

Article

Natural Radionuclides in Soils of Natural-Technogenic Landscapes in the Impact Zone of Potassium Salt Mining

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Abstract: This study considers the contents of natural radionuclides (^{226}Ra , ^{232}Th , and ^{40}K) in the soils of natural–technogenic landscapes in the Verkhnekamskoe potassium–magnesium salt deposit (Perm Krai, Russia). The purpose of the study is to assess the lateral activity distribution of natural radionuclides (^{226}Ra , ^{232}Th , and ^{40}K) in the soils of natural–technogenic landscapes of the mining areas on the territory of the Berezniki–Solikamsk industrial hub. Seventy-five soil samples from depths of 0–0.4 m are examined. The specific activities of the natural radionuclides ^{226}Ra , ^{232}Th , and ^{40}K in the soil samples are measured using gamma spectrometry. The average values of the activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K are 11.78 Bq/kg, 8.11 Bq/kg, and 246.9 Bq/kg, respectively. A gamma survey is conducted using the MKS/SRP-08A search dosimeter–radiometer over a control point framework covering 50,000 hectares. The research shows that the migration of the studied natural radionuclides is limited to a radius of 10 km. The results of the gamma-radiation measurements in the study area fall within a range of 0.06–0.25 $\mu\text{Sv/h}$, which does not exceed the permissible limits. According to the findings, there is an increase in the concentration of natural radionuclides in transit supra-floodplain terrace landscapes and accumulative floodplain landscapes in the area affected by potash mining.

Keywords: soils; ^{226}Ra ; ^{232}Th ; ^{40}K ; potash industry; landscapes



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1. Introduction

Natural radionuclides have existed on Earth since its inception. They are distributed throughout the biosphere and accumulate in the atmosphere, soil, bottom sediments, water, and plants. The abiogenic cycle of radionuclide migration in the biosphere consists of three major stages: (1) matter mobilisation during rock weathering; (2) transfer by a transport medium; (3) sedimentation in local areas and final runoff reservoirs during transportation. The transportation process is the primary source of material that enters the biogenic cycle during soil formation processes [1]. There are numerous radionuclides in the environment containing uranium (U) radioisotopes, thorium (Th) radioisotopes, and natural ^{40}K [2]. Cosmic radiation and radioactive elements in the Earth's crust, water bodies, and atmosphere are the primary sources of the natural radiation background in any area [3]. ^{238}U , ^{232}Th , ^{226}Ra , and ^{40}K are the most common naturally occurring radioactive isotopes in the environment [3,4]. The natural radiation background varies depending on certain territorial aspects, including geological features such as the contents of thorium (Th), uranium (U), and potassium (K) in the rocks [5]; and geochemical features of radionuclide behaviour in different natural environments. The natural radionuclide activity concentrations in the soil range from micro- to thousands of becquerels per kilogram [6–8].

The radiation situation in any territory in modern conditions is formed by the natural and anthropogenic backgrounds. While radionuclides are formed naturally in soils and rocks as a result of radioactive decay, the majority of the environmental phenomena associated with radioactive contamination are caused by mineral extraction and processing. Mining is a considerable source of natural radioactivity [9]. The active development of

industrial production resulted in radical changes in environmental conditions, particularly intense radioactive contamination of the main components of the natural environment (soils, natural waters, rocks, and bottom sediments) [10,11]. Natural radionuclides enter water bodies and accumulate in soils and bottom sediments as a result of migration. Their high concentrations can endanger the habitats of microorganisms, animals, and humans. Natural formations (rocks with a high content of natural radionuclides [12–14]), as well as the mining and processing of coal [15], oil [3], gas [16], and food salt [17–19], can be sources of natural radionuclides that are input into the environment. Mining waste can be a source of radiation [20,21].

Potash production waste is the primary source of environmental component contamination in the study area. The Berezniki–Solikamsk industrial hub is one of the most developed sites in the Verkhnekamskoe potassium–magnesium salt deposit (Verkhnekamskoe Salt Deposit) (Perm Krai, Russia). Environmental pollution caused by ore beneficiation waste is one of the issues posed by mining and processing. The majority of waste is dumped on the ground [22]. As a result of potassium ore enrichment, more than 270 million tonnes of halite waste and more than 30 million m³ of clay–salt slurry are stored on the surface in the development area of the Verkhnekamskoe Salt Deposit [23].

The technogenic impacts of potash production, together with a complex of regional conditions and ecological features of the study area, play an important role in the distribution and accumulation of natural radionuclides in soils of various landscape types. The level of soil radioactivity is an indicator for assessing the territory’s radioecological background [24,25].

The purpose of the study is to assess the lateral distribution of the activity of the natural radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K in the soils of natural–technogenic mining areas within the Berezniki–Solikamsk industrial hub in the area affected by potash enterprises. Naturally, ⁴⁰K is the most relevant of the natural radionuclides studied in the area.

2. Materials and Methods

The study area is located in the northern part of the Verkhnekamskoe Salt Deposit. Soil samples were collected at predetermined points throughout the territory, near mining operations and residential areas. Samples were taken from the upper soil layer at 0–0.4 m intervals. A total of 75 soil samples were collected and examined.

The soil samples were dried to air-dry at room temperature in the laboratory. Organic matter was removed from the dried samples. The samples were prepared for the examination of the specific activity of natural radionuclides (⁴⁰K, ²²⁶Ra, and ²³²Th). An MKS-01A MULTIRAD device (Amplituda, Zelenograd, Russia) was used for the gamma spectrometry. The “Progress” software package (Amplituda, Zelenograd, Russia) was used to process the gamma spectra. The specific activity error range was approximately ±(20%–30%) of the measured values.

An energy calibration of the spectrometer was carried out with a ¹³⁷Cs and ⁴⁰K reference source. The source was a 1 L Marinelli beaker filled with KCl and sealed with a lid. The source had a cylindrical deepening area at the base with the fixed ¹³⁷Cs source (a disc 29 mm in diameter with an active spot in the centre sealed with two 50-µm-thick polyamide films). The source activity was 1500 Bq, with 20% error at a confidence level of 0.95. The spectrum from the gamma spectrometer’s calibration source showed the maxima of the ¹³⁷Cs (energy 662 keV) and ⁴⁰K (energy 1461 keV) total absorption peaks, as well as the count rate in a specific energy range of 620–700 keV. After calibration, the background was measured (for 1800 s) to control the intrinsic noise of the gamma spectrometric tract and to record the contribution of the external gamma radiation to the device’s emission spectrum of the studied sample. The soil samples were placed in the Marinelli beaker up to a 1 L scale marking.

The allowable exposure standards for natural radionuclides were determined in accordance with the radiation safety standards. Their biological impact on the human body was also considered using the calculated value of the specific effective activity A_{eff} .

This reflects the total specific activity of natural radionuclides in a studied material. It was calculated using the following formula:

$$A_{\text{eff}} = A_{\text{Ra}} + 1.31A_{\text{Th}} + 0.085A_{\text{K}} \quad (1)$$

where A_{Ra} and A_{Th} are the specific activities of ^{226}Ra and ^{232}Th (Bq/kg), which are in equilibrium with the rest of the uranium and thorium series members, and A_{K} is the specific activity of ^{40}K (Bq/kg).

Because of the uneven distribution of natural radionuclides in the soil, the homogeneity of radiation exposure was measured based on the radium equivalent (Raeq) in Bq/kg to compare the specific activity levels of materials containing varying amounts of ^{226}Ra , ^{232}Th , and ^{40}K [26–28]. The activity levels were calculated using the following formula:

$$\text{Raeq} = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}} \quad (2)$$

As part of the gamma survey, reconnaissance was carried out. The natural conditions of the area were detailed, specifying the thickness and nature of the deposits, and the gamma background for the rocks was determined. The ambient dose equivalent rate of continuous gamma radiation was estimated during the gamma survey. A search dosimeter-radiometer MKS/SRP-08A (Amplituda, Zelenograd, Russia) was used.

The landscape structure of the study area was investigated using a landscape geochemical approach aimed at exploring the migration of natural radionuclides (^{40}K , ^{226}Ra , and ^{232}Th) in a landscape. This approach considers the role of natural factors in the transformation of matter flows affected by the regional and local parameters of the area. This part of the study was based on landscape descriptions and soil sampling data from autonomous and secondary landscapes in key areas along the landscape's geochemical profile. This profile reflects the successive changes in soils in middle taiga landscapes while considering the locations of potash industry facilities.

The maps were created using the ArcGIS software package (version 10.6.1, ESRI, Redlands, CA, USA) via interpolation.

3. Results and Discussion

Subsoil use has a large impact on the transformation of natural complexes. Mining has a long-term negative impact on the environment. The enrichment of ores and the storage of waste on the surface have a negative impact on the soil. This changes the chemical and physical properties of the soil and increases the contents of salts and microelements [29].

The study area is part of boreal East Europe's middle taiga landscapes. They are located on the denudation plain of the Middle Cis-Urals within the eastern margin of the Russian Platform and the Ufa–Solikamsk depression of the Cis-Ural foredeep. The area consists of nearly horizontal Permian rocks that are covered by a thin layer of anthropogenic deposits [30,31]. These geosystems have been heavily impacted by anthropogenic activity over the last eight decades as a result of active mining.

The assessment of the environmental consequences of mining and the development of environmental protection measures are among the priority tasks aimed at ensuring both radiation protection for the population and the sustainable functioning of natural ecosystems. Soil is the main buffer of the natural environment, bearing the cumulative effect of long-term anthropogenic impacts [32–34]. The radiation characteristics of the upper soil layer (up to 5 cm) is the most important aspect in a soil analysis, since it takes on 95%–98% of the anthropogenic impact.

The average activity concentrations for ^{226}Ra , ^{232}Th , and ^{40}K are 11.78 Bq/kg, 8.11 Bq/kg, and 246.9 Bq/kg, respectively, while the standard deviations are 7.26 Bq/kg, 6.51 Bq/kg, and 114.28 Bq/kg, respectively. Details are provided in Supplementary Table S1.

Table 1 shows the results of the natural radionuclide's content measurements in the studied areas, as well as published data from around the world. The specific activity of ^{226}Ra (A_{Ra}) found in the study area corresponds to the global average level and is closest to

the levels found in Egypt and Brazil. In the study area, isolated cases of ^{226}Ra exceeding the world average levels are found (noted exceedance is up to 9%), as well as isolated exceedances of the world average levels of ^{40}K (exceedances range from 2 to 25%). At the same time, the ^{232}Th activity (A_{Th}) in the study area is significantly lower than the global average values and specific activity values for other studied areas of the world.

The average levels of ^{226}Ra , ^{232}Th , and ^{40}K in the soils in the central part of the study area are 14 Bq/kg, 17 Bq/kg, and 369 Bq/kg, respectively. The formation of an area with higher concentrations of natural radionuclides is caused by meteorological conditions, specifically the wind regime of the study area. Southerly winds occur frequently throughout the year. Taking into account the prevailing wind direction, one can predict the considerable removal of elements with atmospheric flows in the northern direction (Figure 1).

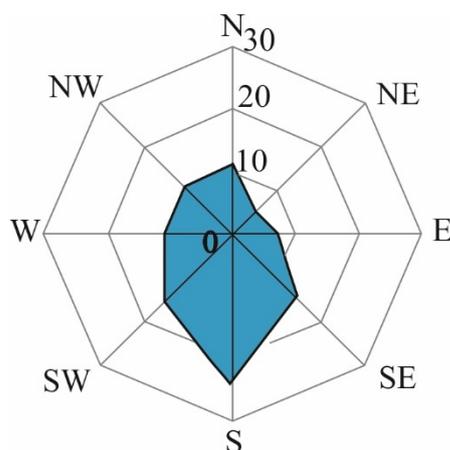


Figure 1. Wind activity in the study area.

Increased values of the specific effective activity (A_{eff}) and specific activity of ^{40}K were observed in the soils of the south-eastern part of the study area. This is associated with the transfer of the element by airflow in the northern and north-eastern directions from the potash mining territory located south of the study area (Figures 2 and 3). This potash enterprise is distinguished by a higher volume of allowed potassium chloride emissions into the atmosphere and the high hypsometric position of the considered area.

The specific effective activity (A_{eff}) is one of the parameters describing the radiation situation. According to the published studies [35,36], the gradation of A_{eff} is defined by the following values (in Bq/kg) based on the level of danger: especially dangerous (more than 3300); dangerous (1101–3300); potentially dangerous (101–1100); safe (less than 100). The average value of the total specific effective activity of natural radionuclides for the soil in the study area is 43 Bq/kg, which corresponds to a safe level. At the same time, the obtained distribution pattern of the A_{eff} of the studied natural radionuclides corresponds to the distribution pattern of the ^{40}K activity (Figures 2 and 3).

The results of the gamma radiation measurements in the study area fall within the range of 0.06–0.25 $\mu\text{Sv/h}$ (Figure 4), which is less than the established ambient dose equivalent rate of continuous gamma radiation for residential and industrial areas (0.3 and 0.6 $\mu\text{Sv/h}$, respectively) [37].

The average radium equivalent activity concentration index (Ra_{eq}) for soils in the study area is 42.39 Bq/kg, which is less than the standard's maximum allowable index (370 Bq/kg) [6]. Details are provided in Supplementary Table S1.

Table 1. Summary of the natural radionuclide activity concentrations in soils within the study area and in other parts of the world in Bq/kg [6,38].

Activity	Natural Radionuclide Activity in the Study Area (Mean Value for 75 Samples)	China [6]	USA [6]	Egypt [38]	Brazil [38]	Global Average Values [6]
A ₂₂₆ Ra	3–36 (17)	1–360	4–130	31–40	29.2	16–116 (33)
A ₂₃₂ Th	1–28 (11)	2–690	4–140	52–61	47.8	7–50 (45)
A ₄₀ K	20–562 (298)	9–1800	100–700	3149–3210	704	100–700 (420)

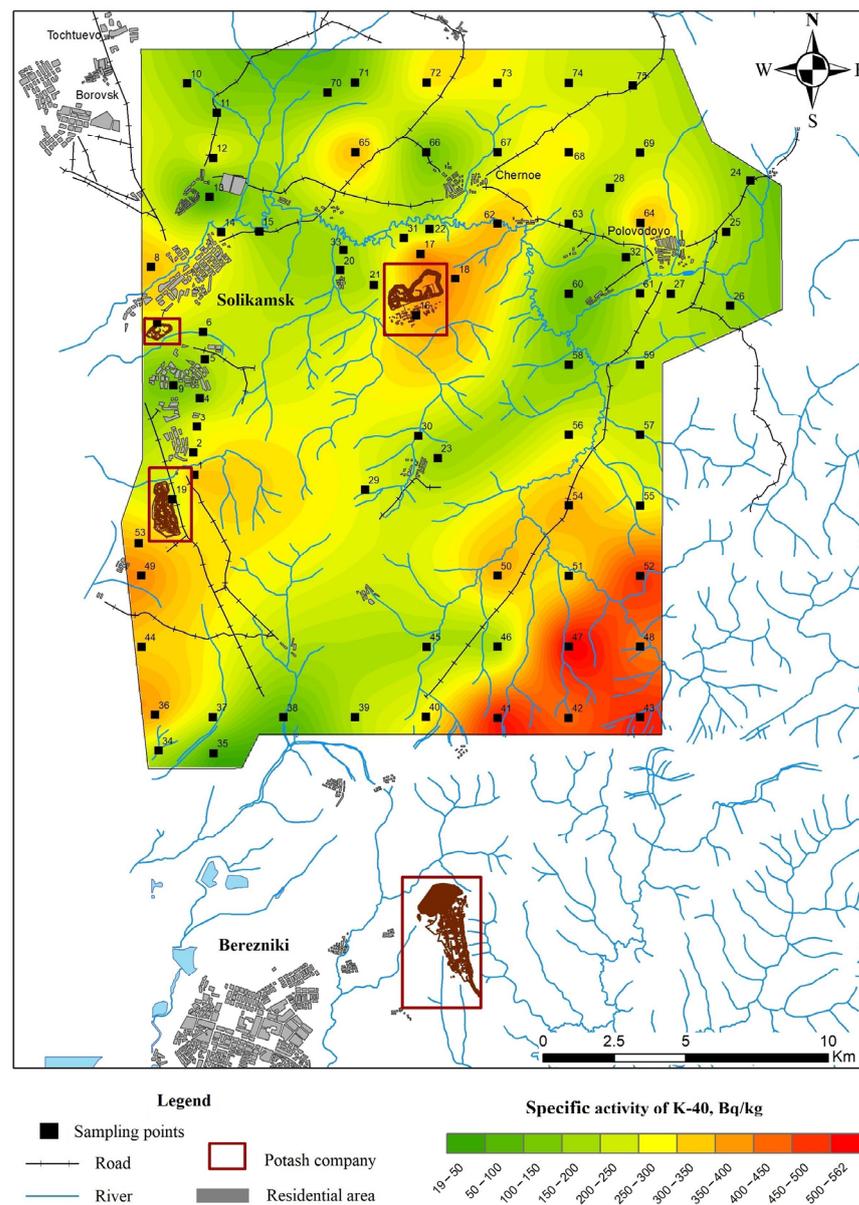


Figure 2. Map of the specific activity of ⁴⁰K in soils within the study area.

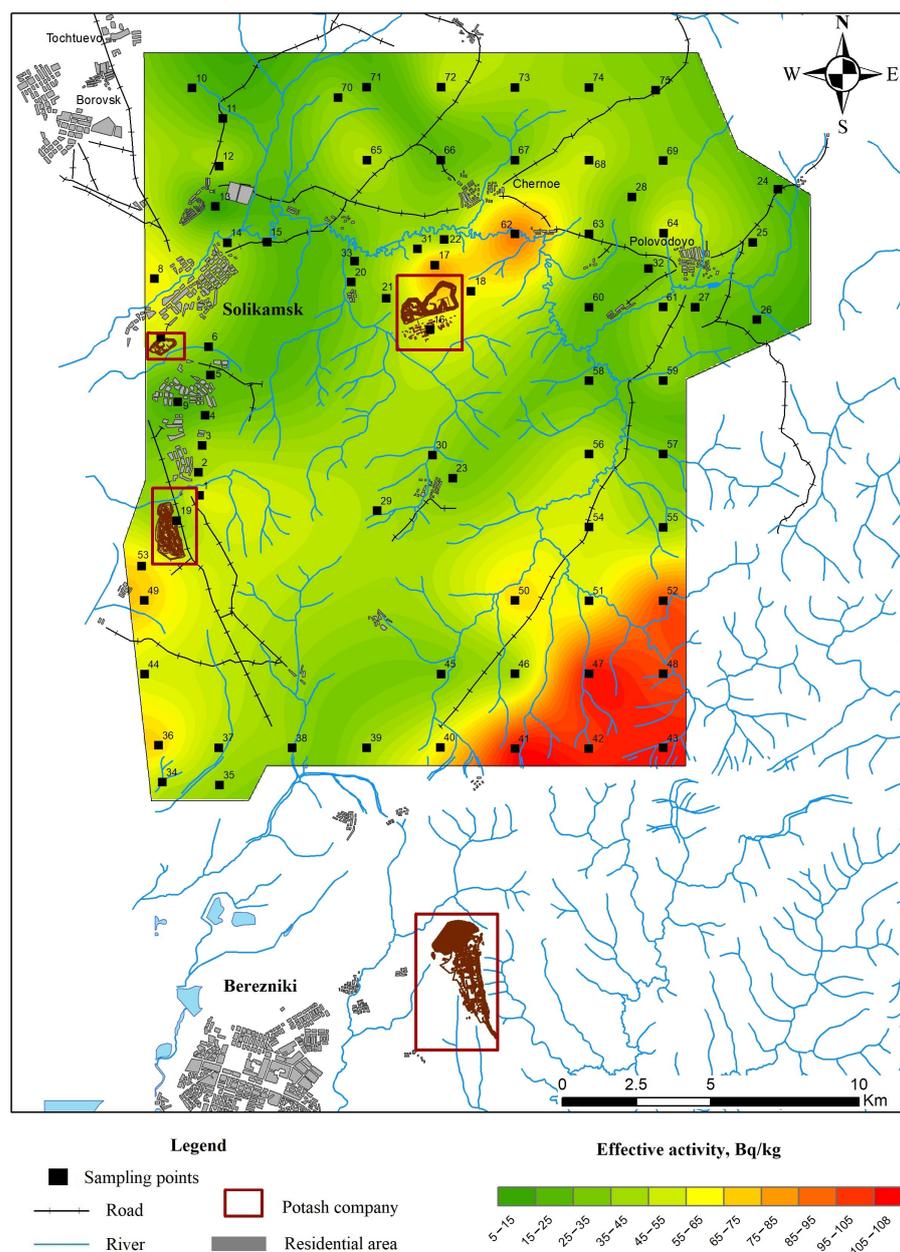


Figure 3. Map of the specific effective activity (A_{eff}) in soils within the study area.

3.1. Natural Radionuclide Differentiation in the Middle Taiga Landscapes Based on the Potash Enterprise Location

All geochemical processes that occur in landscapes are reflected in the soils. In the study area, alluvial and podzolic soils are developed under forest and grass communities within various types of landscapes. According to the geochemical map of the landscape (Figure 5) and the results of the specific efficiency analysis of natural radionuclides (Figure 3), the products of weathering and soil formation in an autonomous landscape come to the lower parts of the relief with the surface runoff and affect the formation of secondary surface and underwater landscapes. It is well known that radionuclide leaching is a common process in soils found in eluvial landscapes [39].

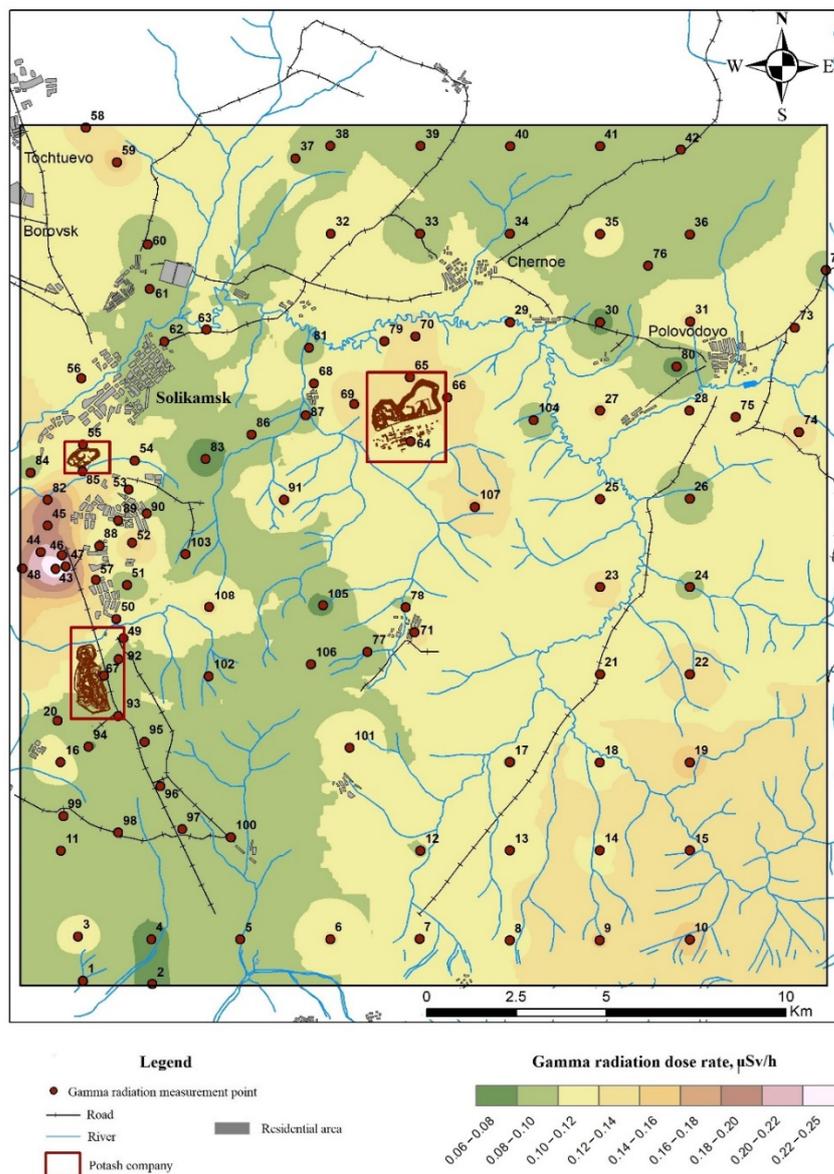


Figure 4. Map of the gamma background of the study area.

Additionally, there is a link between the distribution of various radionuclides in a landscape. This is especially noticeable in hydromorphic and semi-hydromorphic soils within accumulative landscapes. They are distinguished by a relatively higher accumulation of ^{226}Ra compared to ^{232}Th . Additionally, ^{226}Ra is washed out of the upper horizons (A0, A1, and A2) and accumulated in illuvial horizon B. The podzolization and gleying processes lead to ^{226}Ra and ^{232}Th leaching from the upper horizons A1 and A2 and their accumulation in the illuvial horizon B and gley horizons G and Bg [1].

According to the research findings, there is an increase in the concentration of natural radionuclides in transit supra-floodplain terrace landscapes and accumulative floodplain landscapes within the area affected by potash mining. The distribution of ^{226}Ra along the profiles of various soil types is nearly identical to that of ^{232}Th . Increased contents of ^{226}Ra and ^{232}Th were observed in the floodplain soils of the study area (an accumulative type of element distribution), in the area affected by potash mining, and in residential areas. In accumulative landscapes, the average content of ^{226}Ra (14 Bq/kg) is 1.4 times higher than the content of ^{232}Th (10 Bq/kg). The average content of ^{226}Ra is 8 Bq/kg in eluvial landscapes, 11 Bq/kg in transit landscapes, and 14 Bq/kg in accumulative landscapes. The

average ^{232}Th content is 6 Bq/kg in eluvial landscapes, 7 Bq/kg in transit landscapes, and 10 Bq/kg in accumulative landscapes (Table 2).

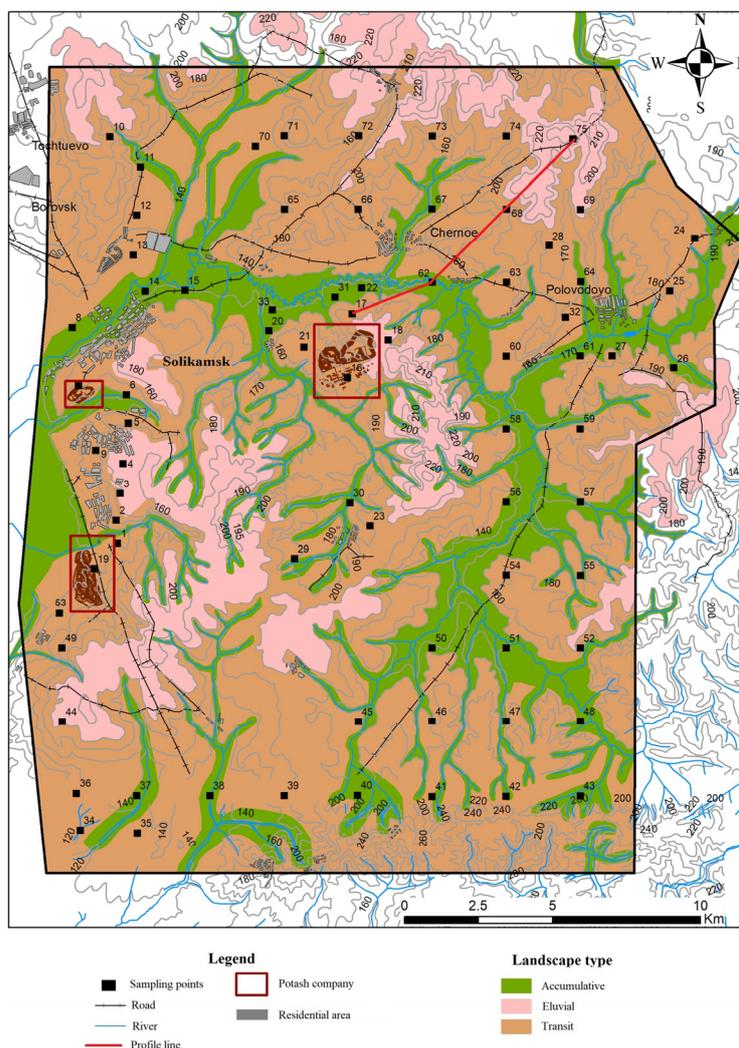


Figure 5. Landscape map of the study area.

Table 2. Average contents of natural radionuclides in different landscapes within the study area in Bq/kg.

Activity	Type of Landscape		
	Eluvial	Transit	Accumulative
A_{Ra}	8	11	14
A_{Th}	6	7	10
A_{K}	247	225	279

According to the findings [7], the activity concentration of ^{40}K in the soil is an order of magnitude greater than for ^{232}Th and ^{238}U . The lateral distribution of ^{40}K mostly depends on the anthropogenic factors, wind regime, landscape type, and geomorphological characteristics. Thus, the maximum concentrations of ^{40}K in the study area were observed in areas affected by potash mining and other anthropogenic activities (including residential territories) (Figure 2). The highest average ^{40}K content was found in accumulative landscapes, where it exceeded 279 Bq/kg (Table 2).

3.2. Lateral Distribution of Natural Radionuclides in Soils Affected by Potash Mining

To study the distribution of natural radionuclides in the area affected by potash mining, soil sampling points were selected at various distances from the potash enterprise in the northern and north-eastern directions on various types of landscape (Figure 5). The data summary indicated the accumulative distribution of ^{226}Ra , ^{232}Th , and ^{40}K in the soils affected by potash mining, taking into account the regional conditions. The highest contents of ^{226}Ra (27 Bq/kg) and ^{232}Th (22 Bq/kg) were recorded at point 62 (accumulative landscape) at a distance of 2.9 km from the industrial site to the north-east (Figures 5 and 6). The contents of ^{226}Ra , ^{232}Th , and ^{40}K decrease with distance from the enterprise. The maximum content of ^{40}K (407 Bq/kg) was observed at a distance of 800 m from the potassium plant within the transit landscape, which is determined here by the active atmochemical impact of the potassium plant.

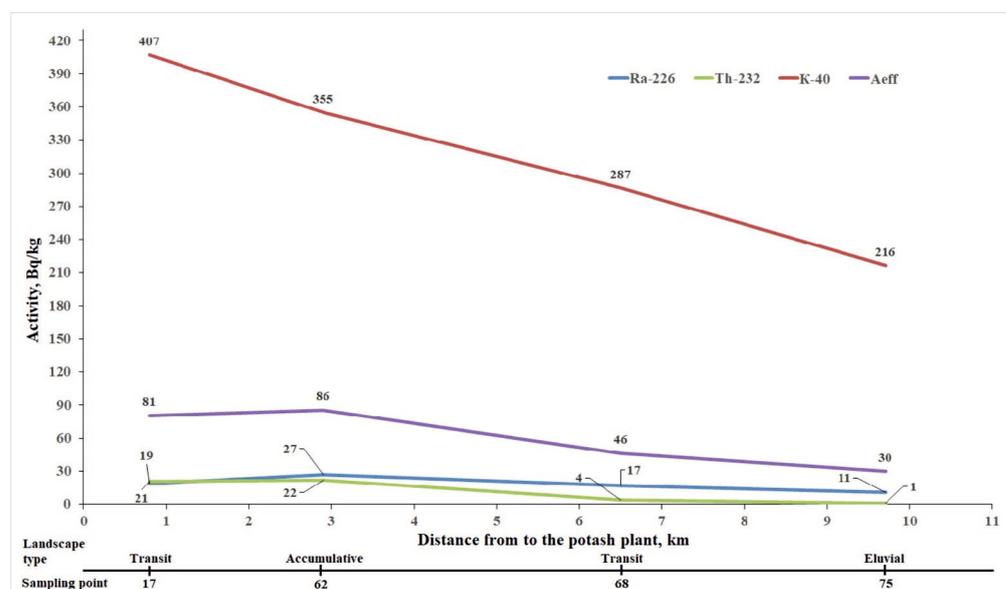


Figure 6. Distribution graph of ^{226}Ra , ^{232}Th , ^{40}K , and A_{eff} at points 17, 62, 68, and 75.

4. Conclusions

The research findings show that the study area in the Berezniki–Solikamsk industrial hub affected by the Verkhnekamskoe Salt Deposit’s development does not have an impact on the radiation situation. The ^{226}Ra , ^{232}Th , and ^{40}K activities in the top soil layers range from 3 to 36 Bq/kg, from 1 to 28 Bq/kg, and from 20 to 562 Bq/kg, respectively. These levels do not exceed the global average values. The results of the gamma-radiation measurements in the study area range from 0.06 to 0.25 $\mu\text{Sv/h}$, which is less than the established ambient dose equivalent rates of continuous gamma-radiation in residential and industrial areas (0.3 and 0.6 $\mu\text{Sv/h}$, respectively). The average A_{eff} index for the soil within the study area corresponds to the safe level.

The lateral activity distribution analysis of the natural radionuclides ^{226}Ra , ^{232}Th , and ^{40}K showed elevated ^{226}Ra and ^{232}Th contents in the floodplain soils and mining-affected areas of the natural–technogenic landscapes within the Berezniki–Solikamsk industrial hub. The effects of the potassium enterprises, the wind regime of the territory, the genetic type of the landscape, and geomorphological characteristics all have a great impact on the lateral distribution of ^{40}K . From the research, we found the accumulative distribution of ^{226}Ra , ^{232}Th , and ^{40}K in soils affected by potassium salt mining enterprises, considering the regional conditions. The future research will be focused on examining the distribution of natural radionuclides in soil horizons within areas with the highest concentrations.

The obtained results provide useful information on the natural radionuclide content in the area affected by mining facilities. The research findings are useful for a comparative

assessment of the contribution of potash mining to the radiation situation of an area, considering the landscape's structure.

The research findings can be used in radiation monitoring and developing practical recommendations to improve the ecological situation of a territory. Thus, a joint analysis of the natural radionuclide content and the landscape structure of a territory may serve as the basis for planning the survey grid when conducting monitoring studies of anthropogenic impacts.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/min12111352/s1>, Table S1. Activity concentrations of ^{226}Ra , ^{232}Th ^{40}K and Aeff in soil samples.

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