

## Sources of Cesium-137 Pollution of Bottom Sediments in the Kara Sea Continental Catchment, Russia

Ivan Semenov, Alexey Miroshnikov

The Organization of Russian Academy of Sciences Institute of Geology of Ore Deposits Petrography, Mineralogy and Geochemistry  
semenkov@igem.ru

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**Abstract:** *The aim of the present research is to evaluate the contribution of  $^{137}\text{Cs}$  global fallout to the contamination of bottom sediments in the mixing zone of the Ob fresh water and the Kara sea salt water. Sources of cesium-137 pollution were explored in the Kara sea continental catchment based on modeling and published data. The main sources are global radioactive fallout (24.8 – 25.2  $10^{15}\text{Bq}$ ), Semipalatinsk nuclear test site (1.3 – 37.0  $10^{15}\text{Bq}$ ), PA ‘Mayak’ (liquid radioactive wastes in the Techa river 1.9 – 13.0  $10^{15}\text{Bq}$  and Karachay lake 200 – 444  $10^{15}\text{Bq}$ , Kyshtym disaster 2.7  $10^{15}\text{Bq}$ ), Krasnoyarsk mining and chemical industrial complex (0.01 – 0.11  $10^{15}\text{Bq}$ ). Cesium-137 inflow to the Kara sea from the Ob river basin (including decay) is 0.04 – 0.44  $10^{15}\text{Bq}$ . Global fallout  $^{137}\text{Cs}$  runoff forms 48 – 73% of total  $^{137}\text{Cs}$  runoff. Cesium-137 runoff from the landscapes contaminated due to activity in the Semipalatinsk nuclear test site and PA ‘Mayak’ is 2 – 42% and 10 – 26% correspondingly.*

**Keywords:** *radioactive contamination; radioecology; cesium 137; river basins; radioactive global fallout; PA ‘Mayak’; Siberian Chemical Combine; Krasnoyarsk Mining and Chemical Industrial Complex.*

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### 1. INTRODUCTION

Estuary and deltaic ecosystems are the final components in the transfer system of fluvial sediments. In deltas suspended sediments can precipitate and thus deltas function as marginal filters for the oceans (Lisitsyn, 1994). Cesium-137 precipitation on the “river-ocean” barrier was investigated in the Rhone (Charmasson, 2003), the Ob and the Yenisei (Standing et al., 2008; Miroshnikov, 2013) river estuaries. Strontium-90 precipitation was explored in the Nile delta (Shawky, El-Tahawy, 1999). Trace elements precipitation was studied in the continental shelf of the Basque Provinces (Martins et al., 2012).

Zones of higher  $^{137}\text{Cs}$  activity are being formed in the bottom sediments of the Ob and the Yenisei rivers and in the Kara sea mixing zone due to several sources of sediment transfer. Global cesium-137 fallout runoff supposedly makes a considerable contribution to the Kara sea radioactive contamination. This contribution has not been estimated properly (Aarkrog, 1979; Sayles et al., 1997; Miroshnikov, Semenov, 2012; Miroshnikov, 2013). There are lots of  $^{137}\text{Cs}$  activity sources in the past. As far as recent sources of  $^{137}\text{Cs}$  higher activity in the Kara sea catchment area, they are the following: the Mayak Production Association (PA ‘Mayak’), Siberian Chemical Combine (SCC), Chemical Industrial Complex (CIC), Semipalatinsk and other nuclear test-sites and global radioactive fallout.

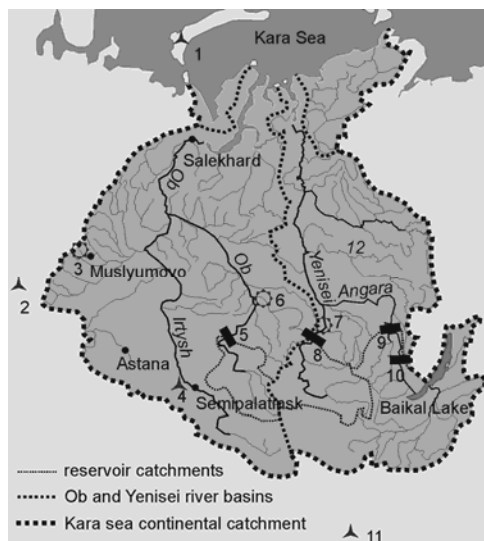
The purpose of the present research is to evaluate the contribution of  $^{137}\text{Cs}$  global fallout to the contamination of bottom sediments in the mixing zone of the Ob fresh water and the Kara sea salt water. For that purpose several research tasks were generated: (i) identifying sources of cesium-137 higher activity in the Kara sea continental catchment, (ii) estimating cesium-137 being transferred in suspended sediments and dissolved form along the Ob fluvial net.

### 2. STUDY AREA

Zones of  $^{137}\text{Cs}$  higher activity are located in the most shallow south-east part of the Kara sea. The morphology of this area is being formed by floods of the Ob and the Yenisei fluvial nets. Maximum levels of cesium-137 activity amount to 100 Bq kg<sup>-1</sup> in the Ob estuary and to 260 Bq kg<sup>-1</sup> in Yenisei estuary (Miroshnikov, 2013).

The largest fresh water runoff (1350 km<sup>3</sup> y<sup>-1</sup> or 56% of the total runoff of the rivers in the Siberian sector of the Arctic ocean catchment) from the biggest sea catchment is typical of the Kara sea. The Ob

runoff is  $530 \text{ km}^3 \text{ y}^{-1}$  and the Yenisei runoff is  $630 \text{ km}^3 \text{ y}^{-1}$  (Ivanov, 1976; Dobrovolsky, Zalogin, 1992). The Ob and the Yenisei river basins are constituent parts of the Kara sea continental catchment (located in Russia, Kazakhstan, Mongolia and China). The Ob and the Yenisei river basins cover 48% and 42% of the Kara sea continental catchment correspondingly (Fig. 1).



**Fig1.** Study area location. Nuclear test sites: 1 – Novaya Zemlya, 2 – Totsky, 3 – Semipalatinsk, 4 – Lop Nor. Nuclear industry: 5 – PA ‘Mayak’, 6 – Siberian Chemical Combine, 7 – Krasnoyarsk Mining and Chemical Industrial Complex. Reservoirs of hydro-electric power stations (HEPS): 8 – Novosibirsk, 9 – Krasnoyarsk, 10 – Bratsk, 11 – Irkutsk. Study areas: 12 – Purpe, 13 – Noyabrsk, 14 – Salym, 15 – Turtas, 16 – Vagai, 17 – Shadrinsk.

The vast Ob river basin includes the rivers entering in the Ob gulf, and spreads from tundra in the north to steppe in the south. It covers 3.5 mln.  $\text{km}^2$ . We define all this area as ‘the Ob river basin’ hereafter.

Cesium-137 runoff decreases from tundra and wetland landscapes (taiga and semi-arid landscapes) to forest-steppe and steppe landscapes in the range of drainage basins according to 10th order river basin classification (R. Horton (1945) coding system). This classification is based on geochemical properties of topsoil and sediment movement rate (Miroshnikov, Semenov, 2012; Semenov, Miroshnikov, 2014; Semenov et al., 2015). It was determined that cesium-137 runoff dramatically increases in the arable land (Belyaev et al., 2013). Mobility and sorption of  $^{137}\text{Cs}$  primarily depend on the presence of clay minerals. Cesium exists in natural water in two forms – water-soluble form and colloidal form. There are no organic colloids with cesium in natural water; only mineral colloids.

The Yenisei river basin is characterized by the small area of boggy grounds (3%) as against the Ob river basin (18%). It can be explained as a consequence of recent bog spreading in Western Siberia (Revenga et al., 1998; Semenov, Miroshnikov, 2014).

The Yenisei river basin includes catchments of the all the rivers flowing into the Yenisei gulf. The basin's territory covers 2.9 mln.  $\text{km}^2$ . Its landscapes are similar to the landscapes in the Ob river basin.

Only 20 – 40% of cesium-137 global fallout are being transferred with suspended matter in the Yenisei, in the Angara and in the Podkamennaya Tunguska river basins that covered by taiga landscapes. This index amounts to 50 – 80% in the Don and in the Seym rivers which basins are located in the arable steppe landscapes. The higher ratio of a drainage lake watershed in the river basin is, the smaller ratio of cesium-137 transferred with suspended matter by rivers is. For example, only 8 – 19% of cesium-137 global fallout are being transferred with suspended matter in the Neva and in the Narva rivers (taiga landscapes in the north-west of the Russian plain) which drainage basins include vast lakes (Bochkov et al., 1983). Suspended matter movement in HEPS reservoirs (Fig.1) is similar to the same one in drainage lakes. In these lakes suspended material can precipitate (Vakulovsky et al., 2008). Therefore,  $^{137}\text{Cs}$  adsorbed on suspended particles can precipitate too.

The total area of the HEPS reservoirs catchment is 1.1 mln.  $\text{km}^2$  in the Yenisei river basin and 0.23 mln.  $\text{km}^2$  in the Ob river basin. Several hydroelectric power stations were built before the most

intensive global radioactive fallout – Irkutskaya (1956), Bratskaya (1957), Novosibirskaya (1961) and Krasnoyarskaya (1963). All these power stations are located in the south of the Kara sea continental catchment (Fig.1). Nowadays all steppe and semi-arid landscapes in the Yenisei river basin are located in the reservoir catchments of hydroelectric power stations.

Study of factors of cesium-137 pollution is essential to profound investigation of the Ob river basin as a main source of radionuclide contamination in the Kara Sea as  $^{137}\text{Cs}$  global fallout accumulation is less in the Ob river basin as compared with the Yenisei river basin.

### 3. MODELING OF GLOBAL FALLOUT $^{137}\text{Cs}$ TRANSFER FROM LANDSCAPES OF THE OB RIVER BASIN TO THE KARA SEA

Different sources of radioactive contamination of environment are located in the north and in the central part of Asia. Potential sources of  $^{137}\text{Cs}$  contamination of the Kara sea continental catchment were chosen based on Perelman's data (Perelman et al., 1996). Then the contribution of the sources to  $^{137}\text{Cs}$  contamination of the Kara Sea continental catchment was evaluated according to different published material.

Study areas of the 6 first-order river basins (Horton encoding system (Horton, 1945)) were chosen based on the classification of the 10<sup>th</sup> order river basins, developed by the authors (Miroshnikov, Semenov, 2012; Semenov, Miroshnikov, 2014). ‘Purpe’ study area is covered by natural landscapes with permafrost, pine forests and peat bogs (Fig.1). Landscapes covered by pine forests and peat bogs are typical of ‘Noyabrsk’ and ‘Salym’ study areas (Semenkov, Usacheva, 2013; Semenov et al., 2015). ‘Turtus’ study area is covered by pine forests. ‘Vagai’ and ‘Shadrinsk’ study areas are located in arable lands replaced by forest-steppe landscapes (Miroshnikov, Semenov, 2012; Semenov, Miroshnikov, 2014; Semenov et al., 2015).

Fixed-size soil samples (n=400) were collected from soil profiles. The soil samples were dried in the oven to constant weight at the temperature of 105°C and then they were analyzed. Cesium-137 activity was measured by the  $\gamma$ -spectrometric method in the Laboratory of Radiogeology and Radiogeocology IGEM RAS. Measurements were carried out on the low-background  $\gamma$ -spectrometric complex with a semiconductor Ge(Li)-detector GEM-4519 (GLP-25300/13) and a 8000-channel amplitude analyzer 919 EG&G ORTEC, followed by processing  $\gamma$ -spectrometric data. The exposure time was 7200 s. In order to control measurement accuracy, a scintillation  $\gamma,\beta$ -spectrometric complex, equipped with NaI(Tl)-detectors 160×160 mm with traps 55×110 mm was used. The exposure time was 3600 s.

Average  $^{137}\text{Cs}$  inventory (E) was calculated for watershed, slope and bottom of the first-order river basin, according to the following formula:

$$E = \frac{\sum_n \left( \sum_i \frac{A_i * m_i}{S_i} \right)}{n} \quad (1)$$

where  $A_i$  is specific  $^{137}\text{Cs}$  activity in soil samples (Bq kg<sup>-1</sup>) collected in  $S_i$  area (m<sup>2</sup>) from n cross-overs,  $m_i$  is weight of a  $i$ -bulk-core soil sample.

Then we calculated  $^{137}\text{Cs}$  total inventory in each first-order river basin that amounts to the sum of  $^{137}\text{Cs}$  inventory in watershed, slope and bottom. Cesium-137 runoff from the first-order river basin was calculated as the difference between decay corrected estimation of total global fallout  $^{137}\text{Cs}$  in the latitudinal band (Sources., 2010) and average  $^{137}\text{Cs}$  inventory (E) in the first-order river basin.

Global fallout  $^{137}\text{Cs}$  runoff from the Ob River basin was calculated, according to the following the formula:

$$R = \sum_{n=1}^{14} [(((S_1 * r) + S_2 * r) * k_2 + \dots + S_n * r) * k_n + \dots + S_{14} * r] k_{14} \quad (2)$$

where  $S_n$  is the total area of n-order river basins; r is total activity of  $^{137}\text{Cs}$  (Bq) entered in a  $n^{\text{th}}$  order river bed from it slopes;  $k_n$  is  $^{137}\text{Cs}$  delivery ratio from a  $(n-1)^{\text{th}}$  order river to a  $n^{\text{th}}$  order river, which varies from 0.1 to 0.95. Cesium-137 delivery ratio for n-order river drainage basins were not calculated before. Based on the assumption, that there are no geochemical barriers on the ways of migration of water-soluble and colloidal  $^{137}\text{Cs}$  forms in the rivers of the Ob basin, we assume  $^{137}\text{Cs}$

delivery ratio from the  $n$ -order basin to the  $(n+1)$  order basin to be '0.95' (Semenkov, Miroshnikov, 2014).

Transfer of  $^{137}\text{Cs}$  tightly adsorbed forms depends on sediment flow. The most intensive  $^{137}\text{Cs}$  migration is characteristic for land use areas. When the river order increases, the slope of long-edge profile decreases (Golosov, Panin, 2006). As a result, suspended sediments' flow decreases and consequently  $^{137}\text{Cs}$  concentrations increase. On plowed territories the process of silting-up of minor rivers (which refer to 5<sup>th</sup> – 7<sup>th</sup> orders according to Horton's classification) takes place. Suspended sediments transferred by the river are also accumulated in the upper ponds of reservoirs and drainage lakes. Thus,  $^{137}\text{Cs}$  is accumulated in the areas of suspended sediments' deposition. These areas are the 5<sup>th</sup> – 7<sup>th</sup> order rivers, the upper ponds of reservoirs and drainage lakes. In this regard we assume cesium-137 delivery ratio as 0,95 for all the rivers which watersheds are not plowed up and plowed up watersheds (including the 2<sup>nd</sup> – 5<sup>th</sup> order and the 7<sup>th</sup> – 14<sup>th</sup> order rivers). For the plowed watersheds of the 6<sup>th</sup> order rivers we assume  $^{137}\text{Cs}$  delivery ratio as 0.1 (Semenkov, Miroshnikov, 2014).

An area ratio of the  $(n-1)$ <sup>th</sup>,  $n$ <sup>th</sup> and  $(n+1)$ <sup>th</sup> order river basins (Horton encoding system (Horton, 1945)) is 6:3:1 (Horton, 1945; Simonov, 2008). We calculated total area of river basins with an order from the 1<sup>st</sup> ( $S_1$  in the equation 2) to the 9<sup>th</sup> ( $S_9$ ) and from the 11<sup>th</sup> ( $S_{11}$ ) to the 14<sup>th</sup> ( $S_{14}$ ) based on GIS approach to the Ob River basin and area ratio calculating. The total square of river watersheds was calculated for the 1<sup>st</sup> – 9<sup>th</sup> order rivers ( $S_1$  and  $S_9$  correspondingly in the formula 2) and for the 11<sup>th</sup> – 14<sup>th</sup> order rivers. As mentioned above, the present approach is based on GIS analysis of the Ob watershed and area ratio (the ratio of watershed squares of different orders).

## **4. RESULTS & DISCUSSION**

### **4.1. Cesium-137 Sources in the Kara Sea Continental Catchment**

Lots of cesium-137 sources such as nuclear test sites and nuclear industry are located in the Kara sea catchment.

#### *4.1.1. Nuclear Test Sites and Global Fallout*

After nuclear weapon tests 1,480  $10^{15}\text{Bq}$  of cesium-137 were released in the environment (Sources..., 2010). Two nuclear test sites are located in the Kara sea catchment near Semipalatinsk (Kazakhstan) and in the Novaya Zemlya archipelago (Russia). The Lop Nor nuclear test site is located in China, in about 600 km from the Ob river watershed (Fig. 1).

6.7 – 6.9  $10^{15}\text{Bq}$  and 5.2  $10^{15}\text{Bq}$  of decay corrected cesium-137 has been deposited in the Ob and the Yenisei river basins respectively, according to the present estimations (Semenkov, Miroshnikov, 2014) based on  $^{90}\text{Sr}$  global fallout in latitudinal zones and the  $^{90}\text{Sr}/^{137}\text{Cs}$  ratio in the surface air (Sources..., 2010).

In 1954 it was carried out nuclear testing in the air in the Totsky test site (the USSR, South Ural, 300 km from the Ob river watershed) (Kagirov, 2013). Based on different assessments, that testing could have produced 0.23  $10^{15}\text{Bq}$  of cesium-137 (Chasnikov, 1996; Ihnashov et al., 2007) which could have deposited around the place of the nuclear bomb testing (Kagirov, 2013).

On the Novaya Zemlya archipelago about 592  $10^{15}\text{Bq}$  of radionuclides were generated after nuclear weapon tests (Chasnikov, 1996; Mihailov, 2006; Ihnashov et al., 2007). It is known that those tests were carried out at the time of strong southern and southwestern winds (Ihnashov et al., 2007). The winds negligibly contributed to radioactive contamination of the Ob river basin (Shcherbov et al., 2001).

The total power of nuclear explosions near Semipalatinsk from 1949 to 1963 amounted to 50 Mt (Chasnikov, 1996; Ihnashov et al., 2007). Therefore 0.31 – 0.33  $10^{15}\text{Bq}$  of cesium-137 could be produced according to estimations (a test of 1Mt nuclear bomb produces 0.0058 – 0.0068  $10^{15}\text{Bq}$  of  $^{137}\text{Cs}$  (Chasnikov, 1996; Ihnashov et al., 2007)). Other estimations show that 888  $10^{15}\text{Bq}$  of cesium-137 were released into the atmosphere in Semipalatinsk nuclear test site since 1949 (Spiridonov et al., 2005).

According to Yu. Izrael (1998) and P. Strand (1998) 12% of radionuclides released into the atmosphere after nuclear weapon tests have formed local radioactive fallout. Therefore, 35.2 – 37.0  $10^{15}\text{Bq}$  (Chasnikov, 1996; Ihnashov et al., 2007) or 10.7  $10^{15}\text{Bq}$  (Spiridonov et al., 2005) of cesium-137 could have deposited in the Ob river basin. These calculations don't include radioactive nuclides that can be transferred by the rivers.

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Several nuclear testing in the air were carried out in the Lop Nor test site from 1964 to 1980. As long as the test site was located far from the Ob river basin and it was separated by the mountains of Central Asia, there are not any estimations of radioactive contamination from Lob Nor tests in the Ob river basin (Zhumadilov et al., 2011).

### 4.1.2. Nuclear Industry

**Table1.** Cesium-137 input into the Kara sea continental catchment

Sources			Time frame of the main input or distinguish time frame	Activiry		References
				Total, 10 <sup>15</sup> Bq	Decay corrected (2014), 10 <sup>15</sup> Bq	
Nuclear weapon tests	Nuclear weapon test sites	Novaya Zemlya archipelago	1954 – 1962	negligible		Chasnikov, 1996; Shcherbov et al., 2001; Mihailov, 2006; Ilnashov, 2007
		Semipalatinsk	1949 – 1963	35.1 – 37.7	8.7 – 12.5	Chasnikov, 1996; Ilnashov, 2007
				1.3	0.33 – 0.44	Aarkrog et al., 1997
				292	2.7 – 3.6	Spiridonov et al., 2005
	Lop Nur	1964 – 1980	not available			
	Totsky	14.09.1954	negligible		Kagiroy, 2013	
Global fallout	Ob river basin	1945 – 2014	14.0 – 14.4	6.7 – 6.9	Semenkov, Miroshnikov, 2014 based on Sources (2010)	
	Yenisei river basin	1945 – 2014	10.6	5.2	Authors estimation based on Sources (2010)	
Radiochemical plants	PA ‘Mayak’	Liquid radioactive wastes into the Techa river	Before 12.1949	0.10	0.02	Il’in, 2005a, 2005b
			12.1949 – 03.1950	0.48	0.11	
			03.1950 – 27.10.2951	11.7	2.9	
			27.10.1951 – 1952	0.59	0.15	
			1949 – 1952	12.2	3.2	
			1949 – 1956	13.0	3.3	Report., 1991; Trapeznikov et al., 1993, Kryshev et al., 1998
			1949 – 1956	1.9	0.48	Christensen et al., 1997
			Mokrov, 2006			
	Kyshtym disaster	1957	0.27	0.078	Shafikov, 2011	
	Karachai disaster	1967	0.00089	0.00026	Christensen et al., 1997	
Siberian Chemical Combine	Liquid radioactive wastes into the Romashka river	Since 1952	negligible		Gauthier-Lafaye et al., 2008; Shafikov, 2011	
	Disaster	06.04.1993	0.0040	0.0025	IAEA, 1998	
Krasnoyarsk Mining and Chemical Industrial Complex	Liquid radioactive wastes into the Yenisei river	1958 – 1992	0.02–0.11	0.070	Vakulovsky et al., 2008	
		1975 – 2002	0.031	0.25	Vakulovsky, 2003; Semizhon et al., 2010	

Daily functioning of the PA “Mayak” and several breakdowns are the sources of radionuclide contamination of the environment (Table 1). There are several calculations for the Techa river radioactive contamination (Report., 1991, Trapeznikov et al., 1993, Christensen et al., 1997; Kryshev et al., 1998; Alexahin, 20005; Ilin, 2005a, 2005b; Mokrov, 2006; Glagolenko et al., 2007; Kuznetsov, 2012, 2013; Kagiroy, 2013). The results of water radioactivity monitoring from 1951 to 1956 were published in 1997 (JNREG, 1997) and 2005 (Ilin, 2005b). Monitoring consists in monthly, daily and average measuring of β-radioactivity in water. There are 8 monitoring points on the Techa river. Besides, radionuclide analysis of the ‘Mayak’ PA wastes are conducted. According to Ilin's

reconstruction (Ilin, 2005b), 13.0  $10^{15}$ Bq of cesium-137 were discharged into the Techa river from 1949 to 1952 (Table 1).

In contrast to Ilin's data (Ilin, 2005b), Mokrov's reconstruction (Mokrov, 2005) shows that, cesium-137 discharge into the Techa river was only 1.9  $10^{15}$ Bq. According to another data (Kagirov, 2013), PA 'Mayak' discharged 99.9  $10^{15}$ Bq of  $\beta$ -radioactive wastes (predominantly  $^{89,90}\text{Sr}$  and  $^{137}\text{Cs}$ ) from 1949 to 1956. Other radioactive pollution estimations for the Techa river are similar to Ilin's data (Ilin, 2005a, 2005b). Total  $^{137}\text{Cs}$  contamination of the Techa river was 12.2 – 13.0  $10^{15}$ Bq in 1942 – 1952 (Report., 1991, Trapeznikov et al., 1993, Christensen et al., 1997; JNREG, 1997; Kryshev et al., 1998; Kuznetsov, 2012, 2013). Daily  $\beta$ -radioactive wastes released into the Techa river amounted to tens  $10^{12}$ Bq before March 1950 and after March 1950 they reached 0.148  $10^{15}$ Bq (Alexahin, 2005).

Discharge of the liquid radioactive wastes into the Techa River was stopped in October 1951. It was reduced by having built a cascade of reservoirs and having constructed various beheaded river channels at the site. Bog lake Karachay was the first constructed reservoir.

In 1957 the breakdown happened in the PA 'Mayak' - the Kyshtim disaster. An explosion and subsequent continues burning of a tank containing radioactive waste. 0.27  $10^{15}$ Bq of  $^{137}\text{Cs}$  was deposited after the disaster which resulted in the formation of the East-Urals Radioactive Trace (EURT) (0.078  $10^{15}$ Bq decay corrected, according to (Shafikov, 2011)). Cesium-137 inventory was 0.067  $10^{15}$ Bq in the EURT (Mikhaylovskaya et al., 2011).

In 1967 dispersion of radioactive wastes took place around lake Karachay. Wind spread dust contained 0.022 – 0.222  $10^{15}$ Bq of  $^{137}\text{Cs}$  from the bottom sediments of dried up lake Karachay in 1967. This contaminated area is located in the catchment of the industrial cascade of reservoirs isolated from the Ob river network.

Karachay Lake is now largely covered in concrete. Now, the potential for overland flow into nearby water systems or redistribution of contaminated sediment (flowing a dry period as exemplified by the 1967 situation) is arguably vanishingly small.

At present lake Karachay has been filled by concrete constructions, so there is the lowest risk of surface liquid runoff from the lake to the Ob fluvial network or risk of wind spreading of radioactive dust on surrounding areas.

There are two nuclear plants inside the Kara sea watershed apart from PA "Mayak" - Siberian Chemical Combine and Krasnoyarsk Mining and Chemical Industrial Complex. Siberian Chemical Combine functioning resulted in radioactive pollution of the Romashka River (the Tom river tributary, the Ob river basin). However, cesium-137 activity was negligible in radioactive wastes of the Siberian Chemical Combine (Leonova et al., 2006; Gauthier-Lafaye et al., 2008; Shafikov, 2011). Emergency with local consequences was in the Siberian Chemical Combine on 6 April 1993. The total  $^{137}\text{Cs}$  activity released into the environment as a result of the emergency amounted to 0,004  $10^{15}$  Bq (IAEA, 1998). Direct-flow cooling cycle reactor was functioning in the Krasnoyarsk Mining and Chemical Industrial Complex (KMCIC) from 1958 to 1992. The Krasnoyarsk Mining and Chemical Industrial Complex discharged 0.031  $10^{15}$ Bq of cesium-137 into the Yenisei River (Vakulovsky, 2003; Semizhon et al., 2010).

#### 4.2. Sources of Cs-137 Inflow into the Ob and the Yenisei Fluvial Network

Cesium-137 supplies in the Ob river basin are the potential sources of its inflow into the fluvial network and potential pollutants of the Kara Sea catchment. Other sources of  $^{137}\text{Cs}$  inflow will be considered below (Table 1).

##### 4.2.1. Global Radioactive Fallout Runoff

Nuclear testing which released into the atmosphere  $^{137}\text{Cs}$  is the main factor of radionuclide contamination of bottom sediments in the Ob River and the Taz River (according to the  $^{139,240}\text{Pu}/^{137}\text{Cs}$  ratio in the sediments). Contribution of other sources of contamination amounts to less than 25% (Sayles et al., 1997). However, in preceding estimations direct radioactive impact (radioactive fallouts from the Novaya Zemlya test site) and indirect radioactive impact (transfer of the global fallout from the river basins) were not evaluated separately.

Total  $^{137}\text{Cs}$  runoff from the Kara sea continental catchment is 0.11  $10^{15}$ Bq since 1961 according to  $^{90}\text{Sr}$  activity in the Ob River near Salekhard (Fig. 1) and the  $^{90}\text{Sr}/^{137}\text{Cs}$  ratio in river water

(Contamination..., 1995). Based on the global radioactive fallout in latitudinal bands (Sources, 2010) and A. Aarkrog's calculations (Aarkrog, 1979),  $^{137}\text{Cs}$  runoff from the Ob river basin to the Kara sea is  $0.17 \cdot 10^{15}\text{Bq}$  (no decay correction) (Contamination..., 1995). According to GIS-based approach to the Ob river basin and field research in 6 first order river basins (R. Horton (1945) coding system), cesium-137 runoff from the Ob river basin is  $0.04 - 0.44 \cdot 10^{15}\text{Bq}$  (decay corrected  $0.001 - 0.008 \cdot 10^{15}\text{Bq}$  in 2014) (Semenkov, Miroshnikov, 2014).

#### 4.2.2. Runoff from $^{137}\text{Cs}$ Contaminated Areas Forming As A Result of the PA 'Mayak' Functioning and Nuclear Testing Near Semipalatinsk Test Site

Cesium-137 can be transferred to the Kara Sea from the landscapes located in the area of East-Urals Radioactive Trace. Higher levels of  $^{137}\text{Cs}$  activity were identified for that research area. Cesium-137 runoff amounts to 10% of the total inventory in the first order river basin (R. Horton (1945) coding system) covered by taiga landscapes which are typical of the East-Urals Radioactive Trace (Semenkov, Miroshnikov, 2014). Decay corrected total cesium-137 runoff from the first-order river basins to the second order river basins (Horton coding system) amounts to  $0.027 - 0.041 \cdot 10^{15}\text{Bq}$  in the EURT. Cesium-137 runoff from the East-Urals Radioactive Trace is  $0.006 - 0.014 \cdot 10^{15}\text{Bq}$  ( $0.004 - 0.006 \cdot 10^{15}\text{Bq}$  decay corrected) given  $^{137}\text{Cs}$  delivery rate (Semenkov, Miroshnikov, 2014).

The Semipalatinsk test site is located in the area covered by inland lake basins (70 – 90% of area) which is not connected with the Ob river network. Such location conditions are favorable for radionuclide accumulation in that area. According to published material, after nuclear tests near Semipalatinsk  $0.13 - 11.32 \cdot 10^{15}\text{Bq}$  of cesium-137 has entered and flowed into the fluvial network (Chasnikov, 1996; Izrael, 1998; Strand, 1998; Spiridonov et al., 2005; Ilnashov et al., 2007). In semi-arid landscapes (like Semipalatinsk test site) cesium-137 mobility is limited by alkalinity of top soils (Shcheglov et al., 2001). It is also limited by decrease of average percentages of illite-vermiculite minerals in topsoils of the Ob River catchment from the north to the south (Champley, 1989). Cesium-137 runoff amounts to 10% of total inventory from landscapes of the first order river basin with slightly acidic soils (Semenkov, Miroshnikov, 2014) with medium percentages (50 – 70%) of illite-vermiculite minerals in topsoils (Champley, 1989).

According to preliminary assessment  $0.013 - 1.132 \cdot 10^{15}\text{Bq}$  of cesium-137 were transferred to the network of the second order rivers of the territory. Only  $0.007 - 0.592 \cdot 10^{15}\text{Bq}$  ( $0.002 - 0.181 \cdot 10^{15}\text{Bq}$  decay corrected) of cesium-137 deposited near the Semipalatinsk test site could have been transferred to the Kara sea given the  $^{137}\text{Cs}$  delivery rate (Semenkov, Miroshnikov, 2014).

According to A. Aarkrog's data (Aarkrog, 1979), global fallout of  $^{137}\text{Cs}$  runoff from the landscapes is 2% of the total inventory. His data shows that  $0.003 - 0.226 \cdot 10^{15}\text{Bq}$  ( $0.001 - 0.070 \cdot 10^{15}\text{Bq}$  decay corrected) of cesium-137 deposited near the Semipalatinsk test site could have been transferred to the Kara sea.

#### 4.2.3. Runoff from Nuclear Industry

Cesium-137 inflow from the Techa river into the Iset river was  $0.044 - 0.048 \cdot 10^{15}\text{Bq}$  (Fig. 1) from 1949 to 1956 (Ilin, 2005b). It was a period of the largest radioactive wastes discharging.

According to Mokrov's (Mokrov, 2005; 2006) reconstruction, Cesium-137 runoff through the cross section in Muslumovo (Fig.1) was  $0.207 \cdot 10^{15}\text{Bq}$  in 1949 – 1954. 11% of the total inventory of  $^{137}\text{Cs}$  were discharged by the PA 'Mayak' and were transferred from the Techa River to the Iset River.

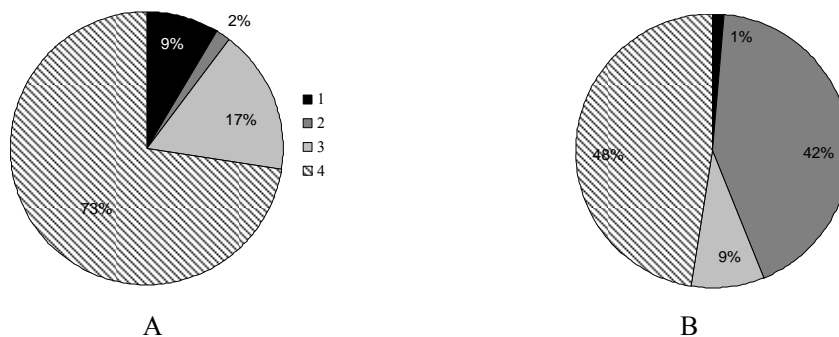
As mentioned above, PA 'Mayak' discharged the largest amount of liquid radioactive wastes ( $0.044 - 0.207 \cdot 10^{15}\text{Bq}$  of cesium-137) to the river network (Table 1). According to Semenov and Miroshnikov data (Semenkov, Miroshnikov, 2014),  $^{137}\text{Cs}$  delivery ratio from the 9<sup>th</sup> order river (such as the Techa River) to the Kara sea amounts to 75%. Therefore, Cesium-137 runoff to the Kara sea is  $0.033 - 0.159 \cdot 10^{15}\text{Bq}$  ( $0.007 - 0.037 \cdot 10^{15}\text{Bq}$  decay corrected).

## 5. CONCLUSIONS

According to the analyzed data and calculations, the smallest cesium-137 runoff from the Kara sea continental catchment amounts to  $0.044 \cdot 10^{15}\text{Bq}$ , the highest one is  $0.444 \cdot 10^{15}\text{Bq}$  with decay correction. Transfer of the global radioactive fallout is the main source of radioactive pollution of bottom sediments in the fresh and salt water mixing zone in the Kara Sea (Fig. 2).

Fig.2. Cesium-137 input to the fresh and salt water-mixing zone of the Kara sea (decay corrected)

estimation). A – the smallest input, B – the highest input from the sources located in the Ob river basin: 1 – the East-Urals Radioactive Trace, 2 – radiochemical plants (PA ‘Mayak’ predominantly), 3 – landscape contaminated due to activity in the Semipalatinsk nuclear test site, 4 – global radioactive fallout.



In order to make more accurate calculations of  $^{137}\text{Cs}$ -pollution additional research is required: (i) monitoring of cesium-137 inventory in the Romashka river floodplain and in the Techa river floodplain (Chesnokov et al., 2000; Linnik, 2011), (ii) identification of the radioactive traces formed as a consequence of nuclear tests in Semipalatinsk, (iii) study of  $^{137}\text{Cs}$  redistribution in the main types of landscapes in the Ob river basin.

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#### AUTHORS' BIOGRAPHY



**Ivan Semenkov**, Junior Research Scientist

*semenkov@igem.ru*



**Alexey Miroshnikov**, Ph.D. Senior Research Scientist

*almir@igem.ru*