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Monitoring and assessment of ^{137}Cs and ^{90}Sr radioactive isotopes in the 'soil – rhizosphere – sedge' system of the Yenisei River floodplain (near impact zone of Krasnoyarsk MCC, Russia)

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Abstract Radiocaesium and radiostrontium contamination in the 'soil – rhizosphere – plants (aerial parts)' system was monitored in the floodplain ecosystem of the Yenisei River in the near impact zone of the Krasnoyarsk Mining and Chemical Combine (MCC). The monitored system included soil, rhizosphere, and sedge vegetation on islands and the river's east bank. The ^{137}Cs and ^{90}Sr specific activities displayed intricate space and time patterns controlled by the river water level, including the time and duration of floods and their correlation with the sedge vegetation season. The specific activities of both radionuclides, especially ^{137}Cs, were above the background in all years of observation, except in a few cases. The soil-to-plant transfer factor (TF) patterns showed continuous ^{137}Cs and ^{90}Sr influx into the system and annual variations in the shares of their bioavailable and fixed forms, especially for ^{90}Sr. The ^{90}Sr distribution in the 'soil – rhizosphere – plants' system observed in 2014–2016 provides evidence for possible local fallout.

Keywords ^{137}Cs · ^{90}Sr · Transfer factor · Floodplain soils · Rhizosphere · Riverside plants

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Introduction

The Krasnoyarsk Mining and Chemical Combine (MCC) is situated on Yenisei River 60 km downstream from Krasnoyarsk city and started operation in 1950. It includes three reactors: AD (operating period 1958–1992), ADE-1 (1961–1992), and ADE-2 (1964–2010). All three are thermal-neutron uranium-graphite channel-type reactors. The AD and ADE-1 reactors produced exclusively weapon-grade plutonium and worked in straight-through mode, discharging cooling waters directly into the river. The ADE-2 reactor had a closed-type cooling system, and aside from plutonium production, it served as a source of steam for electricity production and water heating for heating stations and the electrical grid of Zheleznogorsk city. In 1964, a radiochemical plant began operation as a part of the MCC. Its main task consisted of plutonium extraction from irradiated

uranium. The plant also included a spent fuel regeneration factory. The 'Severny' emplacement area was designed to accept and dispose of liquid low-level and intermediate-level radioactive waste and entered into service in 1967. In 2015, a new commercial mixed oxide (MOX) fuel fabrication facility (MFFF) was launched. Currently, dismantling at AD and ADE-1 is almost completed, and decommissioning activities by radiation-proof on-site disposal methods are in progress. The ADE-2 reactor currently operates in shutdown mode.

The impact of the Krasnoyarsk Mining and Chemical Combine on the Yenisei River floodplain environment has been studied for more than 40 years after ^{137}Cs anomalies of 15 kBq m^{-2} (four to eight times the global fallout) were found in the bottom sediments of the Kara Sea in 1971 (Vakulovsky et al., 1995). Sampling floodplain sediments between 6 and 250 km downstream of MCC in 1973 revealed high contamination with artificial radionuclides, at 460 kBq m^{-2} ^{137}Cs to 90 kBq m^{-2} (Nosov et al., 1993; Vakulovskii et al., 2008; Vakulovsky et al., 1995). The calculated mean annual ^{137}Cs and ^{90}Sr fluxes into the system in 1985–1995 were, respectively, 6.0 and 1.6 TBq year^{-1} in the proximal zone within 15 km away from the MCC and 1.8 and 1.5 TBq year^{-1} in the distal zone at 1360 km downstream (Vakulovsky et al., 1995).

Systematic studies of the MCC-derived radioactive pollution (mainly gamma-emitting ^{137}Cs) of floodplain components began in 1991, shortly before the straight-through reactors were stopped. Then the bottom sediments near the MCC contained 0.3–1.5 kBq kg^{-1} ^{137}Cs (Nosov, 1996). After the reactors had been stopped in 1994, ^{137}Cs decreased slightly to 0.2–1.0 kBq kg^{-1} and 370 kBq m^{-2} (Nosov, 1996). Although the long-lived isotopes in the floodplain decreased to n·10^{-2} Bq L^{-1}, up to 80% of the ^{137}Cs transported to the river system was adsorbed onto particulate matter, while the total contamination depended on secondary release from polluted bottom and floodplain sediment (Nosov, 1996). In general, the total contamination of the floodplain soil and bottom sediments varied little, but for a 5–15% decrease by short-lived isotopes decay (Bolsunovsky et al., 2002; Nosov, 1996).

The floodplain outside the flooded areas has been free from the MCC-related contamination, with the soil ^{137}Cs level about 5–12 Bq kg^{-1} global fallout (Bolsunovsky et al., 1999; Nosov, 1996), except for local anomalies produced by winter effects, such as rafting of mud-bearing ice (Nosov, 1996). The contamination of bottom sediments decreases away from the MCC, and the specific activity of radiocaesium depends on the sediment composition and depth (Bondareva, 2012; Semizhon et al., 2010). Here, ^{137}Cs is higher in the lower strata (Standring et al., 2009) and is the least mobile in shallow sediments (5–15% of mobile forms) among other long-lived isotopes (Bondareva, 2012; Bondareva & Bolsunovskii, 2008).

The Yenisei floodplain sediments contain hot particles (60–70 per km^2), which affect the mobile forms of ^{137}Cs (Bondareva & Bolsunovskii, 2008). The ^{137}Cs specific activity in some hot particles can reach 30 MBq per particle (Bolsunovsky & Melgunov, 2014; Bolsunovsky & Tcherkezian, 2001; Bolsunovsky et al., 2017; Gritchenko et al., 2001; Sukhorukov et al., 2000, 2004, 2009). In natural conditions, hot particles dissolve gradually upon interaction with percolating water or pore moisture, while the ^{137}Cs they carry become involved in secondary migration (~0.5% per flood cycle) (Chuguevskii et al., 2010; Chuguevsky, 2019).

Pollution has also been studied in water biota. Contamination with long-lived gamma-emitting isotopes was found in fish caught upstream and far downstream of the effluent discharge (Bolsunovsky & Dement'ev, 2010; Nosov et al., 1993; Vakulovsky et al., 1995; Zotina et al., 2011). In aquatic plants growing near the MCC, the highest ^{137}Cs activity concentration reached 660 Bq kg^{-1} in 1997–1998, 350 Bq kg^{-1} in 1999–2009, and as low as 70 Bq kg^{-1} in 2010 (Bolsunovsky, 2004; Bolsunovsky et al., 2002, 2011). Cytogenetic and other toxic effects in aquatic plant cells showed a positive correlation with ^{137}Cs in the root layer of bottom sediments (Medvedeva et al., 2014), while ^{137}Cs specific activity correlated inversely with shoot length (Zotina et al., 2014). Aerial plant parts and fungi contained up to 300–520 Bq kg^{-1} of ^{137}Cs in 1993–1994 (Nosov, 1996). Concentration of ^{137}Cs did not decrease significantly in water biota of the Yenisei after the shutdown of the last reactor plant because the discharges of this radionuclide to the Yenisei continued at the same level. On a longer-term scale (since 1973 and since 1991), concentration of ^{137}Cs in fish muscle had significantly decreased, following the decrease in annual discharges of this radionuclide to the Yenisei, and the ecological half-life of ^{137}Cs was estimated as 6.5–12.8 years (Zotina et al., 2019).

Since the beginning of radioactive pollution research, the ^{137}Cs inventory in the Yenisei floodplain sediments has been a subject of considerable interest. It was estimated to be 23 GBq in the Atamanovsky Spit, 5 km downstream of MCC (Sukhorukov et al., 2000), 145 GBq in Beryozovy Island at 19 km, 148 GBq in Mikhin Island at 180 km, and 16 GBq in Cheryomukhovy Island at 250 km (Linnik et al., 2006). The total measured between the MCC and Dudinka (located 1900 km away) in 1972 was 5300 GBq (Tertyshnik, 2007).

Most studies have focused on gamma-emitting radionuclides, meaning data reports on radiostrontium in different floodplain components are limited to fewer publications. Monitoring during 1997–2002 (Sukhorukov et al., 2004) showed an uneven distribution of ^{90}Sr along a soil profile near the MCC. The ^{90}Sr specific activity varied from 8 to 6000 Bq kg^{-1} as a function of depth at sites in close proximity to the MCC and decreased markedly away from the combine to 40–350 Bq kg^{-1}, and down to 10–20 Bq kg^{-1} in soil samples from islands and the river's east bank. The activity was noted to be higher (50 Bq kg^{-1}) in a bay of the Balchug Channel. Relatively high values were measured locally in the near impact zone (620 and 210 Bq kg^{-1}) and in non-flooded islands (110 Bq kg^{-1}), possibly due to atmospheric fallout (Sukhorukov et al., 2004). A similar ^{90}Sr pattern was observed in bottom sediments, uneven spatial distribution and a general decrease away from the MCC, from 100 to 180 Bq kg^{-1}, or locally up to 250 Bq kg^{-1}, to within 10 Bq kg^{-1} (Sukhorukov et al., 2004).

According to modelling, ^{90}Sr influxes to the Kara Sea during 1992–2000 were controlled by run-off from the drainage basin rather than by the operation of the MCC, which was responsible for no more than 12% of total ^{90}Sr flux (Platovskikh et al., 2003). A large portion of ^{90}Sr (20–30%), more than ^{137}Cs, was released from water-logged floodplain soil, i.e. Sr transport into the Yenisei was mostly with intrasoil run-off (Legin et al., 2008). Radiostrontium has high migration mobility in shallow bottom sediments, which carry up to 60–70% of mobile forms (Bolsunovsky & Melgunov, 2014).

Previous studies have revealed an intricate pattern of radionuclide contamination of the Yenisei floodplain. The environment generally recovers as the contaminated floodplain deposits become buried under more recent clean sediments. However, no systematic studies of terrestrial vegetation in the near impact zone of the MCC have been undertaken before, especially in relation to the soil and rhizosphere. Living plants can liberate fixed elements and play an important role, either positively or negatively, in the self-cleaning action of a floodplain ecosystem. In this respect, it is important to constrain the redeposition of radionuclides in the flood plain soil, their partitioning between the soil and rhizosphere, the bioavailability of mobile and fixed forms, and the controls of their distribution among the root layer in the soil, the rhizosphere, and the aerial parts of plants.

Sampling and methods

Sampling sites

During August–September 2004, 2011–2016, we sampled different components of the floodplain ecosystem at five sites on islands and on the banks of the Yenisei in the near-impact zone of the Krasnoyarsk MCC (Fig. 1).

Site A (reference area): Potekhin Island and the Yenisei west bank, situated 200 m away from Shivera Village and 8 km upstream of the discharge. Both sampling points are located on the beach, which is covered with thick grass and becomes flooded systematically with short heavy floods.

Site B: upstream (upper) and downstream (lower) ends of Atamanovskaya Spit (an island), near the right bank of the river, located 6 km downstream of the effluent discharge from the reactor cooling systems. Yearly floods have produced a thick layer of alluvium covered with thick grass and shrubs. The island is located in the impact zone of the MCC wastewaters that the Yenisei current diverges towards the right bank. During the floods, which cover the whole spit area, the flow regime is short and intense.

Site C: upper and lower ends of Atamanovsky Island, 7 km downstream of the discharge. Sampling was close to the river, which becomes flooded at minor rises in water level. The island comprises two zones, grown, respectively, with grass and shrubs and with trees (including pine plantations); the former is flooded yearly or periodically, with short and heavy floods, and the latter never floods, even at the high water levels.

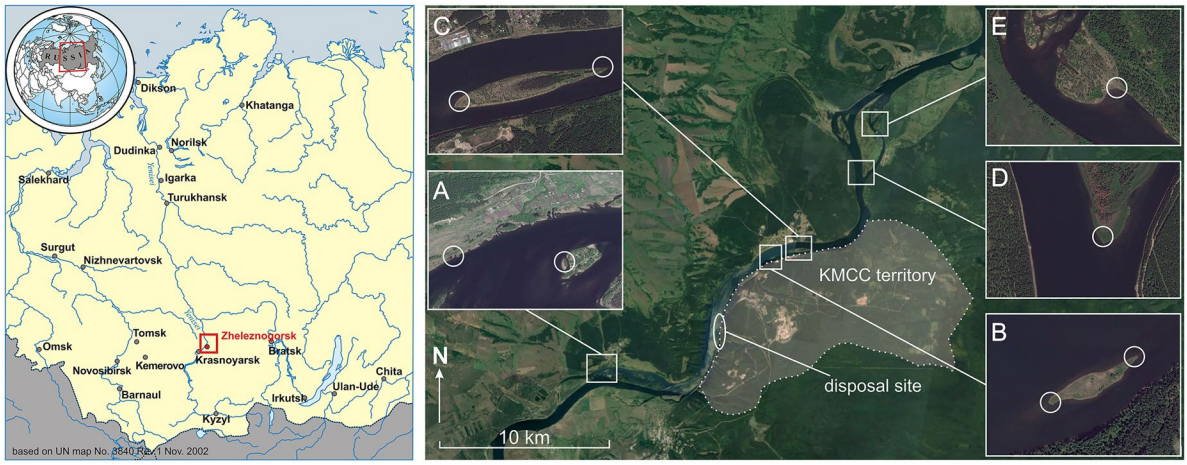

Fig. 1 Study area and sampling sites. **A** Shivera Village area, reference site, 5 km upstream of effluent; **B**: Atamanovskaya Spit, 6 km downstream of effluent; **C**: Atamanovsky Island, 7 km downstream of effluent; **D**: Beryozovy Island, 15 km downstream from disposal site; **E**: Balchug Channel, 18 km downstream of effluent (Google Earth images)

Site D: the upper end of Beryozovy Island, towards the west, 15 km downstream of the discharge. Most of the island is covered with mixed forest and eludes yearly flooding, but the area near the water, with grass vegetation, is flooded more or less systematically, with short heavy floods.

Site E: swampy stagnant bay on a meander of the Balchug Channel, 18 km downstream of the discharge. The channel separates Beryozovy Island from the east bank of the Yenisei. The riverside becomes swampy and accommodates particulate matter during low floods or acts as an additional channel for the stream during short high floods.

Sampling procedure and preconditioning of samples

The sampled plants mainly belonged to the genus of sedge (*Carex* L.): mostly *Carex riparia* Curt. and a minor portion of *Carex versicaria* L. It was simple to select multiple areas with similar conditions as sedge grows profusely in the Yenisei floodplain, on the islands, and riverside. Sedge can tolerate long floods, and as flooding recedes, it can restore its above-ground biomass quickly. Furthermore, sedge has a developed root system that regenerates rapidly after root mass has been withdrawn, and thus, it can be sampled each year at very nearly the same place.

Areas of maximum radioactivity at each site were chosen with an SRP-68–01 scintillation radiometer to ensure that the plants contained enough radionuclides for analysis. All aerial mass of plants was cut over an area of 2–5 m^2, washed in the river water, and rinsed in distilled water on the site. Then two 20 cm × 20 cm to 40 cm × 40 cm cubes of *substrate*, spaced at 60–70 cm, were cut out from the ground and divided manually into soil and roots with rhizosphere. The rhizosphere samples, defined as the soil that clings to roots after being gently shaken (Séguin et al., 2004), were separated from the roots after being dried together. Thus, soil and rhizosphere were collected as bulk samples. Their mineralogy was similar at all sampling sites. The phase composition of soil and rhizosphere consisted of silica and feldspars (plagioclase and K-feldspar). Pelitic minerals, Mg-Fe-chlorite and illite, constituted a sizeable share; there was also a limited amount of amphibole and traces of smectite and siderite.

Since 2011, plants at sites B and C (Atamanovskaya Spit and Atamanovsky Island) were sampled from both the upper and lower ends of the Spit and the Island in order to monitor rapid radioactivity changes. The substrate was sampled in this way from 2015 onwards.

Methods

The specific activity of ^{137}Cs was measured by semiconductor gamma spectrometry, with lead and tungsten shields, on several instruments: a EURISYS

MEASURES EGPC 192-P21/SHF 00-30A-CLF-F coaxial well HPGe detector with a low-background cryostat (France); an AMETEC (ORTEC) GWL-220–15-XLB-AWT well HPGe detector (USA); an IFTP DGDK-100 V coaxial Ge(Li) detector (Dubna, Russia); BDEG-OGK-3 K coaxial HPGe detector (Dubna, Russia). The data were processed using the AnGamma software SPC 'Aspect', (Dubna, Russia) (SPC "Aspect", 2000).

The activity of ^{137}Cs was detected and estimated according to the gamma-ray line $E\gamma = 661.66$ keV (quantum efficiency 0.852), using 250 cm^2 Marinelli beakers for better performance. Measurements, applied to 160–360 g of aliquots homogenised by stirring, lasted from 1 to 24 h depending on ^{137}Cs activity level, to provide a photo peak area accuracy to within 5–10%. The detection limit of ^{137}Cs at this geometry was from 2 to 5 Bq kg^{-1}. Samples of plant ash and low-activity soil and rhizosphere samples (≤ 10 cm^3) were measured on well HPGe detectors in a low-background mode providing 0.01 Bq sensitivity (that corresponded to the measurement 5 g sample at 84,000 s).

The ^{90}Sr specific activity was determined indirectly from the measured radiation of its ^{90}Y progeny in secular equilibrium (VIMS, 2003). The sizes of aliquots were 250–400 g for plants and 50–100 g for their substrate. The substrate samples were annealed at 600 °C in a muffle furnace, while the plant samples were ignited by incremental heating to 480 °C at 50 °C temperature steps, with 30 min exposure at each step. ^{90}Y was obtained from samples with stable strontium dissolved in 6 M HCl and stable yttrium added to the solutions cleaned from all beta emitters that could interfere with ^{90}Sr measurements. The two isotopes attained equilibrium in 14 days corresponding to five 2.67-day half-life periods, to an accuracy no worse than 3%. The chemical output of the Sr carrier (60–95%) was determined by atomic absorption after Sr separation. The radiochemical grade of the ^{90}Y progeny was checked according to repeated measurements after a five-half-life cycle. The ^{90}Y specific activity was measured on a RUB-01P beta radiometer with a BDGB-06P low-background detector in a room with low cosmic and natural background, with additional lead and tungsten shielding. The detection limit of ^{90}Sr was 0.1 Bq kg^{-1}, at a total uncertainty within 20%.

Results and discussion

Soil and rhizosphere were not sampled during 2011, 2013, and 2014 as the plant samples collected in 2011 and 2013 showed decreasing specific activities of ^{137}Cs and ^{90}Sr, despite the effects of the Fukushima nuclear accident felt both globally and in the Yenisei floodplain (Bolsunovsky & Dementyev, 2011; Steinhauser et al., 2014). Soil and rhizosphere monitoring resumed in 2015 and 2016 after markedly higher concentrations of radionuclides were measured in the plants' samples of 2014 (no soil and rhizosphere samples were taken that year due to logistical difficulties).

^{137}Cs and ^{90}Sr in soil and rhizosphere

Soil and rhizosphere (substrate) ^{137}Cs and ^{90}Sr showed time-dependent variations, albeit with long time gaps (Table 1). The substrate samples contained relatively high ^{137}Cs at sites most proximal to the MCC. The content decreased downstream away from the combine and with time but increased abruptly in the Balchug Channel due to favourable conditions for silt deposition (Korobova et al., 2014; Linnik et al., 2004a, 2004b, 2014) and a large amount of organic matter in the soil.

The partitioning of ^{137}Cs between soil and rhizosphere varied both in space and in time. However, the respective pattern in the Balchug Channel was different, possibly, due to high percentages of organic carbon at the site: samples of 2004 from the Atamanovskaya Spit contained 0.82 wt. % C_{org} in soil and 0.66 wt. % in the rhizosphere, while the respective values for the Balchug Channel were 0.59 and 2.99 wt. % (C_{org} was analysed at the Institute of Soil Science, Novosibirsk). As mentioned above, the organic matter of the rhizosphere accumulates most of the radiocaesium and creates favourable conditions for the formation of its mobile and/or bioavailable species. In 2004, at the beginning of the monitoring project, soil ^{137}Cs at the Balchug site was twice the rhizosphere value (1270 ± 130 Bq kg^{-1} against 620 ± 60 Bq kg^{-1}). The difference may result from plant uptake of bioavailable ^{137}Cs from the rhizosphere or, more likely, secondary loss and redistribution of mobile species in soil. In the subsequent years, substrate ^{137}Cs at the site became 2–6 times lower. The difference between soil and rhizosphere persisted but reduced to 410 ± 40

Table 1. ^{137}Cs and ^{90}Sr specific activities in bulk soil and rhizosphere (Bq kg^{-1} dry weight, with measurement error)

			2004	2015	2016
			^{137}Cs		
Shivera village, background		Bulk soil	3.1 ± 0.3	2.0 ± 0.2	1.0 ± 0.1
		Rhizosphere	13 ± 1	5.0 ± 0.5	1.0 ± 0.1
Atamanovskaya spit	Head	Bulk soil	340 ± 34	400 ± 40	110 ± 10
		Rhizosphere	330 ± 33	310 ± 30	110 ± 10
	Downstream end	Bulk soil	-	-	910 ± 90
		Rhizosphere	-	510 ± 50	600 ± 60
Atamanovsky island	Head	Bulk soil	210 ± 20	130 ± 10	140 ± 15
		Rhizosphere	260 ± 26	120 ± 10	180 ± 20
	Downstream end	Bulk soil	-	-	29 ± 30
		Rhizosphere	-	300 ± 30	290 ± 30
Beryozovy island		Bulk soil	190 ± 20	110 ± 10	130 ± 10
		Rhizosphere	220 ± 20	70 ± 7	110 ± 10
Balchug channel		Bulk soil	1270 ± 130	410 ± 40	200 ± 20
		Rhizosphere	620 ± 60	300 ± 30	300 ± 30
			^{90}Sr		
Shivera village, background		Bulk soil	2.8 ± 0.6	16 ± 3	6.5 ± 1.3
		Rhizosphere	4.5 ± 0.9	1.0 ± 0.2	3.5 ± 0.7
Atamanovskaya spit	Head	Bulk soil	13 ± 3	80 ± 16	46 ± 9
		Rhizosphere	21 ± 4	40 ± 8	66 ± 13
	Downstream end	Bulk soil	-	-	84 ± 17
		Rhizosphere	-	55 ± 11	94 ± 19
Atamanovsky island	Head	Bulk soil	6.3 ± 1.3	8.7 ± 1.7	17 ± 3
		Rhizosphere	24 ± 5	3.2 ± 0.6	24 ± 5
	Downstream End	Bulk soil	-	-	11 ± 2.2
		Rhizosphere	-	13 ± 2.6	12 ± 2.4
Beryozovy island		Bulk soil	4.2 ± 0.8	12 ± 2	11 ± 2
		Rhizosphere	3.5 ± 0.7	5.0 ± 1.0	8.5 ± 1.7
Balchug channel		Bulk soil	4.1 ± 0.8	4.8 ± 1.0	18 ± 4
		Rhizosphere	3.9 ± 0.8	4.7 ± 0.9	15 ± 3

'-' is not determined

against 300 ± 30 Bq kg^{-1}, respectively, in 2015 and became even reverse in 2016: 200 ± 20 Bq kg^{-1} in soil against 300 ± 30 in the rhizosphere. The reversal may be due to changes in the way of ^{137}Cs transport to the ecosystem or in the water level that affected the delayed soil run-off. The background ^{137}Cs contents at the reference site likewise showed a descending trend, especially in the rhizosphere where they decreased from 13 ± 1 Bq kg^{-1} in 2004 to 1.0 ± 0.1 Bq kg^{-1} in 2016.

Substrate ^{90}Sr had quite complex space and time patterns. According to 2004 data, its specific activity likewise decreased away from the MCC, but no increase was observed in the Balchug Channel. The reason is that radiostrontium mainly migrates in the dissolved form (Sysoeva et al., 2005), unlike ^{137}Cs, which is adsorbed on suspended particles (Nosov et al., 1993) and stays at accumulations of fine particulate matter. The complex behaviour of mobile ^{90}Sr appears in changes of its rhizosphere–soil difference: 2–4 times higher specific activity in rhizosphere near the MCC and almost equal values far from the combine. In 2015, the specific activity of substrate ^{90}Sr increased both in the reference sample and in samples from the Atamanavoskaya Spit sites and Beryozovy Island. The relatively high ^{90}Sr specific activity at

the reference site indicates atmospheric fallout influents. The greater ^{90}Sr specific activity in soil relative to the rhizosphere value at the reference site can be explained as an effect of the Fukushima event. However, to remain in the soil, atmospheric fallout should be fixed tightly on airborne particles. This idea is inconsistent with a lower percentage of aerosol ^{90}Sr than that of insoluble ^{137}Cs: 18% against 20–50% (Moiseev & Ramzaev, 1975), nor with the selective ^{90}Sr increase at other sites, and with the lack of evidence for the atmospheric fallout of ^{137}Cs. Thus, the fallout episodes, if any, were most likely local and singular and were instead caused by the MCC operation where new facilities were installed in 2015.

^{137}Cs and ^{90}Sr in plants

Time-dependent ^{137}Cs and ^{90}Sr variations in the plants' aerial parts were monitored during 2004, 2011, and 2013–2016, which provided a more complete idea of radioactive pollution in the area. In 2004, the ^{137}Cs in plants from the flooded zones of islands and the Yenisei west bank revealed the same trend decreasing away from the MCC as for substrate (Table 2). However, no ^{137}Cs anomaly was observed at the Balchug site, likely because mobile caesium moved from the rhizosphere, but no plant uptake occurred. This pattern persisted for the whole period of observation.

The situation generally remained the same at almost all sites when monitoring resumed in 2011. The ^{137}Cs activity decreased at sites near the MCC in 2013 but changed little at remote sites. In 2014, ^{137}Cs returned to nearly the previous values at sites proximal to the MCC, but the decreasing trend continued far from the combine facility. Further, ^{137}Cs decrease was observed in 2015 at proximal sites but was especially prominent at distal sites and in the Balchug Channel, where the specific activity became comparable with the background (2.4 ± 0.2 and 1.2 ± 0.1 Bq kg^{-1}, respectively). However, in 2016, all sites showed an increase while the background ^{137}Cs remained stable.

The ^{90}Sr variations over the monitoring period share some similarity with the ^{137}Cs patterns but also have some features of difference. In the same way that ^{137}Cs, the specific activity of ^{90}Sr generally decreased away from the MCC, but the time-dependent variations were different; the ^{90}Sr values were lower during 2011 and 2013, were higher during 2014 and 2015, and fell to the background almost everywhere in 2016. Note that the ^{90}Sr background increased in 2013 to 19 ± 4 Bq kg^{-1}. In the absence of substrate ^{90}Sr data for that year, any explanation for the increase can only

Table 2. ^{137}Cs and ^{90}Sr specific activities in aerial parts of sedge (Bq kg^{-1} dry weight, with measurement error)

		2004	2011	2013	2014	2015	2016
		^{137}Cs					
Shivera village, background		0.8 ± 0.1	0.5 ± 0.05	0.4 ± 0.04	1.0 ± 0.1	1.2 ± 0.1	0.9 ± 0.1
Atamanovskaya spit	Head	240 ± 25	45 ± 5	40 ± 4	60 ± 6	60 ± 6	100 ± 10
	Downstream end	-	240 ± 25	30 ± 3	170 ± 17	120 ± 12	310 ± 30
Atamanovsky island	Head	160 ± 15	180 ± 20	30 ± 3	130 ± 10	90 ± 9	200 ± 20
	Downstream end	-	-	20 ± 2	170 ± 17	30 ± 3	85 ± 9
Beryozovy island		80 ± 8	70 ± 7	50 ± 5	45 ± 5	25 ± 3	65 ± 7
Balchug channel		35 ± 4	55 ± 6	40 ± 4	15 ± 2	2.4 ± 0.2	15 ± 2
		^{90}Sr					
Shivera village, background		2.0 ± 0.4	0.9 ± 0.2	19 ± 4	3.3 ± 0.7	7.0 ± 1.4	1.7 ± 0.3
Atamanovskaya spit	Head	26 ± 5	64 ± 13	17 ± 3	8.9 ± 1.8	19 ± 4	1.7 ± 0.3
	Downstream end	-	26 ± 5	33 ± 7	10 ± 2	74 ± 15	5.5 ± 1.1
Atamanovsky island	Head	12 ± 2	5.5 ± 1.1	26 ± 5	16 ± 3	11 ± 2	1.3 ± 0.3
	Downstream end	-	-	38 ± 8	23 ± 5	20 ± 4	1.5 ± 0.3
Beryozovy island		8.5 ± 1.7	0.9 ± 0.2	8.7 ± 1.7	7.9 ± 1.6	3.6 ± 0.7	0.4 ± 0.1
Balchug channel		1.9 ± 0.4	0.6 ± 0.1	16 ± 3	3.0 ± 0.6	7.1 ± 1.4	1.2 ± 0.2

'-' is not determined. Specific activities below 2 Bq kg^{-1} dry weight are calculated from the specific activities of plant ash

be hypothetical. One cause might lie with the global fallout from the Fukushima nuclear accident, but the ^{90}Sr specific activity in plant samples decreased at all sites instead of increasing. Note that the ^{137}Cs background remained at the same level (0.5 Bq kg^{-1}) in 2013, though much of the isotope was released during the Fukushima disaster (UNSCEAR, 2015). The difference may be due to the different sorption capacities of aerosols and substrates with respect to the two radionuclides: up to 95% of ^{137}Cs is adsorbed on soil (Moiseev & Ramzaev, 1975) while 70–90% of ^{90}Sr exists in the exchangeable form (Sysoeva et al., 2005). On the other hand, the bioavailability of the radionuclides (Burger & Lichtscheidl, 2019; Sysoeva et al., 2005) is different as the release of ^{90}Sr from soil is two or three orders of magnitude greater than for ^{137}Cs (Legin et al., 2008).

The ^{90}Sr increase of 2015, when the monitoring of substrate ^{137}Cs and ^{90}Sr resumed, is worth special consideration. The specific activity of ^{90}Sr in plants was higher near the MCC, including at the reference site with a background of 7.0 ± 1.4 Bq kg^{-1} (though lower than in 2013). On the other hand, the ^{90}Sr specific activity in plants sampled far from the MCC was below or at the background in 2015: 3.6 ± 0.7 Bq kg^{-1} on Beryozovy Island and 7.1 ± 1.4 Bq kg^{-1} in the Balchug Channel. Note that soil and rhizosphere ^{90}Sr values differed more strongly on Beryozovy Island than at the Balchug site in 2015: 12 ± 2 and 5.0 ± 1.0 Bq kg^{-1} against 4.8 ± 1.0 and 4.7 ± 0.9 Bq kg^{-1}, respectively. This provides some evidence of the local spread of the pollution: the amount of dust aerosol that can carry ^{90}Sr was insufficient at remote sites to allow radiostrontium to get into the soil and pass to rhizosphere or plants (case of Beryozovy), or the isotope failed to reach the substrate and remained only on the plant surface (case of Balchug).

Relative ^{137}Cs and ^{90}Sr in the system 'soil – rhizosphere – plants'

The data of 2004, 2015, and 2016 were used to plot specific activity diagrams of the two radionuclides for the 'soil – rhizosphere – plant' system (Fig. 2). The soil/rhizosphere ^{137}Cs specific activity ratios demonstrated positive correlation at the Pearson coefficient r = 0.95–0.99 over the whole period of observation, whereas the respective ratio of plant/soil varied markedly from year to year (R^2 = 0.0043–0.6496), with negative, or near zero, correlation that reached r = 0.81 in 2016 only. The plant/rhizosphere ^{137}Cs ratios likewise vary with time though not as markedly as in sedge and soil (R^2 = 0.0920–0.5255); the correlation was almost absent in 2004 (r = 0.13) but increased to r = 0.562015 in 2015 and to r = 0.72 in 2016. The soil and rhizosphere ^{90}Sr specific activities correlate

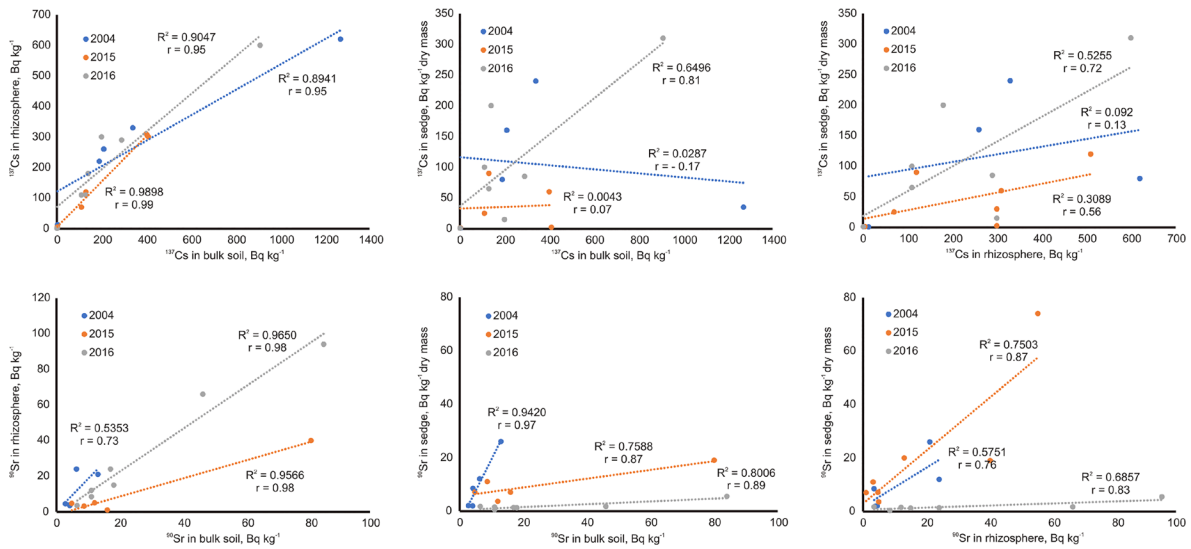

Fig. 2. ^{137}Cs vs. ^{90}Sr correlation in different components of system 'soil-rhizosphere-plants'

quite well (r = 0.73–0.98), the variance being the greatest in 2004 (R^2 = 0.5353) and the smallest in 2015 and 2016 (R^2 = 0.9566 and 0.9650, respectively). On the other hand, ^{90}Sr in sedge and soil correlate better than ^{137}Cs, with moderate variance: r = 0.87–0.97 and R^2 = 0.7588–0.9420; the sedge-rhizosphere correlation is at r = 0.76–0.87 and R^2 = 0.5751–0.7503, respectively.

The behaviour of the two radionuclides in the system changes upon absorption by plants owing to differences in uptake mechanisms. The uptake of ^{137}Cs occurs in the same way as for K, its chemical analogue: transport into the root xylem and stems and leaves, mainly through symplasts via potassium-selective and non-selective ion channels. Conversely, ^{90}Sr, an analogue of Ca, enters mainly via apoplasts and non-selective channels (Burger & Lichtscheidl, 2018, 2019; Ehlken et al., 2002; Roca & Vallejo, 1995; Sokolik et al., 1996). Other dissimilar features may be due to the metabolism of rhizosphere microorganisms which can either immobilise or release the bioavailable forms (Burger & Lichtscheidl, 2018, 2019).

The correlation between ^{137}Cs and ^{90}Sr in the aerial parts of plants was studied (Fig. 3). It was somewhat positive (r = 0.77–0.95) during 2004 and 2014–2016, though at significant variance, especially in later years (R^2 = 0.5855–0.6929). The correlation changes most likely indicate a change in relative influxes of bioavailable ^{137}Cs and ^{90}Sr into the system. In 2011 and 2013, the variance increased (R^2 = 0.0024 and 0.1991, respectively) while the correlation became absent or slightly negative (r = 0.05 and − 0.45, respectively). The ^{137}Cs and ^{90}Sr changes had some other cause from previous years, though the relative specific activities in 2014 approached those of 2004. In general, the revealed patterns do not contradict the hypothesis of local ^{90}Sr influxes.

^{137}Cs and ^{90}Sr in ecosystem components from different parts of islands

The specific activities of ^{137}Cs and ^{90}Sr in ecosystem components sampled in the upstream and downstream ends of the Atamanovskaya Spit and Atamanovsky Island differ markedly (Tables 1 and 2). As determined previously (Korobova et al., 2014, 2016; Linnik et al., 2004a, 2004b, 2005, 2006, 2014), the radionuclides are localised at accumulations of particulate matter carried with water, mainly in the lower ends of islands and in stagnant bays or in places composed of very fine

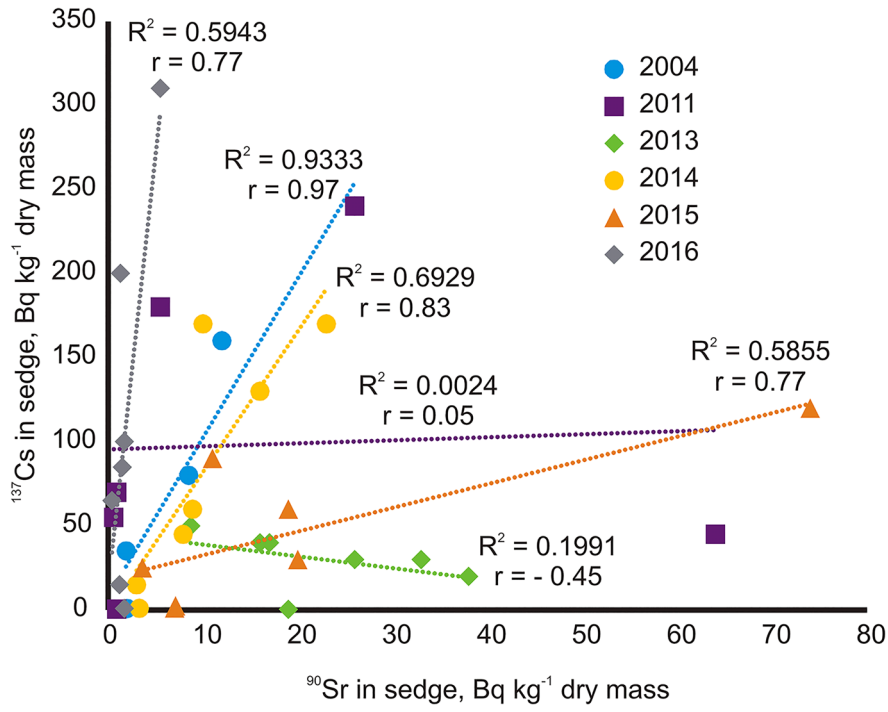

Fig. 3. ^{137}Cs vs. ^{90}Sr correlation in aerial parts of plants in different monitoring campaigns

or fine sand, silt, peat, or light-textured loam. This matches, for instance, the downstream end of the Atamanovskaya Spit located 75–110 m away from the east bank of the Yenisei (108 m away at the upper end and 85 m away at the lower end). Data from 2016, when substrate ^{137}Cs began to increase, show that the amount of ^{137}Cs transported mainly on particulate matter (Nosov, 1996) was much higher in the lower end of the spit than at its head, by factors of 8.8 in soil and 5.5 in the rhizosphere. The respective difference for Atamanovsky Island was smaller (2.1 times for soil and 1.6 times for the rhizosphere). The ^{137}Cs specific activity in ecosystem components in the upper and lower ends of the islands shows the lack of correlation, with $r = -0.17$ and 0.10 for the substrate and plants, respectively (Fig. 4a), which corresponds to the case of transport on suspended particles. Unlike radiocaesium, ^{90}Sr migrates mainly in the dissolved form and is localised preferably at different ends of the spit and the island. On the spit, it is higher in the lower end, whereas in the island, higher values are measured at the head. The difference is by factors of 1.8 for soil and 1.4 for rhizosphere in the spit, and 1.5–2.0 in the island. Note, however, that the setting was reversed in 2015 (though the data are incomplete): ^{90}Sr in rhizosphere sampled in the downstream end of the island was 4.1 times greater than the respective value for the upstream end. Substrate ^{90}Sr values in the upper and lower ends of the islands showed a high correlation at $r = 0.93$, which confirms migration mainly in the dissolved form, but the correlation for plant samples was as low as $r = 0.33$ (Fig. 4b).

Data from the lower end of Beryozovy Island were not considered for several reasons, including the location on the border of the near impact zone of the MCC, the size of the island being much larger than those upstream, complex island geometry and topography due to an abrupt eastward river turn, and complex current patterns around the island (Linnik et al., 2006, 2014).

Excess over background

Excess element content over the background (N) is often used to estimate pollution for different ecosystem components. In our case, the excess was calculated as a ratio of specific activities of the two radionuclides (A) to the respective background value at the reference site (A_{bg}):

$$N = \frac{A\left(\text{Bq kg}^{-1}\text{dry weight}\right)}{A_{bg}\left(\text{Bq kg}^{-1}\text{dry weight}\right)} \quad (1)$$

The specific activity of ^{137}Cs exceeded the background in all components of the 'soil – rhizosphere – plants' system. The excess for the aerial plant organs was the greatest (by 336–433 times) in 2011 and 2016 in the lower end of the Atamanovskaya Spit and the upper end of Atamanovsky Island and remained high in the other years of observations (Table 3). The ^{137}Cs specific activities in soil and rhizosphere (Table 3) were from 14 to 200–300 times greater than the background in different years and at different sites and peaked in 2016 in the lower end of Atamanovskaya Spit (900 and 600 times, respectively).

The excess of ^{90}Sr above the background was much smaller and even negative in some cases. For plants, it was the greatest in 2011 in the upper (74 times) and lower (30 times) ends of Atamanovskaya Spit, reduced to almost zero in 2013, and was within ten times in the subsequent years (Table 4). For soil, a minor excess was observed in 2016 in the lower spit end, while for the rhizosphere, it was the highest in 2015 and relatively high in 2016 (Table 4).

Transfer factor

Plant uptake of radionuclides from the soil is commonly expressed as soil-to-plant transfer factor (TF), normally expressed as a ratio of specific activities in plant tissue and specific activities in soil

$$TF = \frac{A_{plant}(\text{Bq kg}^{-1}\text{dry weight})}{A_{soil}(\text{Bq kg}^{-1}\text{dry weight})} \quad (2)$$

The transfer factor is a key parameter used to predict the effect of radionuclides released in a nuclear fuel cycle (NFC) on the ecosystem and to calculate further the radiation dose the humans receive through the food chain.

The time-dependent TF patterns in the MCC proximal zone differed for ^{137}Cs and ^{90}Sr, which showed increasing and decreasing trends, respectively. The ^{90}Sr TF decreased to 0.036–0.136 at all sites sampled

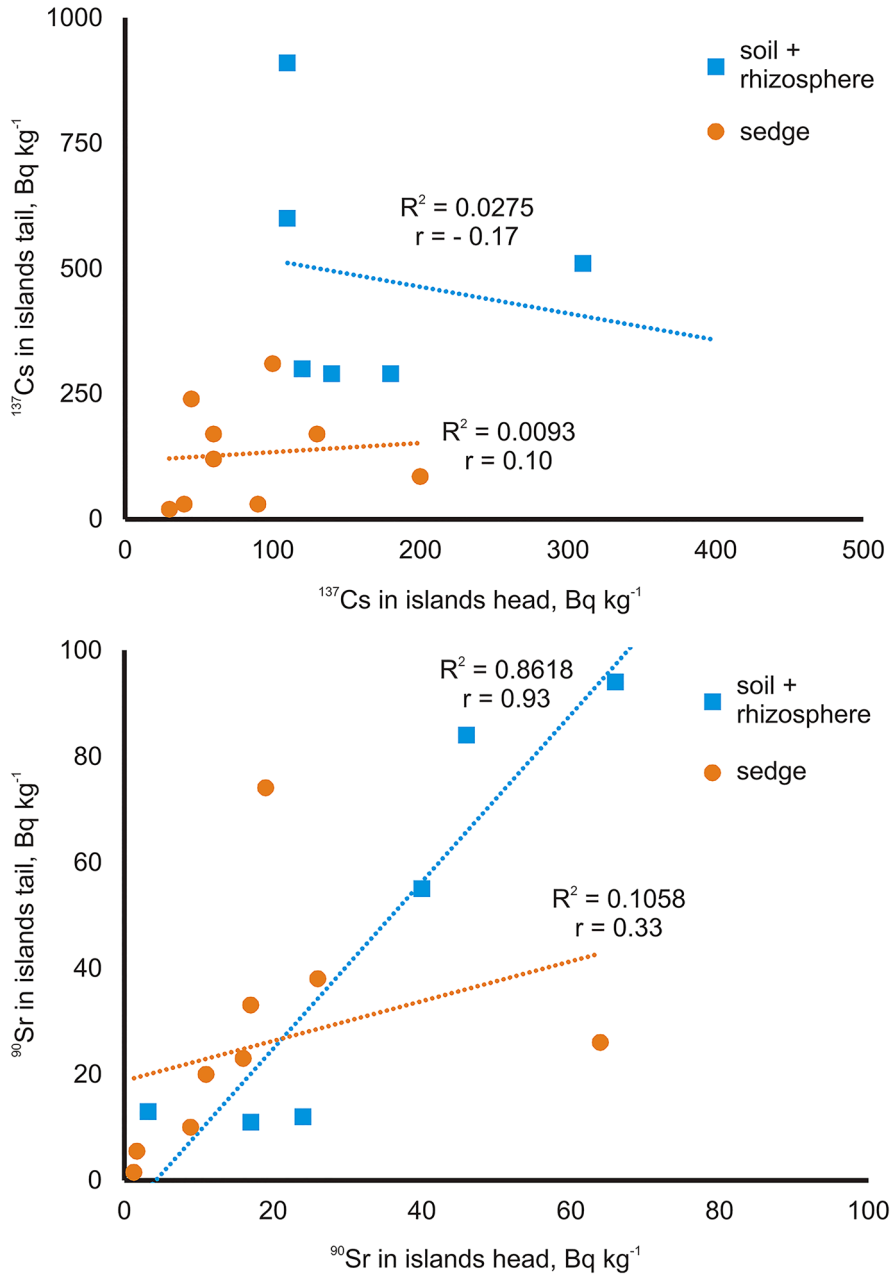

Fig. 4 Correlations of ^{137}Cs and ^{90}Sr in system 'soil-rhizosphere-plants' between upstream and downstream ends of islands

in 2016 (when the background was 0.262) relative to 1.905–2.024 (0.714 background) in 2004 (Table 5). Typically, TF can decrease for several reasons: the absorption volumes change when the nuclides become immobilised in the substrate (ageing effect), isotopes may migrate down the soil profiles to outside the reach of roots (IAEA, 2010), or floods can carry the mobile forms away from the soil which has low sorption capacity (Sysoeva et al., 2005). However, these processes are inconsistent with the fact that the specific activity of substrate ^{90}Sr returned to the level of 2004 in recent years. The TF decrease may indicate that ^{90}Sr became less available for plants because its greater portion was supplied to the system as adsorbed on suspended particles rather than in the dissolved form (Bolsunovsky et al., 1999).

The TF of ^{137}Cs in 2004 and 2015 was notably lower than that of ^{90}Sr and increased from 0.027 to

Table 3. ^{137}Cs excess above background

		2004	2011	2013	2014	2015	2016
Sedge plants							
Atamanovskaya spit	Head	309	79	101	59	50	109
	Downstream end	-	433	85	169	102	336
Atamanovsky island	Head	203	326	79	125	75	221
	Downstream end	-	-	66	166	27	92
Beryozovy island		102	123	147	44	20	71
Balchug channel		43	98	135	12	2	16
Rhizosphere							
Atamanovskaya spit	Head	26	-	-	-	62	106
	Downstream end	-	-	-	-	101	597
Atamanovsky island	Head	20	-	-	-	24	179
	Downstream end	-	-	-	-	59	289
Beryozovy island		17	-	-	-	14	107
Balchug channel		49	-	-	-	59	296
Soil							
Atamanovskaya spit	Head	111	-	-	-	198	109
	Downstream end	-	-	-	-	-	908
Atamanovsky island	Head	68	-	-	-	67	141
	Downstream end	-	-	-	-	-	293
Beryozovy island		61	-	-	-	53	126
Balchug channel		410	-	-	-	207	199

'-' is not determined

Table 4. ^{90}Sr excess above background

		2004	2011	2013	2014	2015	2016
Sedge plants							
Atamanovskaya spit	Head	13	74	0.9	2.7	2.7	1.0
	Downstream end	-	30	1.8	3.1	10	3.2
Atamanovsky island	Head	6.0	6.3	1.4	4.8	1.6	0.8
	Downstream end	-	-	2.1	6.9	2.8	0.9
Beryozovy island		4.3	1.1	0.5	2.4	0.5	0.2
Balchugovskaya channel		1.0	0.7	0.8	0.9	1.0	0.7
Rhizosphere							
Atamanovskaya spit	Head	4.6	-	-	-	40	19
	Downstream end	-	-	-	-	55	27
Atamanovsky island	Head	5.3	-	-	-	3.2	6.9
	Downstream end		-	-	-	13	3.4
Beryozovy island		0.8	-	-	-	5.0	2.4
Balchug channel		0.9	-	-	-	4.7	4.3
Soil							
Atamanovskaya spit	Head	4.7	-	-	-	5.0	7.0
	Downstream end	-	-	-	-	-	13
Atamanovsky island	Head	2.3	-	-	-	0.5	2.6
	Downstream end		-	-	-	-	1.7
Beryozovy island		1.5	-	-	-	0.8	1.7
Balchug channel		1.5	-	-	-	0.3	2.8

'-' is not determined

Table 5. Soil-to-plant transfer factor (TF)

		2004	2015	2016
^{137}Cs				
Shivera village, background		0.254	0.595	0.909
Atamanovskaya spit	Head	0.708	0.149	0.906
	Downstream end	-	-	0.337
Atamanovsky island	Head	0.757	0.673	1.427
	Downstream end	-	-	0.286
Beryozovy island		0.426	0.222	0.514
Balchug channel		0.027	0.006	0.074
^{90}Sr				
Shivera village. background		0.714	0.444	0.262
Atamanovskaya spit	Head	1.970	0.241	0.037
	Downstream end	-	-	0.065
Atamanovsky island	Head	1.905	1.276	0.076
	Downstream end	-	-	0.136
Beryozovy island		2.024	0.300	0.036
Balchug channel		0.463	1.479	0.067

'-' is not determined

0.757, at background values of 0.254 and 0.595, respectively. In 2016, however, it rose above the ^{90}Sr TF at all sites and reached 1.427 (background 0.909). The data of 2016 show the highest ^{137}Cs TF in the upstream parts of the islands, markedly exceeding the values from the lower ends. As noted above, the cause of this difference is that particulate matter, with adsorbed 20–50 % of ^{137}Cs influxes to the river, becomes deposited in the downstream ends of the islands (Nosov et al., 1993; Vakulovsky et al., 1995; Linnik et al., 2006) and is less available for plant uptake (Carver et al., 2007; Ivashkevich & Bondar, 2008). Meanwhile, dissolved ^{137}Cs is more available at the island head.

Furthermore, the TF values were consistently lower in the Balchug Channel than elsewhere, whereas the specific activities of the radionuclides are high in soil but low in plants, possibly, because the soil at the site was rich in highly sorptive organic carbon (see '^{137}Cs and ^{90}Sr in soil and rhizosphere'). However, the ^{90}Sr TF increased to 1.479 in 2015 and exceeded both the background (0.444) and the values from all other sites (0.241–1.276), while the ^{90}Sr specific activities in both substrate and plants were around the background. Data from a water station in Atamanovo Village showed a high water level between late March and late May in 2014 and 2015 and approached the decade maximum in 2014 (Information and Analytical Center of the Register and Cadastre, 2014). The water level in the river was also high in 2016, but rather in January–February and middle-late May, in the periods beyond the sedge spring vegetation season. The high ^{90}Sr TF may result from the water regime providing high soil moisture while ^{90}Sr loss from water-logged soil generally exceeded that of ^{137}Cs, and most of ^{90}Sr, including its bioavailable forms, was carried away with intra-soil run-off (Ovsyannikova et al., 2000; Legin et al., 2008).

Conclusions

The radioactive contamination in the Yenisei floodplain ecosystem in the near impact zone of the Krasnoyarsk MCC has a complex pattern. The specific activity of ^{137}Cs in soil and rhizosphere decreased throughout observations, while ^{90}Sr increased. The river water level fluctuations at some sites (Balchug Channel), especially the time and duration of high stand, may cause a strong influence on the partitioning of isotopes between soil, rhizosphere, and plants (aerial parts). Variations of the soil-to-plant transfer factor (TF) indicate continuing radionuclide fluxes into the system, as well as changes in the shares of bioavailable and fixed species, especially in the case of ^{90}Sr. These changes, including local atmospheric fallouts, apparently controlled the pattern of ^{90}Sr in the 'soil – rhizosphere – plant' system observed

during 2014–2016. The specific activities of ^{137}Cs and ^{90}Sr in different ecosystem components exceeded the background values in all years of monitoring (more strongly for ^{137}Cs), though the specific activity of ^{90}Sr fell below the background in a few cases.

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Author contribution All authors contributed to the study conception and design. Samples collection was performed by Marya Kropacheva, Mikhail Melgunov, and Aleksey Chuguevsky; sample preparation was performed by Marya Kropacheva; analysis was performed by Mikhail Melgunov, Irina Makarova, and Aleksey Chuguevsky. The first draft of the manuscript was written by Marya Kropacheva and Yulia Vosel. All authors read and approved the final manuscript.

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Declarations

Competing interests The authors declare no competing interests.

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