. Radon source contribution in residential places.

which is an equation very similar to that related to soil, the only difference being the introduction of a hyperbolic term to account for the fact that diffusion takes place in a medium of finite thickness.

In this equation R is the area exhalation rate (Bq m^{-2} s^{-1}), F is the emanating power, $\boldsymbol{\rho_{build}}$ is the density of the building material (kg m^{-3}), $\boldsymbol{C_{Build,Ra}}$ is the activity mass concentration of ^{226}Ra in the building material (Bq kg^{-1}), $\boldsymbol{L_{Rn}}$ is the diffusion length (m) and $\boldsymbol{L_h}$ is the half thickness of a slab of building material (m).

The mass activity exhalation rate expressed in Bq kg^{-1} s^{-1} and defined as

$$R_m = \lambda_{Rn} C_{Build,Ra} F_r \tag{2}$$

is the quantity usually determined in laboratory measurements. The area exhalation rate from a wall or a floor made of building material of half-thickness $\boldsymbol{L_h}$, diffusion length $\boldsymbol{L_{Rn}}$ and density $\boldsymbol{\rho_{Build}}$ can then be expressed as

$$R = R_m \rho_{Build} L_{Rn} tagh\left(\frac{L_h}{L_{Rn}}\right) \tag{3}$$

The rate of entry of radon resulting from exhalation from building materials may be expressed as

$$U_{Bm} = \frac{N}{V}(2R_c S_F + R_b S_w) \tag{4}$$

where N is the number of seconds per hour, V is the volume of the reference house (250 m^3), $\boldsymbol{S_F}$ is the surface area of its floor or ceiling (100 m^{-2}), $\boldsymbol{S_w}$ is the surface area of its external walls (100 m^2) and $\boldsymbol{R_c}$ and $\boldsymbol{R_b}$ are the area exhalation rates from concrete and brick, respectively.

3. Measurement of radon-222 in granite building materials

Radon-222 concentration can be measured by calculation of exhalation rate from material surface. We offer two different methods to calculate the radon-222 concentration in building materials: passive method and active method.

3.1. Passive method

3.1.1. Theoretical approaches

In theoretical view, we consider a defined model. In this model, we assume that the release from materials brought into the room is negligible and radon gas is homogeneously mixed with the room air. Then, the concentration in a room can be found by solving the following equation:

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$$\frac{\partial C_i(t)}{\partial t} = E_X \frac{S}{V} + C_0\lambda_v - C_i(\lambda_{Rn} + \lambda_v) \tag{5}$$

where $C_i(t)$ is the ^{222}Rn radioactivity concentration in the home at time t in Bq m^{-3}, E_X is the ^{222}Rn exhalation rate per Bq (m^{-3} h^{-1}), S is the surface area that Rn gas is exhaling (m^2), V is the bulk of home (m^{-3}), λ_{Rn} is the ^{222}Rn decay constant and λ_v is the rate of air exchange at time t in h^{-1}.

This value range is among 0.1 h^{-1} and 3 h^{-1}for residence as air exchange rate value of 0.5 h^{-1} is suggested for residential mechanical ventilation systems (MVSs) according by UNSCEAR report [8]. The value C_0 is the outside ^{222}Rn concentration in the world average value (WAV) 10 Bq m^{-3} in the outside air [9]. At the steady state $\left(\frac{\partial C_i(t)}{\partial t} = 0\right)$, the ^{222}Rn concentration in the room is given by Anjos et al. [10]

$$C_{i=} \frac{E_x S/V + C_0\lambda_v}{(\lambda_{Rn} + \lambda_v)} \tag{6}$$

From the measured values of ^{226}Ra concentration, the radon exhalation rate per unit area can be calculated by

$$E_x = \frac{1}{2} A_{Ra}\lambda\rho\eta d \tag{7}$$

where ρ is the material density (kg m^{-3}), d is the wall thickness (m) and η is the emanation coefficient. The emanation coefficient value was to be reported between 6.04 and 2.54% (average: 4.29%) for granite stones [11].

The presence of water in building materials alters the transport condition; therefore, the above equation is valid only for dry conditions [12].

3.1.2. External γ-ray radiation dose rate

The activity concentration and indoor γ-ray radiation dose rate are calculating for a rectangular source shape with uniform density. The external γ-ray dose rate is calculating in the middle of a standard room with dimension (5.0 m × 4.0 m × 2.8 m) by summing the separately calculated γ-ray dose rates caused by walls and floor. The specific indoor dose rates depended on a large wall thickness and density of materials. It does not depend on the position in the room and dimensions of the room [13].

The density of granites stones is calculating by the experimental method in laboratory. This value approximately is 2580 kg m^{-3}, on average. The granite in markets is usually 3.0 cm thick and (30.0 cm × 50.0 cm) dimension. Also, the dose rate conversion factor is calculated by calculations based on the point kernel integration method for floor covered with 3.0 cm thick granite [14]. The free-in-air absorbed dose value in the middle of the room can be expressed as [10]

$$D(nGy.\, h^{-1}) = K_K A_K + K_{Ra} A_{Ra} + K_{Th} A_{Th} \tag{8}$$

where D is absorbed dose in the center of the room, A_K, A_{Ra} and A_{Th} are the activity concentration (Bq kg^{-1}) of ^{40}K, ^{226}Ra and ^{232}Th, respectively. Coefficients of K_K, K_{Ra} and K_{Th} are their dose conversion factors or specific dose rates in nGy h^{-1} per Bq kg^{-1}.

3.1.3. Experimental procedure

3.1.3.1. Sample preparation

The collected samples are pulverized, sieved through 0.2 mm mesh, sealed in standard 1000-ml Marinelli beakers, dry-weighed and stored for 4 weeks before counting in order to allow the reaching of equilibrium between ^{226}Ra and ^{222}Rn and its decay products. It is assumed that the radionuclides in equilibrium, i.e. the activity of each daughter, were equal to the initial isotope of the series.

3.1.3.2. Sample counting

The γ-ray spectra of the prepared samples are measured using a γ-detector. This detector is a typical high-resolution γ spectrometer based on a coaxial P-type shielded high purity germanium (HPGe) detector. Efficiency of this detector is 80%with a relative photopeak and energy resolution is 1.80 keV full-width at half-maximum (FWHM) for the 1332 keV γ-ray spectra of ^{60}Co, coupled to a high count-rate multi-task 16k multi-channel analysis (MCA) card. Also, the Gamma-2000 commercial software is used for data analysis. For sample counting, each sample is put into the lead shielding container of the HPGe detector and measured for a collect time of 24 hours. Prior to the samples measurement, the environmental background γ-radiation at the counting place site is determined with a blank Marinelli beaker under identical measurement conditions. It is later subtracted from the measured γ-ray spectra for each sample. The ^{232}Th, ^{238}U, ^{226}Ra and ^{40}K activity concentrations are calculated for each of the measured samples together with their corresponding total uncertainties. ^{232}Th activity concentration are measured by taking the mean activity of photopeaks of the decay nuclides ^{228}Ac (968, 911 and 338 keV) and ^{212}Pb (238 keV). On the other hand, ^{226}Ra activity concentrations are calculated from the activity of its short-lived daughter's radionuclide ^{214}Pb at 351 and 295 keV and ^{214}Bi at 609.6 keV. Activity concentrations of ^{40}K are determined directly from its gamma emission at 1460 keV [17].

To compare results of uncertainty, standard materials such as soil-375 and RGK-1 (K_2SO_4), RGU-1 (U-ore) and RGTh-1 (Th-ore) are used, and all obtained results in accordance are completed. The activity levels of ^{226}Ra and ^{232}Th in soil-375 are 424, 20 and 20.5 Bq kg^{-1}, respectively. Also, activity levels of ^{40}K in RGK-1, ^{238}U in RGU-1, ^{232}Th in RGTh-1 are 14,000, 4940, 3250 Bq kg^{-1}, respectively.

For lower limit of detection (LLD), 96% confidence is calculated using a relation of LLD = 4.66 $(F_c)^{1/2}$, where F_c is the Compton background in the region of the selected gamma-ray spectrum.

Figure 3 shows a type of high purity germanium detector (HPGe) set-up and accessories parts.

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Figure 3. A high purity germanium detector (HPGe) set-up used in the passive method.

3.2. Method of active measurement

To measure radon-222 gas in the active measurement method, an alpha GURD model PQ-2000 detector is used. This device works based on the passing of radon-222 gas from a filter to an ionization chamber. Rn-222 surveys are carried out in a special cubic chamber (70 × 50 × 60 cm) with different changeable walls. The walls have different covering materials on the internal surfaces. One set of floor is covered with the most common granite stones. Each sample is put into a special cubic chamber in floor and half walls and measuring for an accumulating time of

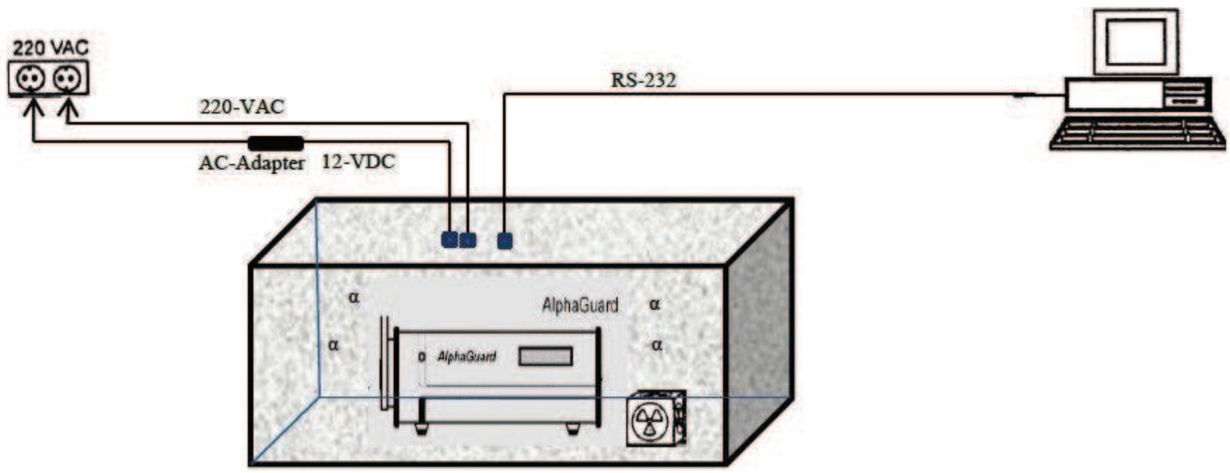

Figure 4. Schematic diagram showing the radon exhalation measurements of granite samples by the active setup method [1].

$t = 90$ min. **Figure 4** shows the schematic diagram of Rn-222 exhalation rate measuring in the active setup method. The background value is subtracted from the measured Rn-222 activity concentration level for each sample. All samples measure in n times to get average results. By Alpha View-Expert software, final activity concentration of Rn-222 gas (A_0) is computed. Rn gas exhalation rate was computed as

$$E_x = A_0 \lambda \left(\frac{V}{F}\right) \tag{9}$$

where E_x is the Rn gas exhalation rate (Bq m^{-2} h^{-1}), A_0 is the final activity of Rn gas (Bq m^{-3}), λ is Rn decay constant (7.567×10^{-3}), V is the emanation container volume (m^3) and F is the area of each sample (m^2).

4. Level of Rn-222 in granite building materials

The level of Rn-222 activity concentrations in dwellings approximately is normal, with a trend for high concentrations to lie above those predicted by this distribution. According to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 2000) [15], the worldwide concentration and population-weighted values of these parameters for dwellings is found to be 25 and 2.5, respectively. Rn-222 concentrations in dwellings are variable between countries because of differences in climate and geology, in techniques and construction materials and in domestic customs. The arithmetic means of radon concentration level for countries various from 12 to 140 Bq m^{-3}. Rn-222 activity concentration levels for some European countries are given in **Table 1** [18].

The rate of entry of radon from the floor and the ceiling of the reference house would amount to about 6 Bq m^{-3} h^{-1}, while the contribution from the external walls would be about 0.4 Bq m^{-3} h^{-1}. The contribution from radon exhalation from the building materials to the radon concentration in the reference house is thus estimated to be 6.4 Bq m^{-3} h^{-1}, if the air exchange rate is taken to be 1 h^{-1} [12].

Considerably greater values of the rate of entry of radon are expected to be obtained when building materials with high ^{226}Ra concentrations and normal emanating power are extensively used. Examples of such building materials are granite, Italian tuff and alum-shale lightweight concrete. Among these materials, Swedish alum-shale lightweight concrete has the highest ^{226}Ra concentrations (about 1300 Bq kg^{-1} on average) and probably the highest ^{222}Rn mass exhalation rate (440 μBq kg^{-1} s^{-1}). Assuming a density of 2000 kg m^{-3} and a diffusion length of 7.4×10^{-2} m with 0.2 m thick slab of 100 m^2 would lead to a rate of entry of radon of about 80 Bq m^{-2} h^{-1} in the reference house [16].

Techniques for reducing the radon entry rate due to exhalation from building materials have been investigated. In Sweden, aluminium foil has been applied to the walls of houses built

Country	Number of houses sampled	Period of exposure	Duration of exposure	Sample characteristics	Radon-222 concentration (average) (Bq m^{-3})
Belgium	300	1984–1990	3 months to 1 year	Population-based (selected acquaintances)	48
Czechoslovakia	1200	1982	Random grab sampling	–	140
Demark	496	1985–1986	6 months	Random	47
Finland	3074	1990–1991	1 year	Random	123
France	1548	1982–1991)	3 months (using open alpha track detectors)	Biased (not stratified)	85
Germany	7500	1978–1984 1991–1993	3 months 1 year	Random	50
Greece	73	1988	6 months	—	52
Hungary	122	1985–1987	2.5 years	Preliminary survey	55
Ireland	1259	1985–1989	6 months	Random	60
Italy	4866	1989–1994	1 year	Stratified random	75
Luxembourg	2500	1991	–	–	65
Netherlands	100	1982–1984	1 year	Random	29
Norway	7525	1987–1989	6 months	Random	60
Portugal	4200	1989–1990	1–3 months	Volunteers in a selected group (high school students)	81
Spain	2000	Winter of 1988–1989	Grab sampling	Random	86
Sweden	1360	1982–1992	3 months in heating season	Random	108
Swaziland	1540	1982–1990	3 months (mainly in winter)	Biased (not stratified)	70
United Kingdom	2093	1986–1987	1 year	Random	20.5

Table 1. Radon-222 levels in dwellings of some European countries [18].

with aerated concrete based on alum shale; the results showed a 50% reduction in the radon entry rate. In the United States, various radon sealants have been tested under conditions representative of normal construction conditions. The radon exhalation rate was reduced from 20 to 80% depending on the surface coating used. Because of the trapping of radon decay products in the wall, there is a relatively small increase in the external gamma dose rate. It was also noted that any cracks that later developed in a sealant such as paint may lead to leaks that negate a large portion of the sealing effectiveness of the paint. Similarly, the radon exhalation rate from any unpainted areas may be increased as they offer to radon a path of least resistance in comparison with painted areas. Radon-222 source with characteristics for building materials is shown in **Table 2** [12].

Material	Country	Number of sampling	Radon-222 mass exhalation rate (μBq kg^{-1} s^{-1})
Concrete			
Heavy concrete	USSR	18	3.2
Light weight concrete	USSR	19	4.1
Ordinary concrete	Sweden	3	11–31
Aerated concrete based on alum shale	Sweden	1	580
Alum-shale concrete	Denmark	1	440
Fly-ash concrete (4%)	USA	8	10
Fly-ash concrete Greece	Greece	4	6.4–20
Concrete	Hungary	100	7.8
Concrete	Denmark	4	4.7
Concrete	Norway	137	9.5–16
Concrete	Greece	–	2.9–5
Concrete	USA	50	2.5–20
(Adopted reference value)	–	–	10
Brick			
Red brick	USSR	12	1.6
Red brick	Hungary	200	3.9
Red brick	Poland	3	18
Red brick	USA	6	1
Brick	Denmark	2	0.17
Brick	Norway	18	4.2
Brick	Greece	5	0.3–7.5
Silicon brick	Poland	3	1.8
Adobe brick	USA	2	3.5
(Adopted reference value)	–	–	2
Gypsum			
Gypsum	USA	12	6.3
Gypsum board	Denmark	1	0.23
By-product gypsum (apatite)	Poland	1	2.6
Lightweight expanded			
Clay aggregate	Norway	12	5.2
Storage rock			
Storage rock	USA	9	5
Wood			
Wood	USA	2	0.2

Material	Country	Number of sampling	Radon-222 mass exhalation rate (μBq kg^{-1} s^{-1})
Sand			
Sand	USA	2	12
Sand	USA	2	3
Gravel			
Gravel	USA	4	2.2

Table 2. Radon-222 source with characteristics for building materials [12].

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