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Environmental modelling of radioactive contamination of floodplains and sorlakes along the Tеча and Iset rivers

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Abstract

The radioactive contamination of the Tеча–Iset rivers downstream from the MAYAK reservoirs has been determined by measurements of ^{90}Sr , ^{137}Cs and transuranic elements in floodplain soils and sorlake sediments. Empirical models for the spatial distribution of the radioactive contamination in the river system were developed by fitting the data to power and exponential functions. The radionuclide inventories calculated from the models were compared with Russian data. The models applied in this study predicted higher inventories than those obtained from the Russian data in the upper reaches of the Tеча; but lower inventories were predicted for the downstream part of the river. Estimation of the ^{90}Sr inventory is encumbered with larger uncertainties than those of ^{137}Cs and $^{239,240}\text{Pu}$ because the distribution of ^{90}Sr does not always follow simple models as do ^{137}Cs and Pu. The contamination downstream of MAYAK of the floodplain and Tеча–Iset river system was estimated to be 0.1 PBq ^{90}Sr , 0.3 PBq ^{137}Cs and 0.8 TBq $^{239,240}\text{Pu}$. While essentially all the ^{137}Cs and Pu outside the MAYAK reservoir are found in the Tеча river, the ^{90}Sr contamination extends further downstream.
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Keywords: Strontium-90; Caesium-137; Plutonium; Inventories; MAYAK

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1. Introduction

The production of plutonium for Soviet nuclear weapons started in 1948 at the “MAYAK” Production Association located in the Cheliabinsk Region in the South Urals near the towns of Kyshtym and Kasli. Medium-level liquid radioactive waste was discharged into the Techa River at Reservoir R3 (in the period 1949–1956) [See Map. (The map is based on satellite photos. It is derived from the ADC WorldMap® and drawn in MapInfo.) In the period 1949–1951 the total release amounted to 106 PBq of β -emitters including 12 PBq of ^{90}Sr and 13 PBq of ^{137}Cs . Between 1951 and 1956 the discharged activity decreased by a factor of 100 (Joint Norwegian–Russian Expert Group, NREG, 1997). The discharge of α -emitters is less well known, but it has been at least 2 TBq. Joint Russian–Norwegian fieldwork in 1994 suggests that the α -discharges may in fact have been an order of magnitude higher.

In 1951 the discharge of radioactive wastes into the Techa river was practically eliminated by introducing a cascade of reservoirs and bypass canals. Before this was accomplished a large part of the floodplain and the bottom of the river were contaminated. Presently, the Asanov Swamps in the upper reaches of the river are an area of permanent contamination of the Techa.

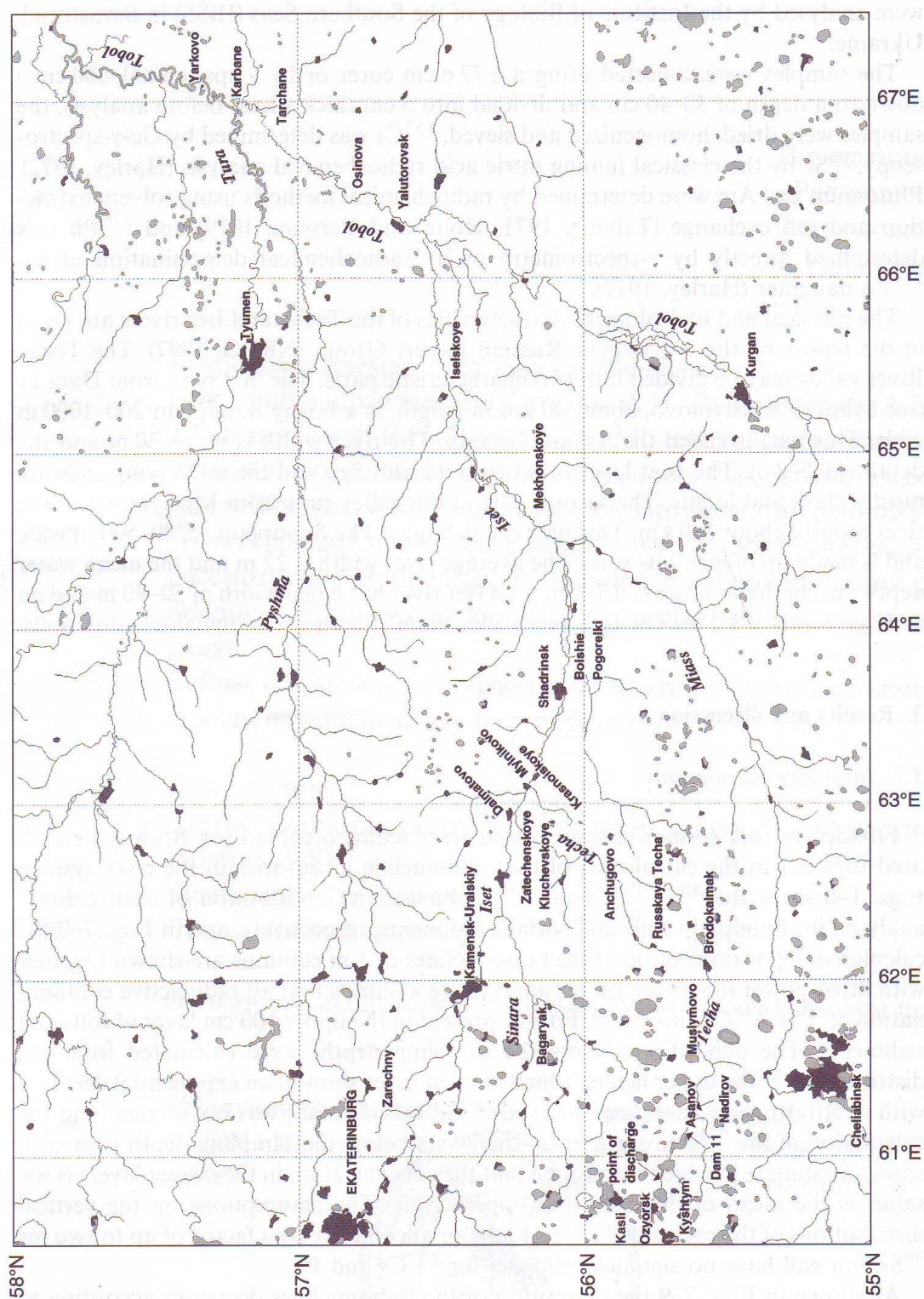
In a joint European–Russian co-operation SUCON (South Ural Contamination) the radioecological impact of the nuclear activities at MAYAK has been studied since 1990 (Aarkrog et al., 1992). The first study of the Techa river in this co-operation was carried out in the summer of 1990 (Trapeznikov et al., 1993). A number of recent Russian studies (Kryshev, Romanov, Sazykina, Isaeva & Blaylock, 1998; Chesnokov, Govorun, Linnik & Shcherbak, 1998; Shcherbak, 1998) have also dealt with the radioactive contamination of the Techa river. Systematic measurements of the radioactive contamination of the Techa river since the early 1950s have been reported (NREG, 1997). These older data do not, however, include information on Pu and other α -emitters.

The aim of the present study has been to estimate today's inventories of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the flooded areas along the Iset and Techa rivers. It has furthermore been our intention to study the spatial distribution of the contamination and thereby to identify the major release years in the sediment columns and to establish the extension of the MAYAK contamination in the river system.

2. Materials and methods

Two sets of samples, floodplain soil and sorlake¹ sediments, were collected (in 1996) by the Institute of Plant and Animal Ecology (IPAE) in Yekaterinburg and analysed by Risø National Laboratory (RISØ) for ^{90}Sr , ^{137}Cs , Pu, Am and excess ^{210}Pb . Most of the samples were also analysed by IPAE for ^{137}Cs and a limited number of samples

¹ A sorlake (also called an oxbow lake) is an annually flooded lake characterized by a low-flow environment and thus expected to preserve the sediment transported there each year (Panteleyev, 1995).



were analysed by the Institute of Biology of the Southern Seas (IBSS) in Sevastopol, Ukraine.

The samples were collected using a 6.77 ϕ cm corer or by a spade (200–600 cm²) down to a depth of 30–40 cm and divided into 5 cm thick slices. Before analysis, the samples were dried, homogenized and sieved. ¹³⁷Cs was determined by Ge- γ -spectroscopy, ⁹⁰Sr by the classical fuming nitric acid radiochemical analysis (Harley, 1972). Plutonium and Am were determined by radiochemical methods using solvent extraction and ion exchange (Talvitie, 1971; Holm and Persson, 1979) and ²¹⁰Pb was determined directly by γ -spectrometry or by radiochemical determination of the ²¹⁰Po daughter (Harley, 1972).

The physical and hydrological characteristics of the Tеча and Iset rivers are given in the report by the Norwegian–Russian Expert Group (NREG, 1997). The Tеча River valley may be divided into two characteristic parts. The first part, from Dam 11 (see Map) to Muslymovo, about 40 km in length, is a boggy floodplain 200–1000 m wide. This part is called the Asanov Swamp. The river width is up to 30 m and the depth is 0.5–2 m. The peat layer is between 0.1 and 3 m and the underlying soils are mostly clays and loams. The second part of the valley runs from Muslymovo to the river mouth, about 160 km. This part is less boggy. The floodplain is 200–500 m wide and is made up of meadow soils. The average river width is 22 m and the mean water depth during the summer is 0.5–1 m. The Iset river has a bed width of 50–70 m and an average depth of 1.2 m. The Iset river valley mainly comprises alluvial-meadow soils.

3. Results and discussion

3.1. Inventory calculations

Floodplain soil, sorlake sediments and river sediments (Nadirov Bridge) were all used together in the calculation of the radionuclide inventories in the river system. Figs. 1–6 show the ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu activity distribution of each column analysed for floodplain soils and sorlake sediments, respectively, and in Figs. 7–9 the calculated depositions of the three radionuclides in 1 m columns are shown together with fitted power functions. In our analysis we assumed that all radioactive contamination of ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu was present in the upper 100 cm layer of soils and sediments. The depositions below the sampling depths were calculated from the distributions in the upper layers, which in most cases showed an exponential decrease with depth. In a few cases, especially for ⁹⁰Sr, this decrease was not evident and the calculation of the ⁹⁰Sr inventories in the layers below the sampling depth is in such cases an estimate in which we assume that the concentration in the deeper layers is the same as the mean of the measured upper layers. The assumptions on the vertical distributions of the radionuclides may add an uncertainty by a factor of up to two for ⁹⁰Sr, but will have no significant impact for ¹³⁷Cs and Pu.

As shown in Figs. 7–9 the deposition of the radionuclides decreases according to a power function ($y = e^ax^b$, see Table 1) with distance from MAYAK. In the case of ⁹⁰Sr the regression was not significant, so for this radionuclide we have used a

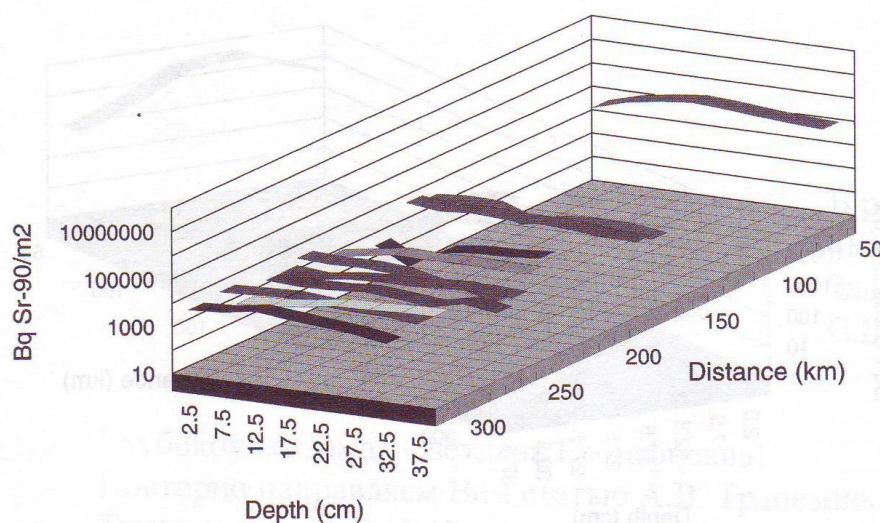


Fig. 1. Vertical and longitudinal distribution of ^{90}Sr (1996) in sorlake and river sediments from the Techa and Iset rivers.

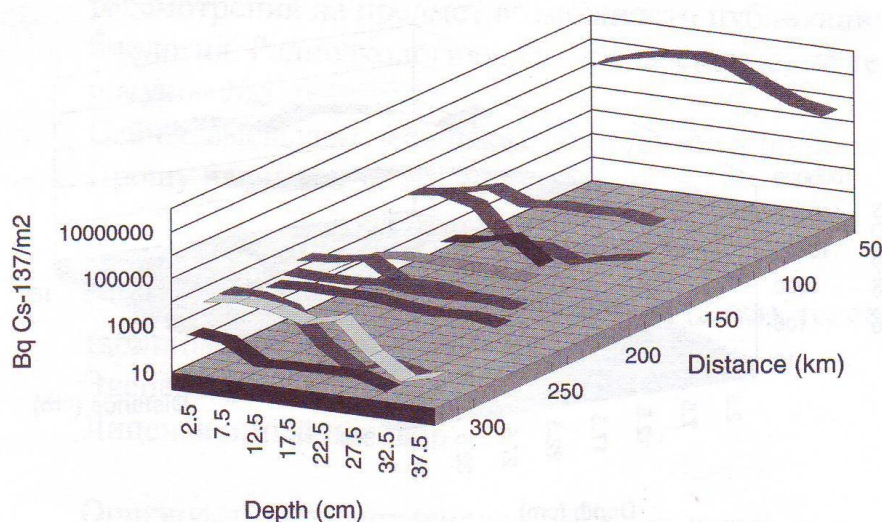


Fig. 2. Vertical and longitudinal distribution of ^{137}Cs (1996) in sorlake and river sediments from the Techa and Iset rivers.

constant deposition density equal to the geometric mean of $\text{kBq } ^{90}\text{Sr m}^{-2}$ in all 1 m columns (179 kBq m^{-2}) between Nadirov Bridge and Bolshie Pogorelki, 49 and 310 km, respectively, from MAYAK. From 310 km and to the outlet of the Ob ($\sim 2800 \text{ km}$ from MAYAK) we assumed that the ^{90}Sr deposit decreased according to the power function shown in Fig. 7 (^{90}Sr), but this assumption is tentative.

The samples used in our inventory calculations were collected in the rivers and floodplain within a belt of approximately 50 m; so we have assumed that the samples

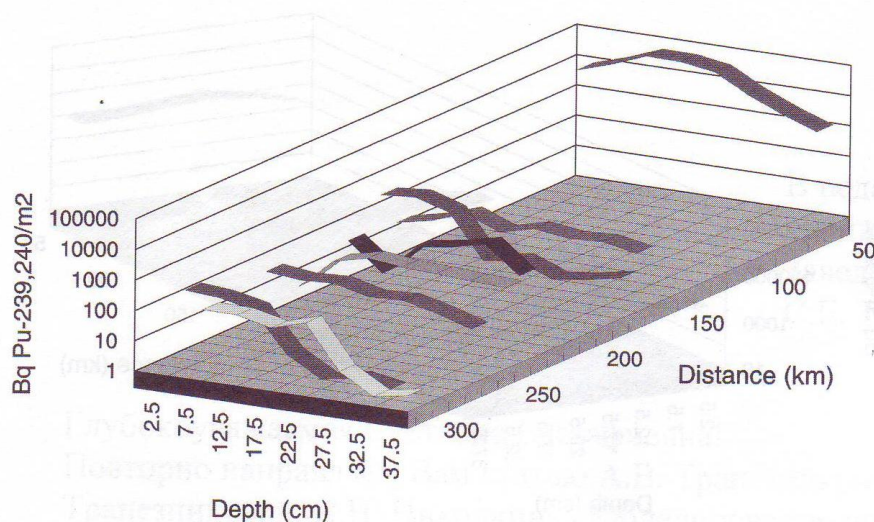


Fig. 3. Vertical and longitudinal distribution of $^{239,240}\text{Pu}$ (1996) in sorlake and river sediments from the Tеча and Iset rivers.

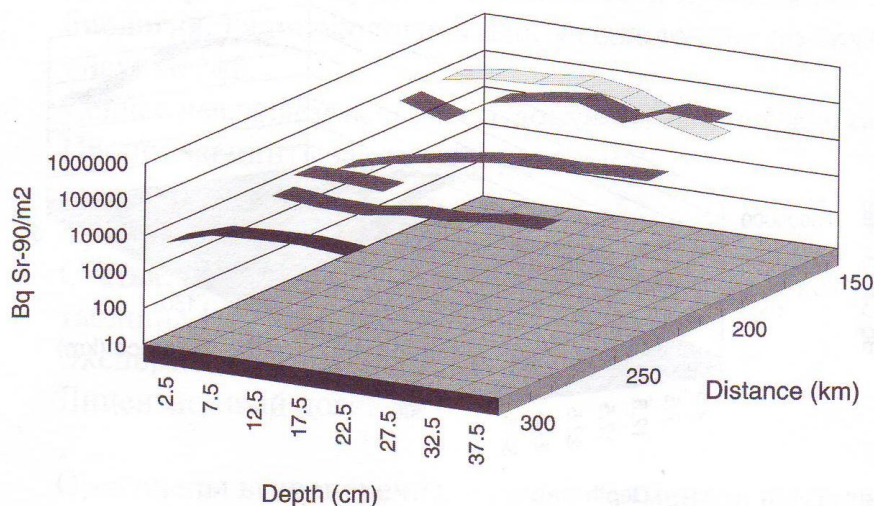


Fig. 4. Vertical and longitudinal distribution of ^{90}Sr in 1996 in floodplain soils along the Tеча and Iset rivers.

represent the contamination to a distance of 25 m on each side of the middle of the riverbed. The contamination extends farther than that but decreases with the distance from the riverbed. The general spatial distribution of radioactive contamination levels in the floodplain has been assessed from gamma radiation dose rates for the Asanov Swamps over a distance of 40 km along the Tеча river (NREG, 1997). The $^{137}\text{Cs}/^{90}\text{Sr}$ ratio in the floodplain was expressed by the power function:

$$^{137}\text{Cs}/^{90}\text{Sr} = 660.7 R^{-1.18}, \quad (1)$$

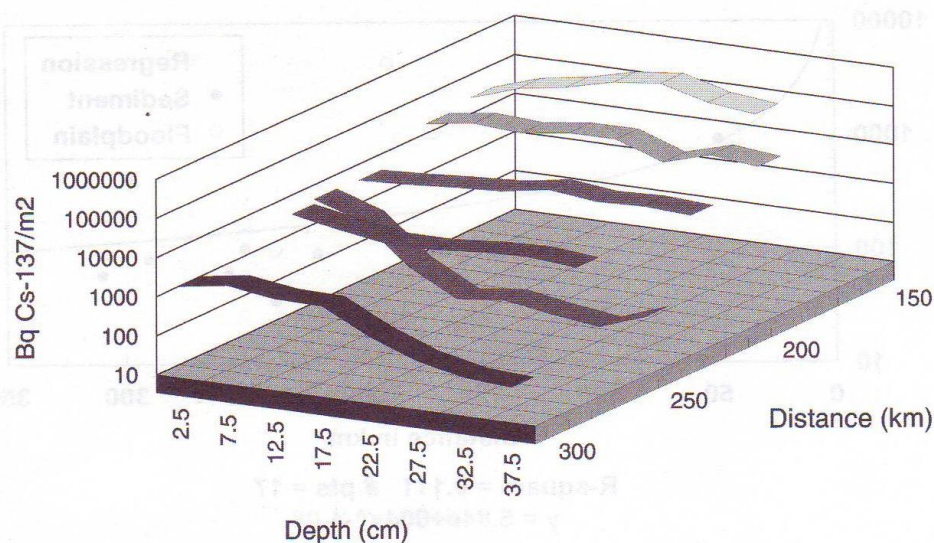


Fig. 5. Vertical and longitudinal distribution of ^{137}Cs in 1996 in floodplain soils along the Tcha and Iset rivers.

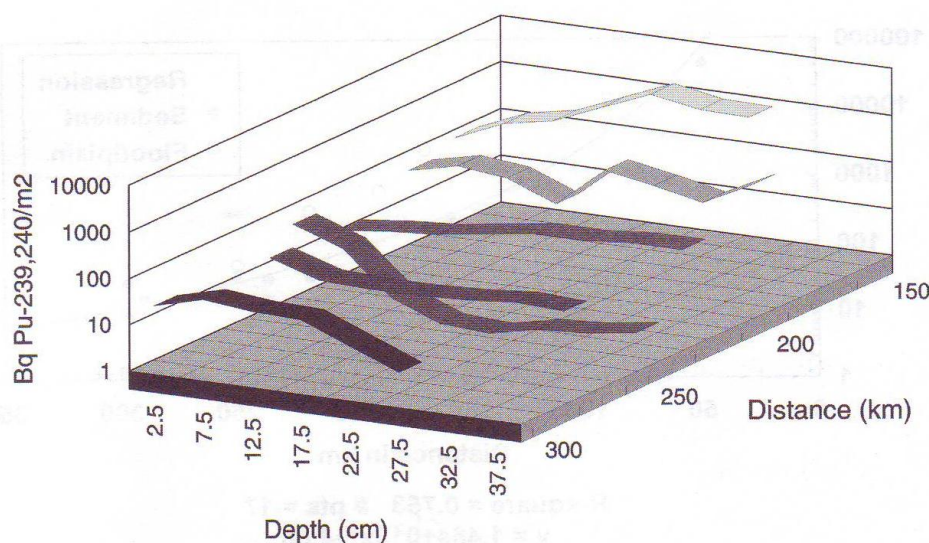


Fig. 6. Vertical and longitudinal distribution of $^{239,240}\text{Pu}$ in 1996 in floodplain soils along the Tcha and Iset rivers.

where R is the distance from the riverbed in meters. From these Russian data we have calculated the following expressions for the horizontal distribution of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ as a function of the distance R from the riverbed:

$$\text{Bq } ^{90}\text{Sr m}^{-2} = A \frac{Be^{-0.0125R}}{660.7R^{-1.18}}, \quad (2)$$

$$\text{Bq } ^{137}\text{Cs m}^{-2} = Be^{-0.0125R}, \quad (3)$$

$$\text{Bq } ^{239,240}\text{Pu m}^{-2} = Ce^{-0.0125R}. \quad (4)$$

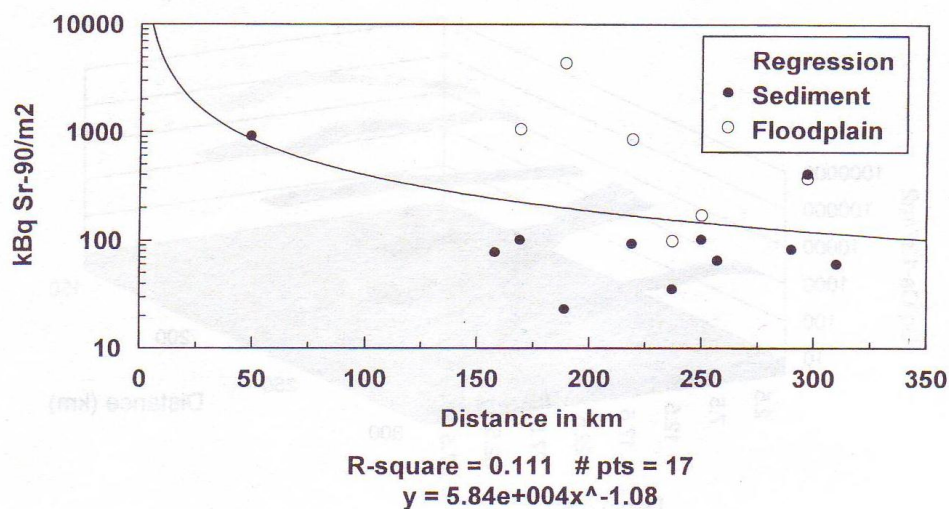


Fig. 7. Cumulative ^{90}Sr in the 0–100 cm sediment and floodplain layer along the Tеча and Iset rivers in 1996.

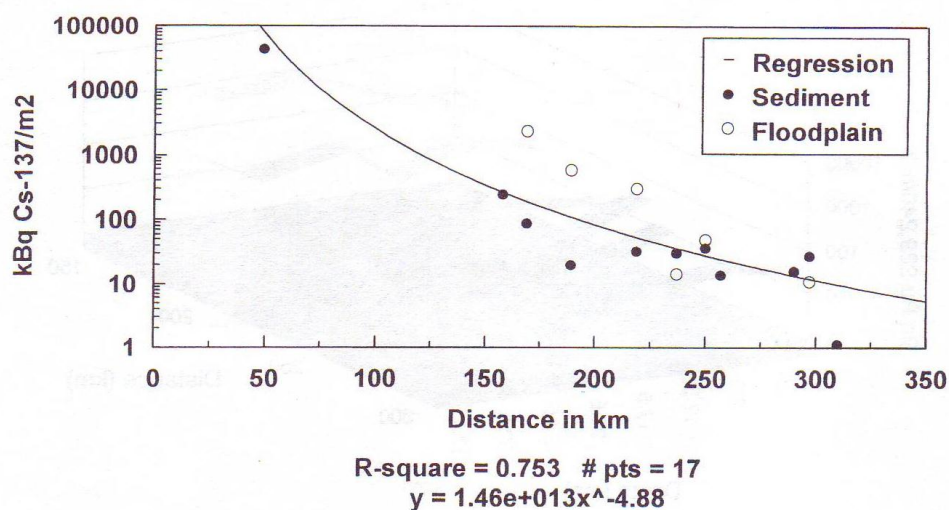


Fig. 8. Cumulative ^{137}Cs in the 0–100 cm sediment and floodplain layer along the Tеча and Iset rivers in 1996.

It appears from Fig. 11 that $^{239,240}\text{Pu}$ may be more mobile than ^{137}Cs ; but over the few hundred meters the floodplain extends from the river bed we find it justified to assume that the horizontal distribution of Pu follows that of ^{137}Cs and that the deposition of ^{137}Cs is proportional to the gamma-dose rate, which decreases exponentially with the distance from the river bed with a half-distance of about 55 ± 10 m (calculated from five transects in the floodplain (Fig. 5.7 in NREG, 1997)). From Eqs. (2)–(4) it is calculated that in order to obtain the total inventories of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the floodplain the deposits found in the 50 m belt should

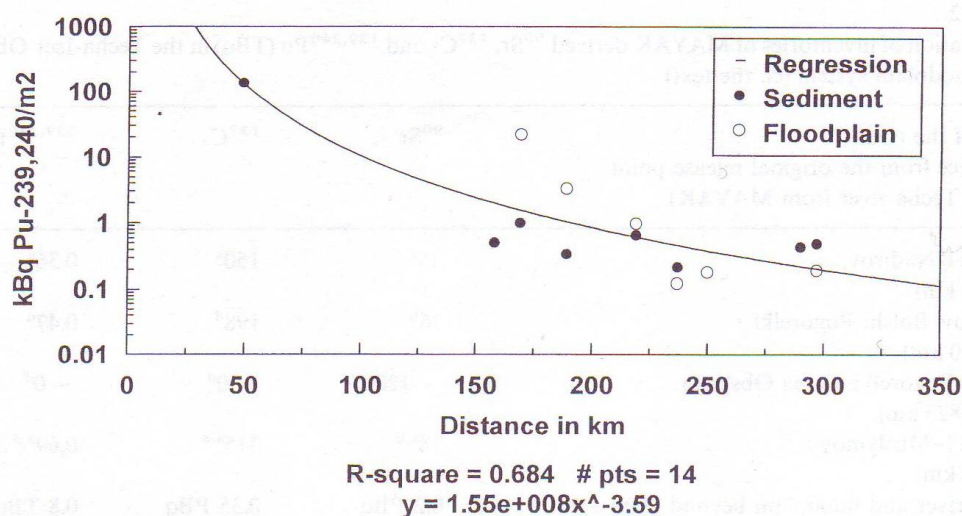


Fig. 9. Cumulative $^{239,240}\text{Pu}$ in the 0–100 cm sediment and floodplain layer along the Techa and Iset rivers in 1996.

Table 1

Coefficients ($\pm 1\text{SD}$) in the equation: $y = e^a x^b$, where y is kBq m^{-2} related to the distance x km from the original discharge point from MAYAK to the Techa river (N : the number of data)

Radionuclide	a	b	N
^{90}Sr	11.0 ± 4.2	$-(1.08 \pm 0.79)$	17
^{137}Cs	30 ± 3.9	$-(4.9 \pm 0.72)$	17
$^{239,240}\text{Pu}$	18.9 ± 3.7	$-(3.6 \pm 0.70)$	14

be multiplied by 32, 3.8 and 3.8, respectively. We have in this calculation assumed that Eqs. (2)–(4) are valid for the entire river system (A, B and C vary with the distance from MAYAK). This assumption may not be correct, but for ^{137}Cs and $^{239,240}\text{Pu}$ most of the activity is deposited in the upper reaches of the river system for which the equations were calculated, so for these radionuclides it has less important implications if the assumption is wrong. In the case of ^{90}Sr the error, which is difficult to quantify, may be greater because ^{90}Sr has moved further downstream than ^{137}Cs and $^{239,240}\text{Pu}$.

The reservoirs built in the upper parts of the Techa river extend to Dam 11, situated 31 km from the original release point at MAYAK. From Dam 11 to Nadirov Bridge we have no samples but Russian data (NREG, 1997) are available from the Asanov Swamp from a location situated 7 km downstream from Dam 11. The levels of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ at this location (1200, 44 000 and 74 kBq m^{-2} , respectively) are so close to ours obtained from Nadirov Bridge (920, 43 000 and 137) that, for the first part of the river (18 km) after the reservoirs, we assume the depositions of the three radionuclides to be represented by the means of these two sets of observations.

Table 2

Calculation of inventories of MAYAK derived ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ (TBq) in the Techa-Iset-Ob rivers and floodplain system (cf. the text)

Part of the river (distance from the original release point to the Techa river from MAYAK)	^{90}Sr	^{137}Cs	$^{239,240}\text{Pu}$
Dam 11-Nadirov (31–49 km)	31 ^a	150 ^a	0.36 ^a
Nadirov–Bolshi Pogorelki (49–310 km)	76 ^b	198 ^d	0.47 ^d
Bolshi Pogorelki–Guba Obskaya (310–2823 km)	~ 120 ^c	~ 0 ^d	~ 0 ^d
Dam 11–Muslymovo (31–78 km)	38 ^{a,b}	315 ^{a,d}	0.69 ^{a,d}
Total river and floodplain beyond Dam 11 (31–2823 km)	0.2 PBq	0.35 PBq	0.8 TBq

$$^a \text{ } ^{90}\text{Sr}: \frac{(920 + 1200)}{2} \times 18 \times 50 \times 10 \times 10^{-6} = 31 \text{ TBq } ^{90}\text{Sr}.$$

$$^b \text{ } ^{90}\text{Sr}: 179(310 - 49) \times 50 \times 32 \times 10^{-6} = 76 \text{ TBq } ^{90}\text{Sr}.$$

$$^c \text{ } ^{90}\text{Sr}: \left(\int_{310}^{2823} 5.84 \times 10^4 \times x^{-1.08} dx \right) \times 50 \times 32 \times 10^{-6} = 120 \text{ TBq } ^{90}\text{Sr}.$$

$$^d \text{ } ^{137}\text{Cs}: \left(\int_{49}^{310} 1.46 \times 10^{13} x^{-4.9} dx \right) \times 50 \times 3.8 \times 10^{-6} = 198 \text{ TBq } ^{137}\text{Cs}.$$

The inventories are then calculated from these mean values by multiplication with the area of the 50 m belt (900 000 m²) and the above-mentioned factors (3.8 for ^{137}Cs and $^{239,240}\text{Pu}$ and 32 for ^{90}Sr).

The uncertainties of the inventory estimates were obtained from a parameter uncertainty analysis carried out using the programme “Crystal Ball”. We have calculated 1 SD ranges for the inventories between Nadirov Bridge and Bolski Pogorelki in Table 2: 51–107 TBq ^{90}Sr , 58–670 TBq ^{137}Cs and 0.13–1.67 TBq $^{239,240}\text{Pu}$. This corresponds to uncertainty factors of 1.45, 3.4 and 3.6 for the three radionuclides, respectively. Due to the above-mentioned assumptions, which in particular influence the ^{90}Sr inventory we estimate the actual uncertainty factor for ^{90}Sr to be no less than those for ^{137}Cs and $^{239,240}\text{Pu}$.

According to Russian estimates (NREG, 1997) 0.27% of the ^{90}Sr in the Techa river system (river + floodplain) between Dam 11 and Muslymovo is to be found in the bottom deposits in the river. For ^{137}Cs the percentage is 2.1. If we assume that this percentage is valid also for Pu, and if we furthermore assume that all the percentages are valid for the entire Techa river, we may calculate by the methods shown above the

radionuclide inventories in the river deposits between Nadirov and Zatechenskoe which were studied in 1990 (Trapeznikov et al., 1993). We find 0.15 TBq ^{90}Sr , 4 TBq ^{137}Cs and 10 GBq $^{239,240}\text{Pu}$. In 1990 we estimated these inventories to be 0.3 TBq ^{90}Sr , > 6 TBq ^{137}Cs and 8 GBq $^{239,240}\text{Pu}$. The apparent agreement between the two estimates suggests that the previous inventory estimate for the river deposits is compatible with the present study.

The above-mentioned joint Norwegian–Russian study (NREG, 1997) has estimated the inventories of ^{90}Sr and ^{137}Cs in the upper reaches of the Techa-river-floodplain between Dam 11 and Muslymovo, i.e. 31–78 km from MAYAK, to 36–44 and 190–230 TBq, respectively. These figures are comparable with those calculated in Table 2 for this part of the river. The two estimates are not fully independent as we in our calculation have used one Russian data set (Asanov Swamp) and furthermore have calculated our horizontal distributions of ^{90}Sr and ^{137}Cs in the floodplain from the Russians' observations (Eqs. (2) and (3)).

In a Russian study (Chesnokov et al., 1998; Shcherbak, 1998), applying collimated scintillator detector techniques for the determination of the ^{137}Cs contamination of the Techa river floodplain, five sites at the river floodplain were mapped: Muslymovo, Brodokalmak, Russkaja Techa, Zatechenskoye and Krasnoitsetskiye (Iset). The study covered about 68 km of the river system and the total number of measuring points was about 20 000. This work is thus probably the most comprehensive investigation of the ^{137}Cs contamination of the downstream parts of the Techa (Iset) river floodplain carried out so far. We may use the Russian inventories from this study to validate our model for ^{137}Cs (Table 3).

According to Table 3 the SUCON model seems to overpredict the ^{137}Cs inventories in the upper parts and underpredict in the lower parts of the river system. About 99% of the ^{137}Cs and $^{239,240}\text{Pu}$ from MAYAK present downstream of Dam 11 is found in the Techa river and floodplains. In the case of ^{90}Sr perhaps half of the MAYAK release may be found beyond the Techa, but the inventory estimate of ^{90}Sr is encumbered with large uncertainties. It should furthermore be taken into account that in regularly flooded areas ^{90}Sr may have penetrated deeper than 1 m (NREG, 1997).

Fig. 10 illustrates the greater mobility of ^{90}Sr than of ^{137}Cs in the river system. From 50 to 300 km from MAYAK the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio increases by a factor of 600. Fig. 11. shows that Pu also is more mobile than ^{137}Cs . The $^{239,240}\text{Pu}/^{137}\text{Cs}$ thus increases by a factor of 5.4 from 50 to 300 km from MAYAK. The vertical migration of both ^{90}Sr and $^{239,240}\text{Pu}$ in the floodplain soils and sorlake sediments is also greater than that observed for ^{137}Cs (see Figs. 1–6).

3.2. Dating of sediment cores

In an earlier paper (Aarkrog, Chen, Dahlgaard, Nielsen, Trapeznikov & Pozolotina, 1997) the Nadirov sediment sample from 1992 was dated by the ^{210}Po method. It was found that the sedimentation rate was $1.22 \text{ kg m}^{-2} \text{ y}^{-1}$ or 0.33 cm y^{-1} . Hence, the maximum layer (14–16 cm) could be dated back to 1944–1950 in reasonable agreement with the early discharges from the MAYAK plant around 1948–1952.

Table 3

Comparison of ^{137}Cs inventories of Chesnokov et al. (1998) and Shcherbak (1998) with SUCON model calculations (see Table 2)

Location	Part of river (distance in km from MAYAK)	Chesnokov and Shcherbak measurement (TBq)	SUCON model calculation (TBq)
Muslymovo	70–87	6.5	28
Brodokalmak	107.5–122.5	2.9	3.7
Russkaya Techa	134–160	1.24	1.99
Zatechenskoye	234–240	0.32	0.044
Krasnoiskoye	245–249	0.074	0.022

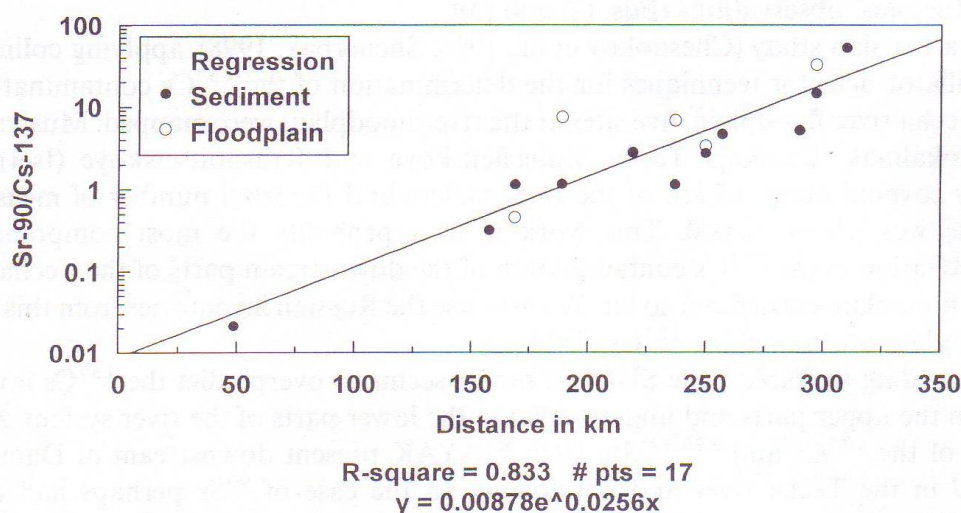


Fig. 10. The longitudinal distribution of the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio in sediments and floodplain soil along the Techa and Iset rivers in 1996.

In the present study we were able to date a sediment column from a sorlake collected 169 km from MAYAK (Fig. 12). The results are discussed below.

3.3. Anchugovo (169 km from MAYAK) Techa

^{210}Pb and ^{210}Po dating showed a sedimentation rate of 0.3 cm yr^{-1} . Hence, we would expect the MAYAK discharge in 1948–1952 to show up in the 10–15 cm layer, but it appears in the 5–10 cm layer, where both ^{137}Cs and $^{239,240}\text{Pu}$ have their maxima. The low inventory ratios ($^{239,240}\text{Pu}/^{137}\text{Cs} = 0.0103$ and $^{241}\text{Am}/^{239,240}\text{Pu} = 0.038$) suggest that most of the contamination comes from the early MAYAK releases rather than global fallout. We believe that the deposition at Anchugovo is delayed by about 15 yr so the 1948–1952 discharge of ^{137}Cs and transuranics first appeared around 1965. The source may have been the floodlands which after flooding events have

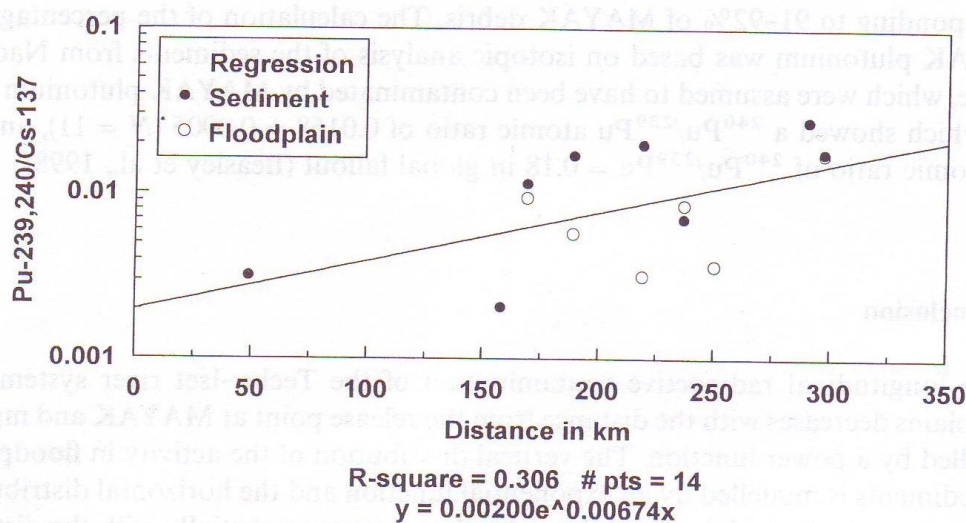


Fig. 11. The longitudinal distribution of the $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratio in sediments and floodplain soil along the Techa and Iset rivers in 1996.

River Sediments from Anchugovo, August 1996.

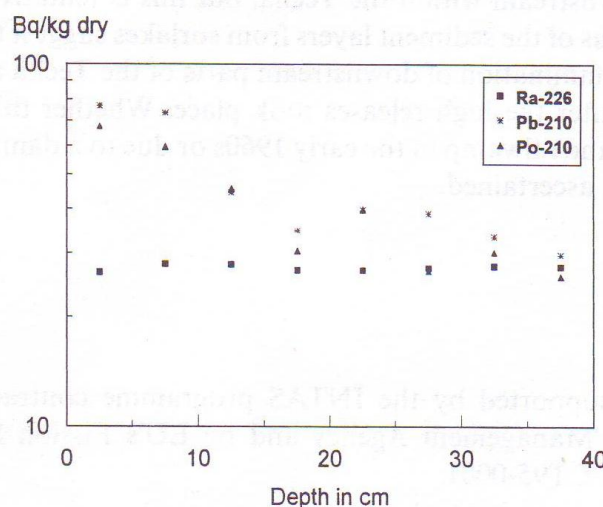


Fig. 12. Ra-226, ^{210}Pb and ^{210}Po concentrations in sorlake sediments collected in 1996 from Anchugovo (Techa River).

released some of their contaminated soils into the river, but the failure of dam 11 (see Map) in the early 1960s may be another explanation.

Isotopic determinations of ^{239}Pu and ^{240}Pu by ICPMS showed an atomic ratio $^{240}\text{Pu}/^{239}\text{Pu} = 0.02$ in the 0–5, 5–10 and 10–15 cm layers (Dahlgard, Chen, Aarkrog, Stürup & Nielsen, 1998). Hence, 98–99% of the plutonium in the upper 15 cm of the sediment column was MAYAK derived. In the 15–20 cm layer $^{240}\text{Pu}/^{239}\text{Pu}$ was 0.03

corresponding to 91–92% of MAYAK debris. The calculation of the percentages of MAYAK plutonium was based on isotopic analysis of the sediments from Nadirov Bridge, which were assumed to have been contaminated by MAYAK plutonium only and which showed a $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio of 0.0168 ± 0.0005 ($N = 11$), and on the atomic ratio of $^{240}\text{Pu}/^{239}\text{Pu} = 0.18$ in global fallout (Beasley et al., 1998).

4. Conclusion

The longitudinal radioactive contamination of the Techa–Iset river system and floodplains decreases with the distance from the release point at MAYAK and may be modelled by a power function. The vertical distribution of the activity in floodplains and sediments is modelled by an exponential function and the horizontal distribution of the contamination of the floodplain also decreases exponentially with the distance from the riverbed. Based on these model assumptions, the inventories of the Techa river floodplain and river sediments downstream the reservoir system at MAYAK were calculated to be 0.1 PBq of ^{90}Sr , 0.3 PBq of ^{137}Cs and 0.8 TBq of $^{239,240}\text{Pu}$. The uncertainty of these estimates is a factor of 3–4. While essentially all MAYAK-derived ^{137}Cs and $^{239,240}\text{Pu}$ are to be found in the Techa river system, about half of the ^{90}Sr may be present downstream within the Techa, but this is tentative.

Age determinations of the sediment layers from sorlakes suggest that the major part of the MAYAK contamination of downstream parts of the Techa and Iset rivers first occurred 10–15 yr after the high releases took place. Whether this has been due to a flooding of the Asanov Swamp in the early 1960s or due to a dam failure in reservoir 11 has not yet been ascertained.

Acknowledgements

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