

RADIOACTIVE CONTAMINATION OF THE TECHA RIVER, THE URALS

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Abstract—The Techa River in the Urals was contaminated with high-level radioactive waste from the MAJAK nuclear installation around 1950. The total discharge to the river amounted to 100 PBq with ⁹⁰Sr and ¹³⁷Cs contributing ~10 PBq each. This study has shown that the river presently contains ~0.3 TBq ⁹⁰Sr, >6 TBq ¹³⁷Cs, and ~8 GBq ^{239,240}Pu. The estimates were made for the part of the river starting 50 km from the point of discharge and ending 240 km downstream at the confluence with the Iset River. Radioactivity was measured only in the upper 0.10-m sediments layer. The external dose rates from the contamination range from 0.1–30 μ Gy h⁻¹. The activity concentrations decrease exponentially or by power functions with distance. *Health Phys.* 65(5):481–488; 1993

Key words: ⁹⁰Sr; ¹³⁷Cs; plutonium; water, surface

INTRODUCTION

THE TECHA River belongs to the Iset-Tobol-Irtysh-Ob river system. In the upper reaches it passes the MAJAK nuclear installation. In 1949–1951, medium- and high-level liquid radioactive waste was released into the open river system 6 km from the Techa source. During that period, 76×10^6 m³ of liquid radioactive waste was discharged, with a total activity of 100 PBq (Academy of Science 1991). Most of the activity (~95%) was discharged in 1950 and 1951. ⁹⁰Sr and ¹³⁷Cs contributed 11.6% and 12.2%, respectively. Now 42 y later, the environmental contamination from this source has decayed to 4.3 PBq ⁹⁰Sr and 4.6 PBq ¹³⁷Cs.

In September 1951, discharge of wastes was eliminated by introducing a cascade of reservoirs and bypass canals. This permitted the removal of significant radioactivity from the open drainage network (Nikipelov et al. 1990); however, before this was accomplished, a large part of the floodplain and the bottom of the river, especially upstream, were contaminated. According to Nikipelov et al. (1990), the major portion (~99%) of

the radionuclides were deposited upstream from Muslumovo in the river floodland and bed. The Techa was withdrawn from economic use and the population of several villages was evacuated. Presently the Asanov swamps in the upper reaches of the river is the area of permanent contamination of the Techa. Migration of radionuclides from the cascade of reservoirs and canals has been observed. This system contains 7.1 PBq ⁹⁰Sr and ¹³⁷Cs (193 kCi) (Academy of Science 1991).

The Techa River's source is in Lake Irtyash and joins the Iset River at the right bank (243 km long) (Fig. 1). The drainage area is 7,600 km². Main hydrological characteristics are summarized in Table 1 (Hydrometeo 1973). Seasonal flooding occurs in April; maximal spring high flood is 2.2 m near the village of Muslumovo and 4 m near the village of Zatechenskoye. In the winter, the upper stream freezes 4–6 d earlier than the lower stream and usually freezes through to the bottom.

Techa River runs a twisting course through a valley with low slopes and a width up to 2 km. The floodplain is open meadow, locally bushy. The landscape is hilly plains with kolkas of mixed forest. The river bottom near the banks is silt-sandy; the middle of the river is pebbly or sandy.

Upstream from the village of Muslumovo the river passes bogged-up areas. During 1949–1951 these areas accumulated significant quantities of radionuclides.

Direct discharges of radioactive waste to rivers were also practiced in the early days of the U.S. weapon plutonium production at Hanford when substantial quantities of radioactive materials were released to the atmosphere and to the Columbia River; however, the U.S. releases mostly consisted of short-lived radionuclides (Cate et al. 1970).

The purpose of this study is to investigate the contemporary radioecological situation in the Techa River outside the MAJAK complex and to estimate the total amount of the most biologically significant radionuclides (i.e., ¹³⁷Cs, ⁹⁰Sr, and ^{239,240}Pu) in the sediments, water, and biota of the river. In order to estimate the contribution of radionuclides to the Arctic Ocean from the Ob, we plan to continue the research and to study the radioactive trace from the point of discharge along the whole Techa-Iset-Tobol-Irtysh-Ob river system.

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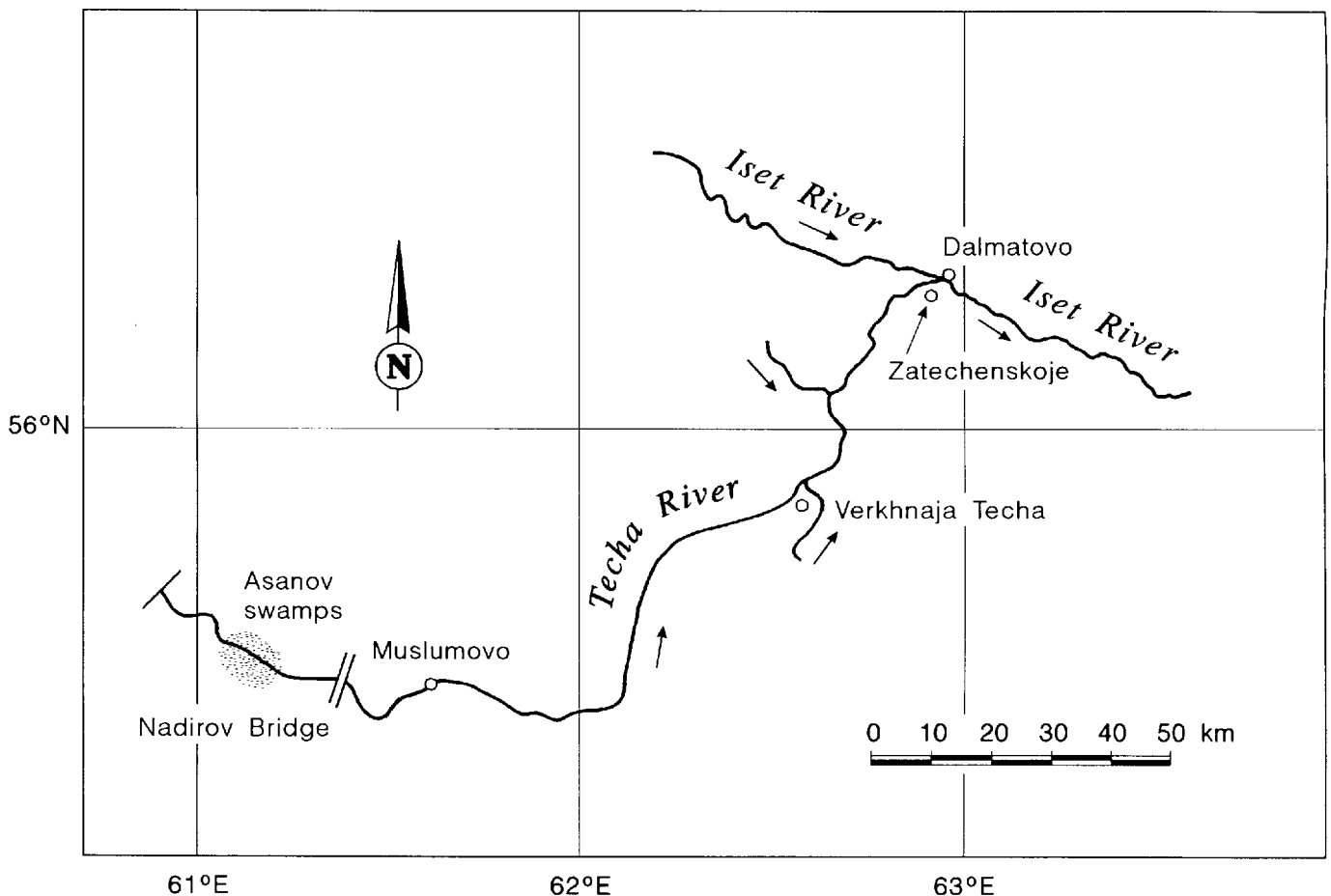


Fig. 1. Techa River.

MATERIALS AND METHODS

Sediments, water, and biota samples were taken in July 1990 from four locations of the Techa River: Nadirov Bridge, Muslumovo, Verkhnjaja Techa, and Zatechenskoje (see Fig. 1) situated 49, 78, 177, and 237 km, respectively, from the point of discharge.

Duplicate water samples were taken at each location. Samples (80–100 L) were filtered through a cotton wool filter used for removing macro admixtures and then evaporated to dryness.

Sediments (1–2 kg) were collected by a dredge which sampled the upper 0–10-cm layer of sediments 3–4 m from the bank. The samples were dried, grinded, and sifted (mesh = 0.1 cm) and aliquots of 100–200 g were gamma measured. These aliquots were used for the radiochemical analysis of ^{90}Sr (50 g) and transuranics (1 g).

Water plants were sampled in the same locations as sediments. The wet weight of each sample was ~2–3 kg. Samples were dried and ashed and 10–20 g ash was used for gamma measurements. Fish was ashed without separation of organs. Gamma background was measured by scintillation detectors.

The samples were analyzed at the Institute of Plant and Animal Ecology (IPAE) for ^{90}Sr (Zvetaeva et al. 1984) and ^{137}Cs and brought to Risø where gamma spectroscopy, plutonium (Talvitie 1971), and americium (Holm et al. 1979) analyses were made. Two sets of ^{137}Cs data were used for an intercomparison between the two laboratories.

The sediment and water inventories in the Techa River may be used for calculating the distribution coefficient (K_d) between water and sediments for the various radionuclides. The inventories were divided by the total amounts of water (4×10^9 kg) and sediments (7×10^8 kg), respectively (calculated from Table 1), in order to get the mean concentrations in Techa sediments and the river water (*cf.* discussion).

RESULTS

River water

The river water (at Pershinskoje) pH = 7.4 and contained 40.3 mg Ca^{++} L^{-1} , 13.2 mg Mg^{++} L^{-1} , 4.3 mg Na^+K^+ L^{-1} , 134.2 mg HCO_3^- L^{-1} , 30.4 mg SO_4^{--} L^{-1} , 16.4 mg Cl^- L^{-1} , 3 mg Fe L^{-1} , 8.8 mg O_2 L^{-1} , and 7.3 mg Si L^{-1} .

The concentrations of ^{90}Sr , ^{137}Cs , and $^{239,240}\text{Pu}$ (Table 2) in the Techa River water decrease with distance (x in km) from the point of discharge according to the exponential model $y = e^{(a+bx)}$, where y is the radionuclide concentration in Bq m^{-3} , as follows:

$$^{90}\text{Sr} \quad a = 8.62 \pm 0.05; \\ b = -(0.0029 \pm 0.0003), \quad (1)$$

$$^{137}\text{Cs} \quad a = 7.03 \pm 0.13; \\ b = -(0.0120 \pm 0.0008), \quad (2)$$

and

$$^{239,240}\text{Pu} \quad a = -(0.38 \pm 0.11); \\ b = -(0.0074 \pm 0.0007). \quad (3)$$

The error term is 1 standard deviation (SD).

The equations show that at the discharge point, the ratio of $^{90}\text{Sr}:^{137}\text{Cs} = 4.9$, and at Zatechenskoje (where the Techa River enters the Iset River), the ratio increased nearly an order of magnitude to 43. The annual water flow at the discharge point is estimated at $2-3 \times 10^7 \text{ m}^3 \text{ y}^{-1}$ and at Zatechenskoje it increased to $3-4 \times 10^8 \text{ m}^3 \text{ y}^{-1}$ (MAJAK 1990). The ^{137}Cs determinations performed by IPAE and Risø were not significantly different.

When we pass from the point of discharge to the end of the river at Zatechenskoje, the amount of ^{90}Sr transported by Techa River increases from $\sim 0.15 \text{ T Bq y}^{-1}$ to $\sim 1 \text{ T Bq y}^{-1}$, while the ^{137}Cs transport seems nearly unchanged ($\sim 0.02 \pm 0.01 \text{ T Bq y}^{-1}$). Thus, there seems to be a net transfer of ^{90}Sr to the river water

during its passage through Techa River. The concentration of ^{60}Co was nearly constant in the Techa River water: 3.2 ± 1.1 ($\pm 1 \text{ SD}$) $\text{Bq } ^{60}\text{Co m}^{-3}$, which also suggests a supply of ^{60}Co .

From Tables 1 and 2, the following expressions for water inventories of radionuclides in the Techa River as a function of distance (x m) were derived as follows:

$$\text{Bq } ^{60}\text{Co} = \int_{49000}^{237000} 9 \times 10^3 x^{0.4} dx \sim 10^7 \text{ Bq}, \quad (4)$$

$$\text{Bq } ^{90}\text{Sr} = \int_{49000}^{237000} 5 \times 10^{10} x^{1.1} dx \sim 2 \times 10^{10} \text{ Bq}, \quad (5)$$

$$\text{Bq } ^{137}\text{Cs} = \int_{49000}^{237000} 4 \times 10^{14} x^{2.1} dx \sim 10^9 \text{ Bq}, \text{ and} \quad (6)$$

$$\text{Bq } ^{239,240}\text{Pu} = \int_{49000}^{237000} 2 \times 10^9 x^{1.7} dx \sim 10^6 \text{ Bq}. \quad (7)$$

Sediments

The sediments in the Techa River contain relatively high ^{137}Cs levels upstream (Table 3), but the levels decrease with increasing distance from the discharge point (see Fig. 2). This is best described by the following power function:

$$y = e^a x^b, \quad (8)$$

where x is the distance (km) from the point of discharge

Table 1. Main hydrological characteristics of the Techa River (Hydrometeo 1973). Water flows are estimated by extrapolation from information given on the flow rate at Muslumovo and Pershinskoje (214 km from the discharge point) (MAJAK 1990). Sediment transport is estimated from information on turbidity of Techa River $\sim 83\text{-g}$ sediments m^{-4} of river water (measured at Pershinskoje).

Location	Distance from point of discharge (km)	Mean width (m)	Mean depth (m)	Water flow ($\text{km}^3 \text{ y}^{-1}$)	Sediment transport (kg y^{-1})
Nadirov Bridge	49	37	2.1	~ 0.02	$\sim 1.66 \times 10^6$
Muslumovo	78	20.5	1.0	0.06	$\sim 5 \times 10^6$
Verknaja Techa	177	25.5	0.5	~ 0.14	$\sim 12 \times 10^6$
Zateschenskoje	237	25.5	1.0	0.35	$\sim 30 \times 10^6$

Table 2. Radionuclides in Techa River water in July 1990 (relative counting error in %).

Location	Radionuclides (Bq m^{-3})						
	^{90}Sr	^{137}Cs		^{60}Co	^{238}Pu	$^{239,240}\text{Pu}$	^{241}Am
		IPAE	Risø				
Nadirov Bridge	5,000 (1)	620 (5)	500 (1)	2.2 (19)	0.0088 (19)	0.52 (6)	0.020 (14)
Muslumovo	4,200 (8)	550 (1)	440 (1)	2.6 (20)	BDL ^a	0.35 (10)	0.012 (15)
Verknaja Techa	3,200 (15)	155 (10)	156 (1)	4.8 (11)	0.0071 (23)	0.174 (7)	lost
Zatechenskoje	2,800 (32)	68 (2)	52 (2)	3.2 (11)	BDL	0.123 (6)	0.005 (29)

^a BDL = below detection limit (i.e., $<0.006 \text{ Bq } ^{238}\text{Pu m}^{-3}$).

Table 3. Radionuclides in Techa River sediments in July 1990 (relative counting error in %).

Location	Type of sediment	Radionuclide (Bq kg ⁻¹ dry weight)						
		¹³⁷ Cs	⁹⁰ Sr	⁴⁰ K (g kg ⁻¹) potassium	⁶⁰ Co	²³⁸ Pu	^{239,240} Pu	²⁴¹ Am
Nadirov Bridge ^a	Peat	280,000 (51)	1,080 (21)	15.3	10.5 (14)	0.50 ± 0.21	42 ± 1	1.6 ± 0.2
	Sand and silt	141,000 (1)	2,050 (48)	9.5	BDL	0.24 (28)	4.7 (8)	0.16 (37)
Musulmovo	Sand	4,000 (16)	1,230 (7)	12.8	BDL	0.51 (18)	43 (6)	1.6 (9)
	Silt	47,000 (12)	670 (28)	12.5	BDL	BDL ^b	0.40 (16)	BDL
Verknaja Techa	Sand	150 (1)	39 (26)	12.5	BDL	BDL	1.04 (16)	0.15 (39)
	Silt	550 (11)	150 (40)	10	BDL	BDL	0.18 (23)	BDL
Zatechenskoje	Sand	34 (6)	22 (9)	13.9	BDL	BDL	0.43 (20)	BDL
	Silt	200 (2)	200 (36)					

^a Double determinations were carried out for transuranics at Nadirov Bridge; the error term is ± 1 SE.

^b BDL = below detection limit (i.e., <1–2 Bq ⁶⁰Co kg⁻¹, <0.03 Bq ^{239,240}Pu kg⁻¹, <0.06 Bq ²³⁸Pu kg⁻¹, <0.1 Bq ²⁴¹Am kg⁻¹).

and y is the radionuclide concentration (Bq kg⁻¹). The exponents a and b are given with 1 SD, as follows:

$$^{90}\text{Sr sand: } a = 23 \pm 2; \quad b = -(3.7 \pm 0.5), \quad (9)$$

$$^{90}\text{Sr silt: } a = 12 \pm 3; \quad b = -(1.2 \pm 0.6), \quad (10)$$

$$^{137}\text{Cs sand: } a = 28 \pm 2; \quad b = -(4.5 \pm 0.4), \quad (11)$$

$$^{137}\text{Cs silt: } a = 33 \pm 1; \quad b = -(5.1 \pm 0.2), \quad (12)$$

$$^{239,240}\text{Pu sand: } a = 14.4 \pm 0.3 \quad b = -(3.0 \pm 0.1), \text{ and } (13)$$

$$^{239,240}\text{Pu silt: } a = 22 \pm 2 \quad b = -(4.2 \pm 0.3). \quad (14)$$

It should be emphasized that these equations are valid only between the points of observations, i.e., within 49–237 km from the point of discharge. The equations predict unrealistically high concentrations close to the point of discharge. The ¹³⁷Cs determinations performed by the IPAE and Risø were generally in good agreement, with the exception of the sample from Nadirov Bridge, where we believe that inhomogenities (hot particles) may have been present.

From Tables 1 and 3, the following expressions for sediment inventories of radionuclides as a function of distance (x m) were derived assuming a mean sediment layer of 150 kg m⁻² of riverbed corresponding to a mean sediment layer thickness of 10 cm:

$$\text{Bq } ^{99}\text{Sr} = \int_{49000}^{237000} 8 \times 10^{17} x^{-2.3} dx \sim 3 \times 10^{11}, \quad (15)$$

$$\text{Bq } ^{137}\text{Cs} = \int_{49000}^{237000} 13 \times 10^{31} x^{-4.8} dx \sim 6 \times 10^{12}, \quad (16)$$

and

$$\text{Bq } ^{239,240}\text{Pu} = \int_{49000}^{237000} 10^{24} x^{-3.9} dx \sim 8 \times 10^9. \quad (17)$$

Biota

The ¹³⁷Cs concentrations in *Ceratophyllum demersum* (Table 4) decreased with distance x (km) from the discharge point according to the power function $y = e^a x^b$, where y is Bq ¹³⁷Cs kg⁻¹ dry weight, $a = 20 \pm 1$, and $b = -(2.9 \pm 0.2)$ (see Fig. 3).

Other radionuclides in *Ceratophyllum* (⁶⁰Co, plutonium, and Americium) also showed a decrease with distance, but ⁹⁰Sr did not. A single fish sample contained a surprisingly low ¹³⁷Cs level (*cf.* the following discussion).

The ¹³⁷Cs results on biota obtained by the IPAE and Risø, were not significantly different.

Activity ratios

If we compare the ratios between transuranic isotopes in the Techa River with those found in U.S.-produced plutonium, we would find some differences. The ²³⁸Pu:^{239,240}Pu ratio was 0.012 in the two most contaminated Techa sediment samples from Nadirov Bridge and Muslumovo but was 0.12–0.25 in Savannah

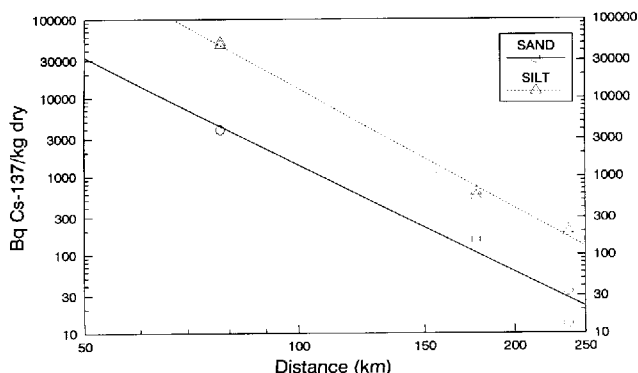


Fig. 2. ¹³⁷Cs in sediment samples collected from the Techa River in July 1990. The concentrations are related to the distance from the outlet from MAJAK to the river.

River sediments. ²⁴¹Am; ^{239,240}Pu was 0.038 in the two samples, but 0.15–0.22 at Savannah (Alberts et al. 1986).

In 1968 we observed a ratio of 0.019 (Aarkrog 1971) in weapons-grade plutonium in sediments at Thule, Greenland. The mean ratio between ^{239,240}Pu and ¹³⁷Cs in Techa sediments was $(3.4 \pm 1.7) \times 10^{-3}$ [± 1 standard error (SE), $N = 7$], and in water, we found $(1.3 \pm 0.35) \times 10^{-3}$ (± 1 SE, $N = 4$). The Great Lakes in the U.S. show a water ratio of $(12 \pm 1) \times 10^{-3}$ (± 1 SE, $N = 5$) (NRCC 1983). The expected ^{239,240}Pu:¹³⁷Cs in power reactor fuel elements is 9×10^{-3} (Belyayev et al. 1991). Hence, the Techa contamination showed lower ratios than observed in global fallout and in power reactors.

DISCUSSION

Concentrations

The radionuclide concentrations in the Techa River water may be compared with those in the Columbia River in the U.S., which have been polluted from the plutonium production at Hanford (Eisenbud 1987). The downstream contributions in 1984 from Hanford were 0.2 Bq ¹³⁷Cs m⁻³, 1 Bq ⁹⁰Sr m⁻³, and 0.3 Bq ⁶⁰Co m⁻³ (i.e., ¹³⁷Cs and ⁹⁰Sr concentrations were 2 to 3 orders of magnitude higher in the Techa River water, and ⁶⁰Co was 1 order higher). Compared with global fallout concentrations in rivers at northern temperate latitudes, ⁹⁰Sr and ¹³⁷Cs in Techa River water were also 2 to 3 orders of magnitude higher (Aarkrog et al. 1991).

The Techa River sediments may be compared with Savannah River sediments, which have been contaminated from the plutonium production at the Savannah River Plant (SRP). Alberts et al. (1986) measured 0.01–0.13 Bq ^{239,240}Pu kg⁻¹ sediments in Savannah River outside the SRP. These levels were 1 to 2 orders of magnitude lower than those in Techa sediments at Nadirov Bridge. The Savannah River samples were collected closer to the plant site than the Techa samples. Compared with global fallout levels in sediments, the

Table 4. Radionuclides in biota collected in the Techa River in July 1990. (Relative counting error in %; otherwise, the error term is 1 SE of double or triple determinations.)

Location	Species	⁹⁰ Sr	¹³⁷ Cs		⁴⁰ K (g kg ⁻¹ potassium)	⁶⁰ Co	²³⁸ Pu	^{239,240} Pu	²⁴¹ Am
			IPAE	Riso					
Nadirov Bridge	Ceratophyllum demersum	1,200 ± 1,100	9,200 ± 2,500	10,600 (1)	56 (9)	18.1 (23)	0.128 (14)	14.6 (9)	0.84 (16)
	Thyphalatifolia	850 ± 510	350 ± 220						
	Carassius (fresh weight)	340 (5)	26 (5)	8.7 (2)	0.87 (5)	0.23 (23)	BDL ^a	0.026 (12)	BDL
Mustumovo	Ceratophyllum demersum	420 ± 20	1,640 ± 330	1,400 (1)	49 (2)	9.6 (13)	0.044 (22)	3.8 (6)	0.29 (20)
	Elodea canadensis	9,100 ± 3,900	3,500 ± 40						
Verknaja Techa	Potamogeton pectinatus	2,400 ± 590	880 ± 70	199 (1)	48 (2)	9.5 (12)	BDL	0.34 (9)	BDL
	Ceratophyllum demersum	2,900 ± 420	200 ± 15						
Zatechenskoje	Elodea canadensis	1,840 (5)	190 (5)	142 (2)	59 (2)	4.8 (26)	BDL	0.36 (11)	ND ^b
	Ceratophyllum demersum	1,650 ± 630	62 ± 38						

^a BDL = below detection limit. Carassius BDL (0.57 kg fresh weight sample): <0.002 Bq ²³⁸Pu kg⁻¹; <0.002 Bq ²⁴¹Am kg⁻¹. Ceratophyllum BDL [100–200 g dry weight (d.w.) sample]: <0.03 Bq ²³⁸Pu kg d.w.; <0.03 Bq ²⁴¹Am kg d.w.
^b ND = no determinations.

Table 5. Observed radionuclide concentration ratios in Techa River samples compared with other observations.^a

Radionuclide	References	Sediment (dry)	Plant (fresh)	Fish (fresh)
		water	water	water
⁶⁰ Co	This work	5 × 10 ³	8 × 10 ²	10 ²
	Rhine (Bayer 1982)	1 × 10 ³	—	—
	Danube (IAEA 1976)	(0.5–12.5) × 10 ³	—	—
	Thompson et al. (1972)	—	2 × 10 ²	50
	Kulikov et al. (1988)	(1–80) × 10 ³	(1–3) × 10 ³	60
⁹⁰ Sr	This work	10 ²	2 × 10 ²	70
	Rhine (Bayer 1982)	10	—	—
	Danube (IAEA 1976)	25–75	—	20–3 × 10 ²
	Thompson et al. (1972)	—	5 × 10 ²	30
	Kulikov et al. (1988)	75–500	3 × 10 ²	(1–5) × 10 ²
¹³⁷ Cs	This work	3 × 10 ⁴	1 × 10 ³	30
	Rhine (Bayer 1982)	2 × 10 ³	—	—
	Danube (IAEA 1976)	(1–28) × 10 ³	—	(2–5) × 10 ³
	Thompson et al. (1972)	—	5 × 10 ²	2 × 10 ³
	Kulikov et al. (1988)	2–4 × 10 ³	9 × 10 ²	(1–14) × 10 ²
Plutonium	This work	5 × 10 ⁴	2 × 10 ³	50
	Thompson et al. (1972)	—	3.3 × 10 ²	3.5

^a The dry matter content of river plants is 20%.

Table 6. Dose rates in $\mu\text{Gy h}^{-1}$ along the Techa River.

Location	Water surface	Bottom	Riverbank
Nadirov Bridge	5	28–30	0.90–24
Muslumovo	6.5–16	0.20–15	0.60–28
Verknaja Techa	0.19	0.13	0.15
Zatechenskoje	0.10	0.1	0.2

1940's and early 1950's, the Techa River contains enhanced levels of ⁹⁰Sr, ¹³⁷Cs, ^{239,240}Pu, ²⁴¹Am, and ⁶⁰Co. The levels upstream are several orders of magnitude higher than those expected from global fallout. Water concentrations 240 km downstream from the discharge point are 2 to 3 orders of magnitude higher for ⁹⁰Sr and ¹³⁷Cs than the expected global fallout levels in river water.

This study only accounts for a minor part of the total radionuclide discharges to the Techa River, *viz.* 7×10^{-5} of the ⁹⁰Sr and 1.3×10^{-3} of the ¹³⁷Cs. The activity was preferentially deposited in the sediments. If we include the floodlands and the first 50 km from the discharge point of river sediments, which was not represented in this study, we may have accounted for 5–10% of the total discharge, but this estimate needs further verification. Most of the 4.3 PBq ⁹⁰Sr and 4.6 PBq ¹³⁷Cs discharged from MAJAK are probably found in the cascade of reservoirs and canals within the fence of MAJAK. More than 1 PBq may, however, be present downstream of the Techa River in the Ob river system or even in the Arctic Ocean. The analytical intercomparison between the IEPA and Risø showed that the two institutions produced comparable ¹³⁷Cs data.

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