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Non-destructive measurements of natural radionuclides in building materials for radon entry rate assessment

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Abstract
To control the specific activity of ^{226}Ra in building materials of operated buildings, a non-destructive in situ method consisted in measurements of gamma spectra with a fixed geometry of the detector position in a room is suggested. The calculation of the average specific activity of natural radionuclides in building materials is carried out by comparing the calculated flux density of unscattered gamma quanta normalized to the specific activity of 100 Bq/kg, and the experimentally measured count rates in the photopeak. For the measured values of the average specific activity of ^{226}Ra in building materials, the expected values of radon concentration in rooms were calculated. The developed method has been verified for rooms in modern multi-storey energy-efficient buildings. Concentrations of ^{232}Th and ^{40}K in building materials which contribute to the indoor external radiation exposure can be measured by developed method also.

Keywords Radium · Specific activity · Building materials · Gamma spectrometry · Radon diffusion · Radon concentration

Introduction

According to numerous estimates, natural sources of ionizing radiation provide the highest radiation effective dose to the population [1]. At the same time, people spend most of their time indoors, so the main radiation exposure from natural radionuclides occurs either in dwellings or in ordinary workplaces. The indoor radiation exposure takes place not only from external gamma radiation of radionuclides of the uranium or thorium series and ^{40}K. Indoor radiation doses from inhalation of radon and its decay products is usually many times higher than external doses from natural radionuclides [1]. During many years the soil under the building have been considered as the main source of radon entry in dwellings and ordinary workplaces [1–7]. Diffusion of radon from building materials was not considered as the dominant source of indoor radon. The building materials were regarded as dominating radon source for buildings which constructed from materials with abnormally high concentrations of ^{226}Ra (Sweden, Czech Republic) [8–11].

However, over the past decade, there have been a number of publications devoted to high levels of radon in modern energy efficient multi-storey buildings with a low air exchange rate. Since the radon concentration above 100 Bq/m^3 is impossible to be achieved in a multi-storey building (above nine storeys) due to the radon exhalation from the ground, it was suggested that the dominant source of radon in such buildings is the building materials [12].

Radon survey in different Russian cities demonstrated that average indoor radon concentration in new energy efficient buildings is higher than in elder conventional houses [13]. However, it was noted that the average indoor radon concentration was significantly different by cities when considering the buildings of higher energy efficient classes. It was suggested that such difference is associated with the ^{226}Ra content in building materials.

There are indirect evidences that high radon concentrations in the air of energy efficient buildings can be observed when the concentration of ^{226}Ra in building materials is about 50 Bq/kg [14–16]. Such building materials meet hygienic standards and cannot be considered as materials with an increased concentration of natural radionuclides.

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For further developing a strategy on protection of population against radon exposure in new energy efficient buildings it is necessary to obtain scientifically based and experimentally approved relationship between indoor radon concentration and ^{226}Ra content. While the indoor radon concentration may be directly measured, there is still no accurate non-destructive method of ^{226}Ra concentration measurements in the building materials of the existing building.

The simplest and the most traditional method to measure the specific activity of natural radionuclides in building materials is to take samples of materials, crush them, and measure their activity using a gamma spectrometer. Obviously, the application of such approach is possible only in rare cases, when large-scale renovation work is carried out in the apartment, as a result of which numerous fragments of wall materials and, less often, building floors become available. In most cases of normally operated apartments, this approach is not applicable.

The main objectives of this work are:

- Development of non-destructive method for determining the average specific activities of natural radionuclides in building materials.
- Conducting measurements of the specific activities of natural radionuclides in rooms in which long-term measurements of radon concentration were previously carried out using track detectors or a radon monitor.
- Comparison of radon measurements with the calculated values of radon concentration based on measurements of the specific activities of radionuclides in building materials using a field gamma spectrometer.

Materials and methods

The main measuring instrument in this work was the field gamma spectrometer MKS-AT6101DR, manufactured by ATOMTEX, Belarus [17]. The spectrometer uses a NaI (Tl) crystal with a diameter and a height of 63 mm as a detector. The signal from the detector is processed by a 1024 channel analyzer. This spectrometer is mainly designed for:

- measurement of ^{134}Cs and ^{137}Cs surface contamination and specific activity in soils;
- measurement of ^{137}Cs, ^{134}Cs and ^{131}I specific activity in water, foodstuffs, agricultural and forestry products and liquid radioactive wastes;
- determination of ^{40}K, ^{226}Ra and ^{232}Th natural radionuclides content;
- radionuclide identification: ^{134}Cs, ^{137}Cs, ^{131}I, ^{40}K, ^{226}Ra and ^{232}Th;
- measurement of gamma radiation ambient dose equivalent rate.

Taking into account the specifics of its main application, the spectrometer is calibrated for measurements in the 2π geometry for a semi-infinite space. In some cases, for measurements in near-surface wells, calibration in the 4π geometry can be used to determine the specific activity of the soil. Obviously, standard calibrations of the gamma spectrometer cannot be used for measurements in apartments and offices, where six volumetric gamma radiation sources are located at different distances from the detector (floor and ceiling slabs and four walls). Therefore, it was necessary to use a different measurement scheme and to perform the appropriate calibration for this scheme. The most accurate measurement of the specific activity of natural radionuclides in each of the walls (possibly made of different materials with different specific activity of natural radionuclides) and floor slab could be carried out when the detector, surrounded by a lead shield, is installed close to the wall or floor slab. For multi-storey buildings it can be assumed that the specific activity of radionuclides in the floor slab is equal to the specific activity in the ceiling slab. With such measurement setup, the thickness of the lead shield must be enough to shield radiation from all sources in the room, except for the object under study (wall or floor slab). Calculations have shown that to meet this requirement, the weight of the lead shield must be 120 ± 20 kg. Therefore, the use of this measurement scheme, especially for large-scale studies, was rejected as unrealizable.

Thus, in this work, the approach was proposed, which consists of the following stages:

- Calibration of the photopeak efficiency of the corresponding radionuclide to the flux density of unscattered gamma quanta by standard radionuclide sources.
- Calculation of the flux density of unscattered gamma quanta in a room, normalized to the unit specific activity of the corresponding radionuclide in building materials. The calculation should be made for the real geometry of the room (length, width, height, thickness of walls and floor slabs), taking into account the presence of door and window openings.
- Measurement of gamma spectrum in the examined room with fixing the exact location of the detector during measurements
- Calculation of the average specific activity in the building materials of the studied room by comparing the measured count rate in the photopeak with the calculated flux density of unscattered gamma quanta in the room, normalized to a unit specific activity.
- Calculation of the radon flux into the room due to its diffusion from building materials and assessment of the expected radon concentration in the air for different values of the air exchange rate in the room.

This approach assumes that the specific activities of natural radionuclides in the materials of walls and floor slabs of the floor and ceiling differ insignificantly. In most practical cases, this assumption is fair enough for a general assessment. Internal walls, obviously free of natural radionuclides (wooden walls, glass-metal panels) are excluded from calculations.

Calibration of the photopeak efficiency

Considering the possible range of scientific problems to be solved in buildings using a field gamma spectrometer, it was decided to calibrate its photopeak efficiency for the three main natural radionuclides: ^{226}Ra, ^{232}Th and ^{40}K. For each of the radionuclides, the most characteristic emission lines in their decay chain were selected: ^{226}Ra—609 keV (^{214}Bi), ^{232}Th—2614 keV (^{208}Tl), ^{40}K—1460 keV. For calibration, we used reference ^{226}Ra source with activity of 508 kBq (^{214}Bi/^{226}Ra ratio 0.59) and ^{228}Th source with activity of 23.1 kBq. The uncertainty of the ^{226}Ra activity was 5%, the uncertainty of the ^{214}Bi/^{226}Ra ratio was 3% ($k=2$). The uncertainty of the ^{228}Th activity was 3% ($k=2$). It is well known that it is impossible to create a standard ^{40}K point source. Therefore, the calibration of the spectrometer for sensitivity to gamma quanta with energy of 1460 keV was carried out using the emission line of ^{152}Eu 1408 keV, which is closest to required energy. The influence of the difference in the absorption coefficients for different energies on the detection efficiency in the photopeak was subsequently taken into account. According to our estimates, the difference in energies between the emission lines of ^{40}K and ^{152}Eu result in a decrease in the detection efficiency in the photopeak for ^{40}K in comparison with ^{152}Eu not more than 1.5%. A standard ^{152}Eu source with an activity of 98.6 kBq was used during the calibration. The uncertainty of the activity of this source is 3% ($k=2$). All reference sources were certified at the D. I. Mendeleev Institute for Metrology (St. Petersburg).

When calibrating the gamma spectrometer, the radium source was located at the distance of 100 cm from the center of the detector. For other sources, considering their lower activity, a distance of 50 cm was chosen. Nevertheless, in all cases, the flux of gamma quanta could be considered as monodirectional. The measurements were carried out at different angles to the detector axis. In this case, an angle equal to 0 degrees meant the incidence of radiation on the front of the NaI (Tl) crystal, and an angle of 180 degrees was understood as radiation that passed through the entire body of the spectrometer.

Calculation of the flux density of unscattered gamma quanta in the room

To calculate the flux density of gamma quanta, each of the sources (wall or slab of floor or ceiling) was modeled with a rectangular parallelepiped of the corresponding size. The data on the dimensions of the room and the thickness of the walls and slabs of the floor and ceiling were taken from the documentation for the apartment. The exact location of the detector was determined using a laser rangefinder. To take into account windows and doors in the walls, the fluxes of photons from sources of appropriate sizes were calculated. Further, the calculated values were subtracted from the flux density of gamma quanta from the corresponding source.

The gamma quanta flux density was calculated using the MicroShield 11.2 software [18]. Each volume source (slab or wall) was simulated by a superposition of $10\times10\times1$ cm cells, considered as the point sources, located in a gamma-absorbing medium.

Measurement of the average specific activity of natural radionuclides in the building materials of the room

To test the developed method of non-destructive measurement of specific activities of natural radionuclides in building materials, gamma-ray spectra were measured by gamma spectrometer in some rooms. The measurement duration was from 30 to 60 min. If possible, repeated measurements were taken. When processing the spectra obtained in the course of measurements, the number of counts in the 609, 1460, and 2614 keV photopeaks was calculated and the count rate I_{meas} in the photopeaks was determined. By multiplying the calculated flux density of gamma quanta by the experimentally determined sensitivity of the detector, the calculated count rate I_{calc} in the corresponding photopeak, normalized to the specific activity of 100 Bq/kg of ^{226}Ra, ^{232}Th or ^{40}K was obtained. Further, the calculation of the average specific activity of the corresponding radionuclide was carried out as:

$$\overline{A_m} = \frac{I_{\text{meas}}}{I_{\text{calc}}} \times 100,\ \text{Bq/kg}. \tag{1}$$

Calculation of the diffusion radon flux into the room from building materials

With the diffusion transfer mechanism, the release of radon and thoron from soil or building materials is described by diffusion equations. The rate of radon exhalation from a material with a specific activity of radium C_{Ra} is described by the Eq. (2) [19]:

$$q = C_{\text{Ra}}\rho E\sqrt{\frac{\lambda_0 D_{\text{e}}}{p}}\tanh\left(d\sqrt{\frac{\lambda_0 p}{D_{\text{e}}}}\right), \tag{2}$$

where q is radon exhalation rate, Bq/m^2 s; ρ is the density of the material, kg/m^3; E is the coefficient of emanation; λ_0 is the decay constant of radon, s^{-1}; D_e is the effective diffusion coefficient by volume of the material, m^2 s^{-1}; p is the material porosity; d is half of the thickness of the material layer, m.

If we assume that radon is homogeneously distributed in the room due to air mixing, then the process describing the change in the Rn concentration in the room can be given by the Eq. (3):

$$\frac{dC_{Rn}}{dt} = \frac{\sum_{i=1}^{6} q_i S_i}{V} + C_{Rn}^{outdoor}\lambda_v - C_{Rn}\left(\lambda_v + \lambda_0\right), \quad (3)$$

where S_i is the area of corresponding wall, floor or ceiling; $C_{Rn}^{outdoor}$ is radon concentration in outdoor air; λ_v is air exchange rate with outdoor atmosphere. As a result of solving Eq. (3), we obtain:

$$C_{Rn} = \left(\frac{\lambda_v}{\lambda_0 + \lambda_v}\right)\left(C_{Rn}^{outdoor} + \frac{\sum_{i=1}^{6} q_i S_i}{\lambda_v V}\right)\left\{1 - \exp\left[-\left(\lambda_0 + \lambda_v\right)t\right]\right\} + C_{Rn}^{indoor}(t=0)\exp\left[-\left(\lambda_0 + \lambda_v\right)t\right], \quad (4)$$

where $C_{Rn}^{indoor}(t=0)$ is indoor radon concentration in the moment of air exchange rate change. As a rule, in dwellings, the air exchange rate is within $0.1 < \lambda_v < 2.0$ h^{-1}, i.e. $\lambda_v >> \lambda_0$. Therefore, for steady state conditions Eq. (3) can be written in the form:

$$C_{Rn} = C_{Rn}^{outdoor} + \frac{\sum_{i=1}^{6} q_i S_i}{\lambda_v V}. \quad (5)$$

For fixed values of all considered parameters, calculations using Eqs. (1)–(4) are not difficult. To assess the uncertainty in the estimation of the radon concentration caused by the unknown values of the radon diffusion coefficient, the emanation coefficient, porosity and uncertainty in assessment of the thickness of the floor and ceiling slabs from monolithic concrete, the RESRAD-Build 3.5 software package was used where these parameters were varied randomly [20]. The uniform distribution was assumed for such parameters as emanation coefficient, porosity and slab thickness. For diffusion coefficient the triangle distribution was selected. The RESRAD-Build 3.5 software. The types and numerical values of the distribution parameters are given below in the Results and Discussion section (subchapter Uncertainty Analysis). The program randomly selects the values of the variable parameters in accordance with the given distribution and calculates the radon concentration in the room. The calculation is repeated 180 times and the result is the expected distribution of radon concentration. The final result of the calculations of the RESRAD-Build 3.5 software is the values of the effective dose and the uncertainties of its estimates for a person in a building containing various sources of ionizing radiation. However, in the process of calculations, the program creates intermediate output files in which the values of the radon concentration and its decay products ^{218}Po, ^{214}Pb and ^{214}Bi are given. Therefore, to estimate the concentration of radon in the room, we used not the reverse calculation from the final value—the effective dose, but the results of direct calculations of radon concentration given in the output files.

Measurements of radon concentrations

To compare the calculated values of the radon concentration with the real values in the examined rooms, long-term measurements of the radon concentration were carried out. In two rooms, measurements of radon concentration were performed using a radon monitor AlphaGUARD PQ2000 PRO, in the third room, measurements were made using track detectors RSKS manufactured by RadoSys. The measurements were carried out in winter period; the duration of the measurements was 3 months.

Results and discussion

Calibration of the registration efficiency in the photopeak

Typical calibration spectra for ^{226}Ra and ^{228}Th are shown in Fig. 1. This figure also shows the background spectrum typical for the room in which the calibration was carried out.

During the calibration, the radiation environment in the room was unchanged. The dose rate of the background gamma radiation, measured with a calibrated gamma spectrometer, was 100 ± 14 nSv/h. The background spectra, normalized to measurement duration of 1 h, completely matched (Fig. 2).

To improve the calibration accuracy during the calculation the sensitivity of the spectrometer in the photopeak to quanta of the corresponding energies, the background spectrum, averaged over the entire calibration period, was subtracted from the calibration spectra obtained using reference sources. The angular dependence of the detector sensitivity is shown in Fig. 3. In general, the angular dependence of the detector sensitivity in the photopeak is weak. The effect of shielding by the body of the

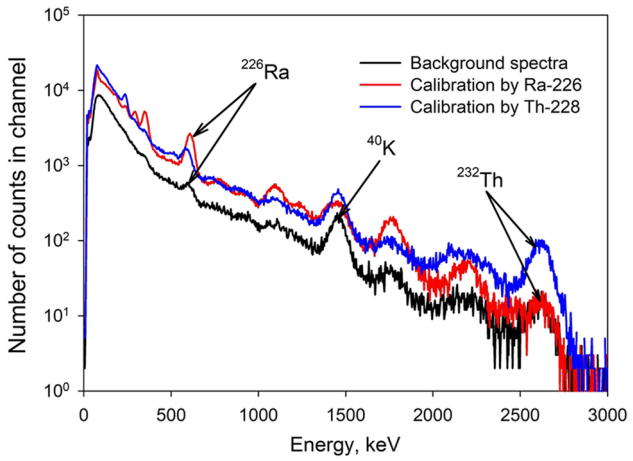

Fig. 1 Typical background spectrum and calibration spectra for ^{226}Ra and ^{228}Th

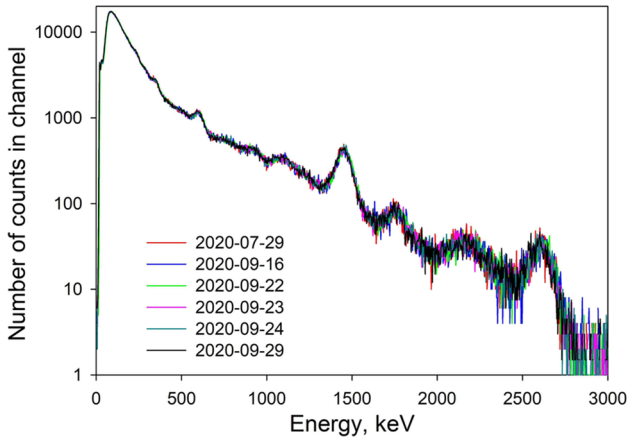

Fig. 2 The background spectra, normalized to 1 h measurement time

spectrometer can be observed to the greatest extent for the 609 keV emission line, which is quite logical to expect.

During the measurements in the room, the gamma radiation interacts with the detector at different angles close to the 4π geometry. Since the exact angular distribution of gamma radiation is practically impossible to determine, in our research we used the averaged values of the sensitivity in the photopeak, normalized to the unit flux density of unscattered gamma quanta of the corresponding energies. The direction corresponding to an angle above 135 degrees (the passage of quanta through the body of the spectrometer) was excluded from consideration because it corresponds to less than 15% of full solid angle 4π. The average values of the spectrometer sensitivity in the photopeak were for ^{226}Ra $\varepsilon = 11.7 \pm 1.0$ counts/cm^2, for ^{40}K $\varepsilon = 4.33 \pm 0.79$ counts/cm^2, for ^{232}Th $\varepsilon = 2.13 \pm 0.09$ counts/cm^2 (one standard deviation is given).

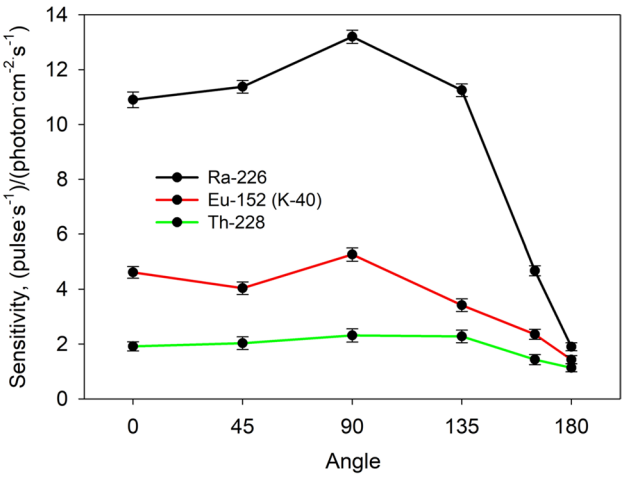

Fig. 3 The angle dependence of the detector sensitivity. The 95% confidence intervals of the extended ($k = 2$) standard total uncertainty due to the influence of the Poisson statistics of the number of counts in the photopeak and the uncertainty of calibration of reference sources are shown

Calculation of the flux density of unscattered gamma quanta in the room

The assessment of the specific activity of natural radionuclides in building materials was carried out in one office and two residential premises in which the average radon concentration was measured using track detectors or a radon monitor AlphaGUARD.

The Room 1, located on the second floor of a two-story building (Ekaterinburg city), was a manager's office in which the calibration of the gamma spectrometer using reference sources was carried out. The building was built in the late 1970s—early 1980s. The dimensions of the room are $6.50 \times 5.84 \times 2.75$ m ($L \times W \times H$). The two walls are all glass panels across the entire width of the wall from floor to ceiling. The other two walls, 15 cm thick, are made of bricks. Each of the walls has a standard 200×100 cm doorway. The floor and ceiling slabs are concrete panels 22 cm thick and contained internal cylindrical voids. The exact position of the detector (near the center of the room at the distance 75 cm from the floor), as well as the position of the doorways, was determined using a professional construction laser distance meter. The dose rate of the gamma radiation, measured by gamma spectrometer, was 100 ± 14 nSv/h.

Considering the presence of voids both in the floor and ceiling slabs and in the bricks, the effective values of the density of building materials were used in the calculations: 1.39 kg/dm^3 for ceilings, 1.60 kg/dm^3 for bricks. For the location of the detector for the room under study, the flux densities of gamma quanta with energies of 609, 1460, and 2614 keV from natural radionuclides in building materials were calculated. The calculated values of

the flux density of gamma quanta normalized on specific activity 100 Bq/kg of each of the radionuclides were: for 609 keV—0.433; for 1460 keV—0.136; for 2614—0.542 photon/(cm^2 s), respectively.

The Room 2 is located in a three-room apartment on the 13th floor of a 16-storey residential building (Ekaterinburg city). The building was built in 2012 using the technology of monolithic concrete. The dimensions of the room are $4.59 \times 3.62 \times 2.54$ m ($L \times W \times H$). The floor and ceiling slabs are 22 cm thick. Two 15 cm thick load-bearing walls are also made of monolithic concrete, one 15 cm thick wall with door is made of bricks, and the opposite wall, 19 cm thick, opening onto the balcony, is made from aerated concrete blocks. During the calculations, a density of 2.4 kg/dm^3 was taken for monolithic concrete, 1.6 kg/dm^3 for brick, as in the previous case, and 0.5 kg/dm^3 for aerated concrete. The dose rate of the gamma radiation, measured by gamma spectrometer, was 147 ± 22 nSv/h.

The measurements were taken near the floor in the center of the room at an equal distance from the walls. The flux densities of gamma quanta with energies of 609, 1460, and 2614 keV were calculated for a given room size and detector location. As in the previous case, the flux densities of gamma quanta were normalized to the specific activity of the considered natural radionuclides 100 Bq/kg. The calculated values of the flux density of gamma quanta were: for 609 keV—0.535; for 1460 keV—0.174; for 2614—0.732 photon/(cm^2 s), respectively.

The Room 3 is located in an apartment on the 7th floor of a 16-storey building (Tyumen city). The building was built in 2016 using the technology of monolithic concrete. The dimensions of the room are $7.44 \times 3.80 \times 2.60$ m ($L \times W \times H$). The room 3 is a living room combined with a kitchen. The outer long wall is made of 30 cm thick aerated concrete. One of the short walls (3.6 m), located at a greater distance from the detector, is made of monolithic concrete 22 cm thick. The other short wall is made of 15 cm brick. The long inner wall is also made of brick, except for a 1.75 m section of 22 cm thick monolithic concrete. The door to the room is in the brick part of the wall opposite the location of the detector of the gamma spectrometer. The dose rate of the gamma radiation, measured by gamma spectrometer, was 38 ± 14 nSv/h.

The location of the detector was on the axis of the room but was offset from the center of the room. As in the previous case, the detector of the spectrometer was located near the floor and the calculations of the flux density were carried out for this geometry. The calculated values of the flux density of gamma quanta normalized to the specific activity of the considered natural radionuclides 100 Bq/kg were: for 609 keV—0.528; for 1460 keV—0.174; for 2614—0.731 photon/(cm^2 s), respectively.

Measurement of the average specific activity of natural radionuclides in the building materials of the room

For the Room 1 during the calibration of the gamma spectrometer, six measurements of background spectra were carried out, which were in good agreement with each other (Fig. 2). Due to the availability of good measurement statistics, the analysis was carried out using the spectrum averaged over all measurements and normalized to the measurement time 3600 s. The experimentally measured count rates in photopeaks for the energies of 609, 1460, and 2614 keV were 1.67, 2.59, and 0.22 counts/s, respectively. Based on the data presented, taking into account the previously performed calibration of the sensitivity of the gamma spectrometer, the average specific activities of natural radionuclides in building materials were calculated: ^{226}Ra—32.9 Bq/kg; ^{232}Th—19.0 Bq/kg; ^{40}K—440 Bq/kg, respectively. These values generally correspond to typical concentrations of natural radionuclides in building materials used in Ekaterinburg (former Sverdlovsk) during the construction of this building (70 s–80 s of the twentieth century) [21].

For the Room 2 the experimentally measured count rates in photopeaks for the energies of 609, 1460, and 2614 keV were 2.77, 3.78 and 0.29 counts/s, respectively. The average specific activities of natural radionuclides in building materials were: ^{226}Ra—44.3 Bq/kg; ^{232}Th—18.6 Bq/kg; ^{40}K—502 Bq/kg, respectively. These specific activities are in good agreement with the typical concentrations of natural radionuclides in building materials used in Ekaterinburg for construction of multi-storey buildings at the beginning of the twenty-first century [14].

For the Room 3 the experimentally measured count rates in photopeaks for the energies of 609, 1460, and 2614 keV were 0.89, 1.76, and 0.142 counts/s, respectively. The average specific activities of natural radionuclides in building materials were: ^{226}Ra—14.4 Bq/kg; ^{232}Th—9.1 Bq/kg; ^{40}K—234 Bq/kg, respectively. It should be noted that there is a large difference in the average specific activities of natural radionuclides in building materials used in the construction of modern multi-storey buildings in Ekaterinburg and Tyumen. These differences are clearly visible at comparison of the spectra measured in the Room 2, Ekaterinburg and the Room 3, Tyumen (Fig. 4).

Calculation of the diffusion radon flux into the room from building materials and radon concentrations

Calculations of the radon flux from building materials and radon concentrations were carried out for a range of air exchange rates from 0.05 to 1 h^{-1}. In the calculations, fixed values of the effective diffusion coefficient $De = 2 \times 10^{-8}$ m^2/s, the emanation coefficient $E = 0.2$ and the porosity of

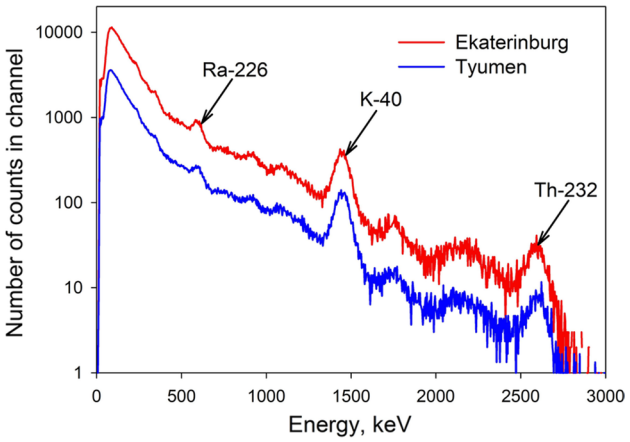

Fig. 4 Gamma ray spectra measured in the Room 2, Ekaterinburg and the Room 3, Tyumen (spectrum measurement time is 1800s)

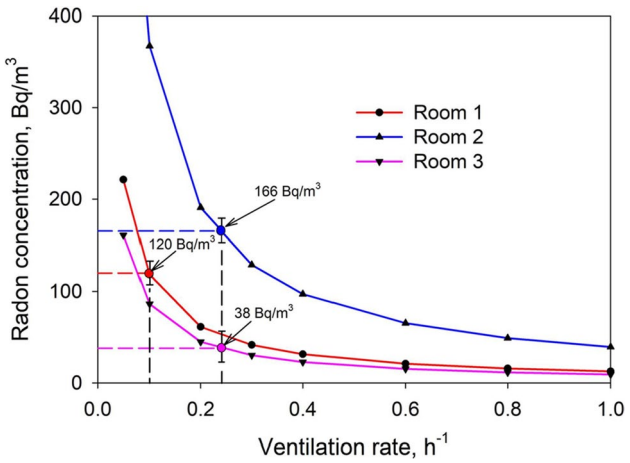

Fig. 5 Comparison of the calculated values of radon concentration, based on measurements of the flux of gamma quanta, with the results of measurements of the average values of radon concentration

the material $p = 0.1$ were taken. The values of the emanation coefficient and porosity are the values set in the program RESRAD-Build 3.5 by default. All these values are within the typical range for these parameters [2, 3, 22–25]. The calculation results are presented in Fig. 5. The figure also shows the average values of radon concentration measured using a radon monitor or track detectors. For the Rooms 1 and 2, measurements of radon concentration were carried out directly in the rooms where the fluxes of gamma quanta were measured. For the Room 3, measurements of radon concentration were carried out in an identical apartment located in the same residential complex. In all buildings under the study, ventilation is carried out due to air exchange with the outdoor atmosphere. Different ventilation modes in summer and winter considerably influencing on air exchange rate due to change both physical processes and human activity. The dependence of air exchange rate on temperature difference ΔT between indoor and outdoor atmosphere was demonstrated in [16]. At $\Delta T \sim 0$ (summer) the average air exchange rate is nearly 0.75–0.70 h^{-1}, at $\Delta T = 45$ °C (winter) the average air exchange rate is not more than 0.2–0.3 h^{-1}. The main reason of this effect is the decrease of ventilation frequency and its duration during cold season [26, 27].

The data presented in Fig. 5 show that for multi-storey buildings, the calculated values of radon concentration, obtained exclusively for radon diffusion from building materials, correspond to the experimentally measured values of radon concentration for the air exchange rate with outdoor atmosphere of 0.25 h^{-1}. According to the results of our research, this value is typical in cold season for modern multi-storey buildings, built with applying energy-saving technologies [15, 16]. For the Room 1, the observed mean value of radon concentration cannot be explained solely by its diffusion from building materials. The experimentally measured value corresponds to the calculated value at the air exchange rate of 0.10 h^{-1}, which is an unrealistically low average air exchange rate. However, it must be taken into account that the Room 1 is located on the second floor, and below it there is a service technical room with an earthen floor, without windows and mechanical ventilation. In our work, it was shown that for a laboratory room on the first floor, the rate of radon intake per unit volume is 30–35 Bq h^{-1} m^{-3} [12]. In the absence of ventilation, the concentration of radon in a technical room can reach 150–250 Bq/m^3. In this case, it is possible for radon to flow from the technical room into the Room 1 located above.

Thus, for multi-storey buildings, where the main source of radon is its diffusion from building materials, the proposed method based on non-destructive measurement of the average specific activity of radium in building materials and the calculation of the radon concentration using the diffusion model makes it possible to obtain results, well consistent with experimental measurements of radon concentration.

Uncertainty analysis

In the approach proposed above, there are a number of uncertainties that affect the accuracy of the result. Let us consider the main factors affecting the accuracy of the estimation of the radon concentration from the results of measurements of the density of the gamma quanta from ^{226}Ra.

The standard uncertainty of ^{226}Ra source contains two types of uncertainties—the uncertainty in determining the source activity and the uncertainty in the activity ratio (source emanation coefficient). The relative expanded standard uncertainty of the ^{226}Ra activity was 5%, the uncertainty of the $^{214}Bi/^{226}Ra$ ratio was 3% ($k = 2$). Thus, the relative uncertainty of the standard ^{226}Ra source, estimated by type B, is $\tilde{u}_B\,(Ra) = \sqrt{0.025^2 + 0.015^2} = 0.029$. In addition,

the uncertainty due to the statistics of the count rate in the photopeak was estimated. This uncertainty is described by Poisson statistics and is determined by the ratio between the number of accumulated counts in the photopeak and the total number of counts in the channels corresponding to the photopeak boundaries. This type A uncertainty can be calculated as:

$$\tilde{u}_{\mathrm{A}}\,(\mathrm{PhP}) = \frac{\sqrt{N_{\mathrm{PhP}} + N_{\mathrm{Sum}}}}{N_{\mathrm{sum}}}, \tag{6}$$

where N_{PhP} is the number of counts in photopeak; N_{Sum} is the total number of counts in the channels corresponding to the photopeak. In the process of calibrating the gamma spectrometer using a reference source, the relative uncertainty of the count rate in the photopeak, estimated by type A, was $\tilde{u}_{\mathrm{A}}\,(\mathrm{Ra}) = 0.013$.

The Poisson error caused by the statistic of the count rate in the photo peak must also be considered during the measurements in the rooms under study. For a spectrum measurement time of 1800s, this uncertainty, calculated by Eq. (6), varied from $\tilde{u}_{\mathrm{A}}(\mathrm{PhP}) = 0.022$ for the Room 2 to $\tilde{u}_{\mathrm{A}}\,(\mathrm{PhP}) = 0.059$ for the Room 3.

As mentioned above, when calculating the flux density of unscattered gamma quanta, the objects under consideration (floor slabs of the field and ceiling and walls) were divided into elementary volumes of $10 \times 10 \times 1$ cm, each of which was considered as a point source located in an absorbing medium. To assess the influence of the degree of partitioning of the modeled object on the accuracy of the calculations, calculations were carried out when the object was divided into cells of $1 \times 1 \times 1$ cm. The calculations showed that there was no difference within four significant digits in the output file of the MicroShield 11.2 software. Therefore, this type of uncertainty was accepted as insignificant.

The calibration measurements demonstrated that the angular sensitivity of the detector is anisotropic (Fig. 3). The sensitivity of the detector to the registration of gamma quanta in the 609 keV photopeak calculated as average for four directions of radiation incidence was $\varepsilon = 11.7 \pm 1.0$ counts/cm^2 (one standard deviation is given). This value of the standard deviation can be considered as the standard uncertainty, estimated by type A. Accordingly, the relative standard uncertainty of the detector sensitivity coefficient due to angular anisotropy was calculated as $\tilde{u}_{\mathrm{A}}(\mathrm{ang}) = 1.0/11.7 = 0.085$. Analysis of the angular dependence of the detector (Fig. 3) shows that can be additional uncertainty of the detector sensitivity due to decrease of the detector sensitivity at quanta incident angle more than 135 degrees. This uncertainty can be reduced by rotating the spectrometer such way, that the crystal of the detector will remain in place, and the body of the spectrometer will be oriented in different directions (4–8 directions). The time of the spectrum measurement at each position of the detector should be the same. Thus, the influence of the decrease of the sensitivity for the angle of incidence of gamma quanta more than 135 degrees will be eliminated.

Uncertainties due to inhomogeneous distribution of natural radionuclides in building materials. In the proposed approach, based on the results of measuring the spectrum of gamma radiation, the average specific activity of ^{226}Ra in building materials is calculated under the assumption that this value is the same for any materials (concrete, brick, aerated concrete). In practice, this assumption in most cases does not correspond to the real situation. With the total constant total activity of ^{226}Ra in the elements of building structures, in some elements it can be higher, and in others it can be lower than the average value. As a result of changing the location of the detector, the calculated value of the flux density of gamma quanta at the new point will differ from the measured value. And vice versa, the calculated values of the average specific activity of radionuclides in building materials, calculated from the results of measurements at different points in the room, will differ. An experiment to determine the average specific activity of radionuclides in building materials when measured at different points in the room was carried out using the example of Room 1. In addition to the measurements taken on the table in the middle of the room (Point 1), three additional measurements were taken: on the floor, directly below the point of initial measurement (Point 2); at a distance of 5 cm from the glass panel, which is the outer wall of the room (Point 3); at a distance of 5 cm from the inner brick wall of the room (Point 4). The last two measurements were taken at a height of 50 cm from the floor. The measurement results are presented in Table 1.

The average value of the specific activity of ^{226}Ra according to the four measurements was 33.2 ± 6.7 (given one standard deviation). The average value of the coefficient of variation was 0.20. This value was taken as the value of the uncertainty, estimated by type A $\tilde{u}_{\mathrm{A}}$ (inh), associated with the inhomogeneous distribution of natural radionuclides in the elements of building structures and the location of the gamma radiation spectrometer detector in the room. It should be kept in mind that this uncertainty may be partly

Table 1 Estimation of the average specific activity of ^{226}Ra in building materials of the Room 1 based on the results of measurements at various points in the room

	Calculated count rate per 100 Bq/kg, s^{-1}	Measured count rate, s^{-1}	Specific activity, Bq/kg
Point 1	5.07	1.67	32.9
Point 2	5.35	1.37	25.6
Point 3	2.82	1.18	41.8
Point 4	5.82	1.89	32.5

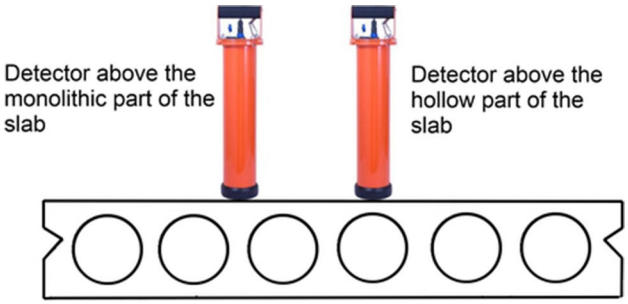

Fig. 6 Various possible locations for the detector on a hollow floor slab

due to the uncertainty of the angular sensitivity of the detector $\tilde{u}_A$ (ang).

It can be noted that the measured values of the count rate of quanta at Point 2 are lower than for Point 1 located 70 cm higher. According to calculations, the flux of quanta at this point should be 5–8% higher than at Point 1. One of the reasons for the observed effect may be the inhomogeneity of the hollow floor slab. When the detector is located on the monolithic part of the slab and above the hollow channel (Fig. 6), the geometry of the measurements and, accordingly, the flux density of quanta may differ significantly. In this regard, in order to avoid the influence of the local inhomogeneities, the installation of the detector on the floor slab should be avoided, if possible.

The specific activity of ^{226}Ra obtained from measurements near the outer glass panel (Point 3) is significantly higher than the results obtained by the measurements inside the room. This is due to the fact, that in the calculations it was assumed that there is no radiation of ^{226}Ra from the direction of the glass outer panel. For a room located on the second floor with glass panel from floor to the ceiling, one cannot exclude the presence of a flux of quanta with an energy of 609 keV from the ground and a three-story building located opposite at a distance of ~ 50 m. Therefore, during measurements, it is necessary to consider the possible presence of external radiation coming through large window or balcony openings.

The above calculations of the expected radon concentration levels at the estimated specific activity of ^{226}Ra in building materials were based on average, typical values for parameters such as emanation coefficient, porosity and diffusion coefficient. In practice, the researcher, as a rule, does not know the exact values of these parameters, unless samples of building materials were taken from the surveyed apartment for laboratory study. To assess this uncertainty in the RESRAD BUILD 3.5 program, the Monte Carlo method was used to calculate the possible range of changes in the concentration of radon when the emanation coefficient, porosity and diffusion coefficient were randomly varied. The Room 2 was chosen as an example. It was assumed that the emanation coefficient is uniformly distributed between the values $E=0.15$ and $E=0.25$, the porosity is uniformly distributed between $p=0.07$ and $p=0.13$, slab thickness uniformly distributed between 20 and 25 cm and the diffusion coefficient values are described by a triangular distribution between 7×10^{-9} and 6×10^{-8} with a mode of 2×10^{-8} m^2/s. For each of the selected values of the air exchange rate from 0.1 to 1.0 h^{-1}, 180 combinations of these parameters were calculated.

The Monte Carlo simulation results are shown in Fig. 7. The confidence interval of radon concentration, calculated as twice the standard deviation over all modeling results, is significantly less than the 95% confidence interval. For comparison, the coefficient of variation of the values obtained during the simulation was 0.079. Uncertainty value corresponding to 95% confidence level, can be calculated as $\tilde{u}_A\,(\text{diff}) = \frac{\overline{C}_{Rni} - C^{0.025}_{Rni}}{2\overline{C}_{Rni}} \approx \frac{\overline{C}_{Rni} - C^{0.975}_{Rni}}{2\overline{C}_{Rni}} = 0.14$. This value was taken as the value of the relative standard uncertainty due to unknown values of the parameters of radon diffusion from building materials.

The combined relative standard uncertainty of radon concentration, estimated by measuring the flux density of unscattered gamma quanta in the room, was calculated as:

$$\tilde{u}_c\,(\text{Rn}) = \sqrt{\tilde{u}_B(\text{Ra})^2 + \tilde{u}_A(\text{Ra})^2 + \tilde{u}_A\,(\text{PhP})^2 + \tilde{u}_A(\text{ang})^2 + \tilde{u}_A(\text{inh})^2 + \tilde{u}_A(\text{diff})^2} = 0.26 \quad (7)$$

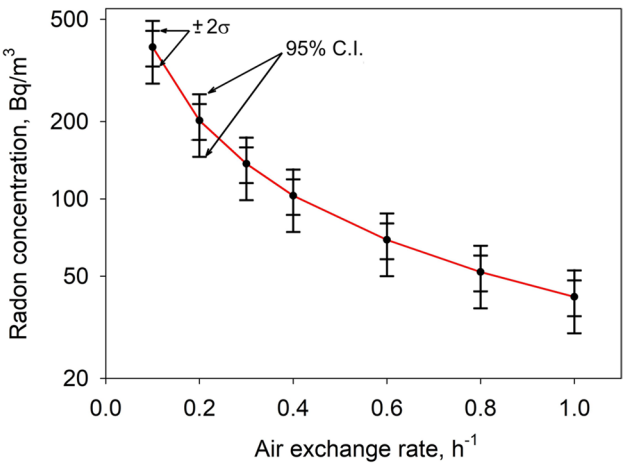

Fig. 7 Calculation of the expected average values of radon concentration in the Room 2, considering the uncertainties of the emanation coefficient, porosity and diffusion coefficient of radon in building materials

Table 2 The budget of uncertainties in the estimation of radon concentration

Uncertainty type	Contribution to the uncertainty budget, %
$\tilde{u}_B$(Ra)	1.23
$\tilde{u}_A$(Ra)	0.25
$\tilde{u}_A$(PhP)	2.34
$\tilde{u}_A$(ang)	10.6
$\tilde{u}_A$(inh)	58.5
$\tilde{u}_A$(diff)	27.1

The budget of uncertainties in the estimation of radon concentration is presented in Table 2. The main sources of uncertainties: uncertainty due to inhomogeneous distribution of natural radionuclides in building materials, uncertainty due to unknown values of parameters of the radon diffusion from building materials, uncertainty of the angular dependence of the detector sensitivity.

Conclusions

The current trend towards the construction of multi-storey residential energy efficient buildings results in the fact that the main source of radon entry is diffusion from building materials. This makes it relevant to control the specific activity of ^{226}Ra in building materials even in cases when it is noticeably below the reference values. For already constructed and operated buildings, the measurement of the specific activity of natural radionuclides in a particular room presents certain organizational and technical difficulties associated with the need to take samples of building materials for laboratory analysis. To solve this problem, non-destructive method is proposed consisted in analysis of gamma radiation spectra of natural radionuclides measured in a selected room by field gamma spectrometer with a fixed geometry of the detector position.

The calculation of the average specific activity of natural radionuclides in building materials is carried out by comparing the flux density of unscattered gamma quanta of a given energy, calculated for the geometry of the room under study and the location of the detector, and the experimentally measured count rates in the corresponding photopeaks. In the calculations, it is assumed that the specific activity of natural radionuclides in building materials is the same for all building structures, regardless of their density (for example, 100 Bq/kg). The presence of door and window openings in the walls is taken into account accordingly—the flux density of quanta from the section of the wall where the door or window is located is calculated and the obtained value is subtracted from the flux calculated for an entire wall.

For a field gamma spectrometer with a 63×63 mm NaI (Tl) crystal, the conversion coefficients from the flux density of unscattered gamma quanta to the count rate in the photopeak of the corresponding energy are calculated. The average values of the spectrometer sensitivity in the photopeak were for ^{226}Ra (609 keV) $\varepsilon = 11.7 \pm 1.0$ counts/cm^2, for ^{40}K (1460 keV) $\varepsilon = 4.33 \pm 0.79$ counts/cm^2, for ^{232}Th (2614 keV) $\varepsilon = 2.13 \pm 0.09$ (one standard deviation is given). The calibration was carried out when the source was removed from the detector crystal at a distance no less than 50 cm, so the flux of quanta could be assumed to be parallel. This allows the results of this calibration to be used for the analysis of preliminary measurements performed on gamma spectrometers of a different type with the same crystal size.

For three different rooms, the flux densities of unscattered gamma quanta were calculated at the point of the detector location. The calculation is made considering the geometry of the room, the thickness and density of building structures (walls and floor and ceiling slabs). Each volume source (slab or wall) was simulated by a superposition of $10 \times 10 \times 1$ cm cells, considered as the point sources, located in a gamma-absorbing medium.

For the selected rooms, the flux density of non-scattered gamma quanta was measured in 609, 1460, and 2614 keV photopeaks. By comparing the measured values with the calculated data, the average values of the specific activity of ^{226}Ra, ^{40}K and ^{232}Th in building materials were calculated. The obtained values are in good agreement with the data on the specific activity of natural radionuclides in building materials used during the construction of the buildings under consideration.

For the calculated values of the average specific activity of ^{226}Ra in building materials, the expected values of radon concentration in rooms were calculated for the air exchange rate from 0.1 to 1.0 h^{-1}. It is shown that for rooms located in modern multi-storey energy-efficient buildings, the calculated radon concentration values correspond to the average experimentally measured values at an air exchange rate of 0.25 h^{-1}, which is in agreement with the estimates of the average air exchange rate for buildings of this type. This confirms the assumption that the dominant source of radon for rooms on the upper floors of multi-storey buildings is the radon diffusion from building materials. It has also been demonstrated that for rooms in low-rise buildings, radon from the ground under the building cannot be excluded.

An analysis of uncertainties affecting the accuracy of estimating the radon concentration from the results of measuring the flux density of unscattered gamma quanta with an energy of 609 keV is carried out. The combined relative standard uncertainty of radon concentration, taking into account all sources of uncertainty was $\tilde{u}_c(Rn) = 0,26$. Uncertainty budget analysis showed that the main sources of uncertainties: uncertainty due to inhomogeneous distribution

of natural radionuclides in building materials, uncertainty due to unknown values of parameters of the radon diffusion from building materials, uncertainty of the angular dependence of the detector sensitivity.

The developed non-destructive method of ^{226}Ra effective content in building materials may be applied to study ^{226}Ra as a source of radon both in new and old buildings in situations of unknown specific activity of natural radionuclides in building materials. Obtained relationship between ^{226}Ra average activity and indoor radon concentration may be used to improve the system of radiological protection in the situation of radon exposure with regard to the current trends in building construction.

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