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Radioactive Pollution of the Ob River System from Urals Nuclear Enterprise 'MAJAK'

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ABSTRACT

The Techa river belongs to the Iset–Tobol–Irtysh–Ob river system. Around 1950 the Techa was contaminated with medium and high level radioactive waste from the MAJAK nuclear installation. The total discharge amounted to 100 PBq; ^{90}Sr and ^{137}Cs contributed 11·6% and 12·2% respectively. Presently the Techa contains about 0·3 TBq ^{90}Sr, more than 6 TBq ^{137}Cs and about 8 GBq $^{239,240}Pu$. The levels of the radionuclides upstream are several orders of magnitude higher than those expected from global fallout. The activity concentrations decrease exponentially or by power functions with distance. The study has shown that the contamination of the soil of the Techa flood plain is 15–85 times higher than global fallout levels. There is an unexplained radioactive contamination in the Iset river after confluence with the Techa.

INTRODUCTION

The Ob in the west-Siberian part of Russia is 3 700 km long, its drainage area is 2 990 000 km^2 and flows to the Kara Sea in the Arctic Basin. There are several sources of radioactive contamination of the Ob river system: (i) global fallout from nuclear weapons testing; (ii) local fallout from explosions at Novaja Zemlya and Semipalatinsk; (iii) discharges of liquid

radioactive waste from nuclear enterprises at Tomsk and Cheljabinsk and from the nuclear power plant near Ekaterinburg, which are all situated on the Ob tributaries.

The major input of radioactivity came from the MAJAK nuclear installation in the Cheljabinsk region. Between 1949 and 1951 medium and high level liquid radioactive waste from MAJAK was released to the Techa river, which belongs to the Irtysh–Ob river system. During that period 76×10^6 m^3 of liquid radioactive waste was discharged with a total activity of 100 PBq. Strontium-90 and ^{137}Cs contributed 11·6% and 12·2% respectively (Academy of Science, 1991). Now, 42 years later, the environmental contamination from this source has decayed to 4·3 PBq ^{90}Sr and 4·6 PBq ^{137}Cs.

Between 1951 and 1964 discharge of the wastes was eliminated by building a cascade of reservoirs and by-pass canals in the upper reaches of the Techa. Presently this system contains 7·1 PBq ^{90}Sr and ^{137}Cs (Academy of Science, 1991). However, before this was accomplished, a large part of the flood plain and bottom of the river, especially upstream, was contaminated. Today 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 too; the volume of filtered water which enters the Techa, is about 10^7 m^3 per year (Genesis and conception, 1993).

The aims of this study are the following:

- to continue the measurements of ^{137}Cs, ^{90}Sr, ^{239,240}Pu in the principal components of the Techa river as reported in Trapeznikov *et al.* (1993), in order to improve the inventory estimates;
- to investigate the level of contamination of the Techa flood plain in the lower reaches and to estimate its significance as a source of additional pollution of the river system;
- to study the radioactive contamination of the Iset river after confluence with the Techa.

MATERIALS AND METHODS

Sediments and water samples were collected between 1990 and 1992 from the Techa, Iset and Ob rivers (Figs 1–3). Duplicate water samples (80–100 litres) were filtered through a cotton wool filter in order to remove macro admixtures and then evaporated to dryness.

Sediments (1–2 kg) were collected 3–4 m from the bank by dredging which sampled the upper, 0–10 cm, layer of sediment. Some sediments

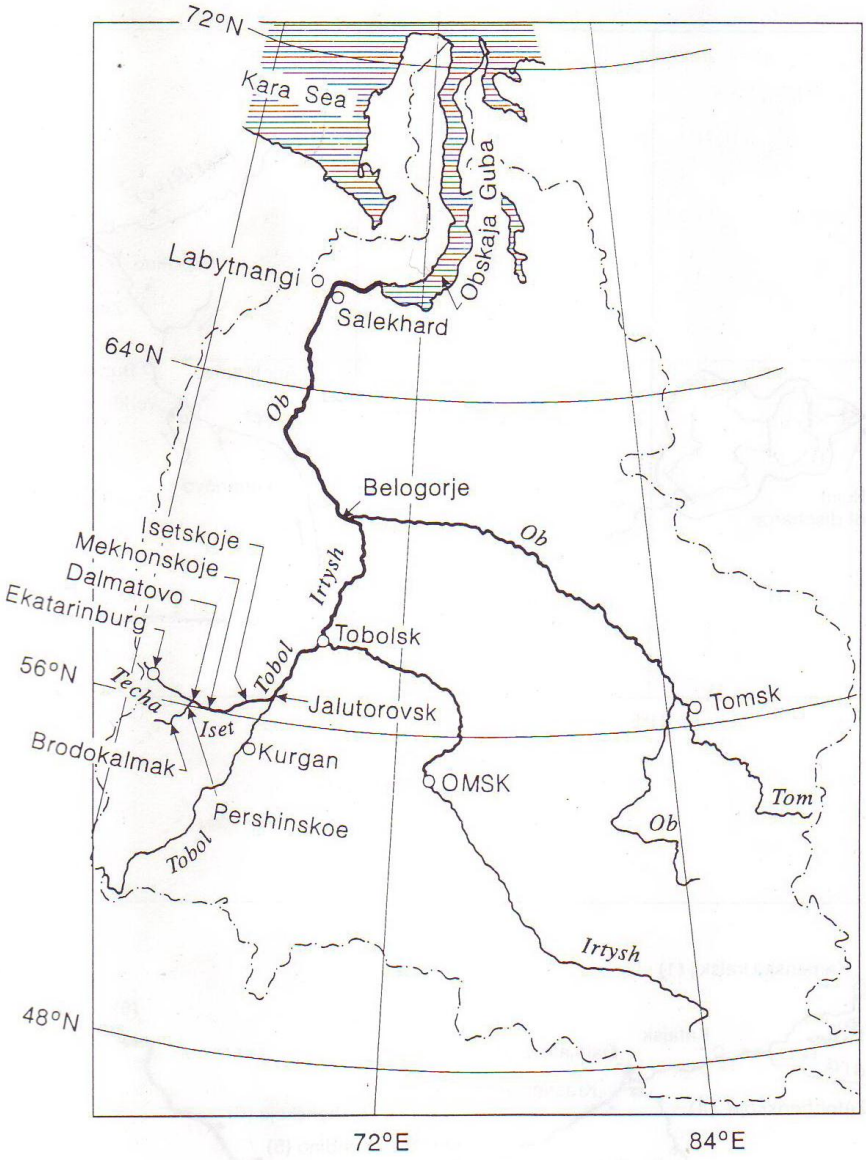


Fig. 1. Drainage area of the Ob river ($2{\cdot}9 \times 10^6$ km^2). (See also Table 1.)

were sampled by a 33·2 cm^2 coring device in 2-cm layers down to about 40 cm. The samples were dried, ground and sifted (sieve 0·1 cm).

The flood plain soil was collected at distances of 3–4 m and 30–40 m, respectively, from the canal of the Techa river down to a 30 cm depth. The soil sections were made by the 'envelope method' with a side of about 10 m.

The aliquots of samples were analysed at the Institute of Plant and Animal Ecology (Russia) for ^{90}Sr (Zvetaeva *et al.*, 1984) and ^{137}Cs. At the Risø National Laboratory (Denmark) γ-spectrometry was used for ^{239,240}Pu (Talvitie, 1971) and ^{241}Am (Holm & Persson, 1979) analyses.

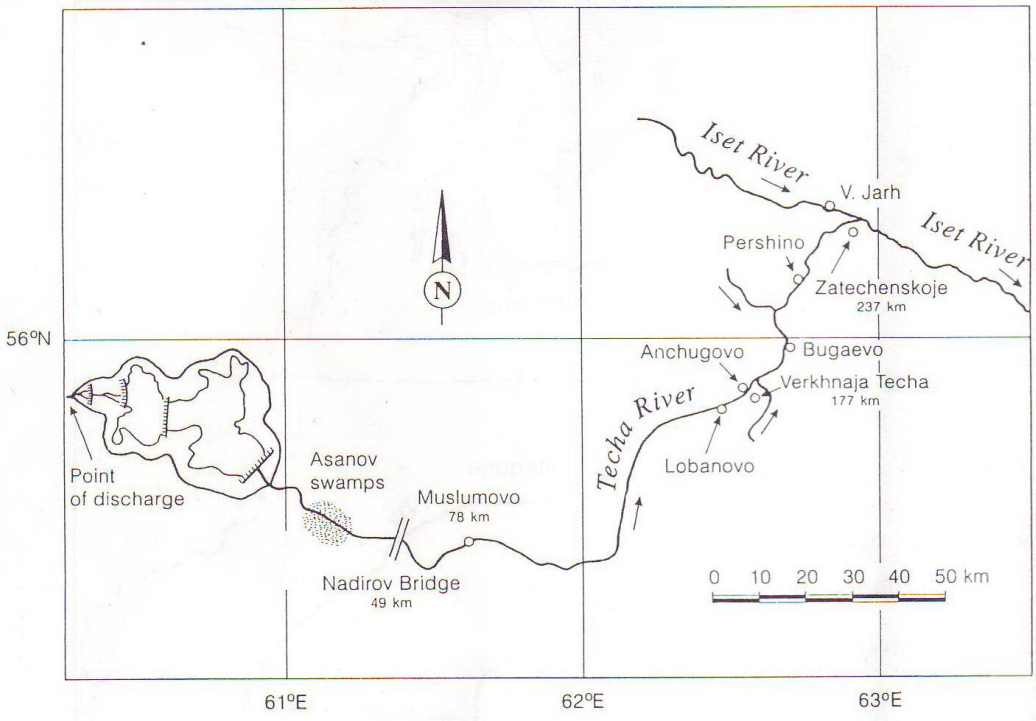


Fig. 2. Techa river.

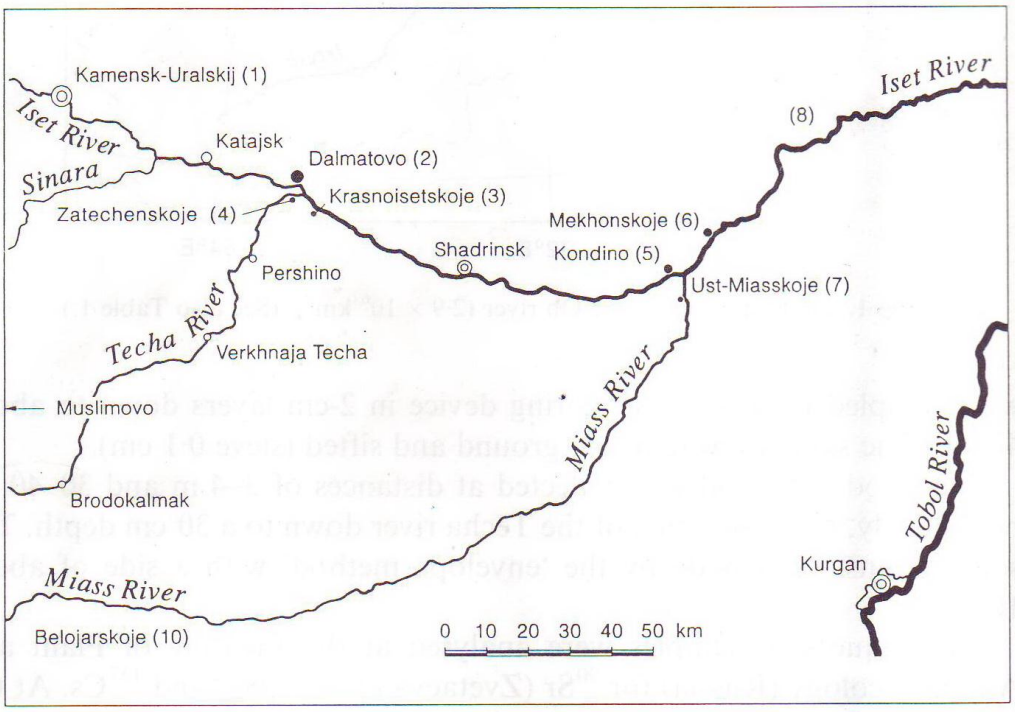


Fig. 3. Iset river.

RESULTS AND DISCUSSION

Techa sediments

Based on samplings at four locations in the Techa river in July 1990 (Trapeznikov *et al.*, 1993) it was found that the sediments contained relatively high ^{137}Cs levels upstream, but the levels decreased with increasing distance from the discharge point. The power function, $y = e^a x^b$, best described the ^{137}Cs concentrations in sediments. The variable, x, is the distance in kilometres from the point of discharge. The exponents a and b are given in Table 1 with ±1 SD.

It should be emphasized that the above equation is valid only between the points of observations within 49–237 km. Compared with the global fallout levels in sediments, the Techa samples showed ^{137}Cs and ^{90}Sr concentrations to be one to four orders of magnitude higher (Aarkrog *et al.*, 1991).

In order to study the distribution of radionuclides in the sediment profile a special sediment sample was collected at the Nadirov Bridge (Fig. 2), where 2-cm layers were taken, down to 40 cm depth (Fig. 4). A maximum occurred at a depth of about 15 cm for concentrations of ^{137}Cs as well as for ^{239,240}Pu. If this depth corresponds to the discharge year 1950, the sedimentation rate would be 150/42 = 3.6 mm per year. The mean ratio between ^{239,240}Pu and ^{137}Cs in the sediment was $(4.1 \times 1.0) \times 10^{-3}$ (±1 SD; $N = 8$), ^{238}Pu/^{239,240}Pu = $(6{\cdot}3 \pm 3{\cdot}8) \times 10^{-3}$ and ^{241}Am/^{239,240}Pu = $(2{\cdot}9 \pm 0{\cdot}4) \times 10^{-2}$. The maximum ^{90}Sr concentration occurred in the 18–20 cm layer, illustrating the greater migratory ability of ^{90}Sr. The total deposition, down to 40 cm, was 0·95 MBq ^{90}Sr m^{-2}, i.e. 2·5% of the ^{137}Cs deposition.

Techa water

The concentration of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in the Techa river water decreases with distance, x (km) from the point of discharge according to the exponential model, $y = e^{(a+bx)}$, where y is the radionuclide concentra-

TABLE 1

Exponents a and b for the Power Function $y = e^a x^b$ (±1 SD) for Sand and Silt

	Sand		*Silt*	
	a	b	a	b
^{90}Sr	23 ± 2	3·7 ± 0·5	12 ± 3	1·2 ± 0·6
^{137}Cs	28 ± 2	−(4·5 ± 0·4)	33 ± 1	−(5·1 ± 0·2)
^{239,240}Pu	14·4 ± 0·3	−(3·0 ± 0·1)	22 ± 2	4·2 ± 0·3

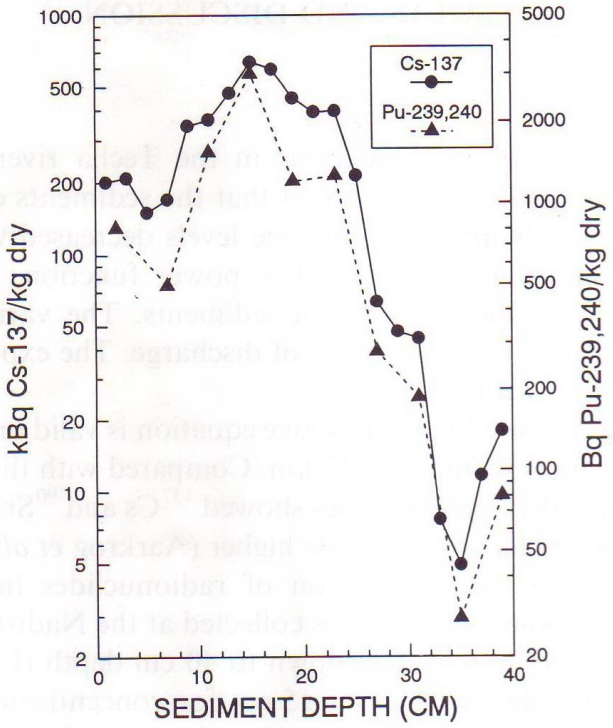


Fig. 4. Caesium-137 and ^{239,240}Putonium in sediments (2-cm layer) collected at Nadirov Bridge from the Techa river in 1992, 49 km from the discharge point. Total deposits in sediments: 37·4 MBq ^{137}Cs m^{-2}; 0·15 MBq ^{239,240}Pu m^{-2}; 0·8 kBq ^{238}Pu m^{-2}; 4·5 kBq ^{241}Am m^{-2}.

tion in Bq m^{-2} (Trapeznikov *et al.*, 1993). Values of a and b are given in Table 2.

Radionuclide concentrations in the Techa river water may be compared with those in the Columbia river, polluted by plutonium production at Hanford (Eisenbud, 1987), which are two to three orders of magnitude lower. Compared with global fallout concentrations of ^{90}Sr and ^{137}Cs, concentrations in Techa water are two to three orders of magnitude higher (Aarkrog, 1991).

TABLE 2

Exponents a and b for the Exponential Function, $y = e^{(a+bx)}$ (±1 SD) for the Techa River Water

	a	b
^{90}Sr	8·62 ± 0·05	−(0·0029 ± 0·0003)
^{137}Cs	7·03 ± 0·13	−(0·0120 ± 0·0008)
^{239,240}Pu	−(0·38 ± 0·11)	−(0·0074 ± 0·00007)

TABLE 3
Radionuclides in Sediment and Water of the Iset and Miass Rivers, in July 1992

Location	*Sediment*		*Water*	
	^{90}Sr *Bq* kg^{-1}	^{137}Cs *Bq* kg^{-1}	^{90}Sr *Bq* m^{-3}	^{137}Cs *Bq* m^{-3}
No 2 Dalmatovo	27	1·2	120	1·3
No 3 Krasnoisetskoje	14	3·1	910	13
No 5 Kondino	10	1·5	620	2·6
No 6 Mekhonskoje	38	9·1	430	15
No 7 Ust-Miasskoje	51	8·3	49	23
No 10 Belojarskoje	15	10·1	490	65

Our study has shown that the ^{90}Sr concentrations have decreased by about a factor of two with water flow through the Techa river while the volume of flowing increases more than ten times from the Nadirov Bridge to the end of the Techa (Table 3). This means that radionuclides are released to the river water during its passage through the Techa. Possible sources of additional pollution are the sediment and the flood plain of the river.

Techa flood plain

A considerable part of the radioactive disposal is contained within the flood plain of the Techa river. The density of contamination with ^{90}Sr and ^{137}Cs, in some of the flood plain areas (3–4 m from the river bank), exceeds the global fallout level by one or two orders of magnitude (Table 4). We used the flood plain of the Iset river near the V. Jarth village (Fig. 2) as a control location as it was considered to be uncontaminated from sources other than global fallout.

The radioecological situation within the central flood plain of the Techa (within 30–40 m from the canal) changes with the distance from the canal. If the flood plain is on open meadow (as seen at Zatechenskoje or at the right bank of the river near Pershino) the contamination level decreases gradually with distance from the river, though the level at 30–40 m from the canal is still 10 times higher than the control. If the landscape is hilly (as at Bugaevo or on the left bank near Pershino) there is a geochemical barrier to the migration of the radionuclides. The total deposition, down to 30 cm and at 30–40 m from the canal, at these hilly locations, is higher than the deposition near the Techa river (Table 4).

The distribution of radionuclides in the soil profile varies as shown in Fig. 5. At some locations strontium and caesium are distributed evenly. In

TABLE 4
Radionuclides in the Soil of the Techa Flood Plain in July 1992

Location. Distance from Techa outlet (km)	*River bank*	*Distance from the river (m)*	*^{90}Sr kBq m^{-2}*	*^{137}Cs kBq m^{-2}*
v. Lobanovo	right	3–4	83	169
77	left	3–4	88	200
v. Anchugovo	right	3–4	230	235
65	left	3–4	96	145
v. Bugaevo	right	3–4	87	88
48		30–40	610	470
	left	3–4	230	167
v. Pershino	right	3–4	122	94
27		30–40	34	55
	left	3–4	290	155
		30–40	380	123
v. Zatechenskoje	right	3–4	230	83
3		30–40	25	58
v. V. Jarh	left	3–4	3·4	5·9
9				

other places, a maximum may be displaced towards the deeper layers, and at others, most of the radionuclides may be absorbed in the upper layers of the soil. The situation with the Techa flood plain is very dynamic. The following events occur periodically: (i) transference of small particles with spring high-waters and rain water; (ii) drift of soil in the river; (iii) covering of soil layers with river alluvium; (iv) vertical migration of radionuclides with subsoil waters. The majority of these processes promote the additional contribution of radionuclides to the river water.

Iset river

The contamination of the Techa river influences the ^{90}Sr and ^{137}Cs concentrations of water and sediments in the Iset river (Table 3). It appears that the Miass river (Fig. 3) contributes to the contamination of the Iset river with ^{90}Sr and ^{137}Cs. The source of this pollution has not been identified in this study. An estimate of the annual transport of radionuclides in the Iset river, at Mekhonskoe, is 1·1 TBq ^{90}Sr and 0·04 TBq ^{137}Cs. This may be compared to the transport of radionuclides in the Techa river at Pershinskoe.

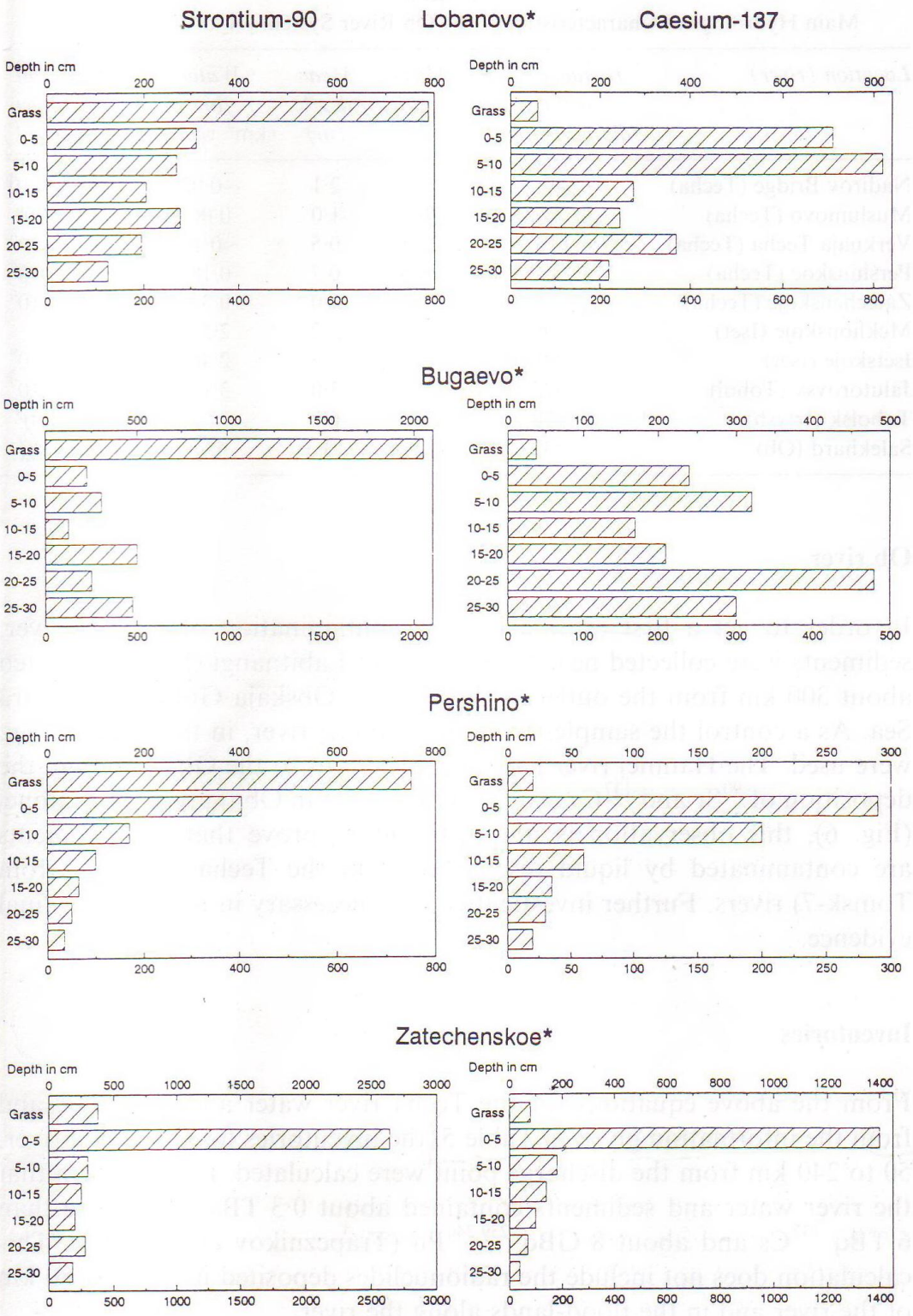


Fig. 5. Concentrations of ^{90}Sr and ^{137}Cs in the Techa flood plain soil, Bq kg^{-1} dry weight. *Name of location (see map on Fig. 2).

TABLE 5
Main Hydrological Characteristics of the Ob River System (Hydrometeo, 1973)

Location (river)	*Distance from point of discharge (km)*	*Mean width (m)*	*Mean depth (m)*	*Water flow* ($km^3\ year^{-1}$)	*Sediment transport* ($kg\ year^{-1}$)
Nadirov Bridge (Techa)	49	37	2·1	~0·02	$\sim 1.66 \times 10^6$
Muslumovo (Techa)	78	20·5	1·0	0·06	$\sim 5 \times 10^6$
Verknaja Techa (Techa)	177	25·5	0·5	~0·14	$\sim 12 \times 10^6$
Pershinskoe (Techa)	214	78·5	0·7	0·18	$\sim 15 \times 10^6$
Zatechenskoje (Techa)	237	25·5	1·0	0·35	$\sim 30 \times 10^6$
Mekhonskoje (Iset)	356	82·9	1·2	2·57	—
Isetskoje (Iset)	480	131	2·6	2·48	$0{\cdot}05 \times 10^9$
Jalutorovsk (Tobol)	1023	164	3·0	3·11	$0{\cdot}11 \times 10^9$
Tobolsk (Irtysh)	1670	681	6·8	77·3	$7{\cdot}9 \times 10^9$
Salekhard (Ob)	2542	2894	13·3	421	15×10^9

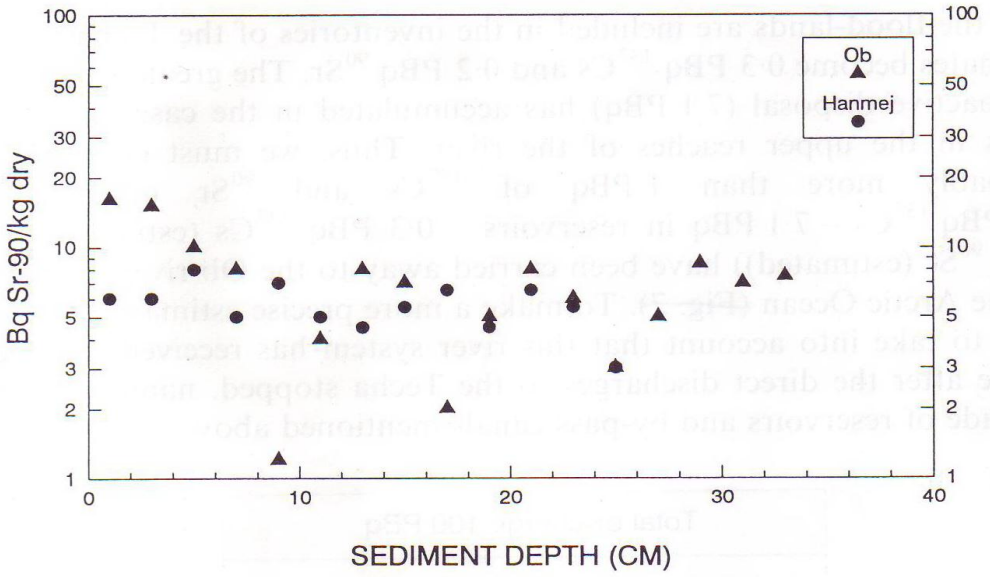
Ob river

In order to get a first estimate of the contamination of the Ob river, sediments were collected near Salekhard and Labitnangi (Fig. 1), situated about 300 km from the outlet of the Ob into Obskaja Guba at the Kara Sea. As a control the samples from the Hanmej river, in the same region, were used. The Hanmej river has no connections to the Ob. Although the deposition of ^{90}Sr and ^{137}Cs was 20–40% higher in Ob than in the Hanmej (Fig. 6), this observation is not sufficient to prove that Ob sediments are contaminated by liquid discharges from the Techa or Tom (from Tomsk-7) rivers. Further investigations are necessary in order to get final evidence.

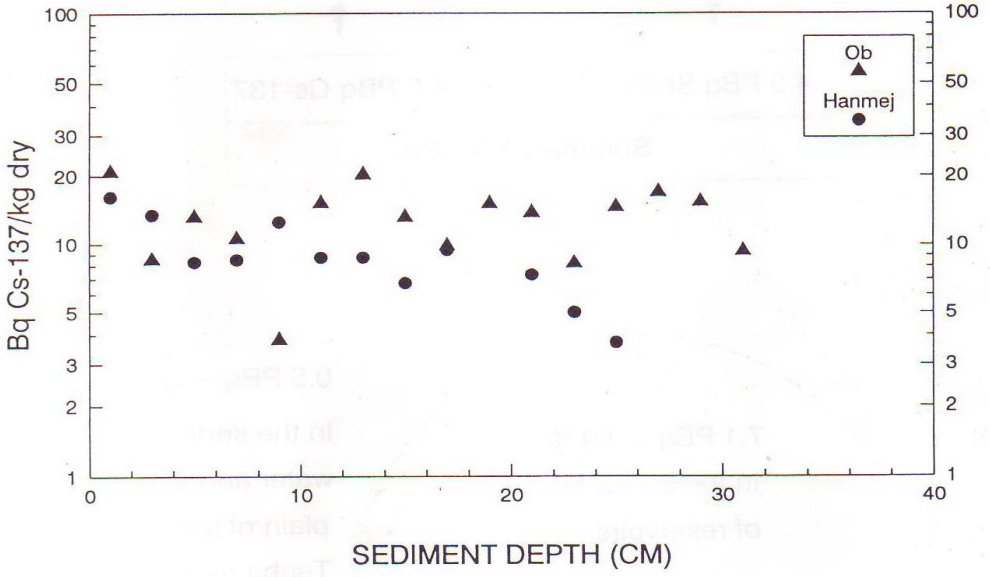
Inventories

From the above equations for the Techa river water and sediments and from the information given in Table 5, the inventories for the Techa river, 50 to 240 km from the discharge point were calculated. It was shown that the river water and sediments contained about 0·3 TBq ^{90}Sr, more than 6 TBq ^{137}Cs and about 8 GBq $^{239,240}Pu$ (Trapeznikov *et al.*, 1993). This calculation does not include the radionuclides deposited in the first 50 km of the river and in the flood-lands along the river.

The amount of radionuclides in the flood plain is estimated at 115–235 TBq ^{137}Cs and 79–189 TBq ^{90}Sr (MAJAK, 1990). Of this, 5 TBq ^{137}Cs and 9 TBq ^{90}Sr are deposited downstream of Muslumovo. Assuming



(a)

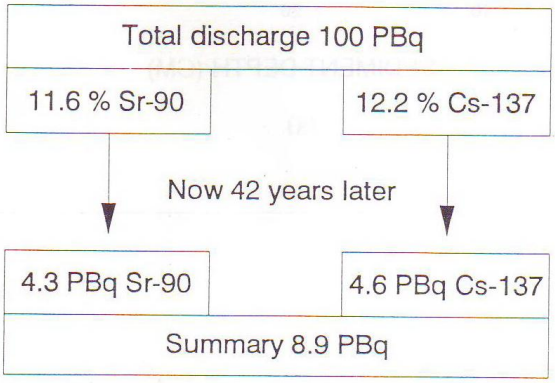


(b)

Fig. 6. (a) Strontium-90 and (b) caesium-137 in sediment samples (2-cm layers) from the Ob and Hanmej rivers in 1992 (Bq kg^{-1} dry weight).

proportionality between the inventories in flood-lands and in river sediments, a rough estimate of the deposition in the river sediments in the first 50 km can be derived. This amounts to 0·1 PBq ^{137}Cs and 2 TBq ^{90}Sr, i.e. an order of magnitude higher than that found downstream of the Nadirov Bridge in Techa river sediments.

If the flood-lands are included in the inventories of the Techa river the estimates become 0·3 PBq ^{137}Cs and 0·2 PBq ^{90}Sr. The greatest part of the radioactive disposal (7·1 PBq) has accumulated in the cascade of reservoirs in the upper reaches of the river. Thus, we must conclude that probably more than 1 PBq of ^{137}Cs and ^{90}Sr (4·3 PBq ^{90}Sr+ 4·6 PBq ^{137}Cs – 7·1 PBq in reservoirs – 0·3 PBq ^{137}Cs (estimated) – 0·2 PBq ^{90}Sr (estimated)) have been carried away to the Ob river system and to the Arctic Ocean (Fig. 7). To make a more precise estimate, it is necessary to take into account that this river system has received radioactive waste after the direct discharges to the Techa stopped, namely from the cascade of reservoirs and by-pass canals mentioned above.



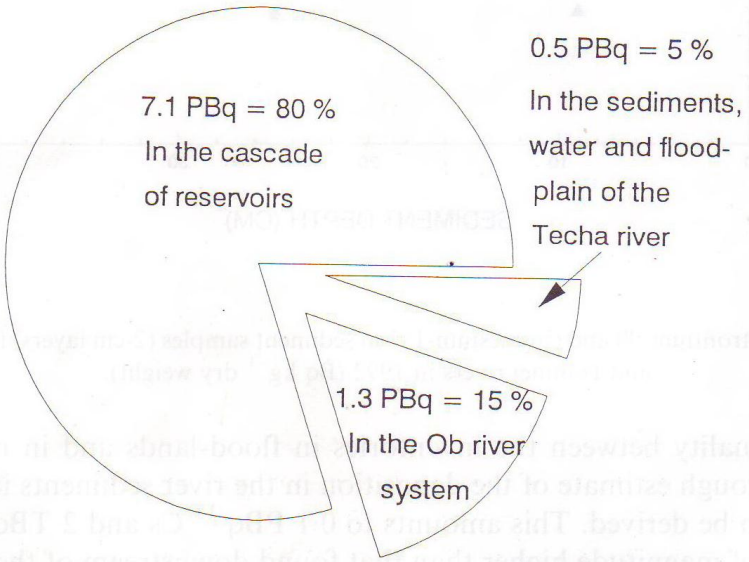


Fig. 7. Inventories.

CONCLUSIONS

Radioecological investigations of the Techa river, polluted with radioactive waste from MAJAK nuclear installation around 1955, have shown, that presently the level of long-lived radionuclides (^{90}Sr, ^{137}Cs, ^{239,240}Pu, ^{241}Am) in the principal components of the river, are several orders of magnitude higher than those expected from global fallout. The Techa flood plain is a source of permanent contamination of the river water.

About 5–10% of the total discharge of radionuclides are found in the water, sediments and flood plain of the Techa river. The major part (80%) of the ^{90}Sr and ^{137}Cs discharged from MAJAK are found in the cascade of reservoirs in the upper reaches of the Techa, within the territory of MAJAK. Over 1 PBq radionuclides can probably be found outside the Techa river, probably in the Ob river system and in the Arctic Ocean. This estimation needs further verification.

Data from the Iset river have shown that this river is contaminated by the Techa as well as by the Miass river. The preliminary results from the Ob river suggest enhanced levels of artificial radionuclides in the sediments, but further analyses are necessary for a final conclusion.

ACKNOWLEDGEMENT

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REFERENCES

Aarkrog, A., Bøtter-Jensen, L., Chen, Q. J., Dahlgaard, H., Hansen, H. J. M., Holm, E., Lauridsen, B., Nielsen, S. P. & Søgaard Hansen, J. (1991). Environmental radioactivity in Denmark in 1988 and 1989. Report Risø-R-570, Risø National Laboratory, Roskilde, Denmark, 212 pp.

Academy of Science. (1991). Conclusion of the Commission on the estimation of the ecological situation in the region of production association 'Majak', organized by the direction of Presidium of Academy Science. No. 1140–501//Russia. *J. Radiobiology*, **31**(3), 436–52.

Eisenbud, M. (1987). *Environmental Radioactivity*. Academic Press, New York, 475 pp.

Genesis and conception of the national programme of the Russia on the radiation reabilitation of the Urals. Ekaterinburg, 1993, 66.

Holm, E. & Persson, B. R. R. (1979). Behaviour of natural (Th, U) and artificial (Pu, Am) actinides in coastal waters. In *Marine Radioecology, Proc. of the 3rd NEA Seminar*, 1–5 October 1979, 237–243.

Hydrometeo (1973). *Resources of surface water. Volume 11.* Hydrometeo Publishing, Leningrad, 848 pp.

MAJAK. (1990). Material for the Supreme Council of the USSR, Cheljabinsk 65, 36 pp.

Talvitie, N. A. (1971). Radiochemical determination of plutonium in environmental and biological samples by ion exchange. *Analyt. Chem.*, **43**, p. 1827.

Trapeznikov, A. V., Pozolotina, V. N., Chebotina, M. Ya., Chukanov, V. N., Trapeznikova, V. N., Kulikov, N. V., Nielsen, S. P. & Aarkrog, A. (1993). Radioactive contamination of the Techa river, the Urals. *Health Physics*, **65**, 481–8.

Zvetaeva, N. A., Filin, V. H., Ivanova, L. A., Revnov, V. N., Rodionov, E. P., Rudaja, N. J., Suslin, I. A. & Shapiro, K. J. (1984). Use of monoiso actyl methylphosphoric geid and 'Miomphat' Fe(III) for the determination of contents of radionuclides in liquid waste. *Nuclear Energy,* **57**, 114–7.