IRSTI 29.15.15; 34.49.23 https://doi.org/10.26577/phst2024v11i2b01

Monitoring the distribution of radon isotopes and their decay products in Almaty

Yu. Zaripova*, A. Yushkov, N. Amangeldiyeva, K. Dyussebayeva and A. Shaidollina

Al-Farabi Kazakh National University, Almaty, Kazakhstan
*e-mail: zarypova.iulyiya1@kaznu.kz

(Received September 5, 2024; received in revised form October 7, 2024; accepted October 16, 2024)

This paper examines the relationship between indoor radon accumulation and the concentration of its decay products in the soil to comprehensively assess the impact on public health, as high indoor radon concentrations have been clearly shown to increase the risk of lung cancer, making it a serious public health problem. Indoor radon concentration, gamma and beta activity of soil samples were studied in Almaty at a distance of 27 m to 1500 m from the tectonic fault. It was found that indoor radon concentration C_{Rn} decreases with increasing distance to the tectonic fault. The indoor radon activity concentration ranged from 6.23 to 405.21 Bq/m^3 with a geometric mean of 38.9 Bq/m^3. The specific activity in the selected soil samples was measured using the SPUTNIK-99 spectrometric setup. The results showed that beta activity varied in the range from 191.67±28.75 to 275.32±41.30 Bq/kg and gamma activity from 112.51±16.88 to 451.60±67.74 Bq/kg. Radon concentration also shows a good correlation with the gamma activity of the collected soil samples. The correlation coefficient was 0.79 for "C_{Rn}-Gamma" and 0.58 for "C_{Rn}-Beta", and the correlation analysis was done using Pearson's correlation tools.

Key words: indoor radon, radon concentration, radon decay products, exposure to radon, gamma radioactivity, beta radioactivity, radioecology.
PACS number(s): 23.40.−s.

1. Introduction

The UN's 17 Sustainable Development Goals call for healthy lives and well-being for all at all ages. As part of task 3.4, monitoring radioactive gases (particularly radon) is recommended to assess their impact on public health [1].

The radioactive gas radon is a daughter product of natural uranium and thorium decay in the soil. It can spread from the source and accumulate in the air, water sources, and plants (flora) by molecular diffusion, gas diffusion, erosion, and dissolution processes [2, 3]. At the same time, its daughter products of decay are heavy metals, which pose the main danger when they enter the body. WHO and IARC classify radon as a Group I carcinogen [4, 5]. It has been clearly established that the impact of radon and its decay products on the human body increases the probability of developing lung and bronchial cancer, especially for non-smokers [6, 7]. And according to data [5], it ranges from 3% to 14%. Moreover, studies [8-10] show a correlation between radon levels and the probability of developing lung cancer. According to statistics for the Republic of Kazakhstan [11], mortality from lung cancer in the country is in first place. An analysis of global studies has shown that there are studies that radon exposure may be associated with other types of cancer [4, 12-14]. However, the results obtained in these studies were not so clear and convincing that further research in this area is relevant.

Radon is colorless and odorless, so it is not detected by the senses, which makes it dangerous for the population [5]. In open spaces, radon quickly dissipates in the air, while in closed spaces, it can accumulate and reach high values, creating a serious danger to human health [15]. This is especially true if we consider that according to data [16], the population spends more than 80% of its time in closed spaces every day.

In Almaty and the Almaty region, the problem of radon hazard in the territory, despite the presence of a large number of tectonic faults (while radon levels in rooms near and above faults can reach high values

[17, 18]), has not been sufficiently studied [19, 20], and comprehensive studies on the impact of radon on public health are not being conducted.

Although indoor radon exposure is the main contributor to the annual radiation dose from natural sources, it is also necessary to consider radon's ability to accumulate in the human environment. The decay of radon isotopes produces alpha, beta, and gamma radionuclides, which are easily absorbed by substances in various phase states [21]. Radionuclides can be found in air, water, and soil as a result of natural and artificial pollution [22], which can increase short-term and/or long-term effects on human health. Therefore, searching for and monitoring local radon "flares" in Almaty is a relevant area.

The relevance of the study in the direction of increasing the risk of cancer is associated with internal irradiation caused by inhalation and food consumption of radon and its decay products, which can cause damage to the respiratory tract, as well as the intestinal tract [12, 23, 24]. The ICRP [15] recommends that all countries establish a reference range for radon volume activity of 100÷300 Bq/m^3. In this regard, the need to monitor radon concentration measurements in residential and administrative buildings [25], where the population spends a lot of time, is becoming more urgent.

At the same time, 80% of the total radiation dose for the population is formed by natural long-lived radionuclides and their decay products, accumulating in soil, water, and air, making them an urgent research problem [26]. Soil plays a significant role in radioactive contamination of the environment, being one of the main sources of radiation exposure to the population, as it serves as the main means of transporting radionuclides into biological systems [27, 28]. When radon decays, radionuclides such as ^{210}Po, ^{210}Pb, ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po are formed. ^{210}Pb has a long residence time in the body and makes a large contribution to the radiation dose [29]. ^{210}Pb and ^{210}Po accumulate for decades in small air passages in the lungs (bronchioles) and gastrointestinal tract and are subsequently absorbed into the blood. Then, there is a transition to other organs and tissues, causing local radiation doses to these organs and tissues.

Since radon is characterized by seasonal, daily, and multi-day variations [30-33], which is associated with differences in pressure, temperature, and other environmental parameters, then in order to determine areas exposed to radon, it is recommended to establish patterns of concentration of its decay products in the soil in order to predict its accumulation in a given area depending on the physical and chemical characteristics. At the same time, according to [34], using the results of gamma studies of soil samples can improve the mapping of radon hazard zones. This is due to the fact that most gamma-emitting nuclides, as well as radon isotopes, are daughter products of decay in natural decay chains from ^{235}U, ^{232}Th, and ^{238}U. Therefore, comprehensive radiation monitoring of environmental objects can be used to assess the expected annual dose from natural radiation sources and construct maps of radon potential in the areas under study.

2. Materials and methods

In this work, radiometric measurements of the equivalent equilibrium volumetric activity (EEVA) of radon in buildings located at different distances from tectonic faults and beta- and gamma-spectrometric measurements of soil samples were performed.

The measurements were carried out at different distances from tectonic faults using the radon radiometer "RAMON-02" in temporary and long-term buildings. Radiometers "RAMON-02" are used to measure the EEVA of radon in the range of 1÷5•10^5 Bq/m^3. The EEVA values of radon and its decay products were obtained by measuring the RAMON-02 device at the positions indicated in Figure 1. For this study, buildings located in Almaty, southeastern Kazakhstan, in the foothills of the Zailiyskiy Alatau, were selected. Radon monitoring measurements were carried out from January 2024 to July 2024 at different distances from tectonic faults. The latest seismic microzonation map, taking into account all known faults, was developed and approved in 2021 by the Institute of Seismology [35].

The Faculty of Physics and Technology (43°13'25.88"N 76°55'29"E) was identified as the first study object (No. 1), located 235 m from the nearest tectonic fault. The second study object (No.2) was the Student House (43°13'08.7"N 76°55'30.2"E), located 27 m from the tectonic fault. The third study object (No. 3) was the Student House (43°13'06"N 76°55'14"E), located 274 m from the fault.

The next object (No. 4) was the Student Service Center building (43°13'11.7"N 76°55'19.4"E), located 386 m from the fault. Measurements were taken in the basements. The fifth object (43°12'35.9"N 76°53'18.5"E) is 1,500 meters from the nearest tectonic fault. The five-story building has four entrances, and the house is a privatized dormitory. The sixth object (43°13'56.7"N 76°57'34.4"E) is 917 m from the fault and is an office building of the Koktem Towers business center. The seventh object of study (No. 7, 465 m from the fault) was the Faculty of Biology and Biotechnology (43°13'25"N 76°55'15"E).

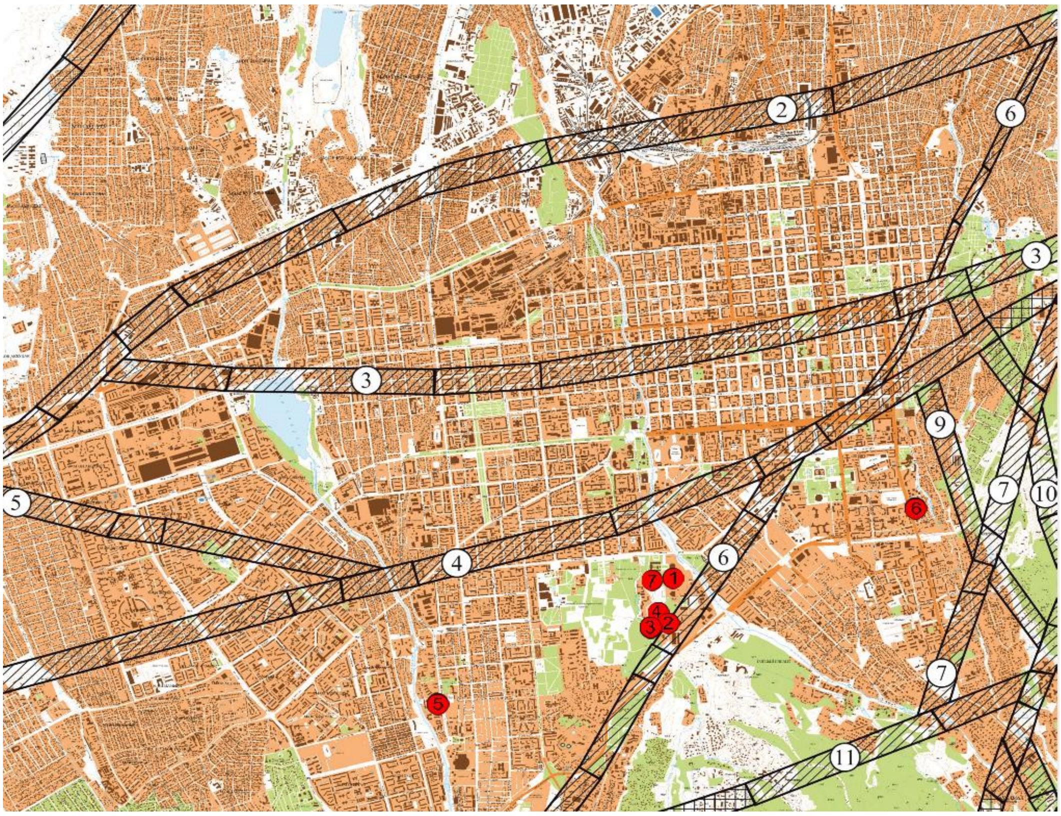


Figure 1 – Radon measurement positions (red dots) on the map of active tectonic faults [36]

All radon concentration measurements were taken in closed, unventilated rooms to determine the highest potential for radon exposure. This is because closed spaces without the influence of air flows have higher radon levels than rooms with good natural or artificial ventilation systems [5]. Three radon EEVA measurements were taken in each position using a RAMON-02 radiometer (exposure time for each measurement was 128 seconds) per day, with subsequent conversion to average daily values for each measurement position.

Despite the fact that radon isotopes are alpha emitters, most of their decay products are beta and gamma emitters with fairly long half-lives: ^{214}Pb ($T_{1/2}$ = 26.8 min), ^{210}Pb ($T_{1/2}$ = 22.2 years), ^{214}Bi ($T_{1/2}$ = 19.9 min), ^{212}Bi ($T_{1/2}$ = 61 min), ^{210}Bi ($T_{1/2}$ = 5.01 days), ^{210}Po ($T_{1/2}$ = 138 days). In this regard, within the framework of this work, beta and gamma spectrometry of soil samples taken at different distances from the fault was also carried out to search for a relationship between the level of accumulated activity and the level of radon concentration. The measurements were carried out on beta and gamma spectrometric installations SKS-99 "SPUTNIK" with scintillation types of detectors based on CsI (45x50 mm) for measuring the activity of gamma-emitting radionuclides and based on ZnS with plastic for measuring the flux density of beta particles.

Soil sampling for gamma and beta measurements was carried out in Almaty at the coordinates indicated in Table 1. Nine samples were collected at different positions located from the nearest tectonic fault between 144 m and 1190 m. About 1 kg of soil was collected for the soil samples by removing 5 cm of the surface soil layer. The measurement positions are shown schematically in Figure 2.

Table 1 – Spectrometric measurement positions of soil samples

No.	Coordinates of measurement positions	Distance to fault, m
1	43°13'09.8"N 76°55'12.6"E	394
2	43°13'24.2"N 76°55'29.0"E	233
3	43°13'20.7"N 76°55'30.5"E	177
4	43°14'37.5"N 76°54'04.4"E	371
5	43°13'08.8"N 76°52'49.9"E	613
6	43°13'38.4"N 76°53'41.7"E	144
7	43°13'38.8"N 76°53'06.8"E	307
8	43°15'46.3"N 76°56'39.2"E	913
9	43°15'58.5"N 76°56'38.6"E	1190

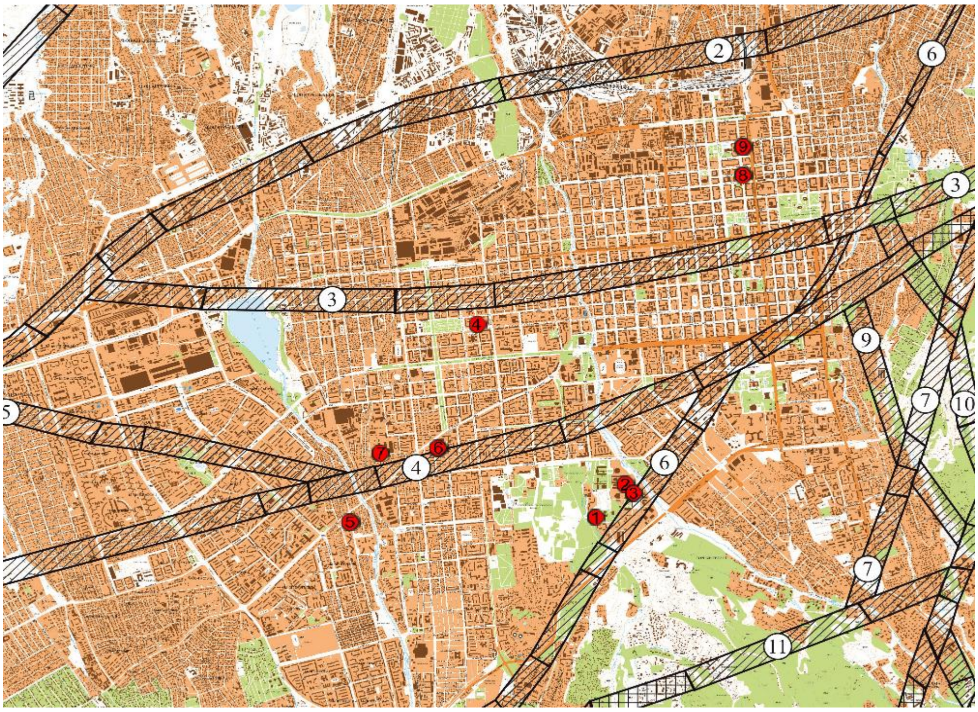


Figure 2 – Positions of beta and gamma activity measurements (red dots) on the map of active tectonic faults [36]

The beta spectrometer was calibrated using the Sr-Y-90 beta source, and the gamma spectrometer was calibrated using Cs-137 gamma sources. Figure 3 shows the calibration curve of the beta spectrometer using the Sr-Y-90 beta source.

After soil sample preparation, a quantitative assessment of the natural beta and gamma activity concentration in the selected samples was performed. Each sample was pre-dried for at least 24 hours (Figure 4a). Manual grinding was performed (Figures 4b and 4c) to increase the concentration of the samples and obtain a homogeneous mixture. Weighing the samples was performed using a digital scale SF-400 with an accuracy of ±0.01 g. Table 2 shows the net weight of the selected soil samples for beta and gamma spectrometry.

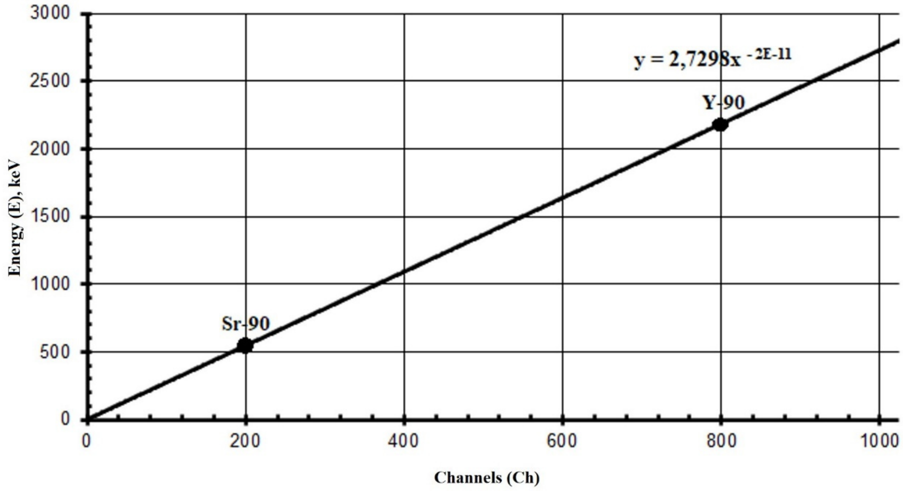


Figure 3 – Beta-spectrometer energy calibration curve

a) b) c)

Figure 4 – Soil sample preparation for spectrometric measurements:
a) preparation of soil samples for drying; b) the process of manual grinding of samples for gamma-spectrometry analysis; c) the process of manual grinding of samples for beta-spectrometry analysis

Table 2 – Sample mass for beta and gamma spectrometry

Sample Code	Position (coordinates) of sample selection	m_β, g	m_γ, g
1	43°13'09.8"N 76°55'12.6"E	22	632
2	43°13'24.2"N 76°55'29.0"E	25	467
3	43°13'20.7"N 76°55'30.5"E	22	416
4	43°14'37.5"N 76°54'04.4"E	21	501
5	43°13'08.8"N 76°52'49.9"E	17	376
6	43°13'38.4"N 76°53'41.7"E	17	459
7	43°13'38.8"N 76°53'06.8"E	16	456
8	43°15'46.3"N 76°56'39.2"E	18	462
9	43°15'58.5"N 76°56'38.6"E	19	503

The exposure of measurements on the SKS-99 "SPUTNIK" spectrometers was no less than 10,000 events per sample to reduce statistical uncertainty. Background measurements were performed before and after each sample measurement. The background spectra were used to correct the net areas of the measured samples' spectra.

The equipment used for measurements has been verified by SOLO LLP and the Almaty Certification Bureau (verification certificate No. BA.17-04-47195 dated September 28, 2023, and is valid until September 28, 2024).

3. Results and discussion

The analysis of the obtained results showed that the geometric mean value for the measured positions was 38.9 Bq/m^3. The radon concentration fluctuated between 6.13 and 405.21 Bq/m^3 (Figure 5). The analysis of the results showed that the radon EEVA decreases exponentially according to the following pattern: $\boldsymbol{C_{Rn}=499.85 \cdot e^{-0.0078 \cdot R}}$, where $\boldsymbol{R}$ is the distance to the nearest tectonic fault. In addition, for some measurement positions in rooms close to a tectonic fault, values exceeded the standard values for radon concentration in indoor air (200 Bq/m^3) [5].

Figure 5 shows that the highest radon concentration value was found at a distance of 27 m in building No. 2 (Figure 1). The results show the influence of the proximity of a tectonic fault on the level of radon concentration inside the premises, as in the works [37, 38]. This is most likely due to the fact that faults are a favorable migration path for radon gas since faults have higher permeability. Thus, if there are many cracks in tectonic faults, which form high permeability, radon from the depth can easily rise to the surface from the depth, leading to higher radon concentrations near the fault.

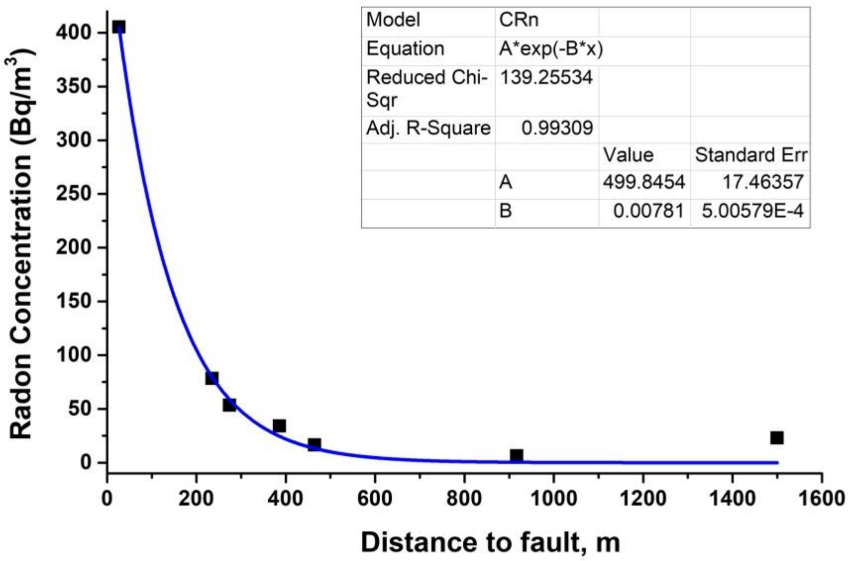
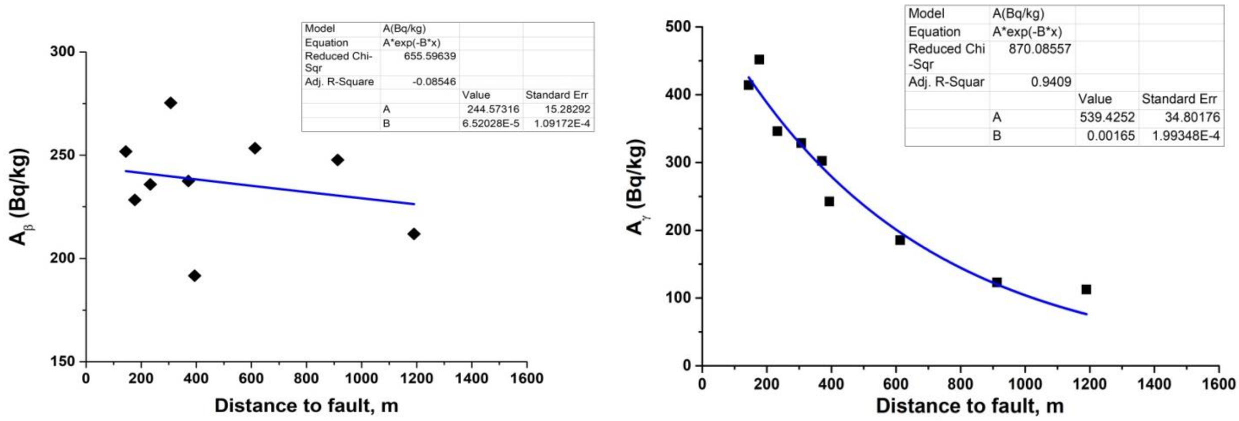


Figure 5 – Average indoor radon concentration depending on the distance from the tectonic fault

Based on the obtained experimental spectrometric data, the specific integral beta and gamma activities were assessed in the studied soil samples. The specific activity of the samples (Bq/kg) was calculated using the following formula [26]:

$$A = \frac{cps}{I_{Y,\beta} \cdot \varepsilon_{\gamma,\beta} \cdot m_{\gamma,\beta}} \qquad (1)$$

where $cps = \frac{N}{t} - \frac{N_0}{t_0}$ – net counts per second; I_γ and I_β –the gamma and beta emission probabilities; $\boldsymbol{\varepsilon}$ – is the detection efficiency; $\boldsymbol{m_\beta}$ – the mass (dry weight) in kilogram of the beta sample (in kg), $\boldsymbol{m_\gamma}$ – the mass (dry weight) in kilogram of the gamma sample (in kg). An exponential pattern of decrease in beta and gamma activities with distance to the tectonic fault $\boldsymbol{R}$ was found (Figure 6): for beta – $\boldsymbol{A_\beta = 244.57 \cdot e^{-6.52E-5 \cdot R}}$, for gamma – $\boldsymbol{A_\gamma = 539.43 \cdot e^{-0.00165 \cdot R}}$.



a) b)

Figure 6 – Concentration of a) beta and b) gamma activities of samples depending on the distance from the tectonic fault

The concentrations of beta activity in soil samples varied from 191.67±28.75 to 275.32±41.30 Bq/kg, and gamma activity from 112.51±16.88 to 451.60±67.74 Bq/kg. The results obtained do not exceed the established world standard of 465 Bq/kg for the activity of natural gamma radionuclides [39] and are in the same ranges for the obtained beta and gamma activities as in the works [40-44]. However, it is worth noting that the maximum value for the gamma activity of the soil (451.60 Bq/kg) obtained in this work exceeds the standard established for the Republic of Kazakhstan (397 Bq/kg) [39].

Pearson correlation analysis was used to calculate correlation coefficients [45]:

$$r_{xy} = \frac{\Sigma(x_i - \bar{x}) \times (y_i - \bar{y})}{\sqrt{\Sigma(x_i - \bar{x})^2 \times \Sigma(y_i - \bar{y})^2}} \qquad (2)$$

where x_i – values taken by variable X; y_i – values taken by variable Y; $\bar{x}$– average by X; $\bar{y}$ – average by Y. Correlation analysis showed that the beta activity concentration weakly correlates with the radon concentration in the air (correlation coefficient – 0.58). A strong correlation was found between the concentrations of gamma activity in the soil and the radon concentrations in the air (correlation coefficient – 0.79). Interpretation of the results of the correlation analysis leads to the following conclusions: 1) beta-emitting radionuclides may have less penetrating ability than gamma radionuclides; 2) spectrometric gamma analysis of radon decay products can be used as an indirect method for measuring radon concentration.

4. Conclusions

The concentration of radon activity and its decay products was measured by spectroradiometric methods for soil and air samples collected in Almaty at a distance of 27 m to 1500 m from the tectonic fault. The results show that the higher the indoor radon concentration, the closer the building is to the tectonic fault. The average radon concentration in the basement of building No.2 in Figure 1 (located 27 m from the tectonic fault) exceeds by 305% the value of 100 Bq/m^3 defined by WHO as requiring action [5] and by 102.5% the value of 200 Bq/m^3 defined in the Republic of Kazakhstan for buildings in operation [46]. This demonstrates the need to take corrective measures, for example, by improving the building's ventilation system.

In the soil samples examined in this study, the average concentration of beta and gamma activities is approximately in the same range as in other countries [47-49]. At the same time, the positive correlation between the concentration of gamma activity and the indoor radon concentration showed that the content of gamma radionuclides in the soil has a similar pattern of decreasing with distance from the tectonic fault. In other words, a high concentration of gamma radionuclides in the soil is a harbinger of increased levels of radon gas in the premises at present or in the near future.

It is important to identify the presence of positions with higher concentrations of radon and its decay products, as well as the causes of their occurrence, for an accurate assessment of the corresponding radon risk. These results will allow the development of protective measures to reduce the entry of radon into indoor air in the future. In particular, understanding the relationship between radon concentration and the activity of radon decay products will allow the development of risk maps and identify areas requiring constant radiation monitoring. This is especially true for areas with a high gamma background level, where urban planners can develop and implement radon-resistant construction methods associated with the potential danger of elevated radon levels.

Acknowledgments

This research is funded by the Science Committee of the Ministry of Science and Higher Education of the Republic of Kazakhstan (Grant No. AP23486701).

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Information about authors:

Zaripova Yuliya, PhD, Senior lecturer at al-Farabi Kazakh National University (Almaty, Kazakhstan) e-mail: zarypova.iulyiya1@kaznu.kz

Yushkov Alexandr, doctor of science, professor at al-Farabi Kazakh National University (Almaty, Kazakhstan) e-mail: yushkov.prof@kaznu.kz

Amangeldiyeva Nazerke. Laboratory assistant at al-Farabi Kazakh National University (Almaty, Kazakhstan) e-mail: amangeldiyeva.nazerke26@gmail.com

Dyussebayeva Kuralay, Senior lecturer at al-Farabi Kazakh National University (Almaty, Kazakhstan) e-mail: kuralay.diusebaeva@kaznu.kz

Shaidollina Aknur. 2nd year Master student, laboratory assistant at al-Farabi Kazakh National University (Almaty, Kazakhstan) e-mail: akojank02@gmail.com