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TRITIUM IN SURFACE WATER OF THE YENISEI RIVER BASIN

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ABSTRACT

The paper reports an investigation of the tritium content in the surface waters of the Yenisei River basin near the Mining-and-Chemical Combine (MCC). In 2001-2003 the maximum tritium concentration in the Yenisei River did not exceed 4±1 Bq/L. It has been found that there are surface waters containing enhanced tritium, up to 168 Bq/L, as compared with the background values for the Yenisei River. There are two possible sources of tritium input. First, the last operating reactor of the MCC, which still uses the Yenisei water as coolant. Second, tritium may come from the deep aquifers at the "Severny" testing site. For the first time tritium has been found in two aquatic plant species of the Yenisei River with maximal tritium concentration 304 Bq/Kg wet weight. Concentration factors of tritium for aquatic plants are much higher than 1.

Keywords: *"Tritium", "the Yenisei River basin", "Surface and ground waters", "Nuclear reactor", ""Severny" testing site", "Aquatic plant", "Concentration factor"*

INTRODUCTION

The Yenisei River is one of the largest waterways in the world. It is about 4000 km long. Its water catchment area is 2605 thousand km^2. The mean annual water discharge in the Yenisei estuary is about 600 km^3.

Tritium is produced by a variety of processes in nuclear power plants such as fission processes, neutron capture and decay processes. At the present time the Yenisei River receives both the tritium routinely released by the Mining-and-Chemical Combine of the Russian Ministry of Atomic Energy (with the direct release of the Combine effluent into the river and due to the removal of tritium from the sanitary-protective zone of the Combine by the river network) and the global fallout tritium (removed from the water catchment area of the Yenisei River basin).

The Mining-and-Chemical Combine is located at Zheleznogorsk in the Krasnoyarsk Territory, on the bank of the Yenisei River (Fig. 1). The nuclear reactors and the radiochemical plant of the Mining-and-Chemical Combine (MCC) have been in operation for more than 40 years and have contaminated the Yenisei River floodplain with radionuclides [1-3]. Two flow-through reactors were shut down in 1992, but the third reactor is still working. It must be put out of service in 2007. The Combine produced weapons-grade plutonium during many years and, as a result, there are considerable amounts of radioactive wastes, which are partly deposited in the storage facilities and open ponds on the Combine's territory, but the greater part has been injected into the deep aquifers of the "Severny" testing site. It is known from the published data of the MCC specialists [4, 5] that the "Severny" testing site is located 12 km north of the MCC radiochemical plant, at the watershed between the Yenisei and the Bolshaya Tel Rivers (Fig. 1).

There may be several potential sources of tritium found in the Yenisei water:

1) the tritium of the water-catchment area contaminated as a result of nuclear weapons tests;
2) the tritium of the water-catchment area contaminated by aerosol discharges of the MCC;
3) the tritium of the reactor coolant released into the Yenisei;
4) the tritium of the effluents of low-activity liquid wastes of the radiochemical production;
5) the tritium migrating from the open settling ponds located in the production area of the MCC; and
6) the tritium migrating from the deep aquifers of the "Severny" testing site.

No other water environment in the world receives tritium from so many sources of contamination. These unique conditions of the Yenisei River can be used to investigate various pathways of tritium migration in the environment.

The purpose of this study was to detect the sources from which radionuclides come into the Yenisei River, based on the detailed investigation of the tritium content in samples (water, sediments and aquatic plants) collected from the surface waters of the Yenisei River basin.

EXPERIMENTAL

Objectives

In the period from July to September 2001-2003 the researchers of the Radioecology Laboratory of the Institute of Biophysics SB RAS collected water samples from the Yenisei River near the MCC and at different distances from it downstream, from the mouths of the streams draining the MCC sanitary-protective zone, and from the Bolshaya Tel River (Fig. 1). Reference samples were collected upstream of the MCC, near Krasnoyarsk. In September 2001-2002 the researchers collected samples of sediments from the Bolshaya Tel River and upstream of the MCC, near Esaulovo, to determine tritium in the pore water. The sediment samples were collected at a depth up to 10-20 cm. In September 2002 and from June to September 2003 we collected samples of aquatic plants: the shining weed (*Potamogeton lucens*) and the Canadian pondweed (*Elodea canadensis*).

Methods

Within the first few hours after collection, the samples were filtered to remove suspended particles and placed into air-tight glass containers. Immediately before analytical investigations, the water samples were subjected to chemical treatment in accordance with the recommendations of the Environmental Measurements Laboratory of the United States [6, 7]. The water samples were distilled with $KMnO_4$ (under alkaline conditions) to eliminate coloration, organic matter, and salts that might interfere with the determination of tritium. Pore water was extracted from sediment samples and aquatic plants by distilling the azeotropic mixture with toluene. The distillation of the azeotropic mixture with toluene separated the solution of total dissolved tritium including both ^3H_2O and organically bound tritium. Depending on the composition of sediments, the water content was 15-30% of the initial mass. The water content of *Potamogeton lucens* was 45% and that of *Elodea canadensis*, 77%. Upon distillation, the resulting samples of aqueous extract were subjected to double purification.

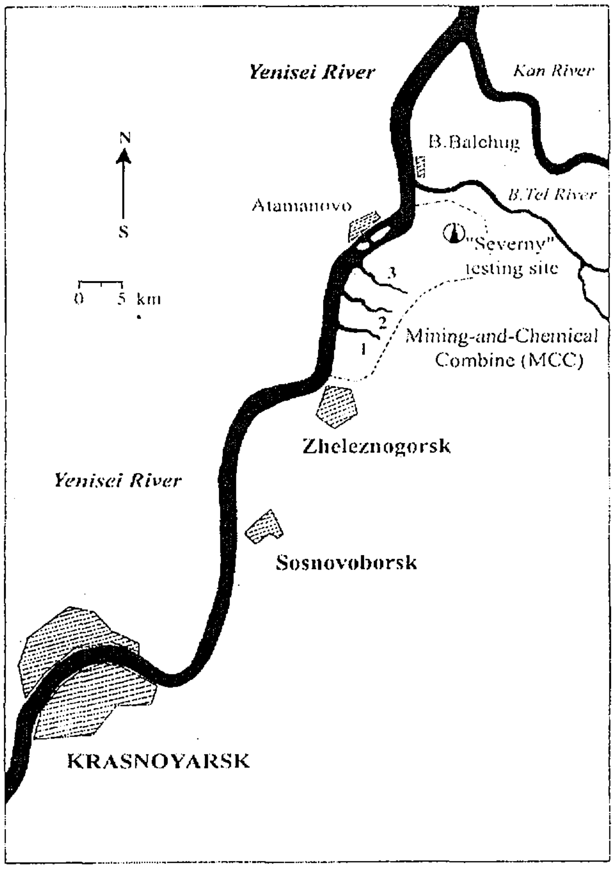

Fig. 1. Diagrammatic map of the south of the Krasnoyarsk Territory near the Mining-and-Chemical Combine of the Russian Ministry of Atomic Energy (Zheleznogorsk): surface waters of the Yenisei River basin: 1 – the Shumikha River; 2 – Stream No. 2; 3 – the Ploskii Stream

- - - - The boundary of the MCC sanitary-protective zone

Measurements

Analytical investigations of the samples for tritium were conducted at the Institute of Biophysics SB RAS (Krasnoyarsk) and at the Centre for Information Processes and Technologies of the RPA RADON (Moscow). Subsamples of water distilled from the oxidation process were assayed for tritium using liquid scintillation counters. At the Institute of Biophysics in Krasnoyarsk purified sample aliquots were mixed with an LS-8 scintillation cocktail in 20-ml low ^{40}K glass scintillation vials at a ratio of 1 to 9. Samples were measured at a temperature of ~6-8°C on a liquid scintillation spectrometer made in Russia. The spectrometer was efficiency calibrated with standard reference tritium solutions. The efficiency of tritium registration was 30%.

At the RPA RADON in Moscow a 10-ml aliquot of the purified sample was placed into a polypropylene vial and mixed with 10 ml of a scintillator solution – an Ultima-Gold AB cocktail. The resulting samples were measured using a TRI-CARB 2550 TR/AB (Canberra, USA) liquid-scintillation spectrometer during 1000 minutes. Tritium and other β-emitters, including ^{14}C, were identified and their activities calculated using the "SpectraDec" program for spectrum processing. The program involves a comparison of the experimental spectrum with the model one, which is obtained by calculation using the library of the spectra for certain isotopes [8]. The spectrometer efficiency in the Low-Level Counting Mode (LLC) was calibrated using standard reference tritium solutions. Sample aliquots with a low tritium content were measured in the Low-Level Counting Mode. This special option is used to measure background concentrations of low-energy radionuclides. The total duration of measurements was 2000 minutes. Between the measurements of two samples a background sample was measured.

RESULTS AND DISCUSSION

Table 1 lists the data on tritium content in water samples of the Yenisei River.

It follows from Table 1 that in the period of our investigations the tritium concentration in the Yenisei River water did not exceed 4 Bq/L and this is consistent with the results of the 1998 expedition to this region (4.3±0.2 Bq/L). Previously, the data on the tritium content in the Yenisei River water, based on the measurements made by the Hydrometeorological Service of Russia, were published annually in the records reporting the radiation situation in Russia [9]. So, it was reported that during 1998-99 the mean yearly tritium concentration in the Yenisei water was 3.7-4.1 Bq/L and did not exceed the average global fallout tritium content in water bodies of Russia [5]. Thus, no contribution of the MCC to the contamination of the Yenisei with tritium has previously been detected. In August 1998, during the joint radioecological expedition to the floodplain of the Yenisei River, which involved the MCC specialists, water samples were collected for the determination of the tritium content. The investigations have shown that at the MCC discharge site, in the mouths of the Bolshaya Tel and the Kan Rivers, the tritium concentration does not exceed 4.3±0.2Bq/L. This corresponds to the background tritium content in the surface waters of Russia [5].

We found that in the mouths of the streams and rivers whose catchment areas are within the MCC sanitary-protective zone the tritium concentration was enhanced versus the background values: in the Shumikha River the maximum tritium concentration was 81 Bq/L, in the Ploskii Stream – 168 Bq/L, and in Stream No. 2 – 32 Bq/L (Table 2). Our data are similar to those obtained in 1998: in the Ploskii Stream and the Shumikha River the tritium concentrations exceeded the background values and amounted to 56 and 125 Bq/L, respectively. Other authors did not suggest any explanation for the higher tritium values and assumed that the tritium had been washed off the soil surface in the MCC sanitary-protective zone [5]. We conducted γ-spectrometric measurements of the water samples collected from the mouths of the streams and rivers and revealed the presence of activation radionuclides in the mouths of the Shumikha River and the Ploskii Stream. This suggests that the source of tritium in these surface waters is the last operating reactor at the MCC, which still uses the Yenisei water as coolant. It was assumed earlier that there is only one outlet of the cooling water into the Yenisei River and that this is the Ploskii Stream. However, our measurements of tritium and γ-spectrometry of water samples suggest a conclusion that there are two outlets of the coolant into the Yenisei River.

Table 2 lists the data on tritium content in water samples of tributaries of the Yenisei Rive at different distances downstream from Krasnoyarsk.

In all the samples collected from the Bolshaya Tel River during the sampling period the tritium content was higher than the background values (1.5-2 times in July and August and almost 10 times in September). The maximum tritium concentration in the water of the Bolshaya Tel River was 35 Bq/L.

Table 1. Tritium content (Bq/L) of the water in the Yenisei River (2001-2003)

Sampling position	Sampling time		
	July	August	September
Krasnoyarsk, 0 km*	2.4±0.8	2.8±0.7 (2±1)	2.5±0.6 (2.6±0.5)
Village of Esaulovo, 45 km*	-	4.8±0.9	5±1
Village of Atamanovo, 86 km	3.1±0.4	2.6±0.5	2.7±0.4
Upstream of B. Tel mouth, 94 km	-	3.1±0.3	3.4±0.5
Downstream of B. Tel mouth, 95 km	-	3.9±0.5	4.7±0.6
Village of B.Balchug, 97 km	2.6±0.5	2.5±0.3	3.3±0.4
Tankin Island, 170 km	-	-	3.0±0.4
Village of Zakharovka, 278 km	-	-	3.2±0.3
Village of Abalakovo, 350 km	-	-	2.9±0.4
Krasikov Island, 422 km	-	-	2.8±0.3
Village of Novonazimovo, 600 km	-	-	3.1±0.3

In brackets are the data of the RPA RADON (Moscow).

* *The distances to sampling points are in km, downstream from Krasnoyarsk*

There are two possible sources from which the Bolshaya Tel River may receive tritium. First, it is the soil surface runoff at the water catchment area near the MCC. Second, tritium may come from the deep aquifers at the "Severny" testing site. As reported by the MCC specialists [4, 5], the "Severny" testing site is located 12 km north of the MCC radiochemical plant, at the watershed between the Yenisei and the Bolshaya Tel Rivers (Fig. 1). Liquid radioactive waste is injected into Horizons I and II located at depths of 370-465 m and 180-280 m, respectively. The MCC specialists report that Horizon II is connected to the valley of the Bolshaya Tel River and Horizon I is presumably connected to both the Kan River valley (12-14 km north of the testing site) and the valley of the Bolshaya Tel River, by way of a slow overflow into Horizon II [4, 5]. The testing site was established in 1967. By now about 5 million m^3 of liquid radioactive wastes with the total decay corrected activity of $1*10^{19}$ Bq have been injected into the two aquifers [10, 5]. In addition to long-lived radionuclides, including transuranic elements, tritium is also injected into the underground horizons. Rybalchenko and co-authors reported that in some wells of the testing site the tritium activity concentration amounted to 330000 Bq/L [4]. However, no elevated tritium content was registered either outside the testing site or in the monitoring wells, as reported by the MCC specialists. As we mentioned above, in August 1998, during the expedition involving the MCC specialists, water samples were collected for the determination of the tritium content in order to find out whether there was an exchange of water between the Yenisei tributaries and the "Severny" testing site. The investigations showed that in the mouth of the Bolshaya Tel River the tritium concentration did not exceed the background values, 4.0±0.2 Bq/L [5], and, hence, the river and the "Severny" testing site were not hydrologically connected. Only in one of the Kan River tributaries was the tritium concentration found to be 1.3 times higher than the background – 5.3±0.2 Bq/L [5]. Based on this, the authors of that work suggested a hydrological connection between the Kan River water and the underground horizons of the "Severny" testing site.

Table 2. Average tritium content (Bq/L) of the water in tributaries of the Yenisei River

Sampling position	Sampling time		
	July	August	September
The Shumikha River - mouth, 81 km*	70±10	81±8	75±11
Stream No.2- mouth, 83 km	32±6	27±4	30±5
The Ploskii Stream- mouth, 85 km	168±25	120±21 (46±5)	120±10
The B. Tel River: 94.5 km mouth	-	5.6±0.6	34±5 (55±6)
50 m upstream from the mouth	-	21±3	-
300 m upstream from the mouth	9±1 (5.1±1.5)	6.4±0.7 (6±2)	21±3 (28±4)
500 m upstream from the mouth	-	4.2±0.8	30±10 (15±3)
1000 m upstream from the mouth	-	6±2	28±3
1200 m upstream from the mouth	-	7±2	33±4

In brackets are the data of the RPA RADON (Moscow).

* *The distances to sampling points are in km, downstream from Krasnoyarsk*

To determine the source from which tritium comes into the water of the Bolshaya Tel River, we collected samples of sediments from the Bolshaya Tel and analyzed the pore water. The measurements of the pore water samples at the Institute of Biophysics SB RAS (Krasnoyarsk) showed an elevated tritium content, up to 58 Bq/L (Table 3). The measurements at the RPA RADON (Moscow), which were conducted using the latest Canberra-Packard (USA) instruments, confirmed the high tritium content in the samples, up to 42 Bq l^{-1}, and also registered the artificial radionuclide ^{14}C, with the maximum activity of 14 Bq l^{-1} (Table 3).

Table 3 shows that in the sediment layer 10 to 20 cm the tritium content is several times higher than in the layer 0 to 10 cm. Tritium is known to be very weakly retained by rock-forming structures, clays in particular [11]. That is why sediments accumulate only insignificant amounts of the tritium that has been washed into the river off the contaminated soil surface. In our case the main source of tritium for sediments is the ground water of the "Severny" testing site horizons, which presumably exchange water with the Bolshaya Tel River [4]. Then the tritiated pore water of the upper sediment layer continuously exchanges with the adjacent water layer of the surface water. As a result, the tritium content in the water of the Bolshaya Tel River increases, and our results confirm this (Table 2).

Table 3. The content of the radionuclides ^{3}H (Bq/L) and ^{14}C (Bq/L) in samples of the pore water from sediments (2001-2002)

Sampling position	Depth of layers	September 2001 ^{3}H, Bq/L	September 2002 ^{3}H, Bq/L	^{14}C, Bq/L
The Yenisei River				
Village of Esaulovo	0-10 cm	-	3.8±1	
Village of B.Balchug,	0-10 cm	3.7±2	4±2	
The Bolshaya Tel River				
mouth	0-10 cm	46±5		
300 m from the mouth	0-10 cm 10-20 cm	49±6 (16±3)	25±5 51±6	(14±1.4)
500 m from the mouth	0-10 cm	58±6 (42±6)	11±2	(1.4±1.0)
800 m from the mouth	0-10 cm	24±3	-	
1000 m from the mouth	0-10 cm	19±3	-	

In brackets are the data of the RPA RADON (Moscow).

The specialists at the Ministry of Atomic Energy of the Russian Federation stated [4] that the low velocity of ground water, 5-6 m/year, moving northward, prevents the radioactive waste from moving beyond the boundaries of the "Severny" testing ground. In the same work, however, they reported instances of radionuclides or thermal anomalies being registered in the remote monitoring wells. The proposed reasons were the elevated pumping pressure and the presence of enhanced permeability zones in the stratum. Thus, if radioactive wastes can migrate faster than estimated, it seems obvious that the real velocity of tritium migration under pumping may significantly exceed the calculated velocity. The "Severny" testing site has been in operation for 35 years and by now tritium has reached the surface waters. The Bolshaya Tel River receives significantly more tritium when the level of the ground water is elevated, as was the case in the autumn of 2001, when the Yenisei water was high.

In the book by Rybalchenko et al. [4] it is reported that radioactive wastes are also injected into deep aquifers in the territory of another facility of the Ministry of Atomic Energy – the Siberian Chemical Combine (SCC) at Seversk of the Tomsk Province. In the same publication it is noted that components of radioactive waste, including tritium, were registered in the monitoring wells of the SCC testing site. This fact gives more support to the possibility of tritium migration outside the boundaries of the testing site.

The reference samples of sediments were collected from the Yenisei upstream of the MCC, near Esaulovo. In the pore water of these sediment samples, the tritium content did not exceed the background values – 4 Bq/L (Table 3).

Thus, our data showing that the tritium content in the pore water of the Bolshaya Tel River sediments is at least 10 times higher than the background values for the Yenisei River, again confirm that the Bolshaya Tel River receives tritium from sediments rather than from the water catchment area. So, we can state that there is water exchange between the surface waters and the radioactively contaminated underground horizons of the "Severny" testing site.

To estimate tritium accumulation by aquatic plants, we collected samples of aquatic plants of the Yenisei River both upstream of the MCC near the village of Esaulovo and in the area affected by MCC discharges (the villages of Atamanovo and Bolshoi Balchug). The sampled aquatic plants were of two species: the shining weed (*Potamogeton lucens*) and the Canadian pondweed *(Elodea canadensis*). The data on the tritium content in the Yenisei samples of these aquatic plants are listed in Table 4.

The data obtained suggest that tritium content of the reference plant samples collected near the village of Esaulovo varied between 3 and 13 Bq/L, depending on the sampling season and plant species. Tritium content of the plant samples collected downstream of the MCC (near the village of B. Balchug) was also higher than tritium content of the Yenisei water.

The Canadian pondweed and the shining weed samples collected in the area affected by the MCC effluents in September contained over 100 times more tritium than the water samples collected in the same positions – 410 Bq/L and 490 Bq/L, respectively (Table 4). The explanation for the rather high tritium content of aquatic plants can be as follows.

Tritium in biological samples is usually categorized as free water tritium and tissue-bound tritium, where the latter means organically bound tritium. It is known that organisms incorporate tritium from exposure to ^3H_2O into tissue-free water very rapidly and reach concentrations near that of external medium. Incorporation of tritium into organic matter of cells occurs at a slower rate than incorporation into the tissue-free water and typically reaches a concentration of about half that in the external medium [12]. However, some tritiated organic compounds do have the potential to accumulate in cell organic matter above levels in the tissue-free water. In our study, the organically bound tritium content was operationally defined as the tritium associated with biological macromolecules [13]. This is the tritium that, as a result of biochemical processes in the analyzed samples, was transformed from the free form to the tissue-bound one and was almost completely extracted from the samples by distilling the azeotropic mixture with toluene.

Table 4. Tritium content and tritium concentration factors for the aquatic plants of the Yenisei River

Sampling position	Aquatic plant	Tritium content: in the aquatic plants, Bq/L	Tritium content: in the aquatic plants, Bq/kg wet weight	Tritium content: in the Yenisei River water, Bq/L	Tritium concentration factors for the aquatic plants, L/kg
Village of Esaulovo	*Potamogeton lucens*	7±2	2.2	5±1	0.44
Village of Esaulovo	*Elodea canadensis*	13±4	10	5±1	2
Village of Atamanovo	*Elodea canadensis*	409±10	304	6 29	52 10
Village of Atamanovo	*Potamogeton lucens*	488±25	222	6 29	37 7
Village of B.Balchug	*Elodea canadensis*	7±3	5.4	4±1	1.4
Village of B.Balchug	*Potamogeton lucens*	6.5±2	2.8	4±1	0.7

We have calculated the concentration factors of tritium for the analyzed plant species. The results are listed in Table 4.

If we take the average tritium content in the Yenisei water equal to 4 Bq/L and calculate the concentration factor (CF) of tritium, for the Canadian pondweed it will be more than 1 even in the reference samples collected near the village of Esaulovo, indicating that tritium concentrates in plant biomass and that it seems to concentrate as organically bound tritium [12]. Assuming that the tritium content in the water near the MCC discharge site can vary from 6 to 29 Bq/L, we find that the calculated CF of the Canadian pondweed can be within the range of 10-52 and the CF for the shining weed – 7÷37. Based on the results obtained, we concluded that the concentration factor of tritium for plants can depend on their water content. So, the maximum concentration factor of the Canadian pondweed was higher than that of the shining weed. The water content of the Canadian pondweed was 77% and that of the shining weed – 45%.

CONCLUSIONS

1. The detailed investigation of the Yenisei River samples conducted over the last three years showed that the maximum tritium concentration in the Yenisei water did not exceed 5 Bq/L, which is consistent with the earlier obtained data.
2. There are waterways in which tritium exceeds the background values for the Yenisei River. For instance, in the mouths of the Ploskii Stream and the Shumikha River the tritium concentration amounts to 168 and 81 Bq/L, respectively. In water samples of the B. Tel River, tritium content is 10 and more times higher than the background concentrations of the Yenisei River. The results obtained indicate that besides the global fallout tritium the surface waters of the Yenisei River basin also receive the tritium from other sources:
· the last operating reactor of the MCC, which still uses the Yenisei water as coolant. Activation gamma-radionuclides found in water samples prove the contribution of this source.
· the radioactively contaminated underground horizons of the "Severny" testing site hydrologically connected with the surface waters. Tritium found in the sediments of the Bolshaya Tel River, which exceeds tritium content in reference samples, proves the contribution of this source. The measurements conducted at the RPA RADON (Moscow) revealed not only tritium but also the artificial radionuclide ^{14}C in the Bolshaya Tel samples of the pore water.
3. For the first time tritium has been found in two aquatic plant species of the Yenisei River: the shining weed (*Potamogeton lucens*) and the Canadian pondweed (*Elodea canadensis*). Maximal tritium concentrations were registered in the samples collected at positions near the radioactive discharge site of the MCC – 410 Bq/L (304 Bq/Kg wet weight) for the Canadian pondweed and 490 Bq/L (222 Bq/Kg wet weight) for the shining weed. Tritium concentration factors for the two plant species were also determined for the first time. Concentration factors of tritium for aquatic plants are much higher than 1, indicating the presence of tritium in the plant biomass as organically bound tritium.

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