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Investigation of radionuclides in the Yenisey River floodplain systems: Relation of the topsoil radionuclide contamination to landscape features



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ABSTRACT

Landscape–radiometric survey and soil sampling performed in the islands of Beriozovy and Balchug (20 km downstream the Krasnoyarsk Mining and Chemical Combine), and in the Mikhin Island (180 km downstream) showed that the distribution of technogenic radionuclides depends upon the history of contamination and landscape features of the floodplain. Contamination densities of $^{137}\mathrm{Cs}$ appeared to be significantly higher than could be expected from global fallout (1.75–2.5 kBq/m²): in 2000 the maximum value for $^{137}\mathrm{Cs}$ in the Beriozovy Island equaled 663 kBq/m², in the Balchug site - 577 kBq/m² and in the Mikhin Island - 518 kBq/m²). $^{137}\mathrm{Cs}$ contamination density was practically independent of the remoteness from the KMCC that proved its considerable migration in the water-soluble or fine particulate forms. Vertical distributions of man-made nuclides in soil cores depended upon the different half-life of the studied radionuclides, the soil profile relative altitude, its structure and texture. The two main burial depths of $^{137}\mathrm{Cs}$ activity depended upon the intensity of sedimentation and varied from 5 cm to 20–25 cm. In 2000 maximum contamination by $^{60}\mathrm{Co}$ and $^{152,154}\mathrm{Eu}$ isotopes was associated with the top layer and decreased exponentially with depth.

Obtained data is believed to be important for ecological monitoring of the flood plains subjected to radionuclide contamination.

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1. Introduction

The Krasnovarsk Mining and Chemical Combine (KMCC) was constructed for the production of weapons-grade plutonium and is located in the hard-rock area on the right side of the Yenisey River 60 km downstream of Krasnovarsk. The startup of the first water cooled channel reactor was in 1958, the second one in 1961, and the third in 1964. The KMCC enterprise also included a radiochemical plant. Cooling water was discharged to the Yenisey River. In 1992 two reactors producing plutonium were shut down, and in 2010 the last reactor used also for the production of electric power, hot water supply and for heating the town of Zheleznogorsk was also decommissioned. KMCC operation in the period from 1958 to 1992 led to the contamination of the Yenisey floodplain and bottom sediments by artificial radionuclides including radioisotopes with different half-lives including ¹³⁷Cs, ⁶⁰Co, ¹⁵²Eu, and ¹⁵⁴Eu (the corresponding half-lives are equal to 30.2, 5.3, 13.3 and 8.6 years). After the shutdown of the two once-through reactors radionuclide discharge to the river decreased by a dozen times and for the period from 1975 to 2000 ¹³⁷Cs disposal approximated 19,636 GBq (530 Ci) accounting for radionuclide decay (Vakulovsky et al., 2001).

Initial information on the considerable radionuclide contamination of the Yenisey River was obtained at the beginning of the '70s of the previous century (Vakulovsky et al., 1995), but the results of these studies at that period were not available to the scientific community for security purposes. By now there is no published data on annual discharges to the Yenisey River basin in the 1960-1975 period. Radioactivity measurements in the floodplain areas performed in the '70s have been sporadic (Tertyshnik, 2007). It was not earlier than the '90s when the large-scale systematic radionuclide studies of the Yenisey River floodplain from the town of Zheleznogorsk to the Kara Sea (covering the distance of over 2000 km) started. The results allowed the conclusion that maximum contamination of the bottom sediments and floodplain soils took place in the '60-'70s of the last century in areas adjacent to KMCC (Bolsunovsky, 2004; Linnik et al., 2000; Nosov and Martynova, 1996; Nosov et al., 1993; Sukhorukov et al., 2000, 2004; Vakulovsky et al., 1994, 1995).

Radioactive contamination in the lower reaches of the Yenisey River down to its inflow into the Kara Sea was firstly registered at the beginning of the '70s (Vakulovsky et al., 1994), its traces in this segment of

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the Yenisey floodplain were found at the beginning of the 1990s (Kuznetsov et al., 1994) and in 2001 (Korobova et al., 2007, 2009; Linnik et al., 2013).

The main argument in favor of the suggestion concerning maximum disposal particularly in the '60s was the burial of the layer with peak ¹³⁷Cs activity at the depth of 15–20 cm and even deeper in some cases. Maximum contamination by the technogenic radioisotopes within the river basin is usually related to the extremal high water period of 1966 which led to inundation and the accompanying radionuclide contamination of all the islands in vicinity of the enterprise. After the building of the Krasnoyarsk hydroelectric power station (HEPS) in 1970 the regulation of the water regime of the Yenisey River started and since then the areas of high floodplain have practically never been submerged or submerged but only for a few days per annum. Therefore these parts of the floodplain have preserved the traces of the early contamination of 1958–1970. The medium-level and low floodplain has been subjected to contamination of alluvial deposits until the reactors were shut down in 1992.

Landscape and radioecological studies carried out in the Vernadsky Institute in the framework of a series of international projects (STREAM, ESTABLISH) helped to reveal the distribution of technogenic radionuclides depending upon the geomorphology, lithology, and vegetation cover in both near the KMCC impact zone (Linnik et al., 2004a,b, 2005) and in remote areas (Korobova et al., 2007, 2009). Patterns and inventories of radioactive contamination of island sites of the Yenisey River were presented in Linnik et al. (2006).

Radiation monitoring of the nearby zone of the KMCC is still essential due to the presence of highly radioactive "hot" particles buried in soils (Bolsunovsky and Tcherkezian, 2001; Gritchenko et al., 2001; Sukhorukov et al., 2009). After the termination of radionuclide discharges and in the absence of the extreme flooding natural processes start "deactivating" the flood plain by burial of the contaminated layers that lead to a decrease of the surface radioactivity and exposition dose. However in case of a catastrophic flooding and washing of the soil there exists a real possibility of the transport of "hot" particles to the river basin. That is why considerable attention is paid to a mathematic modeling of the radiation situation depending upon landscape factors of radionuclide distribution within the floodplain (Linnik and Potapov, 2009).

It is worth mentioning that unlike the Techa River basin that has also been subjected to radionuclide contamination due to discharges of the radiochemical plant (Chesnokov et al., 2000), the Yenisey floodplain is studied to a minor extent. Definite concern is related to the Yenisey contamination with radioisotopes of ^{239,240}Pu, that may be estimated on the basis of contamination with ¹³⁷Cs, ⁶⁰Co, ¹⁵²Eu, and ¹⁵⁴Eu (Kropatcheva et al., 2012).

The main goal of this study was to analyze landscape differentiation and burial patterns of radionuclides in floodplain facies of the Yenisey in the nearby and remote impact zones of KMCC in both lateral and vertical directions with due regard to lithology related to hydrological regime and periods of contamination.

2. Study area

The Yenisey flood plain area is characterized by a varying width ranging from 2 km to 30 km. It narrows at places of outcropping Archean metamorphic granulite and amphibolite rock complex then being presented only by thin colluvial deposits. A metamorphic complex is formed by pyroxene–biotite, garnet–biotite and pyroxene–garnet–biotite gneisses substituted for biotite, garnet–biotite, spar–biotite gneisses and granite–gneisses (Geologic map, 1978).

According to the dated neolithical findings the middle Yenisei upper flood plain 6 to 8 m high has started to form 14–18 kya to 4–5 kya since the Noril'sk glaciation period and the following interglacial stage (Zubakov, 1965).

The study area characterizes the two floodplain fragments: 1) the island of Beriozovy with the adjacent right riverside massif Balchug, and 2) the island of Mikhin (close to the left riverside floodplain) located 20 km and 180 km downstream of the KMCC discharge point correspondingly.

Investigated islands are situated in the widened segments of the flood plain. In the region of the Bol'shoi Balchug settlement the flood plain is formed on Middle Jurassic parent rocks. The bottom of the parent rocks is formed of pebbles with beds of oblique laminated sandstone covered by coarse-grained oblique sandstone subsequently replaced by fine-grained deposits. On the top layers argillite layers are inter-bedding with aleurolite and medium-, small-, and fine-grained sandstone.

3. Materials and methods

A field study of radionuclide contamination of the Yenisei River performed in 1999–2000 included the landscape cross section of the flood plain with description of landscape structure, relief, soil and vegetation cover, and alluvium composition to analyze landscape peculiarities responsible for the distribution of radionuclides (137Cs, 60Co, 154Eu, 152Eu) discharged by KMCC. Despite a rather short half life of 60Co and 152,154Eu a study of the distribution of these radioisotopes in the near zone of KMCC remains important nowadays (Kropatcheva et al., 2012). The soil and landscape field survey data were used to create facies' maps.

Soil vertical profiles (identification of the types using Russian classification, Egorov et al., 1977) have been sampled at the test plots selected on the basis of the landscape hydrological analysis of radionuclide deposition to verify the field radiometric data and to analyze the soil geochemical properties. The procedure of soil and sediment sampling included: 1) a detailed visual description of the structure, thickness, composition, density, texture, and organoleptic features of the soil layers and beds opened in the pit; 2) manual and continuous sampling from a fixed square (15 \times 15 cm) in increments of 2 cm to 5–10 cm increasing with depth with due regard to lithological composition of the layers. Sampling of the low-level floodplain layers has been limited to the depth of the shingle layer often found 40 cm to 100 cm below the surface.

Radioactivity measurements were carried out along landscape cross-sections to reveal correlation between gamma-emitter distribution and landscape features of the floodplain. Gamma-emitters' inventory and their depth migration was determined with the help of a field radiometer "CORAD" based on a collimated scintillation detector $(NaI(T1) \varnothing 50 \times 50 \text{ mm})$ (Chesnokov et al., 1997, 1999). The developed technique makes it possible to estimate the thickness of a soil layer containing 80% of gamma-emitters' inventory that indirectly shows the depth of radionuclide migration. The thickness of the soil layer is calculated on the basis of a measured parameter Z corresponding to free paths of γ -quanta with the energy of 662 keV in the examined soil (Chesnokov et al., 1997). If the contaminated layer is buried under a conditionally clean one, Z may be presented as $Z = L_0 + L_1$, where L_1 is the thickness of the contaminated layer (Chesnokov et al., 1997, 1999) and L_0 is the thickness of the clean layer. Registration protocol included all three parameters (Z, L₀, and L₁). In case of maximum contamination in the surface layer $L_0 = 0$ (Chesnokov et al., 1997, 1999). The procedure started with L₀ measurement to evaluate alluvium sedimentation rate after the reactors' shutdown and termination of radionuclide discharges.

"CORAD" verification against both the 137 Cs contamination density and its depth distribution basing on parameters Z, L_0 , L_1 performed in the Yenisey floodplain landscapes is described in Stream (2002), Linnik et al. (2005).

Sampling and landscape-radiometric survey were carried out in August 2000 when the river water level was 0.9 m above the low-water level. Radionuclide determination was carried out using a low

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