



Mechanisms of the formation of radioactive soil contamination in the waterstream zone from the tunnel in the area of underground nuclear tests at the Degelen site, Semipalatinsk test site

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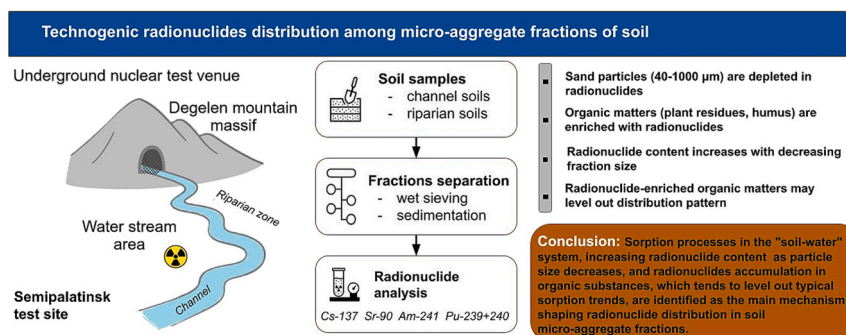
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HIGHLIGHTS

- Sand particles (40–100 µm) are significantly depleted in radionuclides
- Organic matter (plant residues, humus) is substantially enriched with radionuclides
- Radionuclide content increases with decreasing soil particle size due to sorption
- Radionuclide-enriched organic matter levels out the typical sorption patterns
- ¹³⁷Cs, ⁹⁰Sr and ²⁴¹Am, ²³⁹⁺²⁴⁰Pu exhibit different spatial distribution patterns

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigates the distribution of ¹³⁷Cs, ⁹⁰Sr, ²⁴¹Am, and ²³⁹⁺²⁴⁰Pu in soil micro-aggregate fractions of various sizes in a waterstream area emerging through a tunnel cavity at the Degelen site of the Semipalatinsk Test Site, where an underground nuclear test was conducted. The objective was to identify radionuclides distribution patterns in soil micro-aggregate fractions and key mechanisms of radioactive soil contamination resulting from prolonged contact with contaminated water. Results show that radionuclides distribution among soil micro-aggregate fractions is uneven and depends on fraction size and composition. Mineral components (40–1000 µm sand particles) are significantly depleted in radionuclides, while organic matter (partially decomposed plant residues, humus) demonstrates high accumulation, especially of ²³⁹⁺²⁴⁰Pu. A general trend was observed indicating that radionuclides concentration, especially ¹³⁷Cs, increases with decreasing particle size. However, this pattern is less expressed for ⁹⁰Sr, ²⁴¹Am, and ²³⁹⁺²⁴⁰Pu due to their significant enrichment in organic matter. Thus, sorption processes in the "soil-water" system, increasing radionuclide accumulation as particle size decreases, and the formation of organic substances with uniformly high radionuclide content, which tends to level

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out typical sorption trends, are identified as the main mechanisms shaping radionuclide distribution in soil micro-aggregate fractions. Additionally, the presence of highly radioactive individual particles (“hot” particles) may locally enhance enrichment of particular fractions.

1. Introduction

Radioactive contamination of natural environments with artificial radionuclides continues to pose one of the most complex environmental challenges, as released radionuclides enter the biogeochemical cycles and accumulate in ecosystem components over extended periods (Aleksakhin, 2009; Salbu, 2009; Salbu et al., 2018). Consequently, research into radionuclide behavior remains highly relevant, particularly regarding the mechanisms governing their migration and redistribution within environmental components. Advancing this understanding is essential for assessing and forecasting the radioecological situation in contaminated areas, evaluating potential risks to ecosystems and human health, and supporting the development of remediation and long-term monitoring strategies.

Intensive nuclear weapons testing during the second half of the 20th century significantly contributed to the release of long-lived artificial radionuclides including transuranic elements, fission products, and activation products. The Semipalatinsk Test Site (Kazakhstan), where 456 nuclear tests of various types were conducted between 1949 and 1989, was one of the sites that contributed to large-scale radioactive contamination of the environment. Radioactive soil contamination at the site continues to be a focus of extensive scientific research, offering valuable insights into contamination patterns and radionuclide behavior under varying environmental conditions. (Baklanova et al., 2020; Kabdyrakova et al., 2018; Krivitskiy et al., 2022; Kunduzbayeva et al., 2022; Panitskiy et al., 2022).

Underground nuclear tests of medium and low yield (up to several tens of kilotons) were carried out at the Degelen test site – a mountainous massif approximately 17–18 km in diameter – where 209 tests were conducted within 181 tunnels from 1961 to 1989 (Lukashenko, 2015). Following the dismantling of the nuclear weapons testing infrastructure in 1996–1998, all tunnels were sealed. Radioecological studies have shown that radioactive contamination across most of the site is negligible, as the majority (>90 %) of the radioactivity generated by the nuclear detonations confined within the tunnel voids. However, elevated radionuclide levels persist near certain tunnels where groundwater transports contaminants from explosion cavities to the surface forming spatially restricted meadow ecosystem characterized by radionuclide accumulation in riparian soils and bottom sediments (Lukashenko, 2010; Panitskiy et al., 2010; Panitskiy and Lukashenko, 2015; Yermakova et al., 2024).

The primary mechanism responsible for soil contamination under prolonged exposure to radioactive water may involve the sorption of radionuclides by soil particles from the liquid phase, including precipitation and co-precipitation processes. Similar mechanism is assumed to underlie the contamination of riparian soils and bottom sediments of water bodies affected by nuclear fuel cycle (NFC) facilities. Numerous studies have demonstrated that soils contaminated via sorption and precipitation/co-precipitation of radionuclides from a liquid media (contaminated water, liquid radioactive waste, etc.) are characterized by the highest accumulation of radionuclides in fine-grained (silty, clay) soil fractions. Radionuclide concentration increases with decreasing particle size, which is attributed to the greater specific surface area and enhanced sorption capacity provided by clay minerals. Thus, according to Nyhan et al., 1976, the highest specific activities of ²³⁸Pu and ²³⁹⁺²⁴⁰Pu in alluvial channel soils receiving liquid radioactive discharges in Los Alamos (New Mexico, USA) were found in the fine-grained fractions (<53 µm). These fractions had a surface area 7 times greater and a cation exchange capacity 3 times higher than the coarser fractions (2–23 mm). A positive correlation was observed between

plutonium concentration and particle surface area. Similarly, Livens and Baxter, 1988, observed a clear increase in radionuclide activity (¹³⁷Cs, ²⁴¹Am, ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu) with decreasing particle size in soils and estuarine silts contaminated by radioactive liquid discharge from the Sellafield nuclear facility (UK). The highest specific activities were detected in the clay-sized fractions (<2 µm), with enrichment levels ranging from 1.5 to 40 times those found in bulk samples. The authors noted the important role of particle sorption capacity, mineralogical composition, and organic matter content in the retention of actinides in soils and sediments. Linnik et al., 2004, Volosov et al., 2011 and Korobova et al., 2007, 2014 emphasized the key role of clay and silt fractions (<5–10 µm) in the accumulation of ¹³⁷Cs in alluvial soils and bottom sediments of the Yenisei River floodplain, impacted by discharges from the Krasnoyarsk Mining and Chemical Combine (Russia), as well as in the Iput and Buldynka rivers within the broader Chernobyl accident influence zone. This accumulation is attributed to the high surface area and sorption capacity of clay minerals, particularly illite and smectite. Radiocaesium retention is thought to occur primarily through specific sorption and ion-exchange processes. Moreover, highly active “hot” particles are presumed to contribute to the elevated radionuclide concentrations observed in certain particle-size fractions. Elevated radionuclide inventories in coarser soil fractions were also observed by the authors, which may result from their mass dominance in the soil matrix and the aggregation of fine particles. Therefore, these insights underscore the complex interplay of physical and chemical factors influencing radionuclide distribution in soils, emphasizing the importance of ongoing research to deepen our understanding of these processes.

This study examines the distribution of artificial radionuclides (¹³⁷Cs, ⁹⁰Sr, ²⁴¹Am, and ²³⁹⁺²⁴⁰Pu) across soil micro-aggregate fractions of varying sizes within the area of the waterstream originating from the tunnel where an underground nuclear test was conducted. The aim is to elucidate the primary patterns, quantitative characteristics, and mechanisms of radioactive soil contamination resulting from prolonged exposure to radioactively contaminated water.

2. Materials and methods

2.1. Object of study

The study was carried out at the Degelen site in the area of a waterstream flowing from the cavity of tunnel No. 177, where an underground nuclear test was conducted. The waterstream flows along a mountain terrace where the slope becomes gentler, flatter and more elongated. There are signs of anthropogenic disturbance near the water surface. The course of the waterstream is sinuous and braided. At the beginning of the waterstream, the channel splits into two parts, which join after 30–35 m to form a single stream. At a distance of 50–60 m, the waterstream overflows considerably and there are small areas with backfilled soils. The width of the channel does not exceed 1 m and its total length is about 485 m. At a distance of 400–450 m, the channel widens, becomes swamped and then dries up. The main radioactive contamination of the soil in the area of the tunnel is confined along the channel and floodplain of the waterstream.

The intensity of water discharge in a waterstream depends on seasonal changes. During heavy precipitation and snowmelt, the water may overflow and flood the riparian zones.

2.2. Study of waterstream relief

Absolute elevation was determined along the waterstream channel using a high-precision geodetic satellite positioning system Epoch50 (USA). The measurement of elevation above sea level was carried out along three profiles through the center of the waterstream channel and along the banks. The distance between profile points in the longitudinal direction was 5 m, while the distance between points in the transverse direction (from the point in the waterstream channel to the points on the banks) depended on the width of the channel, averaging 0.5 m.

2.3. Soil sampling

Soil samples were taken directly from the waterstream channel and from the flooded riparian zones (floodplain) as it moved away from the point where the water reached the ground surface (source). Soil samples from the channel were taken at the water's edge, i.e. at the point where the water surface intersects with the soil, on both banks. Floodplain (riparian) soils were sampled along the left bank at a distance of 1–5 m from the channel. 30 samples were taken along the waterstream channel and 8 samples along the floodplain (Fig. 1).

Soil samples were taken using a steel rectangular scoop-shaped sampler measuring 5, 10 and 10 cm in height, length and width, respectively. Sampling depths ranged from 5 to 25 cm depending on the bank height, with a sampling area of 200–300 cm². The weight of the samples ranged from 2.5 to 3.0 kg.

The geographical coordinates of the sampling point were determined using a GPS receiver. Exposure dose rate (EDR) and beta particle flux density were measured at the center of the sampling point using the MKS-AT6130 dosimeter-radiometer (Belarus). The EDR values varied from 0.88 to 2.86 $\mu\text{Sv/h}$, the beta particle flux density values ranged from 117 to 723 part./($\text{min} \times \text{cm}^2$).

2.4. Sample preparation for laboratory analysis

Soil samples were dried in a drying cabinet at a temperature not exceeding 60 °C and thoroughly homogenized. Large stones and plant inclusions were removed from the dried soil sample, which was then sieved through a 1 mm mesh sieve. The fraction >1 mm was discarded and the fraction <1 mm was used for further analyses (GOST 29269-91, 2005).

2.5. Separation of samples into micro-aggregate fractions

Samples were separated into micro-aggregate fractions using two methods: sieving with water washing ("wet sieving") and settling in still water (sedimentation). A 300 g soil sample was wetted with distilled water to a doughy state and then kneaded in a porcelain mortar for 10–15 min using a rubber-tipped pestle to disperse soil micro-aggregates (Vadyunina and Korchagina, 1986). The soil suspension obtained was then transferred in portions to a column of sieves with mesh sizes of 500, 250, 100, 63, 40 μm arranged in order of decreasing mesh size. The fractions on each sieve were thoroughly washed with a stream of water using a washer. Then purified soil fractions were quantitatively transferred to pre-weighed porcelain cups, dried and weighed. Thus, samples were separated to fractions of 40–63, 63–100, 100–250, 250–500 and 500–1000 μm by sieving. The fraction passing through a 40 μm sieve was further sedimented.

The soil suspension obtained after sieving (<40 μm) was transferred to a 1000 cm³ measuring cylinder (water column height ~ 32 cm), shaken until the bottom sediment was completely stirred up and allowed to stand for the required time. At the end of the settling time, the supernatant suspension was removed by decantation and the bottom sediment was collected in a separate container, dried and weighed. Sedimentation times averaged 18 min, 1 h 10 min and 20 h 28 min and were adjusted according to the temperature of the slurry according to the procedure (GOST 12365-14, 2015). The decanted suspension was put back to the cylinder and the water was made up to the same level and the above procedure was repeated. The particles that had not settled after 20 h 28 min were separated by centrifugation.

It should be noted that the investigation of the riparian and channel soils was carried out at different stages of the research, which resulted in the use of slightly different schemes for separating micro-aggregate fractions. Riparian soils were fractionated using a more detailed scheme delineating the following size classes: 1000–500 μm , 500–250 μm , 250–100 μm , 100–63 μm , 63–40 μm , 40–8 μm , 8–5 μm , 5–1 μm , and < 1 μm . A simplified scheme was applied to channel soils which included the following fractions: 1000–250 μm , 250–63 μm , 63–40 μm , 40–8 μm , 8–5 μm , <5 μm . Despite the difference in resolution, both schemes incorporated key size boundaries corresponding to major aggregate and textural classes (macro- and microaggregates, as well as sand, silt, and clay fractions), ensuring the comparability of results. For instance, in various classification systems, 250 μm and 63 μm are commonly considered thresholds for microaggregates and are typically associated with sand fractions (Filippova et al., 2019; Six et al., 2000) while the 5 μm boundary was recognized as the transition between silt

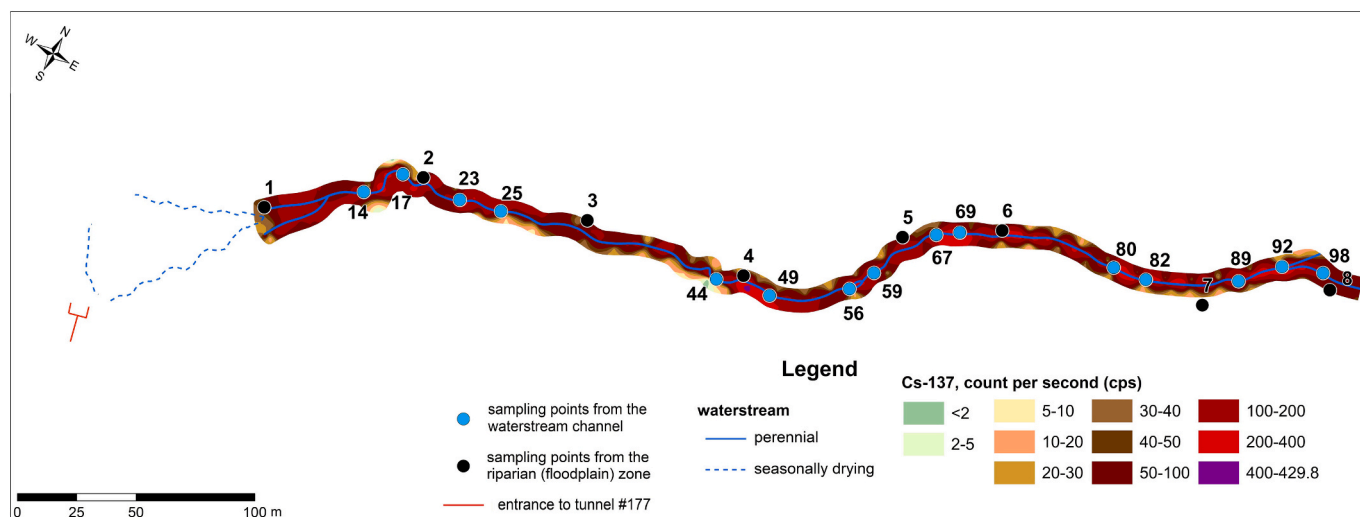


Fig. 1. - Location map of soil sampling points within the waterstream area of tunnel No. 177.

and clay fractions (Martín et al., 2018). Therefore, although the degree of fractionation detail differed, the selected schemes are methodologically sound and did not affect the interpretation of radionuclide distribution patterns among microaggregate fractions.

Fractions between 40 and 1000 μm were further separated into “organic” and “mineral” fractions. After sieving, the fractions retained on the sieves were transferred to a separate porcelain cup. The separation of the “organic” and “mineral” fractions was then carried out by gradually decanting the “organic” fraction, which floats and rises above the sand particles due to its low density. During this manual separation, the separated fractions could contain impurities from each other, but by batch processing of samples and repeated decanting, this risk was reduced to a possible minimum. The external view (photos) of the fractions is shown in Fig. 2.

All separated fractions were dried, weighed and the mass percentage of each fraction was determined by the ratio of fraction mass to total mass of sample taken for analysis.

All soil fractions ranging from 40 to 1000 μm collected from the waterstream channel were separated into “organic” and “mineral” components. For soils from the riparian zone, this separation was conducted on one representative sample due to the labor-intensive nature of the fractionation procedure and the large number of subsamples requiring subsequent radionuclide analysis.

2.6. Radionuclide analysis

Specific activity of the radionuclides ^{137}Cs , ^{241}Am , ^{90}Sr and $^{239+240}\text{Pu}$ was determined in soil samples and separated fractions. The specific activity of ^{137}Cs and ^{241}Am radionuclides was determined using a Canberra GX-2020 gamma spectrometer with a germanium semiconductor detector (BE 2020), (USA) (Activities of radionuclides in counting samples, 2014.). The measurement time varied from 20 min to 2 h, depending on the expected specific activity of the radionuclides in the samples. The detection limit was 1 Bq/kg for ^{137}Cs and 0.3 Bq/kg for ^{241}Am . The relative total uncertainty was up to 20 %.

The specific activity of ^{90}Sr was determined by direct measurement using a scintillation beta spectrometer with the “Progress” software (Russia). Polystyrene-based scintillators served as the scintillation material in the spectrometric system. For the preparation of counting samples, a cuvette with manufacturer-certified geometry was used. An aliquot of the soil sample or its micro-aggregate fractions was placed into a pre-weighed aluminum cuvette with a diameter of 70–80 mm. The sample was then leveled, compacted using a manual press, and sealed with polyethylene film to prevent dispersion. The cuvette containing the sample was reweighed using an electronic balance with a resolution of 0.01 mg, and the sample mass was determined by weight difference. Sample masses ranged from 15 to 20 g, depending on their density. Prior to measurement, the spectrometer was energy-calibrated using a $^{90}\text{Sr} + ^{90}\text{Y}$ reference source with an activity of 3.0 kBq. Each sample was

measured for 20 min (Procedures, 2004).

Radiochemical extraction of $^{239+240}\text{Pu}$ included several phases: extraction of radionuclides from the soil matrix by complete ‘wet’ mineralization (decomposition), purification from interfering radionuclides, preparation of counting samples and determination of specific activity of radioisotopes. To account for losses during the radionuclide analysis the isotopic label for ^{242}Pu was introduced into the sample. The amount of isotopic label for ^{242}Pu was 50 % of the expected content of $^{239+240}\text{Pu}$ in the sample, estimated based on an assumed activity ratio of ^{241}Am to $^{239+240}\text{Pu}$ equal to 1:10.

Complete “wet” mineralization (digestion) of the sample was carried out using concentrated hydrofluoric acid (HF) and a mixture of concentrated hydrofluoric and nitric acid (HNO_3). Chromatographic purification and separation of Pu stabilized as Pu(IV) from the dissolved sample was carried out using a strongly basic anionite resin based on a styrene-divinylbenzene copolymer (trade mark AB-17-8, Russia) (Measurement Procedure, 2016).

A counting sample (α -source) was obtained by electrolytic deposition of Pu on a steel substrate. The specific activity of $^{239+240}\text{Pu}$ in the sample was determined using an alpha-spectrometer with a semiconductor detector “Alpha – Analyst” (CANBERRA, USA) with a measured alpha particle energy range of 3.6–6.0 MeV, an energy resolution of not >23 keV and a detection efficiency for alpha emission of up to 25 %.

To control the quality of the analyses, one “control sample” and one “blank sample” were included in each batch of nine samples. The control sample was randomly selected from the samples in the batch. The blank sample was prepared in advance from samples taken from areas with background levels of man-made radionuclides. The control and blank samples were analyzed simultaneously with all other samples. The control sample was used to check the quality and consistency of the analytical results, and the blank sample was used to check for cross-contamination of the samples.

2.7. Particle-size analysis of soil

The principles of particle-size analysis of soils and micro-aggregate fractionation (item 2.5) are similar. However, the main differences are the dispersion methods, i.e. the destruction of soil aggregates, and the selection of fractions. The pyrophosphate method with boiling was used to disperse soil aggregates in particle-size analysis. To a 5 g soil sample, 10 ml of 4 % sodium pyrophosphate solution and 250 ml of distilled water were added, and then the suspension was boiled for 1 h. The suspension was cooled to room temperature and transferred to a 1000 cm^3 glass cylinder through a 0.1 mm sieve. The soil particles retained on the sieve were washed thoroughly with water, transferred to a porcelain cup and dried in a drying cabinet at 105 °C to constant weight. The dried soil particles were sieved through sieves with mesh sizes of 0.5, 0.25 and 0.1 mm.

The suspension passing through the 0.1 mm sieve was further

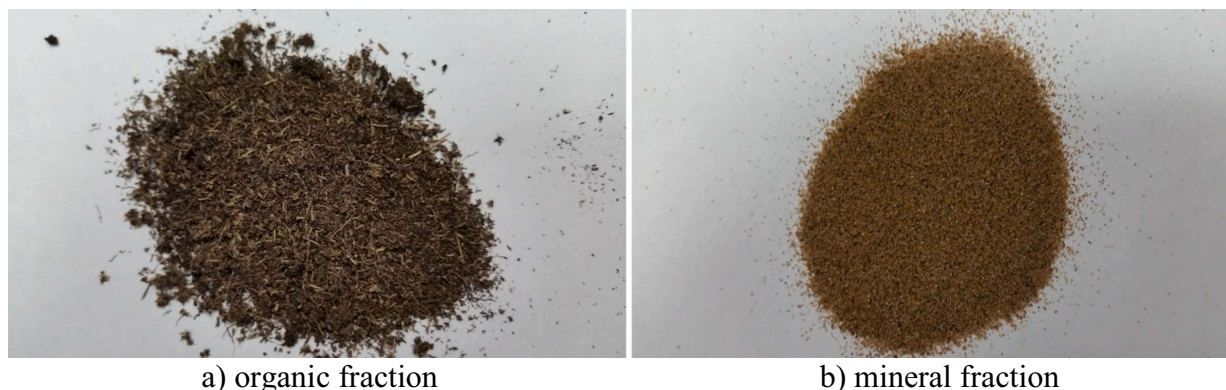


Fig. 2. – External view of “organic” and “mineral” fractions.

analyzed by the pipette method, which is based on the separation of particles by size when they fall into a still liquid (sedimentation). The volume of the suspension in a measuring cylinder was brought to 1000 cm³ with distilled water, the suspension was shaken until the sediment was completely agitated from the bottom and allowed to stand for the required time. At the end of the time, a sample of the suspension was taken with a 25 cm³ pipette from a specified depth.

The time of suspension sampling after the start of sedimentation was based on the estimated specific gravity of the soil particles and the temperature of the suspension. The depth of suspension sampling was: for particles smaller than 0.05 mm - 25 cm; for particles smaller than 0.01 mm - 10 cm; for particles smaller than 0.001 mm - 7 cm.

The suspension sample was transferred from a pipette into a pre-weighed porcelain crucible, evaporated, and then dried in a drying cabinet at 105 °C until a constant weight was reached. It was then cooled to room temperature in a desiccator filled with granulated calcium chloride (KCl).

All the fractions, separated by sieving and selected using a pipette were weighed on analytical balances with a precision of 0.0001 g after drying. The mass percentage of each fraction was calculated by the ratio of the mass of the fraction to the total mass of the sample taken for analysis, with correction for hygroscopic moisture (GOST, 2015).

2.8. Calculation of enrichment factor

The enrichment factor was used as the parameter for distribution of artificial radionuclides (¹³⁷Cs, ²⁴¹Am, ²³⁹⁺²⁴⁰Pu, and ⁹⁰Sr) in micro-aggregate fractions of soil. The enrichment factor was calculated as the ratio of the specific activity of a radionuclide in a given micro-aggregate fraction to its specific activity in the bulk soil, using the formula below: (1):

$$E_f = \frac{A_{sa}^{fr}}{A_{sa}^{sl}} \quad (1)$$

where,

E_f – enrichment factor of fractions;

A_{sa}^{fr} – specific activity of radionuclides in fractions, Bq/kg;

A_{sa}^{sl} – specific activity of radionuclides in the soil, Bq/kg.

The enrichment factor indicates the degree to which particle-size fractions are enriched or depleted in radionuclides relative to their average content in the bulk soil.

3. Results

3.1. Relief of the studied area

The studied waterstream has a slight slope from its source to its terminus. At the source of the waterstream, the elevation is 654.3 m

above sea level and decreases to 639.9 m at the terminus (Fig. 3). The steepness of the slope was about 1.8° (Osipov, 2016), which allows characterizing the area as slightly sloping. The longitudinal profile of the waterstream is smoothly curved, steeper at the headwaters and becoming gentle towards the terminus, with a slight stepping.

3.2. Level and type of radioactive contamination of the soil in the waterstream zone

Soils in the waterstream zone are contaminated with both fission products (¹³⁷Cs, ⁹⁰Sr) and transuranic radionuclides (²⁴¹Am, ²³⁹⁺²⁴⁰Pu). Nevertheless, fission products make a dominant contribution to the radioactive contamination of soils, as evidenced by the ratio of the specific activity of ¹³⁷Cs and ²⁴¹Am radionuclides (Table 1).

The dynamics of changes in the content of fission products and transuranic radionuclides along the waterstream channel vary. Thus, along the water stream, as the distance from its source increases, the specific activity of ¹³⁷Cs and ⁹⁰Sr in the soil at the water's edge increases, while the content of ²⁴¹Am remains almost constant.

To assess the significance of the observed spatial trends in radionuclide content, Spearman's rank correlation coefficient (r_s) was applied, as it is non-parametric and suitable for small datasets (Gravetter and Wallnau, 2015). Table 2 summarizes the r_s values reflecting the relationship between radionuclide activity in channel soils and the distance from the waterstream source, calculated separately for the right and left banks.

For ¹³⁷Cs and ⁹⁰Sr, a positive correlation is observed between specific activity and distance from the waterstream source. In all cases, the calculated Spearman's rank correlation coefficient exceeded their critical values, indicating a statistically significant trend of increasing activity with increasing distance. On the right bank, the correlation can be interpreted as moderate (Hermans et al., 2011), while on the left bank it is considerably higher ($r_s = 0.546$ and 0.867 for ¹³⁷Cs; $r_s = 0.587$ and 0.763 for ⁹⁰Sr, respectively), thereby confirming a reliable dependence of radionuclide distribution on distance. In contrast, for the transuranic radionuclides (²⁴¹Am and ²³⁹⁺²⁴⁰Pu), the correlation coefficients do not exceed the critical values, indicating no statistically significant spatial pattern in their distribution along the waterstream channel. Overall, the statistical test applied indicates a significant distance-dependent trend in the distribution of ¹³⁷Cs and ⁹⁰Sr, while no such relationship was identified for ²⁴¹Am and ²³⁹⁺²⁴⁰Pu.

The spatial distribution of radionuclides along the riparian zone of the waterstream, except for ⁹⁰Sr, is uneven and is accompanied by alternating significant decreases and increases in the specific activity of the radionuclides. The distribution of ⁹⁰Sr in the riparian zone of the waterstream is uniform, which is evidently due to its high migration capacity. Additionally, in the riparian soils, the ratios of the specific activity of radionuclides ¹³⁷Cs/²⁴¹Am and ⁹⁰Sr/¹³⁷Cs vary significantly, which may result from differences in the intensity of the sorption

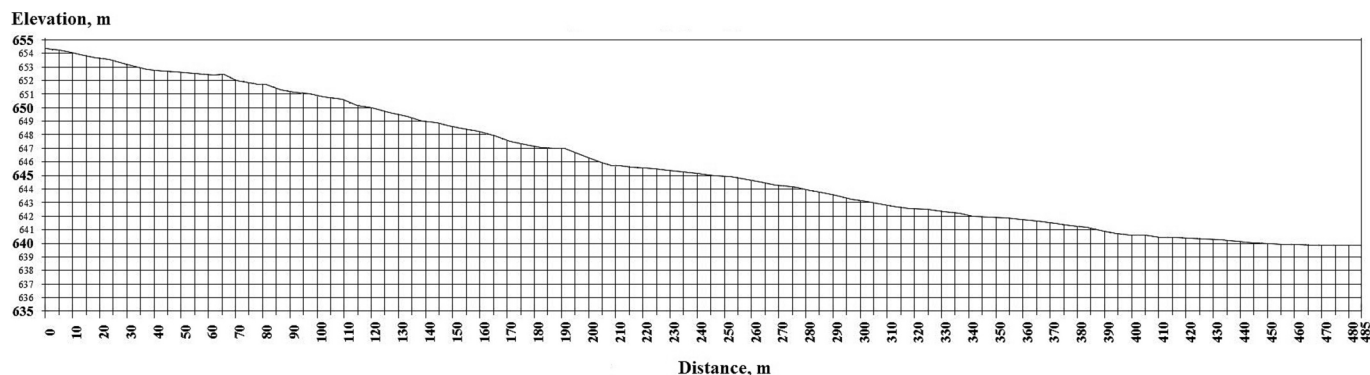


Fig. 3. – Longitudinal elevation section of waterstream, tunnel No. 177.

Table 1
- Specific activity of radionuclides in soils of the water stream zone of tunnel No.177 at Degelen site.

Item No	Sampling point	Distance, m	Specific activity, Bq/kg				Radionuclide specific activity ratio	
			²⁴¹ Am	¹³⁷ Cs	⁹⁰ Sr	²³⁹⁺²⁴⁰ Pu	¹³⁷ Cs/ ²⁴¹ Am	⁹⁰ Sr/ ¹³⁷ Cs
Soil from waterstream channel (at waters' edge)								
	14/1*	38	155 ± 15	1800 ± 180	130,000 ± 180	–	12	72
	14/2		455 ± 45	17,800 ± 1800	120,000 ± 1800	–	39	6,7
	17/1	55	400 ± 40	66,800 ± 6700	85,000 ± 6700	5500 ± 800	167	1,3
	17/2		7 ± 1	12,300 ± 1200	150,000 ± 1200	–	1757	12
	23/1	87	125 ± 10	5500 ± 550	170,000 ± 550	4800 ± 500	44	31
	23/2		125 ± 10	12,300 ± 1200	160,000 ± 1200	–	98	13
	25/1	98	190 ± 20	32,400 ± 3200	62,000 ± 3200	–	171	1,9
	25/2		520 ± 50	18,500 ± 1800	130,000 ± 1800	11,000 ± 1000	36	7,0
	44/1	205	195 ± 20	26,300 ± 2600	80,000 ± 2600	–	135	3,0
	44/2		210 ± 20	16,800 ± 1700	120,000 ± 1700	–	80	7,1
	49/1	226	290 ± 30	85,000 ± 8500	160,000 ± 8500	–	293	1,9
	49/2		120 ± 10	59,000 ± 6000	120,000 ± 6000	–	492	2,0
	56/1	264	270 ± 30	27,500 ± 3000	160,000 ± 3000	–	102	5,8
	56/2		170 ± 20	20,100 ± 2000	180,000 ± 2000	–	118	9,0
	59/1	277	230 ± 25	51,900 ± 5000	180,000 ± 5000	–	226	3,5
	59/2		310 ± 30	66,300 ± 6500	160,000 ± 6500	–	214	2,4
	67/1	314	260 ± 25	62,400 ± 6000	180,000 ± 6000	–	240	2,9
	67/2		190 ± 20	25,100 ± 2500	160,000 ± 2500	–	132	6,4
	69/1	326	210 ± 20	60,800 ± 6000	150,000 ± 6000	–	290	2,5
	69/2		190 ± 20	58,400 ± 6000	130,000 ± 6000	6600 ± 700	307	2,2
	80/1	391	260 ± 25	58,400 ± 6000	180,000 ± 6000	–	225	3,1
	80/2		260 ± 25	67,600 ± 7000	210,000 ± 7000	–	260	3,1
	82/1	401	150 ± 15	53,400 ± 5500	91,000 ± 5500	–	356	1,7
	82/2		280 ± 30	80,100 ± 8000	180,000 ± 8000	8600 ± 900	286	2,2
	89/1	440	140 ± 15	37,500 ± 4000	160,000 ± 4000	–	268	4,3
	89/2		220 ± 20	57,600 ± 6000	180,000 ± 6000	–	262	3,1
	92/1	457	290 ± 30	83,000 ± 8500	180,000 ± 8500	–	286	2,2
	92/2		330 ± 35	88,600 ± 9000	190,000 ± 9000	–	268	2,1
	98/1	485	330 ± 35	87,400 ± 9000	210,000 ± 9000	–	265	2,4
	98/2		380 ± 40	92,700 ± 9500	210,000 ± 9500	–	244	2,3
Soils in the riparian zone of the waterstream								
	1	91	840 ± 170	14,000 ± 2500	105,000 ± 20,000	14,000 ± 2800	17	8
	2	157	<10	75 ± 15	110,000 ± 22,000	220 ± 40	>7,5	1500
	3	219	420 ± 80	34,000 ± 6500	160,000 ± 32,000	47,000 ± 9500	81	5
	4	281	<20	7000 ± 1400	180,000 ± 35,000	3400 ± 700	>350	25
	5	349	360 ± 70	36,000 ± 7000	190,000 ± 38,000	21,000 ± 4000	100	5
	6	391	310 ± 60	18,000 ± 3500	190,000 ± 38,000	14,000 ± 2800	56	10
	7	473	470 ± 90	3200 ± 650	155,000 ± 30,000	5300 ± 1000	7	50
	8	528	30 ± 6	20 ± 4	5100 ± 1000	270 ± 50	0,7	255

* Note: after the slash, 1 indicates the right bank, 2 indicates the left bank.

Table 2
– Spearman's rank correlation coefficient (r_s) values.

Parameter	¹³⁷ Cs		⁹⁰ Sr		²⁴¹ Am		²³⁹⁺²⁴⁰ Pu
	Right bank	Left bank	Right bank	Left bank	Right bank	Left bank	
Calculated r _s value	0.546	0.867	0.587	0.763	0.18	0.281	0.6
Critical r _s value (α = 0.05)	0.521 (n = 15)						1.0 (n = 5)

processes that these radionuclides undergo.

3.3. Particle-size and micro-aggregate composition of soils

In this study, soil particle-size composition refers to the relative content of primary (non-aggregated) particles of various sizes, determined using a chemical dispersion method (aggregate destruction). Soil micro-aggregate composition refers to the relative content in the soil of both primary and secondary particles, bound together in aggregates, of various sizes, determined using water as the sole dispersing agent.

The particle-size composition of the channel soils is heterogeneous and is characterized by the following average ratio (% by mass) of elementary soil particle fractions: sand and gravel fraction (>1 mm) - 14 %, sandy fraction (1–0.05 mm) - 18 %, silt fraction (0.05 mm–0.001 mm) - 63 %, and clay fraction (<0.001 mm) - 5 %. The soils are

predominantly silty, especially with coarse silt particles (0.05–0.01 mm). The content of clay fractions is low.

Along the waterstream channel, the particle-size composition of the soils undergoes significant changes, which are most clearly seen on the left bank of the waterstream (Fig. 4). Compared to the source, the proportion of coarse silt particles increases towards the terminus of the waterstream, while the proportion of gravel and sand fractions decreases, in some places reaching barely detectable amounts.

From the perspective of micro-aggregate composition, the channel and riparian soils exhibit similar characteristics. Both are predominantly composed of fractions in the 250–1000 μm and 8–40 μm size range. In addition, the soils contain a substantial amount of organic matter, represented by plant debris, turf, etc. (Figs. 5 and 6).

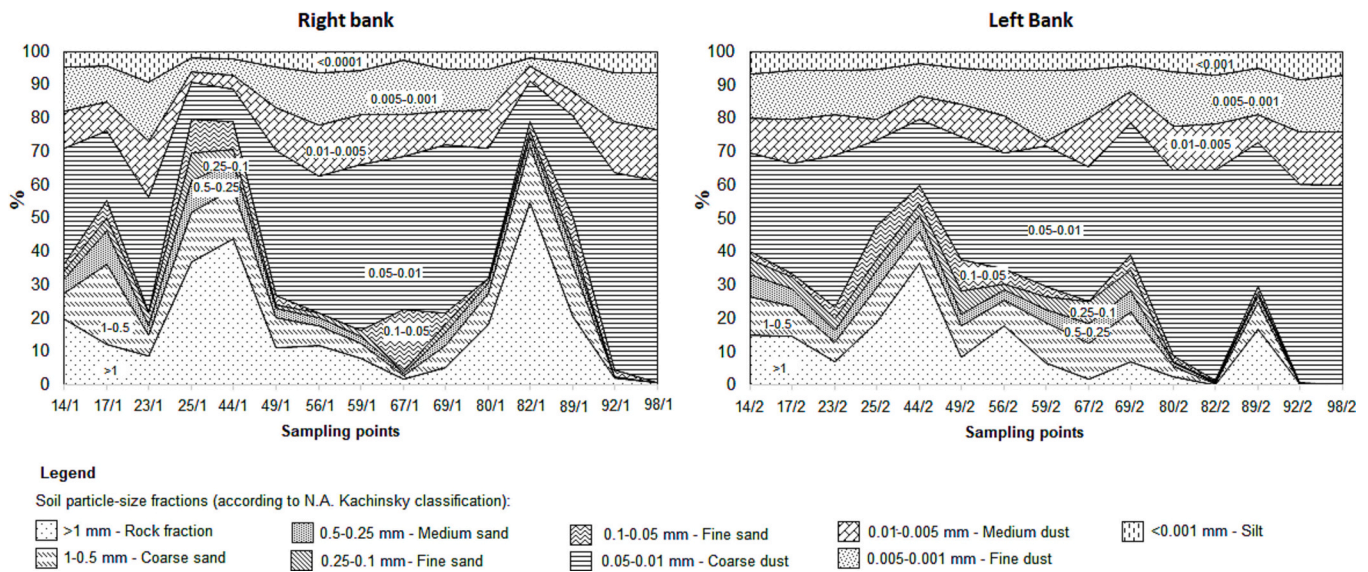


Fig. 4. – Particle size (mm) composition of the waterstream soils.

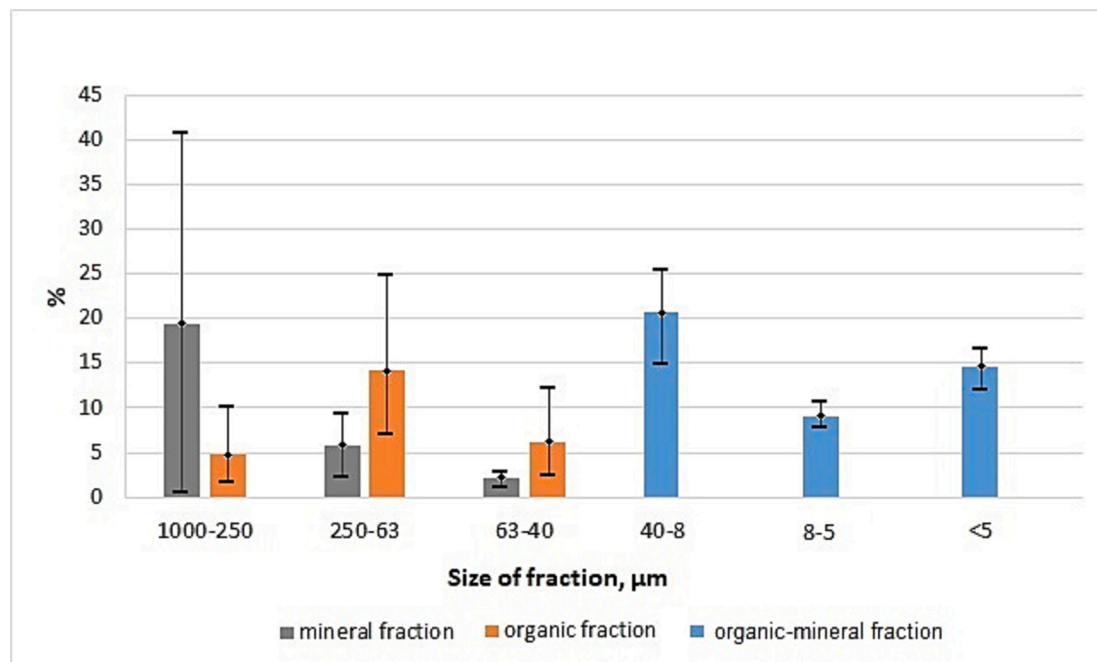


Fig. 5. – Mass-percentage of micro-aggregate fractions in soils from the waterstream channel (Bars represent the average mass percentage; error bars show the range (minimum-maximum) observed across samples).

3.4. Distribution of artificial radionuclides in micro-aggregate fractions of soil

Data on the content of radionuclides ^{137}Cs , ^{90}Sr , ^{241}Am , and $^{239+240}\text{Pu}$ in the micro-aggregate fractions of channel and riparian soils are presented in Tables S1 and S2 (Supplementary materials).

The distribution of radionuclides in the micro-aggregate fractions of channel soils is uneven. The highest specific activity of ^{137}Cs was found in the fraction with a size of $<5\ \mu\text{m}$, while the lowest was in the “mineral” part of the fraction sized 1000–250 μm . These values differ by a factor of 10–30. It should also be noted that in all fractions, divided into “mineral” and “organic” parts, the “organic” part contains 1.5 to 16 times more ^{137}Cs than the “mineral” part.

The minimum content of other radionuclides was also observed in

the “mineral” part of the 1000–250 μm fraction. However, the highest specific activity values were found in the “organic” parts of various fractions. In all fractions ranging in size from 40 to 1000 μm , the “organic” part contains significantly higher concentrations of ^{90}Sr , ^{241}Am , and $^{239+240}\text{Pu}$ radionuclides than the “mineral” part. For example, the difference in the specific activity of ^{90}Sr in the “organic” and “mineral” parts is 7–44 times, for ^{241}Am it is 1.6–116 times, and for $^{239+240}\text{Pu}$ it is 6–144 times.

In riparian soils, the distribution of radionuclides in micro-aggregate fractions is also uneven. The lowest content of all radionuclides is found in the 1000–500 μm fraction. However, the highest values of their specific activity have been recorded in various fractions. For instance, ^{137}Cs is predominantly concentrated in the fraction sized $<1\ \mu\text{m}$, with the exception of a single case (point 7), where the maximum value of its

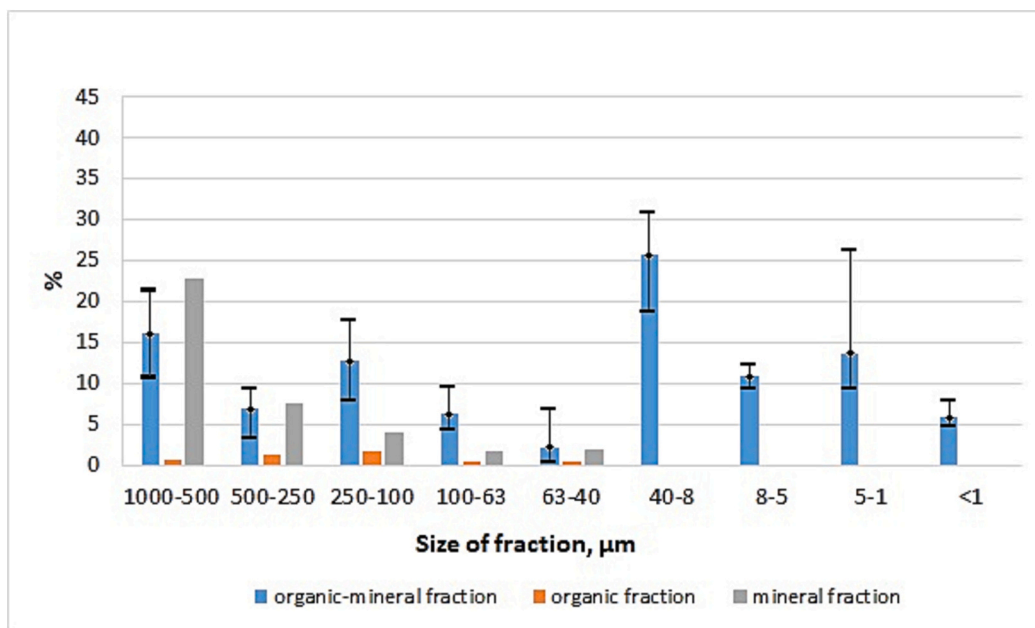


Fig. 6. – Mass-percentage of micro-aggregate fractions in soils from the riparian zone of waterstream (Bars represent the average mass percentage; error bars show the range (minimum-maximum) observed across samples).

specific activity is recorded in the 40–8 μm fraction. The highest specific activity values for ^{90}Sr are also primarily found in the fraction sized <1 μm. Exceptions include a few points (points 6 and 7), where the 63–40 μm fraction dominates in terms of its specific activity.

In the middle and lower course of the waterstream, the 63–40 μm fraction is also characterized by a high content of ^{241}Am . However, in the upper course of the waterstream, the highest values of its specific activity are shifted towards the 100–63 μm and < 1 μm fractions. High values of the specific activity of $^{239+240}\text{Pu}$ are predominantly found in the 100–63 μm fraction; however, there are rare cases where its highest content occurs in the 63–40 μm, 250–100 μm, and 40–8 μm fractions.

The difference between the minimum and maximum values of specific activity of radionuclides in the fractions is: for ^{137}Cs - 3.52 times, ^{90}Sr - 1.6-25 times, ^{241}Am - 2.55 times, $^{239+240}\text{Pu}$ - 2.3-875 times.

It should also be noted that high levels of radionuclides were found in some fractions of both channel and riparian soils.

4. Discussion

4.1. Mechanisms of the formation of radionuclides distribution in microaggregate fractions of soil

Data on the enrichment factors (E_f) for the micro-aggregate fractions of channel and riparian soils are presented in Tables 3 and 4, respectively.

The results show that in soils collected from the waterstream channel, E_f values vary depending on both the fraction size and its composition. Thus, there are significant differences in E_f values between

Table 3

– E_f values of micro-aggregate fractions of soil sampled from the waterstream channel (n = 5).

Value	Fraction size, μm						40–8	8–5	<5
	1000–250		250–63		63–40				
	Mineral	Org.	Mineral	Org.	Mineral	Org.			
¹³⁷ Cs									
Min.	0,0003	0,48	0,24	0,91	0,39	0,68	1,01	1,23	1,64
Max.	0,02	1,22	0,55	1,52	0,68	1,52	1,41	1,90	2,54
Average	0,007	0,88	0,44	1,18	0,51	1,09	1,26	1,53	1,95
²⁴¹ Am									
Min.	0,01	1,15	0,12	1,30	0,04	1,00	0,91	0,72	1,06
Max.	0,72	2,61	0,31	3,21	0,26	2,81	1,50	1,47	1,92
Average	0,21	1,55	0,19	1,89	0,16	1,71	1,26	1,17	1,44
⁹⁰ Sr									
Min.	0,04	1,66	0,07	1,29	0,09	1,00	0,77	0,94	1,19
Max.	0,17	2,92	0,42	2,81	0,27	2,25	1,24	1,57	1,91
Average	0,11	2,10	0,22	1,92	0,17	1,67	1,06	1,21	1,47
²³⁹⁺²⁴⁰ Pu									
Min.	0,0001	0,61	0,07	0,67	0,02	0,77	0,38	0,42	1,12
Max.	0,01	4,78	0,24	3,93	0,19	3,25	1,25	1,45	4,23
Average	0,003	2,19	0,14	1,95	0,10	1,72	0,94	1,11	1,94

Table 4– E_f values of micro-aggregate fractions of soil from the riparian zone ($n = 8$).

Values	Fraction, μm								
	1000–500	500–250	250–100	100–63	63–40	40–8	8–5	5–1	<1
^{137}Cs									
Min.	0,06	0,12	0,67	0,79	0,19	1,08	1	1,26	0,65
Max.	0,62	0,91	0,88	1,13	2,01	1,3	1,38	1,74	2,87
Mean (excluding outliers)	0,36	0,48	0,77	0,96	0,97	1,13	1,24	1,52	1,85
Outliers moderate	–	–	0,22; 1,2	0,24	–	–	0,4	0,56	–
extreme	–	–	–	–	–	0,56; 2,93	–	–	–
^{90}Sr									
Min.	0,2	0,37	0,9	1,01	1,01	0,89	0,84	0,81	1,19
Max.	0,79	1,04	1,36	1,36	1,21	1,13	1,36	1,85	2
Mean (excluding outliers)	0,44	0,74	1,15	1,17	1,13	1,04	1,11	1,35	1,52
Outliers moderate	–	–	–	–	1,48	–	–	–	–
extreme	–	–	–	–	19,5	–	–	–	–
^{241}Am									
Min.	0,06	0,11	0,2	0,16	0,49	0,6	0,53	0,51	0,5
Max.	0,8	1,05	1,43	1,91	1,66	2,98	1	1,37	1,64
Mean (excluding outliers)	0,40	0,57	0,92	1,01	1,06	1,61	0,83	1,00	1,02
Outliers (extreme)	–	–	–	–	5,38	–	–	–	–
$^{239+240}\text{Pu}$									
Min.	0,06	0,36	0,52	1,12	1,11	0,59	0,19	0,92	0,7
Max.	1,21	1,12	1,49	1,67	1,39	1,54	1,49	1,2	1,37
Mean (excluding outliers)	0,49	0,75	1,09	1,45	1,26	1,00	0,85	1,03	1,12
Outliers moderate	–	–	–	–	0,63	–	–	0,52	–
extreme	–	–	–	0,52	3,3; 51,5	–	–	2,14	–

“mineral” and “organic” fractions (Wilcoxon signed-rank test, $T = 0$, $p = 0.031$, one-tailed). The E_f values for mineral fractions are relatively low indicating depletion in radionuclides. The lowest E_f values for all studied radionuclides were observed in the mineral fraction sized 1000–250 μm . This fraction is particularly depleted in ^{137}Cs and $^{239+240}\text{Pu}$, with average E_f values of 0.007 and 0.003, respectively. However, the content of these radionuclides increases significantly in finer mineral fractions. For example, the E_f values of ^{137}Cs increase significantly in the 250–63 μm and 63–40 μm mineral fractions, with average values reaching 0.44 and 0.51, respectively. The minimum (0.0003 in the 1000–250 μm fraction) and maximum (0.68 in the 63–40 μm fraction) E_f values for ^{137}Cs determined in “mineral” fractions of different sizes differ by approximately 2200 times.

A similar trend was observed for $^{239+240}\text{Pu}$. The ratio between the minimum (0.0001 in the 1000–250 μm fraction) and maximum (0.24 in the 250–63 μm fraction) E_f values of $^{239+240}\text{Pu}$ registered in the mineral soil fractions ranging from 40 to 1000 μm is approximately 2400. However, the overall range of E_f values for $^{239+240}\text{Pu}$ is smaller than that for ^{137}Cs . For example, the average E_f values of $^{239+240}\text{Pu}$ in mineral soil fractions of varying sizes range from 0.003 to 0.14, whereas those for ^{137}Cs range from 0.007 to 0.51. Based on the cumulative accumulation of radionuclides in “mineral” fractions ranging from 1000 to 40 μm , it can be assumed that ^{137}Cs exhibits a higher affinity for coarser soil mineral particles, whereas the ability of $^{239+240}\text{Pu}$ to bind with soil mineral components within this size range appears to be relatively low.

In the studied soils, the differentiation of E_f values for ^{241}Am and ^{90}Sr in “mineral” fractions of different sizes is poorly expressed. The average E_f values in the “mineral” fractions sized 1000–250 μm , 250–63 μm , and 63–40 μm vary insignificantly (for ^{241}Am – from 0.16 to 0.21; for ^{90}Sr – from 0.11 to 0.22), which indicates a relatively uniform distribution of these radionuclides in the various fractions of mineral particles in channel soils.

In contrast to “mineral” fractions, a high accumulation of radionuclides occurs in “organic” fractions. The greatest concentration of $^{239+240}\text{Pu}$ is found in the “organic” fractions. The average E_f values of this radionuclide range from 1.72 to 2.19, with the maximum value

reaching 4.78. Compared to $^{239+240}\text{Pu}$, the accumulation of ^{90}Sr and ^{241}Am in the “organic” fractions is somewhat less. The average E_f values for ^{90}Sr and ^{241}Am range from 1.67 to 2.10 and from 1.55 to 1.89, respectively, and the maximum values do not exceed 2.92 and 3.21, respectively. Of all studied radionuclides, ^{137}Cs shows the lowest level of accumulation in “organic” fractions. Average E_f values for ^{137}Cs in “organic” fractions range from 0.88 to 1.18, and the highest value recorded in these fractions reaches only 1.52. In general, the distribution pattern of all studied radionuclides in the “organic” fractions appears fairly uniform, showing no clear dependence on fraction size. The concentration effect of radionuclides in undecomposed organic matter was also reported by Kropacheva et al., 2013, with enrichment of the “organic” fraction (isolated from the >250 μm fraction) relative to the bulk soil observed at approximately 1.2 times for ^{137}Cs and 7.5 times for ^{90}Sr . The detected disparity in accumulation between ^{137}Cs and ^{90}Sr , consistent with our results, may indicate differing interactions of these radionuclides with organic matter.

Among fine (<40 μm) organic-mineral fractions of channel soils, the predominant accumulation of all radionuclides is observed in the <5 μm fraction. A weakly expressed trend of increasing enrichment with decreasing fraction size is present. Meanwhile, within the <40 μm fractions, E_f values for the radionuclides exhibit a notable degree of similarity. Thus, the range of mean E_f values is as follows: 1.26–1.95 for ^{137}Cs , 1.17–1.44 for ^{241}Am , 1.26–1.47 for ^{90}Sr , and 0.94–1.94 for $^{239+240}\text{Pu}$. These results may suggest that fine (dusty-silty) organic-mineral soil particles exhibit little selectivity in the accumulation of different radionuclides. The preferential accumulation of radionuclides in fine soil fractions is well established in the scientific literature. For example, Livens and Baxter, 1988 reported an enrichment of radionuclides in fine particle size fractions (<2 μm) of intertidal sediment, with $^{239+240}\text{Pu}$ enriched by approximately 5 times, ^{241}Am by approximately 4 times, and ^{137}Cs by nearly 5 times. This enrichment is primarily attributed to the elevated sorption capacity of clay and silt particles, resulting from their high specific surface area and the presence of secondary clay minerals in their mineralogical composition.

Statistical analysis of the data set obtained for channel soils, based on

a robust method, did not reveal any markedly deviating E_f values that could be classified as outliers. However, the analysis of data obtained for riparian soils revealed E_f values identified as outliers using the interquartile range method. Values falling outside the lower and upper threshold values corresponding to 1.5 and 3 times the interquartile range were classified as moderate and extreme outliers, respectively.

Outliers in the dataset may arise due to measurement errors, random variation, or as an expression of genuine features of contamination in the studied soil. To objectively assess the nature and potential origins of the anomalous values, a statistical analysis was performed using the Generalized Extreme Studentized Deviate (GESD) test (ISO 16269-4:2010, 2010). Since the initial dataset exhibited pronounced asymmetry, as indicated by skewness coefficients ranging from 2.0 to 2.8 and kurtosis values from 4.3 to 8.0, and given that the GESD test assumes normally distributed data, a Box–Cox transformation was applied. This transformation helped approximate a normal distribution, thereby improving the reliability of the test results. The GESD test showed that, after transformation, the values initially identified as outliers did not significantly differ from the rest of the dataset at the 95 % confidence level. On the one hand, this suggests that these anomalous values are unlikely to result from analytical error or random fluctuations. On the other hand, their notable deviation from the majority of values may indicate substantial heterogeneity in soil contamination.

Thus, Goriachev et al., 2011; Lukashenko et al., 2020 demonstrated, that the presence of “hot” particles is a distinctive feature of soil contamination in the waterstream zone near the tunnels. Two types of radioactive particles have been identified in soil samples: particles representing remnants of wood and vegetation, and fragile, amorphous organo-mineral conglomerates. The activity of these particles can exceed the radioactivity of the surrounding soils by several orders of magnitude. The formation of such particles is associated with the sorption and coprecipitation of transuranic elements with iron and manganese oxides and hydroxides. The presence of such “hot” particles may explain the high radionuclides enrichment observed in certain soil fractions, for example, in the 63–40 μm fraction, where elevated E_f values were recorded: ^{90}Sr – 19.5; ^{241}Am – 5.38; $^{239+240}\text{Pu}$ – 51.5). Although the studies by Goriachev et al., 2011; Lukashenko et al., 2020 provided a comprehensive analysis of “hot” particles in soils from tunnel water stream zones using autoradiography, and their findings are widely acknowledged, the identification of such particles in the studied fractions remains a promising topic for further research, contributing to a better understanding of heterogeneity and localized anomalies in soil contamination.

The data presented in Table 5 provide valuable insight into the distribution of radionuclides between the “organic” and “mineral” fractions of riparian soils. Notably, although the “organic” fractions constitute only a small portion of the total soil mass (Fig. 6), their radionuclide content is substantially higher compared to the corresponding “mineral” fractions. The specific activity of radionuclides in the “organic” fractions exceeds that in the “mineral” fractions by up to 30 times for ^{241}Am and 14 times for ^{137}Cs .

Fig. 7 illustrates the distribution of ^{137}Cs and ^{241}Am in the “organic”

and “mineral” fractions of different size classes. In the “organic” fractions, slight variation in E_f values was observed, particularly for ^{137}Cs , within the range from 0.42 to 1.0 (a 2.3-fold difference), which may indicate a relatively uniform distribution of radionuclides regardless of fraction size. In contrast, a trend of increasing ^{137}Cs content with decreasing particle size was exhibited in the “mineral” fractions: the E_f values rose from 0.05 in the 1000–500 μm fraction to 0.28 in the 63–40 μm fraction (a 5.6-fold increase). A comparable pattern of ^{241}Am distribution was noted in both the “organic” and “mineral” fractions: within the 1000 to 63 μm range, E_f values in the “organic” fractions increased moderately from 0.25 to 0.50 (a 2-fold rise), while in the “mineral” fractions the E_f values increased more sharply from 0.01 to 0.13 (a 13-fold increase) as fraction size decreases. Notably, in the 63–40 μm size class the E_f values for ^{241}Am were markedly higher than in other fractions (2.73 in the “organic” fraction and 7.79 in the “mineral” fraction) deviating strongly from the general trend. One possible explanation for this sharp deviation may be the presence of “hot” particles identified in soils contaminated by tunnel water discharges in previous studies (Goriachev et al., 2011; Lukashenko et al., 2020). Considering this, the 63–40 μm data point for ^{241}Am was excluded from the regression analysis to avoid distortion of the underlying distribution trend.

The observed patterns of ^{137}Cs and ^{241}Am distribution in the “organic” and “mineral” components of riparian soil fractions are supported by the coefficients of determination (R^2) from the regression analysis. In the “mineral” fractions, high R^2 values (0.97 for ^{137}Cs and 0.82 for ^{241}Am) suggest a strong correlation between decreasing particle size and increasing E_f values. In contrast, the low R^2 values observed for the “organic” fractions (0.02 for ^{137}Cs and 0.03 for ^{241}Am) indicate no clear dependence of E_f values on fraction size, which is consistent with the assumption of a more uniform radionuclide distribution across size classes. The relatively constant radionuclide content in the “organic” fraction likely leads to levelling of radionuclide distribution among the micro-aggregate fractions, which is reflected in the average E_f values. The levelling effect of the “organic” fraction was more pronounced for ^{137}Cs .

The graphs in Fig. 8 show the relationship between the size of soil particle fractions and their radionuclide content. When constructing the graphs for the 1000–250 μm , 250–63 μm and 63–40 μm fractions of channel soils, E_f values were used that were calculated based on the estimated mean specific activity of radionuclides in the organic-mineral fraction. This approach ensured internal consistency of the results and enabled a commensurate interpretation of the data on radionuclide content in fractions separated into “mineral” and “organic” components, as well as in unseparated fractions containing both. The integrated specific activity of radionuclides in the organic-mineral fraction, obtained as a mass-weighted value, was calculated according to eqs. (2) and (3):

$$SA_{org-miner}^{fr} = \frac{A_{miner}^{fr} + A_{org}^{fr}}{m_{miner}^{fr} + m_{org}^{fr}} \quad (2)$$

$$A_{miner/org}^{fr} = SA_{miner/org}^{fr} \times m_{miner/org}^{fr} \quad (3)$$

where,

$SA_{org-miner}^{fr}$ – integrated (mass-weighted) specific activity of radionuclides in the organic-mineral fraction, Bq/kg;

A_{miner}^{fr} and A_{org}^{fr} – absolute activities of the radionuclide in the mineral and organic fractions, respectively, Bq;

m_{miner}^{fr} and m_{org}^{fr} – masses of mineral and organic fractions, respectively, kg;

$SA_{miner/org}^{fr}$ – specific activity of the radionuclide in the mineral or organic fractions, determined experimentally, Bq/kg.

As shown by the graphs (Fig. 8), the distribution of radionuclides

Table 5

– Specific activity of radionuclides in “organic” and “mineral” soil fractions from riparian zone of the waterstream (n = 1).

Fraction, μm	Specific activity, kBq/kg			
	^{241}Am		^{137}Cs	
	“mineral” fraction	“organic” fraction	“mineral” fraction	“organic” fraction
1000–500	0,12 ± 0,002	0,37 ± 0,07	0,2 ± 0,04	1,8 ± 0,4
500–250	<0,008	0,24 ± 0,05	0,31 ± 0,06	4,3 ± 0,9
250–100	0,041 ± 0,008	0,21 ± 0,04	0,4 ± 0,08	2,7 ± 0,5
100–63	0,10 ± 0,02	0,42 ± 0,08	0,78 ± 0,16	3,1 ± 0,6
63–40	7,4 ± 1,5	2,3 ± 0,5	1,2 ± 0,6	2,8 ± 0,6

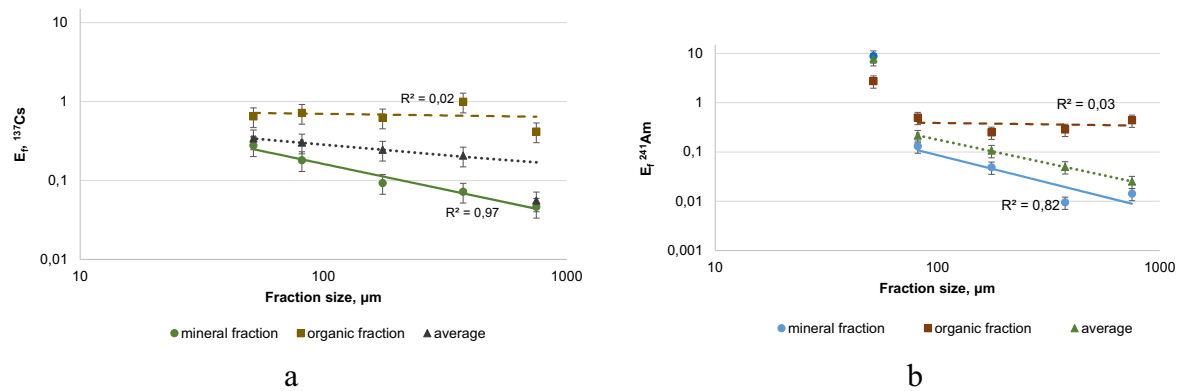


Fig. 7. – Distribution of ^{137}Cs (a) and ^{241}Am (b) in “organic” and “mineral” fractions of soils from the riparian zone.

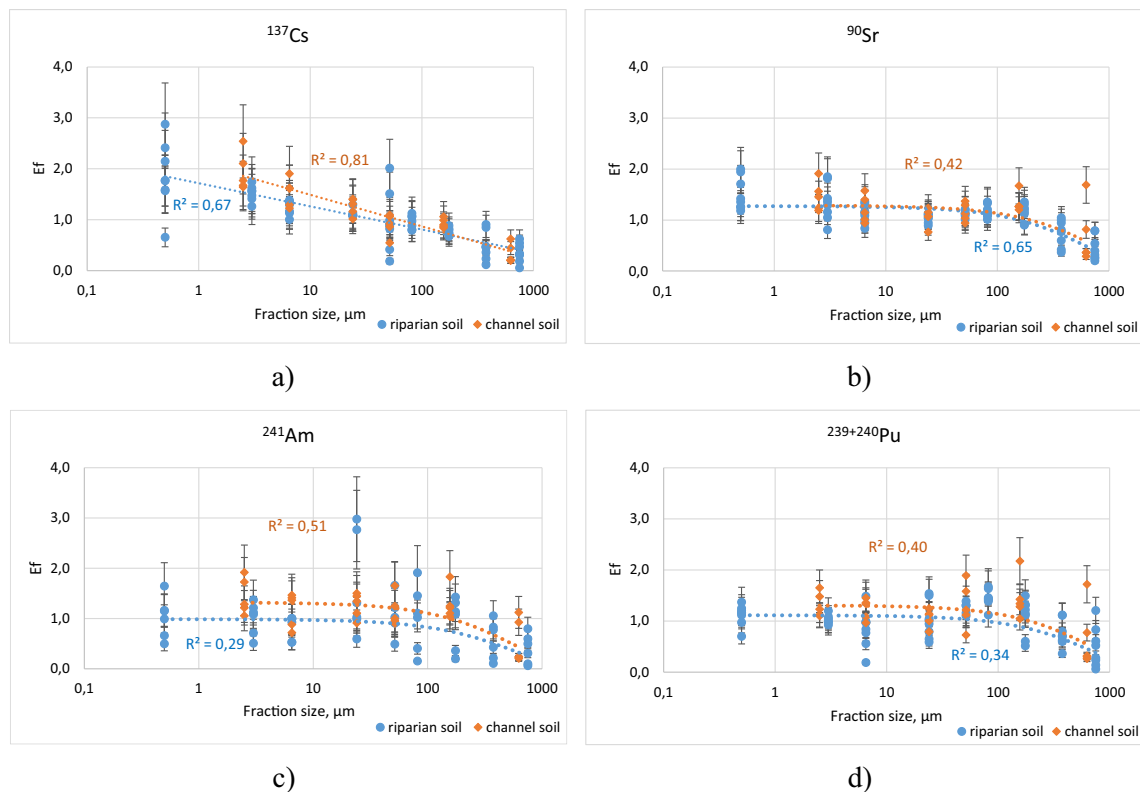


Fig. 8. – Distribution of ^{137}Cs , ^{90}Sr , ^{241}Am and $^{239+240}\text{Pu}$ in soil micro-aggregate fractions (organic-mineral) in the waterstream zone near the tunnel No.177 at “Degelen” site.

among micro-aggregate soil fractions exhibits a decreasing trend in accumulation with increasing particle size, which is most pronounced in the case of ^{137}Cs . This pattern is traditionally attributed to the sorption of radionuclides from the aqueous phase. It is well known that one of the key factors influencing the sorption processes at the solid-liquid interface in heterogeneous media is the specific surface area. Smaller particles have a larger specific surface area, resulting in more efficient sorption. However, some studies (Alberts and Muller, 1979) have shown that this relationship may not always be observed. In certain instances, no significant increase in radionuclide concentration with decreasing particle size has been reported, indicating the potential influence of additional mechanisms such as ion-exchange reactions, interactions with organic matter, particle aggregation, and other factors. This may account for the less pronounced particle-size dependence observed in the distribution of ^{90}Sr , ^{241}Am and $^{239+240}\text{Pu}$. The results may suggest that the distribution pattern of ^{90}Sr , ^{241}Am and $^{239+240}\text{Pu}$ in soil micro-

aggregate fractions is shaped not only by sorption-related mechanisms that promote accumulation in finer particles, but also by interactions with organic substances, which exert a levelling effect on radionuclide distribution. Additionally, the aggregation of silt-sized particles may further enhance this levelling effect.

The observed differences in the distribution of ^{137}Cs and other studied radionuclides among soil micro-aggregate fractions can be attributed to their different geochemical behavior. In freshwater ecosystems, Cs can exist in a truly dissolved state as a monovalent cation (Cs^+), which has the largest ionic radius among simple cations. Due to this larger ionic radius, Cs is effectively retained by the adsorption surface of clay minerals, which are abundant in fine soil particles (Titayeva, 2000).

Strontium (Sr) may also occur in water in its ionic form (Sr^{2+}). However, strontium radionuclides are capable of forming radiocolloids (Titayeva, 2000).

The behavior of plutonium (Pu) is more complex: it can occur in several oxidation states simultaneously in solution and is prone to complexation, including with humus substances. In natural suspensions, plutonium is predominantly adsorbed in its reduced forms (Pu(IV) and Pu(III)). Colloidal organic matter can act as a reducing agent (Titayeva, 2000).

Like plutonium, americium (Am) can form complexes with colloidal organic matter. It has been shown (Fröhlich and Kaplan, 2018) that under neutral and alkaline conditions, the presence of organic substances – particularly humic acids – reduces the sorption of americium by clay mineral particles. This behavior is explained by the possible formation of mixed complexes of Am(III)-humate-carboxylates under environmental conditions.

Earlier studies have shown (Toropov, 2020) that the water in the waterstream from the tunnel 177 is fresh, slightly alkaline and has a hydrocarbonate-sulphate-calcium-sodium composition. Colloidal particles identified as organic colloids of humic and fulvic acids (1–2 nm), as well as inorganic or organic-mineral colloidal particles (100–450 nm) were found in the water. Data on the speciation of radionuclides in the water indicate that the majority of ^{137}Cs (~70 %) and ^{90}Sr (~95 %) are found in the <3 nm fraction, which corresponds to a combination of dissolved species and low-molecular-weight organic colloids. Significant amounts of $^{239+240}\text{Pu}$ were found in all studied forms: approximately 30 % in suspensions (10,000–450 nm), about 20 % in colloids (450–3 nm), and around 50 % in the <3 nm fraction. Under such conditions in water, radionuclides can be effectively sorbed onto and bound with amorphous soil organic matter, such as humus and partially decomposed plant residues, which explains their high degree of enrichment.

Moreover, for radionuclides with high bioavailability in the studied waterstream soils, particularly ^{90}Sr (Larionova et al., 2018; Lukashenko et al., 2020), in addition to incorporation into the organic matter, the biological cycling of plants may also contribute to their retention within soil organic fractions. As plants that have accumulated radionuclides during their life cycle die and shed their biomass, and as this biomass subsequently decomposes, humus enriched by radionuclides may form.

The prolonged duration of this process (several decades) and high productivity of the vegetation likely promote the accumulation of large amounts of organic residues in the soil. In contrast, in case of radionuclides with low mobility and limited root uptake, such as ^{137}Cs , ^{241}Am and $^{239+240}\text{Pu}$, physical and chemical interactions with organic matter (e.g., sorption, complexation, etc.) may play a major role. Thus, the accumulation of radionuclides in the organic phase can result from both sorption by organic residues and the biological cycling of plants, with their relative contributions determined by the specific biogeochemical properties of individual isotopes.

Thus, several key mechanisms can be identified that determine the distribution of radionuclides among micro-aggregate soil fractions:

- 1) sorption processes occurring in the “soil-water” system, which lead to a characteristic increase in radionuclide accumulation with a decreasing soil particle size;
- 2) high accumulation of radionuclides in organic phase (partially decomposed plant residues), which may have a levelling effect on the typical patterns associated with sorption processes;
- 3) formation of highly active individual particles (“hot” particles), which may cause a sharp increase in radionuclide enrichment within certain fractions.

4.2. Mechanisms of formation of spatial distribution of radionuclides in the waterstream zone

Along the waterstream channel, the ^{137}Cs content increases with increasing distance from the waterstream source. A similar but less pronounced trend in specific activity is observed for ^{90}Sr . However, no clear dependence of the lateral distribution of ^{241}Am and $^{239+240}\text{Pu}$ on distance has been established (Fig. 9).

In addition to sorption processes, the transport of solid particles by the water flow (solid runoff) may influence the spatial distribution of radionuclides along the waterstream. To assess the potential impact of solid runoff on this spatial distribution, a detailed analysis of changes in

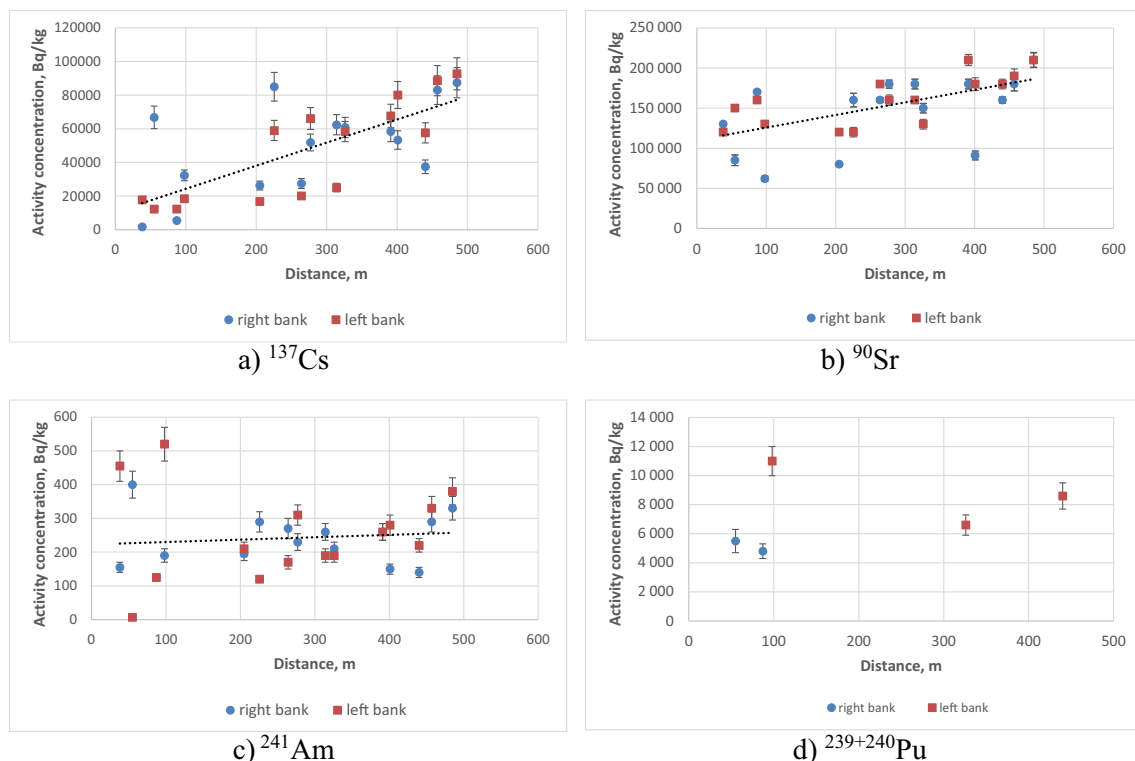


Fig. 9. – Spatial distribution of radionuclides along the waterstream.

soil fractions with distance from the waterstream source was conducted.

Data analysis showed that in the upper and middle reaches of the channel, the soils have a lighter structure, with sandy loams and light loams being more common. As the distance from the source increases, the content of rocky and sandy fractions ranging from >1 mm to 0.05 mm significantly decreases, while the content of fine fractions smaller than 0.05 mm slightly increases (Fig. 10, Table 6). At the lower reaches of the waterstream, the soils are characterized by a heavier structure and are classified as medium and heavy loams based on their particle-size composition.

It should be noted that structural changes in the soils are most pronounced along the left bank of the waterstream.

One of the factors determining the formation of river runoff is geomorphological conditions (relief). In the upper and middle courses of a waterstream, due to the greater elevation drop and steeper slopes, the transport of solid runoff (particles) can occur more intensively. In the lower course of the waterstream, where the terrain becomes gentler, the accumulation of transported material may take place. Thus, the results of the study suggest that the transport of solid runoff may be one of the key processes responsible for the formation of the spatial distribution of fission products – particularly ^{137}Cs – in the waterstream zone. The depletion of the riverbank soils in the lower course of the waterstream with sandy fractions and their enrichment in clay particles contributes to radionuclide accumulation in this zone. It should be noted that the key influence of sediment transport, particle size and hydrodynamic factors on the migration behavior of ^{137}Cs in riverine and estuarine systems has also been highlighted in studies of the Kuma River near the Fukushima Daiichi Nuclear Power Plant (Yamasaki et al., 2016).

However, the transport of solid particles likely does not play a

significant role in the spatial distribution of transuranic radionuclides along the waterstream, as their predominant accumulation occurs in the “organic” component of the soil (plant residues, turf, humus, etc.).

5. Conclusion

The distribution of ^{137}Cs , ^{90}Sr , ^{241}Am , $^{239+240}\text{Pu}$ among the micro-aggregate soil fractions was studied in soils contaminated as a result of waterborne migration of radionuclides via a waterstream flowing through the cavity of the tunnel where an underground nuclear test was conducted at the “Degelen” site of the Semipalatinsk test site. The mineral soil components (sandy fractions sized 40–1000 μm) were depleted in radionuclides, while the “organic” substances (semi-decomposed plant residues, humus) accumulated large amounts of radionuclides, showing substantial enrichment.

Radionuclide concentration generally increased with decreasing fraction size, which was more clearly expressed for ^{137}Cs than for ^{90}Sr , ^{241}Am , and $^{239+240}\text{Pu}$. Radionuclides accumulation in the “organic” fractions was relatively uniform, which may mask the effect of sorption processes and smooth out the overall distribution pattern among soil fractions. Rare cases of extreme enrichment of soil fractions with radionuclides were identified in riparian soils, presumably due to the presence of highly active (“hot”) particles, leading to significant heterogeneity in soil contamination.

Thus, the obtained results indicate that radionuclides distribution in the micro-aggregate fractions of the studied soils is governed by several fundamental processes: sorption of radionuclides onto soil particles which increases their accumulation as particle size decreases; high and relatively uniform accumulation of radionuclides in organic matters;

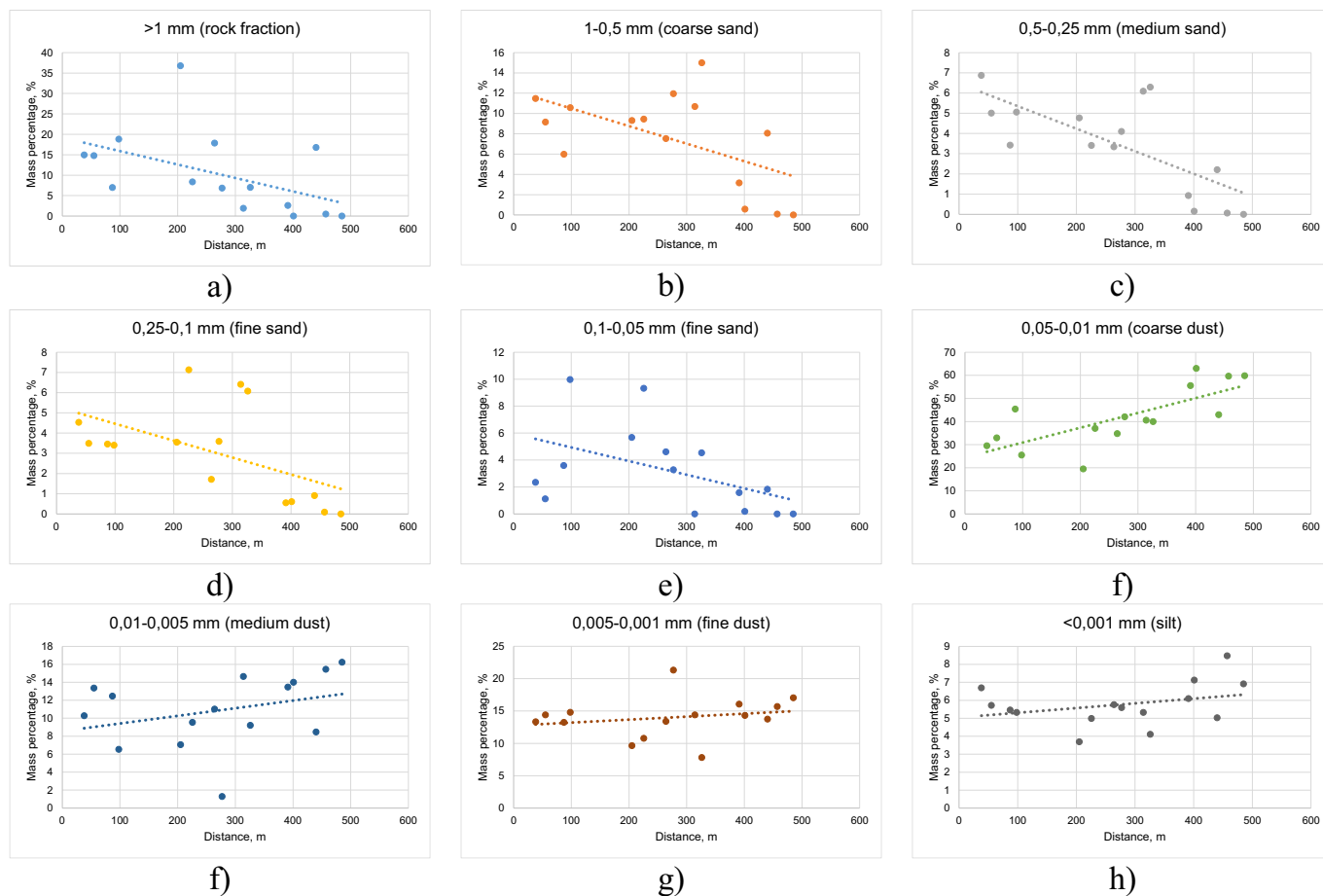


Fig. 10. – Changes in the content of soil particle-size fractions along the left bank of the waterstream (Soil particle-size fractions are named following the classification by N.A. Kachinsky).

Table 6

– Values of correlation coefficients of fraction content change from distance.

Place	Fraction, mm								
	>1	1–0,5	0,5 – 0,25	0,25–0,1	0,01 – 0,05	0,05–0,01	0,01 – 0,005	0,005–0,001	<0,001
Right bank	–0,15	–0,39	–0,65	–0,66	–0,1	0,47	0,17	0,07	0,0007
Left bank	–0,5	–0,56	–0,72	–0,53	–0,48	0,74	0,32	0,22	0,33

and the formation of highly active individual particles (“hot” particles), causing sharp enrichment of certain fractions. These processes interact with each other, collectively determining the extent of radionuclide accumulation in different soil fractions.

¹³⁷Cs and ⁹⁰Sr show the highest accumulation in the lower reaches of the waterstream, apparently due to the effects of terrain relief and the transport of solid runoff. The spatial distribution of ²⁴¹Am and ²³⁹⁺²⁴⁰Pu along the waterstream did not show a clear correlation with distance from the source, likely reflecting their strong association with organic matter.

CRediT authorship contribution statement

A.M. Kabdyrakova: Writing – original draft. **A.Ye. Kunduzbayeva:** Investigation. **A.T. Mendubayev:** Investigation. **E.G. Batyrbekov:** Resources. **S.N. Lukashenko:** Supervision. **V.A. Vityuk:** Resources. **N.V. Larionova:** Project administration. **S.K. Kabdrahmanova:** Supervision.

Declaration of competing interest

The authors declare no conflicts of interest relevant to this study.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2025.180443>.

Data availability

Data will be made available on request.

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