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Gamma-radiation exposure by natural radionuclides in residential building materials on example of nine Russian cities

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Abstract

The objective of the study was to estimate the effective dose rates of external population exposure based on measurements of average specific activity of natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in building materials of existing buildings. The measurements were performed using new developed non-destructive technique in 100 apartments in 9 Russian cities. The effective dose rate is 34 nSv h^{-1} in average and varies from 10 to 102 nSv h^{-1} between cities. Specific factors of conversion from specific activity to effective dose rate were obtained taking into account the real room geometries.

Keywords Natural radionuclides · Specific activity · Building materials · Non-destructive measurement method · Effective dose

Introduction

The issue of ensuring the radiation safety of the population against radiation from natural radionuclides (NRN) contained in building materials has been dealt with since the 1970s [1–12]. Indoor exposure is determined by ^{226}Ra, ^{232}Th, and ^{40}K present in building materials of enclosed structures such as walls, ceilings, and floors. More specifically, the natural radioactivity is caused by gamma-radiation daughter decay products of NRN. Since the most people spend about 80% of their time indoors [7], the exposure by NRN poses an important issue. The external exposure dose rate depends on specific activity of NRN in building materials and geometry of the room.

The construction of modern high-rise buildings involves the use of various building materials. For example, the floor and ceiling can be made of monolithic reinforced concrete, walls of brick or aerated concrete, and interior partitions can be made of gypsum. Determining the dose rate from ^{226}Ra, ^{232}Th and ^{40}K in each building material separately is rather a difficult task.

Conventional gamma spectrometry methods for measuring the specific activity in building materials are destructive [13]. This is especially important for existing residential buildings, where construction has already been completed and sampling with destruction of building materials is not applicable. Recently, a non-destructive method of measuring the NRN specific activities in building materials has been developed [14]. This method can be utilized to study the situation of external exposure under different NRN specific activity in the existing buildings.

The specific air kerma rate is used to convert from the specific activity of the NRN to the air kerma rate. The values of these factors for each of the NRN can be found in the following publications [8–12]. They vary from 0.908 to 0.922 for ^{226}Ra, from 1.02 to 1.10 for ^{232}Th, and from 0.0767 to 0.0806 for ^{40}K. These calculations were performed for a standard room without windows, doors. The wall materials of the standard room are the same and the density of all building materials is 2.32 g cm^{-3}. Obviously, real rooms differ from the standard room. In a series of studies, the geometry of semi-infinite space was used to calculate the air kerma rate [15–19]. In [20, 21] the conversion factor of 1.4 [23] was used for conversion from the geometry of the open half-space to the room geometry. In general, the use of

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simplified geometry leads to incorrect estimation of radiation dose from building materials in residential buildings.

The conventional conversion coefficient recommended by UNSCEAR is used for conversion of air kerma to effective dose. Its value is 0.7 Sv Gy^{-1} in UNSCEAR [22]. Unfortunately, this coefficient doesn't take into account the dependence of conversion coefficient on the ratio of NRN in the material and the presence of scattered radiation. It also doesn't consider the dependence of the coefficient values on the energy spectrum of natural radionuclides. So, it's possible to use the modern and refined dose conversion coefficients from ICRP Publication 116 [24], which take into account the dependence on the energy of the radionuclide emission line.

According to the requirements of the European Union and the IAEA, the reference levels of additional exposure due to NRN in building materials must not exceed 1 mSv year^{-1} [25–27]. In order to limit the external exposure of the population to NRN in building materials, the concept of "specific activity index" (activity concentration index I_γ) has been introduced. This value should be less than 1.

Russia has another approach for limiting external human exposure from NRNs in building materials. The Russian Radiation Safety Standards NRB-99/2009 [28] introduces the effective specific activity of NRN in building materials. It must not exceed 370 Bq kg^{-1}. This value is calculated with weight coefficients corresponding to the specific activity of NRN. Weight coefficients used for ^{232}Th and ^{40}K correspond to rounded values given in [3–5]. At the same time there is not limit of annual external dose from building materials in [28], but this document states that the effective dose rate of gamma radiation in the premises should not exceed the effective dose rate in open areas by more than 0.2 µSv h^{-1}.

An analysis of published data has shown that there are no direct estimates of the external exposure of the population, taking into account the actual content of NRN in the building materials of rooms in existing buildings and their geometric parameters.

The above data allow to formulate the research objective: to perform direct estimates of effective dose from external exposure basing on measurements of the content of NRN in building materials of existing premises.

Research tasks:

1. To carry out measurements of ^{226}Ra, ^{232}Th, ^{40}K specific activity in existing buildings in large cities of Russia located in different climatic, geographical, and geological zones using non-destructive gamma spectrometry.
2. To calculate the specific coefficients that allow the conversion of the NRN specific activity to the air kerma rate or effective dose rate using a new software and updated physical constants for a standard room concept in the calculations.
3. To estimate the effective dose rate from NRN in building materials using the real and model room spectra and the air kerma to effective dose conversion factors for isotropic geometry from the ICPR Publication 116 data [24].
4. To compare effective doses estimated for real and standard rooms.

Materials and methods

The non-destructive technique for measuring the average NRN specific activity in building materials in existing buildings is described in detail in [14].

The non-destructive method is based on the field gamma spectrometry, a detector is placed directly in the room under investigation. An MKS AT6101DR spectrometer was used to perform the measurements. [29]. The range of energies recorded by the spectrometer is from 50 keV to 3 MeV. The NaI(Tl) ∅63 × 63 mm crystal was used as the detector. The spectrometer recorded gamma-ray spectra for 3600 s and was pre-calibrated for the detection efficiency in the ^{226}Ra, ^{232}Th and ^{40}K photopeaks. The energy scale of the spectrometer's measuring path was stabilized with a reference sample before each measurement. Simultaneously, the geometric parameters of the room were measured using a laser rangefinder. The building materials of measured rooms were visually estimated and then are determined using the building technical passport. The common building materials are monolithic concrete, floor slabs with air cavities, brick, aerated concrete, gypsum, etc. The total flux density of unscattered gamma quanta at the detector location was calculated using the MicroShield 11.2 software [30]. The flux densities from windows and doors were subtracted from the total flux density of the corresponding walls. The material type and density of each enclosing structure was considered in the calculation of the total unscattered gamma quanta flux density. Wall thicknesses were taken from the building passport or measured.

During the analysis of the energy spectra of radionuclides, the main characteristic emission lines in the decay chain were choose: ^{232}Th–2614 keV (^{208}Tl), ^{40}K–1460 keV. In contrast to the methodology described in [14], the 1.758 MeV (^{214}Bi) line was chosen for ^{226}Ra as the characteristic emission line. Preliminary analyses showed that when ^{226}Ra and ^{232}Th are simultaneously present in the energy spectrum, there is a contribution of the ^{208}Tl emission line (0.583 MeV) to the ^{226}Ra full absorption peak of 0.609 MeV (^{214}Bi). Therefore, the specific activity of ^{226}Ra was determined using the 1.758 MeV (^{214}Bi) emission line, despite the fact that the gamma radiation quantum yield of this energy for decay is only 15.4%. Experimental I_{meas} count rates were determined for each selected line. The calculated

I_{calc} count rate at the corresponding peak was determined and normalized to the 100 Bq kg^{-1} specific activity of ^{226}Ra, ^{232}Th or ^{40}K or the actual measurements geometry. Then the average specific activity of the corresponding radionuclide was calculated as follows:

$$\overline{A_m} = \frac{I_{meas}}{I_{calc}} 100, \text{ Bq kg}^{-1}. \quad (1)$$

The obtained specific activity values of ^{226}Ra, ^{232}Th and ^{40}K were used to calculate the effective specific activity of the natural radionuclides in building materials according to the following equation:

$$A_{eff} = A_{Ra} + 1.3A_{Th} + 0.09A_K. \quad (2)$$

It should be noted that the specific activity of ^{226}Ra non-emanating fraction in the building material was obtained during the direct measurements of ^{214}Bi with the energy line of 1.76 MeV. Therefore, the total ^{226}Ra specific activity was calculated using the specific activity of ^{214}Bi and the ^{222}Rn emanation factor. The emanation factor has different values in literature [31–34], so it was decided to use the value of 0.2 recommended by UNSCEAR [23].

The relative uncertainty of the average specific activity of ^{226}Ra measurement was calculated in detail in [14] and was estimated as 23%. Preliminary calculations show that for the other considered radionuclides the relative uncertainty of the specific activity measurement can be assumed to be the same.

Calculation of weight coefficients and specific dose rate

The value of dose rate normalized per unit of NRN specific activity depends on both the exposure geometry (infinite or semi-infinite space, a plate of limited thickness and dimensions, room geometry, etc.) and material characteristics (composition, thickness and density). Among the reviewed works, there are no unified values of the coefficients relating the air kerma with the NRN specific activity in building materials (Table 1).

Table 1 Specific air kerma rate in standard room for different radionuclides and different calculation methods

Specific air kerma rate, nGy h^{-1} Bq^{-1} kg			Calculation method	Reference
^{226}Ra	^{232}Th	^{40}K		
0.922	1.02	0.0779	Monte Carlo	[8]
0.914	1.10	0.0776	Numerical integration considering the buildup factor	[9]
0.922	1.10	0.0806		[10, 11]
0.908	1.06	0.0767		[12]

The specific air kerma rate and the specific effective dose rate were calculated for a standard room to compare with the results of reviewed works [8–12]. The standard room has dimensions 5.0×4.0×2.8 m without windows and doors with 20 cm thick walls made of concrete with a density of 2.32 g cm^{-3}. The specific activity of ^{226}Ra, ^{232}Th or ^{40}K was supposed to be 100 Bq kg^{-1} in calculations. The ^{226}Ra and ^{232}Th were assumed to be in equilibrium with their decay products.

The calculations were performed using the specialized software MicroShield 11.2 similar to the calculation method described in [14], the air kerma rate in the room center of standard room was calculated for each elementary volume of walls or ceiling (floor). The buildup factor was also considered in calculations.

Computer calculations were performed separately for all ^{226}Ra (24 energy groups with energies from 0.052 to 2.448 MeV) and ^{232}Th (20 energy groups with energies from 0.039 to 2.615 MeV) emission lines. For ^{40}K, the calculations were done at 1.460 MeV energy. The kerma rate in air was then summed for all ^{226}Ra, ^{232}Th and ^{40}K emission lines, followed by normalization of the resulting values to a specific activity of 1 Bq kg^{-1}.

To convert the air kerma rate to effective dose rate, the data given in ICRP Publication 116 [24] for isotropic exposure geometry were applied. These data were fitted by a smooth function to describe the energy dependence of the conversion coefficient.

Results and discussion

Specific activity of natural radionuclides in building materials

The specific activity of NRN was measured in residential premises located in multi-storey buildings of different years of construction (from 1856 to 2021) and different building envelope materials (monolithic concrete, brick, aerated concrete, panel, etc.). The objects of the study were apartments in the following cities: Moscow, St. Petersburg, Ekaterinburg, Chelyabinsk, Tyumen, Salekhard, Krasnodar, Nizhny Novgorod and Novosibirsk. A total of 100 apartments were chosen. The sample was characterized by an average room height of 2.6 m and a total wall area of 72 m^2 (including floor and ceiling area). The wall thickness varied from 6 to 80 cm and the density ranged from 0.6 g cm^{-3} (aerated concrete) to 2.4 g cm^{-3} (reinforced concrete). The value of effective specific activity of NRN in building materials was calculated according to the Eq. (2) for each room. The arithmetic mean, minimum and maximum values of ^{226}Ra, ^{232}Th and ^{40}K specific activity and the NRN effective specific activity in building materials are presented in Table 2 for surveyed cities.

The data given in Table 2 show that the maximum effective specific activity is observed in St. Petersburg. Somewhat smaller values are observed for Ekaterinburg and Chelyabinsk, even smaller for Moscow and they are minimal for other cities.

It is interesting to compare the obtained average values of effective specific activity in apartments in existing buildings with the results of the NRN monitoring of effective specific activity in different building materials. The last one was conducted by the Russian sanitary authorities and described in detail in [35]. The comparison of results is shown in Fig. 1.

Analysis of the data presented in Fig. 1 shows that for most of the studied cities, the average effective specific activities of NRN measured by the non-destructive method are in good agreement with the data of traditional building materials monitoring. At the same time, it should be taken into account that in [35] the data refer only to Moscow and St. Petersburg. In all the other cases, the results of the sanitary monitoring are related to the region in general. The largest differences in the average effective specific activity are observed for Nizhny Novgorod and Novosibirsk. This case requires additional analysis, but it may be due to the fact that most of the surveyed houses in Nizhny Novgorod and Novosibirsk were built in 2009–2021 (except for four buildings that were built in 1967–1981).

Table 2 An average specific activity of NRN in building materials of multi-storey residential buildings in nine cities of Russia, obtained by non-destructive method (the range of values is given in parentheses)

City	Number of apartments	Specific activity (Bq kg^{-1})			
		^{226}Ra	^{232}Th	^{40}K	A_{eff}
Moscow	11	16.8 (10.9–21.8)	25.6 (15.0–41.6)	365 (253–620)	82.9 (65.9–120.3)
St. Petersburg	13	18.9 (11.6–31.0)	49.5 (31.0–65.2)	805 (580–984)	155.7 (113.7–201.3)
Ekaterinburg	28	31.5 (9.1–49.1)	28.2 (9.9–49.1)	512 (81–771)	114.2 (29.9–180.1)
Krasnodar	7	7.1 (4.7–8.3)	20.8 (17.5–23.2)	282 (241–306)	59.6 (51.9–65.0)
Nizhny Novgorod	10	8.5 (4.3–12.8)	11.1 (6.1–22.6)	166 (117–222)	37.9 (24.0–57.3)
Salekhard	7	9.9 (7.1–12.8)	12.6 (8.6–18.6)	248 (205–306)	48.6 (37.3–61.7)
Chelyabinsk	8	21.3 (12.1–41.4)	39.1 (20.6–113.0)	478 (355–919)	115.2 (74.8–271.0)
Tyumen	6	10.1 (3.8–22.0)	9.7 (4.5–17.7)	274 (185–361)	47.4 (30.6–77.6)
Novosibirsk	10	8.7 (7.0–13.8)	19.9 (10.5–25.4)	382 (230–516)	69.0 (43.4–86.8)

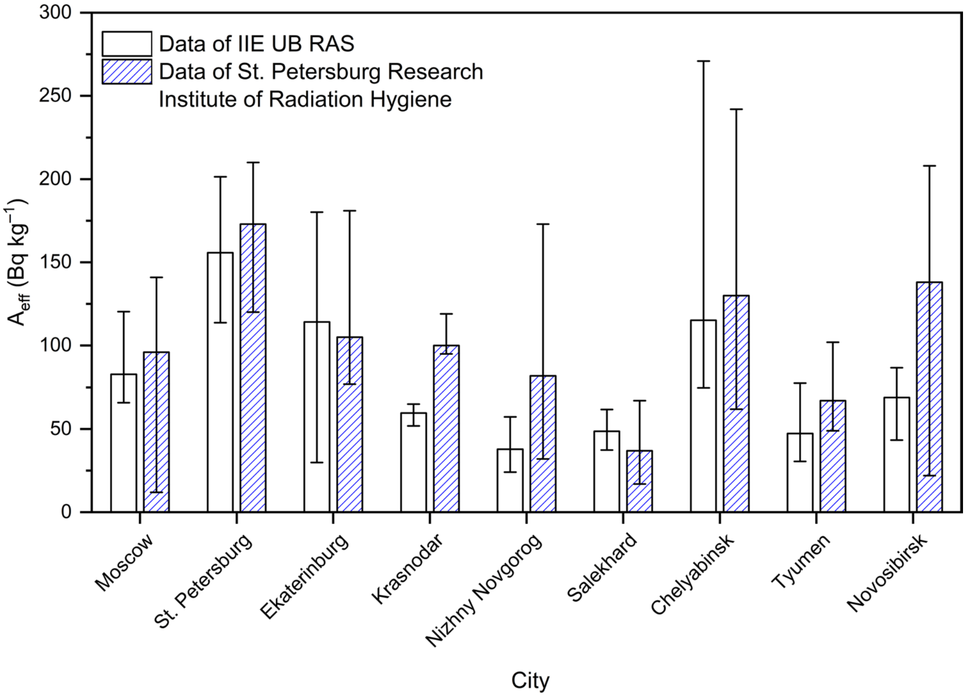


Fig. 1 Comparison of data on direct measurement of the average, minimum and maximum values of the NRN effective specific activity in apartments in existing buildings with the effective specific activity statistics of monitoring in various building materials [35] (value range is indicated)

According to the previous studies, the average effective specific activity in building materials in Russia is 93 Bq kg^{-1} [3, 31]. In our study, this value is 1.3 times lower. The difference can be explained by the fact that most of the buildings in our sample were built after 2000 year (75%). It's assumed that the decrease of content of NRN in new buildings is associated with stricter controls and regulatory requirements to ensure radiation safety.

Estimation of the effective dose rate due to the natural radionuclides in building materials

The effective dose rate caused by gamma-radiation of NRN was calculated for all surveyed rooms based on the estimation of average specific activity of ^{226}Ra, ^{232}Th and ^{40}K in floor slabs and enclosing building constructions, as well as room geometry. The calculation was performed for each radionuclide separately. The results are given in Table 3. In most of the surveyed cities, the dominant source of external exposure in dwellings is ^{40}K, which is present in building materials. The next important radionuclide is ^{232}Th. The percentage contribution of each NRN to the effective dose rate for the cities considered is shown in Fig. 2.

Table 3 Average effective dose rate due to natural radionuclides in building materials of multi-storey residential buildings in nine cities of Russia (the range of values is indicated in parentheses)

City	Effective dose rate (nSv h^{-1})			
	^{226}Ra*	^{232}Th	^{40}K	Total value
Moscow	6.3 (4.4–7.6)	16.7 (9.0–30.0)	15.9 (9.0–27.4)	38.9 (25.4–61.9)
St. Petersburg	7.2 (4.8–10.8)	32.4 (22.9–42.8)	35.4 (21.4–41.9)	75.0 (50.2–88.2)
Ekaterinburg	12.2 (3.6–20.5)	18.5 (6.4–36.9)	22.2 (3.5–37.3)	52.9 (13.5–94.8)
Krasnodar	2.7 (1.7–3.0)	13.6 (12.0–14.8)	12.4 (11.1–13.7)	28.8 (26.1–30.4)
Nizhny Novgorod	3.2 (1.4–4.4)	7.2 (4.6–16.5)	7.2 (5.1–10.9)	17.5 (11.7–30.9)
Salekhard	4.0 (3.0–5.0)	8.6 (5.6–12.5)	11.4 (9.6–15.2)	24.0 (18.4–29.6)
Chelyabinsk	8.2 (4.7–12.5)	24.9 (13.7–58.0)	21.1 (15.9–31.4)	54.1 (35.8–101.9)
Tyumen	3.9 (1.5–7.5)	6.4 (3.0–10.4)	12.4 (8.3–15.8)	22.6 (14.5–32.1)
Novosibirsk	3.4 (2.6–4.8)	13.4 (7.6–17.3)	17.3 (11.2–23.8)	34.0 (22.5–43.7)

*The emanation factor of 0.2 was assumed for ^{226}Ra calculations

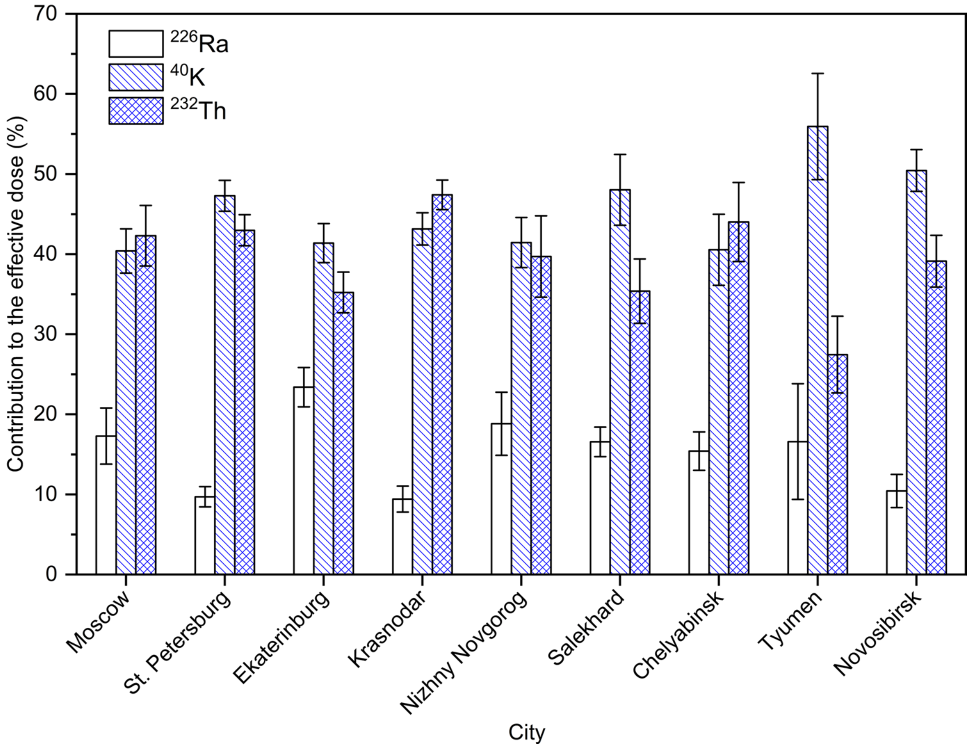


Fig. 2 Average relative contribution of ^{226}Ra, ^{232}Th and.^{40}K to the effective dose rate (95% confidence intervals are given)

Estimation of the dose rate normalized to a unit specific activity

When analyzing the external exposure of the population to NRN in building materials, it is useful to estimate the specific dose rate normalized to a unit specific activity of one or another NRN. In this case, it's necessary to consider both a standard room and real rooms for which all the experimental data are available.

The values of the specific air kerma rate and the specific effective dose rate for the standard room, normalized to a unit of effective specific activity of NRN, are presented in Table 4.

Comparing our calculations of specific air kerma rate for standard room with literature data presented in Table 1, the following can be concluded: the most significant difference (–15%) is observed in the calculations of the specific air kerma rate for ^{226}Ra; for ^{232}Th and ^{40}K the differences are –4 and –10%, respectively. This may be due to refine of the nuclear physics constants, such as dose conversion coefficients [24], used in the calculations.

Taking into account the updated results of the specific air kerma rate and the specific effective dose rate in the room for isotropic exposure geometry, the weighting factors in Eq. (1) were recalculated as $a_1 = 1.326$ and $a_2 = 0.093$.

After rounding to two digits these values of weighting coefficients correspond to the values used in NRB-99/2009, but in some cases they are differ from the values estimated in relatively earlier works [8–12]. Calculations performed for geometries of room surveyed showed that the weight coefficients for ^{232}Th and ^{40}K differ from the values $a_1 = 1.326$ and $a_2 = 0.093$ by no more than ± 0.002.

The values of the specific air kerma rate and specific effective dose rate shown in Table 4 correspond to the standard room. In fact, the values of air kerma rate and the specific effective dose rate for this type of room are overestimated compared to a real one. Direct measurements of average NRN specific activity values for surveyed room make it possible to obtain distributions of specific kerma rate or specific effective dose rate. It's possible to consider the influence of room geometry, real thickness and density of building structures, as well as the presence of windows and doors. The distribution of the specific effective dose rate normalized per 1 Bq kg^{-1} in comparison with the data for the standard room is shown in Fig. 3. It can be seen that the actual values of the specific effective dose rate are in most cases significantly lower than the calculated values for the standard room.

Table 4 Specific air kerma rate and specific effective dose rate calculated for a standard room using a specialized software package MicroShield 11.2 (the building material is concrete)

Value	Radionuclides		
	^{226}Ra	^{232}Th	^{40}K
Specific air kerma rate (nGy h^{-1} Bq^{-1} kg)	0.781	1.026	0.0702
Specific effective dose rate (nSv h^{-1} Bq^{-1} kg)	0.574	0.760	0.053

The difference between the average specific effective dose rate obtained for surveyed rooms and standard room is no more than – 13% for ^{226}Ra, – 8% for ^{232}Th, and – 13% for ^{40}K (Fig. 3). Apparently, that the specific effective dose rate for standard room overestimates one for surveyed rooms for each radionuclide. This is due to the difference between geometries of the rooms (doors and window openings) and the characteristics of the building materials used in real and standard rooms.

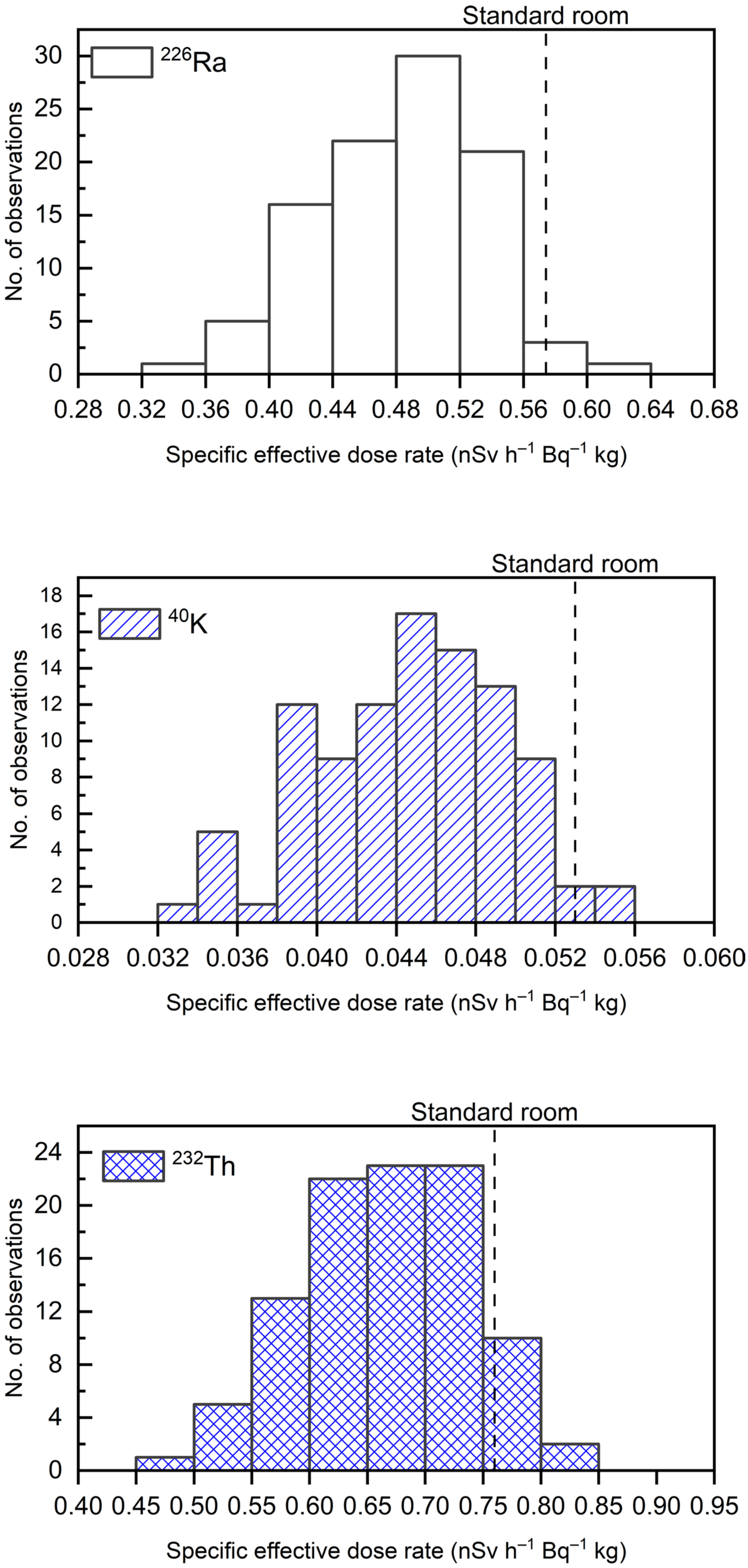
It is more illustrative to present the distribution of the specific effective dose rate of surveyed rooms normalized to the limit of 370 Bq kg^{-1} in comparison with the limit of the increase of the external dose rate due to building materials by 0.2 μSv h^{-1}, as required in the NRB-99/2009. The distribution of the effective dose rate normalized to an average effective specific activity of 370 Bq kg^{-1} is shown in Fig. 4.

Assessment of the external population exposure characteristics due to NRN contained in building materials

In practice, the method of non-destructive measurement of the average specific activity of NRN in building materials of apartments in existing buildings [14] has provided researchers with a new and effective tool. For the first time, estimates of the average specific activities of ^{226}Ra, ^{40}K and ^{232}Th and effective dose rates due to NRN in building materials of rooms in existing buildings were obtained.

The conducted studies have a wide geographical coverage and are applicable to a large part of the territory of Russia. This makes it possible to perform preliminary estimates of the average NRN specific activity levels in materials of existing buildings at the national level. It also allows to estimate the average effective dose rates due to NRN content in building materials. Estimates were made for urban housing stock, since data on NRN specific activity measurements in building materials for rural houses are currently unavailable. Based on statistical data [36], the relative contribution of Moscow, St. Petersburg and Ekaterinburg to the total urban housing stock in Russia was estimated. When calculating the average values for Russia, specific activity of NRN in building materials and effective dose rates obtained for these cities were considered with a weight factor corresponding to the share of each city in the total housing stock. It was assumed that the results of measurements in Chelyabinsk, Tyumen, Krasnodar, Nizhny Novgorod, Salekhard and

Fig. 3 Distribution of specific effective dose rate for surveyed rooms per 1 Bq kg^{-1} compared to standard room data



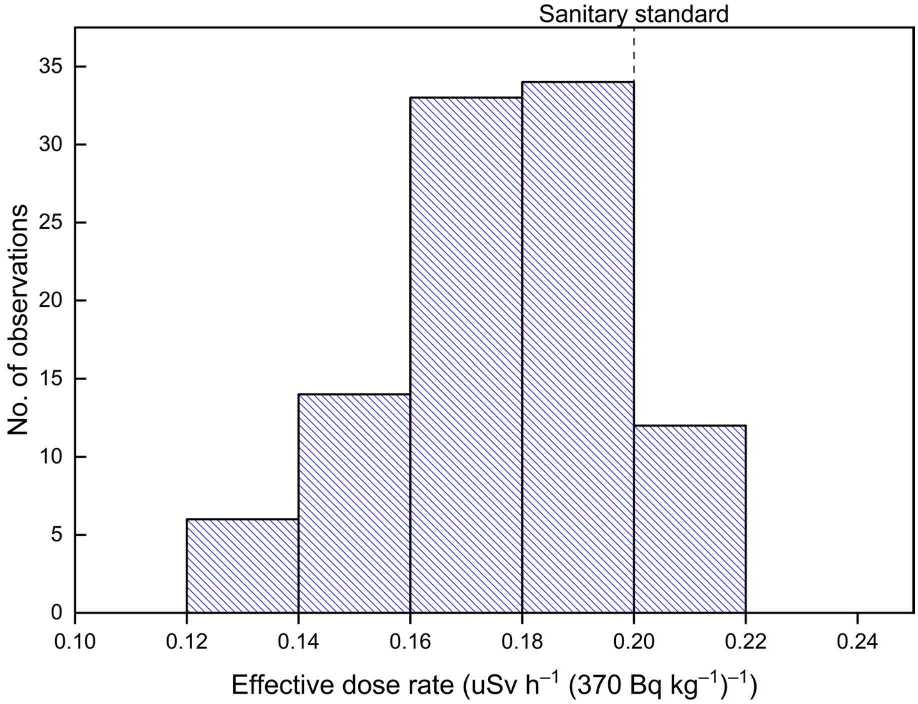


Fig. 4 Effective dose rate distribution for surveyed rooms normalized to the average effective specific activity of NRN of 370 Bq kg^{-1}

Table 5 Relative contribution of various cities to the urban housing stock in Russia

City	Area for 2019 (m^2)	Weight
Moscow	245 800 000	0.085
St. Petersburg	139 968 447	0.049
Ekaterinburg	42 047 619	0.015
Other cities	2 451 688 734	0.851
Sum	2 879 504 800	1.0

Novosibirsk can be considered as a sample characterizing the rest of the urban residential facilities in Russia. A similar approach was used earlier to estimate the parameters of radon concentration in the multi-storey buildings in Russia [37]. Initial data used in calculations and the numerical values of the weight coefficients are given in Table 5.

The NRN specific activity in the urban housing stock in Russia and the resulting effective dose rates were estimated considering the weighting factors (Table 5). The results are given in Table 6. The average annual effective dose of external exposure due to NRN in building materials of multi-storey buildings can be estimated to be about 0.24 mSv year^{-1} (the time spent indoors is 7000 h per year).

Conclusions

1. According to the results of a survey of nine Russian cities carried out using a new non-destructive method of measurement the average specific activities of NRN in real building materials were obtained for existing multi-storey buildings. The measured average values were ^{226}Ra–12 Bq kg^{-1}, ^{232}Th–21 Bq kg^{-1}, ^{40}K–338 Bq kg^{-1}, A_{eff}–70 Bq kg^{-1}
2. The distributions of the specific conversion factor from NRN specific activity to effective dose rate with respect to the real room geometry were obtained. The application

Table 6 Estimated values of the average specific activities of NRN in building materials of urban buildings in Russia and the average effective dose rates due to their presence (minimum and maximum values are given in brackets)

Specific activity, Bq kg^{-1}			
^{226}Ra	^{232}Th	^{40}K	A_{eff}
12.1 (3.8–49.1)	21.2 (4.5–113.0)	337.6 (81.3–984.3)	70.1 (20.3–271.0)
Effective dose rate, nSv h^{-1}			
^{226}Ra	^{40}K	^{232}Th	Sum
4.6 (1.4–20.5)	13.9 (3.0–58.0)	15.0 (3.5–41.9)	33.5 (9.6–101.9)

of specific conversion factors obtained for the standard room overestimates the effective dose in the most of real rooms in multi-storey buildings.

3. The average effective dose rate from NRN in building materials was 34 nSv h^{-1} and varied from 10 to 102 nSv h^{-1} between cities. The average annual effective dose to urban population from NRN in building materials was estimated to be 0.24 mSv $year^{-1}$.

4. In case of $A_{eff} = 370$ Bq kg^{-1}, the effective dose rate may exceed the radiation safety standard 0.2 μSv h^{-1} defined in terms of the permissible increase of the external effective dose rate indoor in approximately 12% of the surveyed rooms in existing buildings.

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Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

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