RESEARCH PAPER

GAMMA-EMITTING ISOTOPES SPECIATION IN FLOODPLAIN SOILS OF THE BALCHUGOVSKAYA CHANNEL TEMPORARY STREAM (THE YENISEI RIVER)

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Received: April 8th 2024 / Accepted: November 27th 2024 / Published: December 31st 2024
https://doi.org/10.24057/2071-9388-2024-3339

ABSTRACT. The paper presents the first data on the ratio of gamma-emitting isotopes (^{137}Cs, ^{152}Eu, ^{154}Eu, ^{60}Co) speciation in the floodplain soil and rhizosphere of floodplain plants in the Balchugovskaya channel. This channel is located in the near impact zone of the Krasnoyarsk Mining and Chemical Combine alongside the Yenisei River. The formation of a temporary stream during high floods affects the spatial distribution of isotope specific activities in the soil and rhizosphere of this area. The gross specific activities of isotopes vary in a very wide range. The highest ones are recorded in the lower outlet of the temporary stream (up to 800 Bq kg^{-1}). The procedure of sequential extraction was applied to obtain data on isotope speciation. The hydrological regime characterized by high floods in this section of the Yenisei River, as well as the biological activity of plants, exerts a noticeable influence on the spatial distribution of isotope specific activities and their speciation ratios. Lower percentages or the absence of mobile forms of isotopes in the plant rhizosphere, as compared to the bulk soil, provide evidence for this. In general, isotopes in the soil and rhizosphere tend to accumulate in the organic and residual fractions. There are noticeably different distributions of isotope speciation in central areas and sides of the temporary stream entrance and outlets. The most diverse speciation patterns were observed for ^{152}Eu and ^{154}Eu isotopes, but under different hydrological conditions.

KEYWORDS: speciation, gamma-emitting isotopes, soil, rhizosphere, the Yenisei floodplain

CITATION: Kropacheva M. Y., Repina A. V., Vosel Y. S. (2024). Gamma-Emitting Isotopes Speciation In Floodplain Soils Of The Balchugovskaya Channel Temporary Stream (The Yenisei River). Geography, Environment, Sustainability, 4(17), 217-222
https://doi.org/10.24057/2071-9388-2024-3339

ACKNOWLEDGEMENTS: This work was supported by a grant from the Russian Science Foundation (project No 23-27-00364). The analytical studies of samples were performed in the Analytical Center for multi-elemental and isotope research SB RAS.

Conflict of interests: The authors reported no potential conflict of interest.

INTRODUCTION

The Yenisei floodplain has been significantly impacted by the Krasnoyarsk Mining and Chemical Combine (KMCC) located in the town of Zheleznogorsk. The main source of this impact were two direct-flow reactors that produced weapons-grade plutonium. The impact on the floodplain biogeocoenosis was significant and was traced all the way to the Kara Sea, where the content of ^{137}Cs in the bottom sediments exceeded global fallout by 4-8 fold (Vakulovsky et al. 1995). When the KMCC direct-flow reactors were turned off in 1991 and 1992, the specific activities of isotopes in the water decreased by a factor of 1000 (Nosov 1996; Nosov et al. 2010). However, the amounts of isotopes in the sediments of the floodplain remained quite high (Linnik et al., 2004; Sukhorukov et al., 2000, 2004).

Radioecological studies in this territory have been conducted over more than 30 years. Part of the research focused on the study of isotope distribution in the Balchugovskaya channel, located in the so-called MCC near impact zone. The floodplain area from the MCC reactor cooling discharge outlet to the Kan River influx into the Yenisei River (~30 km) is considered the latter. The channel has a complex geomorphological structure and hydrological regime (Linnik et al., 2005). Both along the entire Yenisei floodplain and in the Balchugovskaya channel, the riverside zone 5-50 m wide, flooded during floods, exhibits the highest level of isotope contamination (Nosov et al., 1993; Stukin et al., 2002). The isotope distribution in this riverbed section correlated with parameters of hydrological and sedimentation regimes in the 1960-1990s, in particular after the Krasnoyarsk hydroelectric power plant constructed in 1967 (Linnik et al., 2005). Numerous studies have focused on the distribution of isotopes in bottom sediments (Bolsunovsky, 2011; Bolsunovsky and Bondareva, 2007; Bolsunovsky et al., 2021; Bondareva and Bolsunovskii, 2008; Semizhon et al., 2010; Zotina et al., 2014, 2019) of the Balchugovskaya channel.

There were noticeably fewer studies of isotope distribution in floodplain soils. Favorable conditions for the deposition of silt and large amounts of organic matter along the river banks, resulting in a sharp increase in isotope content in floodplain soils, were observed in the Balchugovskaya channel (Korobova et al., 2016; Kuznetsov et al., 1999; Linnik et al., 2005; Linnik et al., 2004). The results of sampling and radiometric mapping at the channel cape led to the identification of various landscape-radiation sections in this part of the floodplain (Linnik et al., 2004; Sukhorukov et al., 2004). The profiles showed that a noticeable amount of radioactive contamination was contained both in the surface deposits and distributed throughout the soil profile, including the underlying gravel layer. The isotope ratios in floodplain sediments were also used to determine the chronology of floodplain contamination (Linnik et al., 2005; Linnik et al., 2004). Several studies have focused on the distribution of isotopes in the granulometric fractions of floodplain soils (Korobova et al., 2014; Kropacheva et al., 2013; Linnik et al., 2004). Thus, the Balchugovskaya channel represents a complex floodplain segment, which is part of the area where the floodplain biogeocenosis continues to be influenced by the nuclear fuel cycle plant.

In models of isotope migration, the main parameter is the proportion of exchange forms of the isotope in the soil or bottom sediment (Bulgakov et al., 2002; Konoplev, 2020; Mikhaylovskaya et al., 2002). Moreover, studies on isotope migration typically treat the soil as a single entity (Tajima et al., 2022; Tsuji et al., 2016). The soil that is part of the plant's rhizosphere and the soil outside the rhizosphere have different conditions for isotope accumulation, as shown by our monitoring studies in 2003-2016 (Kropacheva et al., 2021; Kropacheva et al., 2011; Kropatcheva et al., 2012). However, samples for them were taken at only one site of the Balchugovskaya channel (Fig. 1a, point 1). The remaining territory has not been covered by studies on the distribution of artificial isotopes in the rhizosphere of floodplain plants.

The aim of the study is the determination of isotope speciations in surface sediments (floodplain soil and plant rhizosphere) of the Balchugovskaya channel. The isotopes ^{137}Cs, ^{152}Eu, ^{154}Eu, and ^{60}Co are analyzed in this study because they are the main gamma-emitting contaminants of the Yenisei River floodplain. The procedure of sequential extraction was applied to obtain data on isotope speciation. Determination of the percentage of mobile speciations of isotopes will allow the assessment of isotope removal from floodplain soil and plant rhizosphere during floods of different intensityintensities. The obtained data can be used in the assessment of secondary isotope pollution of the Yenisei floodplain and modeling of isotope migration in the river system.

MATERIALS AND METHODS

Sampling

The Balchugovskaya channel is an anabranch between the Yenisei eastern bank and Berezovy Island. At a distance of 3.5 km from the entrance to the channel, the riverbed rounds a ledge on the eastern bank (Linnik et al., 2005). During high floods, the water flow goes straight through the ledge, forming two temporary streams (Fig. 1b).

In August 2019 floodplain soils were sampled at nine points: at the entrance to the temporary streams (points 1, 6, 7) and at two outlets, the upper (points 2, 3, 8) and lower (points 4, 5, 9). The depth of sampling (~30 cm) was chosen to capture the entire layer of sedge roots. At each point, a cube of soil with sedge roots (~30 cm^3) was extracted and manually disassembled into plant roots with adjacent rhizosphere and bulk soil. The part of the soil that remained adhered to the roots after shaking the sample was considered a rhizosphere; the rest of the soil was taken as bulk soil (Séguin et al., 2004). The rhizosphere was separated from the roots after drying.

Sequential chemical fractionation

To study the association of isotope to different speciation, sequential extraction procedure were applied to soil and rhizosphere samples using a five-step procedure (Table 1). The procedure has previously been applied in other research projects on floodplain soils and bottom sediments in the KMCC near impact zone. (Bondareva, 2012; Semizhon et al., 2010). However, this is the first case of its application to floodplain soils of the Balchugovskaya channel temporary streams.

The point 7 rhizosphere sample wasn't fractionated because it was already being used to find out ^{90}Sr-specific activities by β-radiometry with radiochemical preparation.

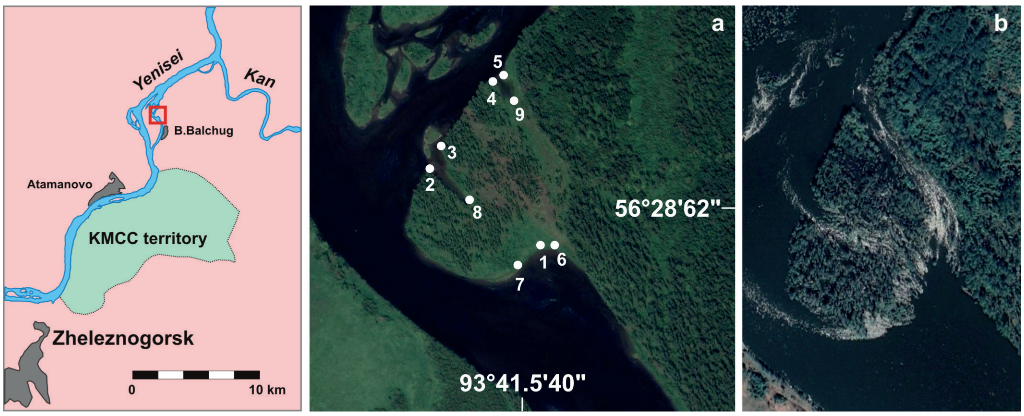

Fig. 1. Balchugovskaya channel: a) sampling scheme (August 2019); b) same place in high flood (June 2021)

Table 1. Schemes of sequential extraction of isotopes from a soil and a rhizosphere

fraction		chemical reagents	mobility	
I	exchangeable ions	CH_3COOH_4 (1M) pH=7	decrease	↓
II	carbonates	CH_3COOH_4 (1M) and HNO_3 pH=5		
III	Fe, Mn oxides and hydroxides	$NH_4OH{\cdot}HCl$ (0,2M) in CH_3COOH (25%)		
IV	organic matter	H_2O_2 (35%) + HNO_3 (1M) pH 1.5, 85°C		
V	residual solids	-		

Measuring procedure

The specific activity of isotopes was measured by semiconductor gamma spectrometry with lead and tungsten shields on an IFTP DGDK-100V coaxial Ge(Li) detector (Dubna, Russia). The data were processed using the AnGamma software SPC "Aspect", (Dubna, Russia) (SPC "Aspect" 2000). The following gamma-lines were used for isotope detection: 661.66 keV (^{137}Cs), 1173 and 1332 keV (^{60}Co); 344 and 778 keV (^{152}Eu), 1274 keV (^{154}Eu). Measurements lasted from 1 to 24 hours, depending on isotope activity level, to provide a photopeak area accuracy to within 5–10%. The detection limit for ^{137}Cs was from 2 to 5 Bq kg^{-1}. All sample activities were recalculated according to the date of sampling.

The Analytical Center for Multi-Elmental and Isotope Research SB RAS conducted the analytical studies of the samples.

RESULTS AND DISCUSSION

The bulk isotopes specific activities vary in a wide range. The ^{137}Cs exhibit the highest values, ranging from 200 to 800 Bq/kg. The levels of isotopes specific activity in the upper outlet are approximately equal to the ones at the entrance to the temporary stream (^{137}Cs) and in some cases exceed them (^{152}Eu and partially ^{60}Co). The lower outlet has significantly higher bulk isotopes specific activities for almost all isotopes than the upper outlet. This outlet has a higher bed at the turning point, which obviously causes less frequent flooding and no isotope leaching. Higher isotope levels have also previously been recorded at this site, although less contamination corresponded to the higher terrain (Linnik et al., 2005). In general, isotopes in the soil and rhizosphere gravitate toward organic and residual fractions.

The ratio of speciation isotope patterns in soils and rhizospheres at the side points of the temporary stream outlets is about the same. Points 2 and 4 on the left side of the temporary stream outlets are more similar than points 3 and 5 on the right side. The distribution of speciation ^{154}Eu isotope forms in the soil and rhizosphere of the central areas of temporary stream outlets (points 8 and 9) is significantly different. In the upper outlet, the isotope is associated with a significant proportion of carbonates (24-34%). In the lower outlet, it is linked to Fe oxides and hydroxides (30%). The ratio of speciation isotopes on the right side of the temporary stream entrance (point 6) is also noticeably different from typical. Obviously, favorable conditions for sediment accumulation are formed here even at high floods, as indicated by the distribution of the speciation ^{154}Eu isotope. This isotope is equally present in the exchangeable and carbonate fractions in soil, in the exchangeable fraction, the Fe and Mn oxides and hydroxides fraction, and the organic fraction in the rhizosphere. Earlier studies have reported markedly higher densities of isotope contamination at this site (Linnik et al., 2005) than the center (point 1) and left side (point 7) of the entrance to the temporary stream.

Lower levels or lack of isotopes in exchangeable fraction in the rhizosphere compared to bulk soil may be a consequence of uptake of these forms by plant roots. The mobile forms of Eu isotopes in these three points (6, 8, and 9) result from the absence of two typical processes for other points and isotopes: 1) the mobile forms of the isotope are not washed out at low floods; 2) Eu isotopes are not uptaken by plants (Kropatcheva et al. 2012). The lack of ^{60}Co in soil or rhizosphere fractions when its total activities are present (points 1, 5, 6, and 7) indicates that the total activities were measured for an active particle that did not fall into the weighed quantity for sequential extraction technique.

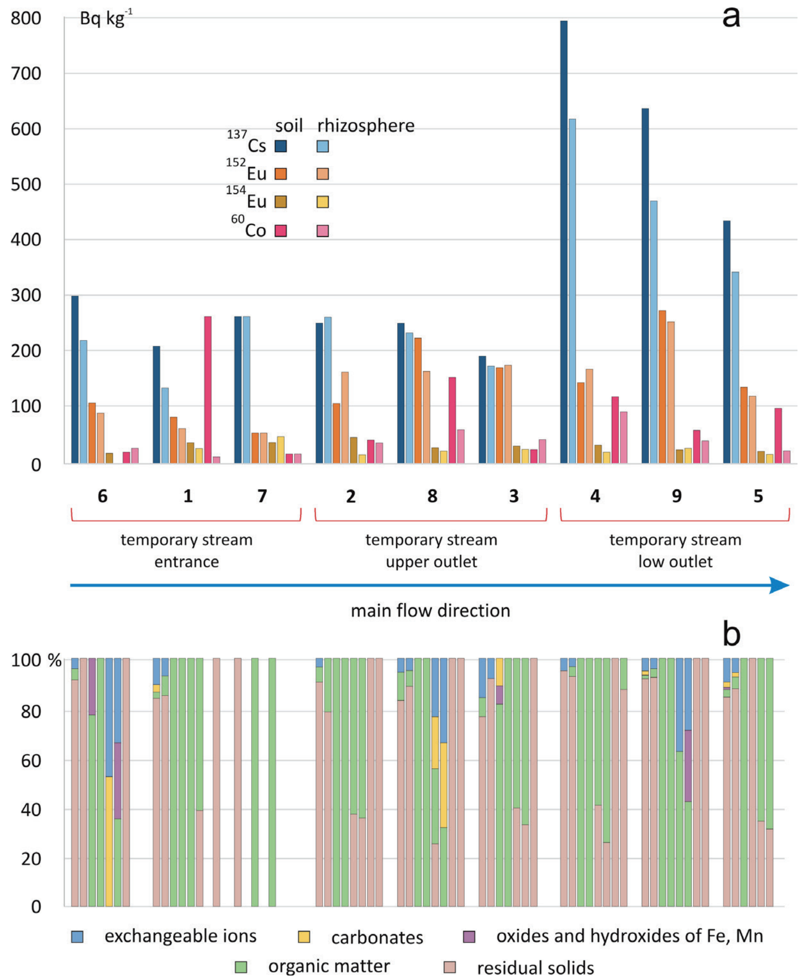

Fig. 2. Isotope specific activity in soil and rhizosphere bulk samples (a) and isotope speciation distribution in the samples (b)

A statistical analysis was conducted based on the obtained data to identify possible correlations between the specific activities of isotopes and their speciation. Since the data samples are not normally distributed, the Spearman rank-order correlation coefficient ρ was used to calculate the correlation coefficient.

The total specific activities of some isotopes had very strong positive (0.75–1.00) and statistically significant ($p<0.05$) correlations with each other. These are ^{137}Cs in the soil and the rhizosphere, ^{152}Eu in the soil and the rhizosphere, ^{152}Eu in the soil and ^{154}Eu in the rhizosphere, ^{154}Eu in the soil, and ^{60}Co in the rhizosphere (Fig. 3). The other ratios, although quite high, are not statistically reliable.

Table 2 shows statistically significant correlations between the specific activities of isotopes in soil and rhizosphere and the percentage of speciation isotopes in soil and rhizosphere.

Very high positive correlations (0.75-1.00) are observed between the specific activities of isotopes in the soil and rhizosphere, between fractions I of soil and rhizosphere, and between fractions IV of soil and rhizosphere (Table 2, Nos. 1, 3, 6). A large R^2 value (0.7436-0.8998) is also characteristic for all these correlations. The R^2 values for many of the ratios (Table 2, Nos. 4, 8–11, and 14) are much lower (0.4094-0.5543), but the correlations are high positive or negative (|0.50 - 0.75|). The remaining ratios (Table 2,

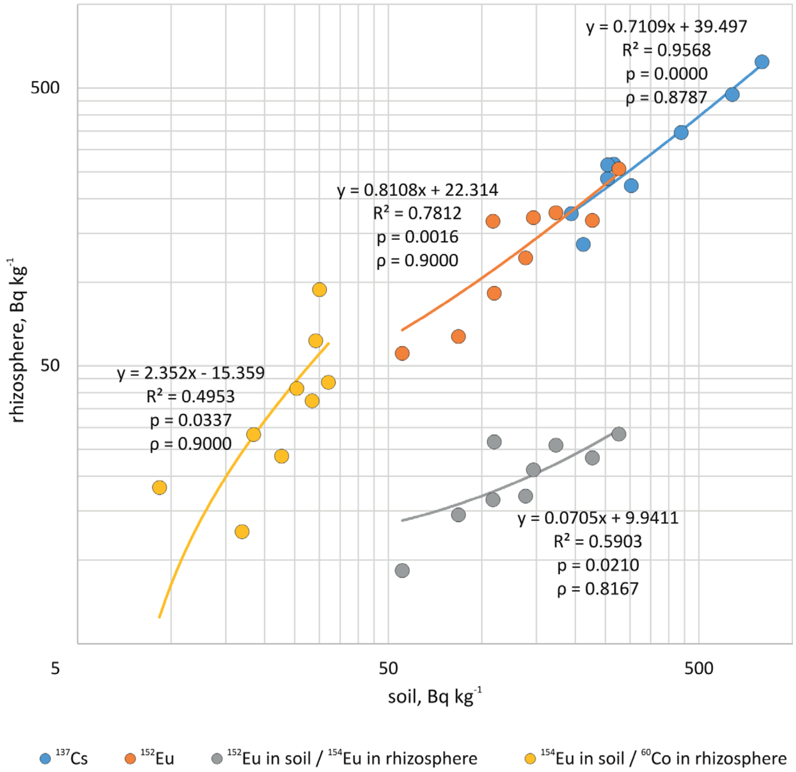

Fig. 3. Correlations between the isotope specific activities in soil and rhizosphere

Table 2. Statistically significant ($p<0.05$) correlations between the specific activities of isotopes and their speciation

No.	Ratios	ρ	R^2	p
1	soil, Bq kg^{-1} vs. rhizosphere, Bq kg^{-1}	0.8657	0.8998	0.0000
2	soil, Bq kg^{-1} vs. V fraction of rhizosphere, %	0.4156	0.2242	0.0062
3	I fraction of soil, % vs. I fraction of rhizosphere, %	0.9345	0.8956	0.0000
4	II fraction of soil, % vs. I fraction of rhizosphere, %	0.6179	0.5409	0.0000
5	I fraction of soil, % vs. III fraction of rhizosphere, %	0.4954	0.7646	0.0000
6	IV fraction of soil, % vs. IV fraction of rhizosphere, %	0.7625	0.7436	0.0000
7	IV fraction of soil, % vs. V fraction of rhizosphere, %	-0.4132	0.3031	0.0011
8	V fraction of soil, % vs. IV fraction of rhizosphere, %	-0.6109	0.4859	0.00001
9	V fraction of soil, % vs. V fraction of rhizosphere, %	0.5320	0.4094	0.00008
10	I fraction of soil, % vs. II fraction of soil, %	0.5660	0.5543	0.00000
11	IV fraction of soil, % vs. V fraction of soil, %	-0.6689	0.5437	0.00000
12	I fraction of rhizosphere, % vs. II fraction of rhizosphere, %	0.4609	0.3299	0.0006
13	I fraction of rhizosphere, % vs. III fraction of rhizosphere, %	0.4939	0.5598	0.00000
14	IV fraction of rhizosphere, % vs. V fraction of rhizosphere, %	-0.6558	0.4916	0.00001

Nos. 2, 5, 7, 12, 13) have medium positive or negative (|0.25 - 0.49|) correlations, with low R^2 values (0.2242-0.5598), except for ratio No. 5 (R^2 = 0.7646).

Positive correlations between the states of isotopes in the soil and rhizosphere (specific activities and speciation) indicate similar migration paths of isotopes in these states. According to correlation ratios 1, 3, and 6, it can be asserted that the total migration of isotopes is primarily determined by the behavior of isotopes in exchangeable and organic fractions. The proportions of isotopes found in V fractions (correlation ratio No. 9) determine the isotope migration pattern, which is significantly weaker.

CONCLUSION

The first data on the distribution of ^{137}Cs, ^{152}Eu, ^{154}Co, and ^{60}Co isotopes in the soil and rhizosphere of floodplain plants in the Balchugovskaya channel temporary streams have been obtained. The sequential extraction technique was employed for the first time in the study of these floodplain soils, with the aim of determining isotope speciation in both the soil and the rhizosphere of floodplain plants. The hydrological regime formed during flooding affects the total specific activities of isotopes and isotope speciation. Migration properties of isotopes are determined mainly by their behavior in exchangeable and organic fractions of soil and rhizosphere.

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