EVALUATION OF DISTRIBUTION COEFFITIENTS AND CONCENTRATION RATIOS OF ^{90}Sr AND ^{137}Cs IN THE TECHA RIVER AND THE MIASS RIVER

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Abstract

Empirical data on the behavior of radionuclides in aquatic ecosystems are needed for radioecological modeling, which is commonly used for predicting transfer of radionuclides, estimating doses, and assessing possible adverse effects on species and cenosis. Preliminary studies of radioecological parameters distribution coefficients and concentration ratios for ^{90}Sr and ^{137}Cs showed not a full agreement with the default values (obtained based on the geological data on stable elements) used in the Erica Tool and the Resrad Biota codes. Unique radiation situation in the Techa River contaminated by long-lived radionuclides (^{90}Sr and ^{137}Cs) in the mid of the last century allow to improve the knowledge about these parameters for the river systems. Therefore the study was focused on the evaluation and of radioecological parameters for the Techa River and the Miass River, viz., distribution coefficients and concentration ratios for ^{90}Sr and ^{137}Cs. To achieve the aim the current contamination of biotic and abiotic components of the river ecosystems was studied; distribution coefficients for ^{90}Sr and ^{137}Cs were calculated; concentration ratios of ^{90}Sr and ^{137}Cs for 3 fish species (roach, perch and pike), gastropods and filamentous algae were evaluated. Obtained results were compared with default values used in the well-known computer codes ERICA Too and RESRAD BIOTA. It was shown that the concentration ratios of ^{137}Cs in fish bodies depend on the predominant type of nutrition (carnivores and phytophagous). The concentration ratios of 90Sr are species-specific and strongly depended on Ca^+ concentration in water. The universal characteristic allowing to combine the data of fish caught in the water with different mineralization was proposed by multiplying CR^{Sr-90} and concentration of Ca^+. The individual variability of concentration ratios for fishes was well-fitted by Generalized Logistic Distribution function (GDL). The GDL can be used for probabilistic modeling of the concentration ratios in fishes.

Keywords - Aquatic ecosystems, Background radiation, Distribution Coefficient, Concentration Ratio, Strontium-90, Cesium-137

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1. Introduction

Empirical data on the behavior of radionuclides in aquatic ecosystems are needed for radioecological modeling, which is commonly used for predicting transfer of radionuclides, estimating doses, and assessing possible adverse effects on species and cenosis. Important indicators of radionuclide transfer are the Distribution Coefficient (*Kd*) and the Concentration Ratios (*CR*). The *Kd* parameter is a factor related to the partitioning of a contaminant between the solid and aqueous phases. The *Kd* metric is the most common measure used in dosimetric and transport codes to describe the extent to which contaminants are sorbed by soils (EPA, 1999). Particularly, for aquatic ecosystems, the well-known computer codes ERICA Tool (Brown et al., 2008) and RESRAD BIOTA (USDOE, 2004) use *Kd* to describe the relative activity concentrations of radionuclides in sediment and water (Eqn. 1). The distribution coefficient is used to predict radionuclide activity concentration in sediment based on that in water, or vice versa, if data on one of them are lacking (Beresford et al, 2008).

$$Kd(l\,kg^{-1}) = \frac{\text{Activity concentration in sediment (Bq kg}^{-1}\text{ dry weight)}}{\text{Activity concentration in filtered water (Bq l}^{-1}\text{)}} \quad (1)$$

The Concentration Ratio values (*CR*) are defined as an equilibrium radionuclide activity concentration in a whole organism divided by that in the media (Howard et al., 2013). For aquatic systems, these are the ratios (Eqn. 2) of species-specific raduonuclide concentrations in organisms (expressed in Bq g^{-1}) and in filtered water (Bq l^{-1}) (Hosseini et al., 2008); sometimes the *CR* is termed the concentration factor (Vanderploeg et al., 1975, Smith et al., 2012) or the bioaccumulation factor (Davies. and Dobbs, 1984).

$$CR(l\,kg^{-1}) = \frac{\text{Activity concentration in organism (Bq kg}^{-1}\text{ fresh weight)}}{\text{Activity concentration in filtered water (Bq l}^{-1}\text{)}} \quad (2)$$

In this study, the data collected on two river ecosystems in the Southern Urals (Russia) were used to investigate these two parameters for radionuclides typical of the Urals radiation situation (^{137}Cs and ^{90}Sr).

One of the rivers under study was the Techa River which is flowing through a complex of hydro-technical constructions of the Mayak production association (the first Russian nuclear plant) in Chelyabinsk region and is slowly leaking to its confluence with the Iset River in the Kurgan region (Fig. 1). The Techa River was highly contaminated in the early period of the Mayak operation (1949-1956) due to both routine and accidental releases of liquid radioactive wastes. The total amount of released activity was equal to 114 PBq; about 98% of the radioactivity was dumped within a short time period from September 1950 to October 1951 (Degteva et al. 2012). The waste cocktail contained both short-lived isotopes (^{95}Zr, ^{95}Nb, ^{103,106}Ru, ^{141,144}Ce, ^{91}Y, ^{98}Sr, ^{140}Ba) and the long-lived ^{90}Sr and ^{137}Cs (their half-life times are equal to 28.79 yrs. and 30.07 yrs., respectively). After 1952, the radiation exposure of aquatic biota was mostly determined by the long-lived radionuclides. Even now, after the expiration of the term of ^{9O}Sr and ^{137}Cs 2 half-lives, the contaminations in the upper river region (about 70 km from the source of releases) is still rather high (Trapeznikov et al., 1993; Akleev et al., 2000; Balonov et. al., 2007; Melnikov et al., 2012). The releases resulted in the deposition of the radionuclides in the bottom sediments and floodplain soils along the stream which became a source of a secondary contamination of the river water. The ^{137}Cs to ^{90}Sr ratio of concentrations as well as the concentrations themselves are decreasing with the distance from the source of

releases. Therefore, in the framework of the current study, the sampling was performed at three sites, which were located at different distances from the site of releases.

The second river under study was the Miass River, which flows mostly in Chelyabinsk and Kurgan regions and joins the Iset River similar to the Techa River. However, the Miass River was contaminated by anthropogenic radioactivity only due to global fallouts from the above-ground nuclear weapons testing. This contamination, which peaked in the mid-1960s, can be assumed as homogeneous throughout the river basin. Therefore, the sampling was done within the stream range from 19 to 86 km downstream of the dam of the Argazi Reservoir (Fig. 1). In the radioecological studies of the Techa River the Miass River is considered as a comparison waterbody. Concentration ratios were evaluated for three species of fishes which are the most common in these rivers, viz. perch (*Perca fluviatilis*), roach (*Rutilus rutilus*) and pike (*Esox Lucius*). Besides fishes, representatives of benthic organisms (the mixture of different species of gastropod mollusks) and aquatic plants (filamentous algae) were also studied.

Thus, to achieve the aim evaluating the Distribution Coefficients and the Concentration Ratios of ^{90}Sr and ^{137}Cs in the river ecosystems, the following tasks were solved:

1) evaluation of the current contamination of biotic and abiotic components of the river ecosystems;

2) evaluation of distribution coefficients for ^{90}Sr and ^{137}Cs (Kd^{Sr-90} and Kd^{Cs-137});

3) evaluation of concentration ratios of ^{90}Sr and ^{137}Cs (CR^{Sr-90} and CR^{Cs-137}) for 3 fish species (roach, perch and pike), gastropods and filamentous algae.

2. Materials and Methods

2.1. Description of sampling sites

The Techa River belongs to small rivers (the Strahler stream order is 3) with an annual mean flow at the mouth about 4m^3/s (0.7m^3/s - 9.3 m^3/ s according to CPL, 1984). Its total length is about 200 km. The Techa River is a right tributary of the Iset River and it has several small tributaries that usually dry up in summer time. The river depth varies from 0.2-0.5 m at the shallows, and up to 3 m in backwaters; its width is in the range of 15-20 m. Some reaches of the Techa River are frozen through to the bottom in cold winters. The Miass River is larger than the Techa River (the Strahler stream order is 4) with an annual mean flow at the mouth about 15.4 m^3/s, and its length is 658 km. There are several small tributaries, ponds and reservoirs in the basin of the river. The depth of the river varies from 0.2 m in the shallows and up to 7 meters in the reaches. The seasonal maximums of the water flow rates for both rivers are usually observed in April. Annex 1 presents different hydrochemical indexes of water at sampling station, measured in summer 2012.

Five fishing expeditions were undertaken. The first one was launched in September 2011; the second and forth ones were accomplished in the spring (late in April and early in May) of 2012 and 2013; the third and fifth ones took place in the fall (late in August and early in September) of 2012 and 2013. Figure 1 shows the locations of sampling sites. The sites along the

Techa River are indicated as RT; the sites on the Miass River are marked as RM. Taking into account the river is a dynamic system whose water course can change and the shoreline is characterized by considerable seasonal and climate-driven differences, the sampling sites were not fixed points.

The RT1 is in the upper part of the stream of the Techa River. It is the place closest to the source of the radioactive releases (5 km below the dump, which is the border of the hydro-technical constructions isolating a higher contaminated area on the river). The positions of sampling places varied within a range of several meters. In the upper reaches of the river the stream passes through the wetlands. The right bank is lower than the left one. Near the river bank a diversity of the phytocenosis species is observed with the predominance of reed (*Phragmites communis L.*) and cattail (*Tipha latiholia L.*). The bottom sediments are sandy at shallows and there is peaty silt at deeper parts of the stream. The depth in the middle of the flow is about 1 m. The bottom of the RT1 is covered with various species of pondweed (*Potamogeton sp.*) and coontail (*Ceratophyllum submersum L.*).

The RT2 is the joint name of several nearby sampling sites in the mid reach of the Techa River located 88 -129 km downstream of RT1. In this area the stream is moderately twisty. The right shore is higher than the left one. The shores are covered with alder trees (*Álnus sp.*) and willow trees (*Salix sp.*). In lowlands, reed beds (Phragmites communis L.) grow densely. The river bottom is covered with gravel and organic sediments. The depth in the middle of the flow is about 1.5 m.

The RT3 is in the low flow of the Techa River. It is located 215 km downstream of RT1 and 30 km upstream of the confluence of the Iset River near Pershinskoye Village. The positions of sampling places varied within a range of several meters. The stream is moderately twisty and the shore plants are the same like in the mid-flow area. The right shore is steep and higher than the left one, which is flat. The bottom of the river is covered with river gravel and silt sediments at deeper parts of the stream. The depth in the middle of the flow is also about 1.5 m.

The RM is the joint name of sampling sites in the upper reaches of the Miass River located at a distance of 19 and 86 km downstream of the Argazi Reservoir. The low banks of the river are covered densely with vegetation. The bottom of the river is covered with sand, river gravel, plant residue and silt sediments at parts of the stream. The depth in the middle of the watercourse is about 1 m.

2.2. Sample collection

At each station, samples of water, bottom sediments, zoobenthos (gastropod mollusks), filamentous algae and fish (perch, roach and pike) were collected. Water samples were taken from the surface level and placed into plastic containers, with a total volume of 60 l.

Fish dwell mainly in small backwaters, where it is easier to catch them. At each of the stations, backwater fish were caught with electric fishing rod SAMUS with the cathode length 1 m, diameter of the anode-net 0.6 m, the distance between cathode and anode was 4 to 7 m. The following parameters were used for catching fish: in spring, power 350 – 480 W, frequency 70 – 90 Hz; in summer, power 100 – 150 W, frequency 60 - 80 Hz. Additionally, in spring time gill nets (at stations RT1 and RT2) and trawl net (at station RT3) were used. All fishes caught were

given the unique numbers (UN). Specimens were weighed, packed individually (with the indication of UN on the packing polyethylene bags) and put into a container with ice for transportation. Samples are stored in a freezer at -18°C at the Experimental Department of the URCRM. Defrosting soft tissues of fish lost about 2% - 17% of the weight. Therefore, after defrosting each fish was weighted again. Table 1 presents the numbers of examined fishes and their mean mass at the time of catching. Average masses of fish species were not correlated with fishing season; however, individual variability of masses was significantly larger in the spring time.

Superficial bottom sediments (first 5 cm) were taken at places of fishing using a ladle bottom dredge with a capture area of 0.025 m^2. The sediments are mainly silt. Benthos organisms were separated by flushing through a hydrobiological sieve. The sediment sample transportation was done using plastic containers with a volume of 1-3 l separated and washed gastropod mollusks were fixed by 10% formalin solution with the addition of Na_2CO_3. Filamentous algae samples were taken from various substrates (stones, snags, etc.) using scrapers.

2.3. Evaluation of ^{90}Sr and ^{137}Cs concentrations

Evaluation of radionuclide concentrations in the components of aquatic ecosystems were performed after preliminary radiochemical separation of ^{137}Cs and ^{90}Y.

2.3.1. Pre-treatment and preparation of samples

Water samples were trickled through a 25-micron filter to weed out the sand, coarse particles of silt, as well as small aquatic organisms. Then, a sample fraction about 5-20 l was additionally trickled through a 0.45-micron filter to separate fine particles of silt, clay and microorganisms.

Sediment samples were air-dried, large stones/rocks and roots of plants were removed. Then, the dried sample was crushed, sifted through 1 mm meshed sieve and mixed to obtain a relatively homogeneous sample. Then, the sample was calcined at a temperature of 400-500^oC for 3-4 hours.

Biological samples were prepared in two ways. Mollusks and filamentous algae samples are air-dried. Fish samples are grounded to a mince, dried under a heat lamp and ashed at 400-500^oC overnight. All biological samples were finally subjected to wet ashing.

2.3.2. Pre-concentration and separation of nuclides from sample matrices

2.3.2.1. Water samples

Fifty mg of Cs carrier and 40 mg of Sr carrier were added to the sample and then the HNO_3 is added to adjust pH to 1. Cesium was concentrated on nickel ferrocyanide ($Ni(NO_3)_2$ 0.1 mol/l + $K_4[Fe(CN)_6]$ of 0.1 mol/l). Thereafter, Sr was concentrated in the form of carbonates by adding Na_2CO_3(20g /l). Then the sample was carefully mixed and settled for a few days. The solution was centrifuged and decant supernatant was discharged. The precipitate was treated by HNO_3 (3mol/l) for dissolution of carbonates and then centrifuged to separate the solute carbonates and ferrocyanide precipitate. The ferrocyanide precipitate then was used for radiochemical separation and purification of ^{137}Cs. Carbonates $(NH_4)_2CO_3$ were precipitated

from the carbonate solution for elimination of Na^+, filtered and dissolve again with HNO_3. The solution is transferred into a 100 ml flask. The 5 ml aliquot of the solution was taken for determination of the Sr chemical yield after the separation from the sample matrix using flame photometry (Flame photometer U4.2, produced in Zagorsk at the optical-mechanical plant, USSR, 1978). The rest of the solution was used for radiochemical extraction of ^{90}Y.

2.3.2.2. Sediment samples

Fifty mg of Cs carrier and 40 mg of Sr carrier were added to the sample. After that, the sample was leached by HCL solution (of which the volume was 3 times larger than a sample amount) of 4 mol/L with 30 min boiling. The leaching procedure had been repeated 3 times. After each boiling the substrate was cooled and filtered. The precipitate was washed with hot water. All liquid phases were combined. Then the oxalates were precipitated by adding $H_2C_2O_4$+NH_4OH to 4÷4.5 of PH. The precipitate was settled for 12-16 hours and then it was filtered. As a result, the solid phase (oxalates) containing ^{90}Sr and the liquid phase containing ^{137}Cs were separated. The oxalate precipitate was dried up under a heating lamp at 700-800^oC for 3-4 hours and dissolved with HNO_3 (4 mol/L) on a hot plate. Then the solution was diluted with 200 ml of water and used for extraction of ^{90}Y. Ten ml aliquot of the solution was used for determination of the Sr chemical yield after the separation from the sample matrix. For this purpose, the method of flame photometry is applied. Cesium was concentrated on nickel ferrocyanide ($Ni(NO_3)_2$ of 0.1 mol/l + $K_4[Fe(CN)_6]$ of 0.1 mol/l).

2.3.2.3. Biological samples

Fifty mg of Cs carrier were added to the sample and all of it was dissolved with HNO_3 while boiling. The solution was filtered, transferred to a flask and diluted with $H_2O$to achieve the volume 250 ml. The normality was set as equal to 0.3-0.4 mol/L. The solution is used for extraction of ^{90}Y and then it is used for concentration of Cs on nickel ferrocyanide ($Ni(NO_3)_2$ of 0.1 mol/l + $K_4[Fe(CN)_6]$ of 0.1 mol/l).

2.3.3. Radiochemical separation and purification of radionuclides

2.3.3.1. ^{137}Cs

Nickel ferrocyanide precipitation was transferred with H_2SO_4 to a pyrex glass and the remainder of the liquids were evaporated. Then the sample was dissolved with 50 ml of HCl (3mol/L) and cooled. Then 4ml of saturated solution of NH_4I was added to convert I^2 into I^+. After the solution was decolorized, 0.5 ml of $SbCl_3$ was added. Antimony cesium iodide was precipitated with vigorous stirring. After settling for 1 hour the water fraction was centrifuged. The precipitate was washed off 3 times by 5-7 ml of concentrated CH_3COOH and once by C_2H_5OH, dried up under a heating lamp and transferred to a steal array (with a known mass) fixed with ethanol. The prepared sample was weighted using an analytical balance to determine chemical yield of Cs separation. Then the sample was placed on a steal array with a diameter of 3 cm (with known mass).

2.3.3.2. ^{90}Y

The solution contaminated with Sr was transferred to a 0.5 l flask. The water was added to achieve 0.25 l of solution volume. The normality was set as equal to 0.3-0.4 mol/L (for

HNO_3). Then 1ml of monoisooctylmethyl ether phosphonic acid (MIOMPhA) and 0.5ml of toluol were added. The solution was shaken for 15-20 min and then 3 ml of $FeCl_3$ was added to it. About 20 min of the next shaking led to the formation of a solid extract with ^{90}Y in the form of carbonates. The precipitate was placed on a steel array with a diameter of 3 cm (with known mass) fixed by ethanol.

2.3.4. Measurements of radioactivity

Measurements of ^{90}Sr and ^{137}Cs activity was performed using low-background α- β-counters UMF2000 and UMF 1500 (produced by SPC-scientific and producing company "Doza", Russia), respectively. Each of the samples was measured 5 times repeatedly. Measurement time was 1,000 s. Calibration was done using standard radioactive solution of ^{137}Cs and ^{90}Sr+^{90}Y. The measurement results were corrected for self-absorption and chemical yield of radionuclides. Performance parameters for radionuclide detection, such as null measurement uncertainty (NMU) and detection limit (DL), are shown in Table 2. Definitions of the parameters were drawn from the JCGM 200: 2012 (JCGM, 2012).

2.4. Statistical analysis

If the data pools are well fitted by normal distribution with similar width, the comparison of the data sets is performed using t-test (α=0.05). If the shapes or widths of the distributions are different, the Mann-Whitney test (α=0.05) is performed. The data below the detection limit (BDL) were analyzed based on the comparison of the distributions of the blank (empty still array) measurements and sample measurements.

The distance between two distributed values was evaluated based on the Monte Carlo simulation. The differences between pairs of randomly drawn values, which belonged to the distributions compared, were analyzed and an average difference was assumed as a mathematical expectation of the measurand. The examples of the data treatment are shown in Annex 2.

The distribution fitting was performed using the EasyFit software (MathWave Technologies, USA).

3. Results and discussion

3.1. Contamination of biotic and abiotic components of ecosystems of the Techa River and the Miass River

3.1.1. Water contamination

Radionuclide concentrations in the water of the Miass River were found to be in the ranges of 0.01-0.05 Bq l^{-1} and 0.004-0.04 Bg l^{-1} for ^{137}Cs and ^{90}Sr respectively and they slightly depend on seasonal river water contents (rainfall, seasonal floods, etc.).

In the Techa River radionuclide concentrations are strongly influenced by the high non-uniformity of radionuclide distribution in sediments and flood plain areas along the water course. Figure 2a,b demonstrates spring and fall concentrations of ^{137}Cs and ^{90}Sr in the water of the Techa River at different sampling sites. The upper border of the shading box corresponds to 95^{th} percentile of radionuclide concentrations measured in the Miass River. As can be seen from Figure 2a, the seasonal difference of concentration of ^{137}Cs in the lower reaches of the Techa River (RT3) is relatively small and can be ignored, and both concentration values are fallen into

the range of the background (the Miass River contamination). Seasonal difference in ^{137}Cs concentrations in the swamped upper streams (RT1) is also statistically insignificant. However, 3.5 times difference between spring and fall water specific activity of ^{137}Cs is observed at RT2. It could be expected that an increase in water contents in the spring should lead to dilution and decrease in the concentrations of the radionuclide. Nevertheless, the inflow of melt water from the riverside catchment area brings about the secondary water pollution with ^{137}Cs in exchangeable form (solutions and colloids). The maximum spring inflow of the radionuclides into the Techa River should be in the upper stream passing through the wetlands. The secondary pollution is somewhat offset by the effect of dilution at RT1, making the spring ^{137}Cs concentrations close to the fall values. In spring, the increases in the water level and flow rate (at RT2 the water level rises by 30 cm, and the flow rate increases 4-fold) lead to slowdowns in cesium sorption in the surface waters. Accordingly, the ^{137}Cs concentrations in the water along the river decrease slower than it can be observed in the fall.

This is confirmed by analysis of water filtered through 0.45-micron filter. After such filtration Cesium can remain in solution only in the form of ions or colloids. Figure 3 presents fractions of ^{137}Cs in the Techa River water sorbed on particles with size above 0.45 microns. As can be seen, at RT1 the sorbed fraction of ^{137}Cs in spring is about 8% smaller than that in the fall. A sorbed fraction of ^{137}Cs at RT2 in spring is more than 5 times smaller than in the fall. While floating along the course with very low spring sorption the surface water ^{137}Cs at RT2 was mainly represented by bioavailable fraction (about 92%). However, colloidal and dissolved cesium eluted from the swamps and floodplains of the upper and middle stream do not practically reached the river mouth. More than 65% of ^{137}Cs in the surface water of RT3 is transferred on fine particles of clay and silt. In the Miass River, which is deeper than the Techa River, a fraction of ^{137}Cs sorbed on the particles (>0.45 mkm) is practically the same and equal to 0.18-0.25.

Concentrations of ^{90}Sr in the Techa River water exceed the global levels by 3 to 4 orders of magnitude. Concentrations of ^{90}Sr in the fall water (Fig 2b) at each of the stations were higher than those in the spring time. This can be simply explained by dilution, as a main factor of seasonal variations of ^{90}Sr concentrations in the water. Strontium circulating in the river ecosystem has existed in the soluble form and it is not strongly dependent on sorption processes.

3.1.2. Bottom sediments

Table 3 presents the concentrations of ^{90}Sr and ^{137}Cs in the bottom sediments collected in different expeditions at different sampling sites. In spite the concentrations of ^{137}Cs in the water of RM and RT3 were practically the same, the bottom sediments of ^{137}Cs at RT3 are, on the average, 6 times higher. In the RT1 the concentration excess of ^{137}Cs in bottom sediments over RM level can reach up to 3 orders of magnitude (water concentrations differed only by order of magnitude).

A large variability of the bottom sediment contamination at each of sample sites is observed for both radionuclides, and it is determined mainly by the fact that a sampling site is not a fixed point. There is a high non-uniformity of radionuclide concentration in bottom sediments across the stream. The stream can form local “spots” with high concentrations which is most typical of backwaters and blink forks. The exchange of water between the main stream and zones with stagnant or slow circulating water adjacent to the main stream, side pockets and bed sediment leads to changes in transport routes, transport times and fate of solutes in flowing water. The exchange with these zones causes a temporary detainment of a portion of the water as compared with the relatively faster moving waters in the main stream. Therefore, much higher

amounts of radionuclides discharged at in the time of massive releases were accumulated in the bottom sediments of stagnant areas comparing with the sediments of the main stream. Moreover, year-to-year the stream course can shift significantly at some river areas. That happened in 2013, when the river flow at RT1 station was shifted coinciding with the stream course position at the time of the major releases. As a result the concentration of ^{137}Cs in bottom sediments at RT1 in the spring of 2013 was found to be much higher than that in the spring of 2012 almost by the factor of 30. Since ^{137}Cs in bottom sediments is water-insoluble it does not influence the water contamination significantly.

Dry weight fractions of silts at RM, RT1 and RT2 were comparable, they varied (at each station) in the range from 33 to 64%. At RT3 the dry weight fractions made up from 75 to 81%.

3.2. Biota

3.2.1. Benthic organizms

Zoobenthos samples were consisted of a mixture of gastropod mollusks. Cesium-137 concentrations in the mollusks of the Techa River varied in the range from the background levels (5±2 *10^2 Bq kg^{-1} of dry weight) up to 2.8*10^4 Bq kg^{-1} at RT1. Concentrations of ^{90}Sr exceeded the background levels (150±90 Bq kg^{-1} of dry weight) by 1-2 orders of magnitude. Table 4 presents the average values of radionuclide concentrations of gastropods sampled in different expeditions. The percentage of dry weight in the samples is equal to 30±6%, on average.

3.2.2. Filamentous algae

The number of samples of filamentous algae available for the study was small (Table 4). However, even such small selection allows to conclude that both ^{90}Sr and ^{137}Cs concentrations in the species are significantly higher than the background levels (40±10 Bq kg^{-1} of dry weight) even at the RT3 (180±20 Bq kg^{-1} of dry weight). The maximal concentration of ^{137}Cs was equal to 1.16 *10^4 Bq kg^{-1} of dry weight. For ^{90}Sr, the minimal and maximal values in the Techa River were 0.98 and 3.2 *10^3 Bq kg^{-1} of dry weight, respectively, that is significantly higher than the concentrations in the Miass River (40±10 Bq kg^{-1} of dry weight). The percentage of dry weight in the samples is on average equal to 11±1%.

3.2.3. Fish

Background levels of ^{137}Cs and ^{90}Sr concentrations in the fishes caught in the Miass River were in average about 9 Bq kg^{-1} and 1.5 Bq kg^{-1} respectively. The body-average concentrations in the fishes from the Techa River were varied from 20 to 250 Bq kg^{-1} of ^{137}Cs and from 300 to 1000 Bq kg^{-1} of ^{90}Sr.

It could be expected that the seasonal differences in the above indicated radionuclide concentrations in water led to differences in the body burdens. However, the statistically significant seasonal differences in both ^{137}Cs and ^{90}Sr concentrations were found only for roach from RT2. The body-averaged concentrations of ^{137}Cs and ^{90}Sr in the fishes are presented in Table 5.

A statistically significant correlation of body mass and ^{137}Cs concentration was found for roaches and perches sampled at RM and RT2 as well as for roaches from RT1. It was found that for perches below 30-40 g of body weight, and for roaches below 80-90 g of body weight the average radionuclide concentrations are higher than those in the bodies of large fishes. Moreover, small fishes demonstrate a higher individual variability of ^{137}Cs concentrations. Figure 4 presents an example of such mass thresholds found at RM for roach and perch (Fig. 4a and 4b, respectively).

The mass dependence for these two species can be explained by age-related changes from one type of nutrition to another. Large roaches live mainly in the deep areas and feed mostly on

mollusks, while small fishes feed mainly on vegetable organisms. Staple food of small perches is plankton and partially insect larvae. Larger individuals are predators that in addition to plankton and insect larvae feed on fish. The tendency of mass dependence for ^{90}Sr concentration was also observed for these two species, however, a large individual variability in the specific activity leads to statistical insignificance of the mass dependence. Thus, for analyzing ^{137}Cs accumulation, in the current study roaches and perches were conventionally divided into carnivores and phytophagous depending on the body mass. It should be noted that the data available do not allow classifying strictly fishes by the type of nutrition and, therefore, such fish grouping is very provisional.

As can be seen from Table 5, the ratios of ^{137}Cs concentrations in carnivore and phytophagous species from the Miass River are about 1.5-1.7 times higher than those registered in fishes from the Techa River. The smaller ratios in the Techa River can be explained by migration of the fish in the non-uniformly contaminated environment. Specifically, for roaches from the Techa River the mass dependence of body-average ^{137}Cs concentration can be ignored, except for one case. It is likely that the phytophagous roaches caught in the spring were the members of the shoal that had just migrated from the upper reaches. Pike, that has no mass (age) dependent food preferences, does not demonstrate mass dependence on radionuclide concentrations.

As can be seen from Table 5, the concentrations of ^{90}Sr in the bodies of roaches are higher than those for perch and pike sampled at the same site by factors 1.5 and 2, respectively. The concentrations of ^{137}Cs in the bodies of large individuals of different species sampled at the same site were similar (no statistically significant difference); however, the small perches and roaches showed significant inter-species difference. On the average, the specific activity in small fishes was twice higher compared to that shown by large fishes. The concentrations of both radionuclides decreased consistently with distance downstream. The levels of Cs content in the body of fish from RT3 became comparable to the background levels. Concentrations of Sr in the lower reaches exceeded significantly the background levels.

4. Distribution coefficients

Distribution coefficients for ^{90}Sr and ^{137}Cs were calculated as an average of all combinations of sediment-to-water concentration ratios (for specific sampling site). As it was shown above, the spring and fall differences in ^{137}Cs at RT2 concentration in water was non-negligible, for this site two seasonal *Kd*s are calculated. Table 6 presents the results of estimations of *Kd* for ^{90}Sr and ^{137}Cs. Table 6 shows mean coefficients and the ranges of particular values of sediment-to-water concentration ratios in comparison to default parameters (obtained based on the geological data on stable elements) used in ERICA Tool and RESRAD BIOTA.

For the Techa River, *Kd* values (for both ^{137}Cs and ^{90}Sr) show the trend to decrease with distance downstream. This can be due to the fact that insoluble fraction of the radionuclide in bottom sediments is practically not interrelated with the soluble one which is circulating in water and the biotic components of the ecosystem.

As can be seen from Table 6, the upper limits of Kd^{Cs-137} for the Techa River are comparable to default value of the ERICa Tool. However, minimal values could be by two orders of magnitude lower than maximal ones. Such a considerable difference is associated with

the high non-uniformity of the contamination of the bottom sediments with ^{137}Cs insoluble compounds.

For the Miass River, where radionuclides are of the global origin, Kd^{Cs-137} values are much lower than the default values used in ERICA Tool, but they are approaching minimal values of *Kd* for the Techa River. In contrast, the *Kd* values for ^{90}Sr in the Miass River show a good agreement with the default values used in the ERICA Tool. However, the *Kd* values for ^{90}Sr in the Techa River are 10-100 times lower.

By contrast, the default Kd^{Cs-137} of the RESRAD BIOTA is comparable to minimal values detected at RM and significantly less than the respective Techa River data, whereas the Kd^{Sr-90} is in good agreement with the Techa River data.

Because of the non-uniformity of the spatial radionuclide distribution in the sediments, the distribution coefficients *Kd* evaluated using randomly selected samples for both ^{137}Cs and ^{90}Sr may vary by several orders of magnitude at a sampling site. Therefore, the correct *Kd* evaluation for the Techa River should be performed based on sampling over a large area at each of sampling site, which is quite labor-intensive. Thus, the *Kd* parameter is not appropriate as an radio-ecological parameter for the non-uniform contamination of the bottom sediments.

5. Concentration ratios

5.1. Benthic organisms

The concentration ratios calculated individually for each of the samples collected in different rivers were similar for ^{137}Cs and significantly differed for ^{90}Sr. CR^{Sr-90} for gastropods from the Miass River is on the average higher by an order of magnitude than that in gastropods from the Techa River. In other words, the bioaccumulation of ^{90}Sr in the gastropod mollusks is probably river-specific because of the sensitivity to hydrochemistry, contamination of bottom sediments and/or differences in the age (shell volume) of the mollusks. Therefore, the dependence of ^{137}Cs concentration in the samples on the water concentration is presented in Fig. 5a for both rivers, but an analogous dependence for ^{90}Sr shown in Fig. 5b was plotted based only on the Techa River data (the ^{90}Sr in the Miass River was below the detection limit and, therefore, it was useless for an individual analysis). Figures 5a and b illustrate the linear regression for ^{137}Cs (r= 0.765, p= 0.0002) and ^{90}Sr (r=0.418, p=0.13), respectively. The dashed lines border 90% prediction interval. In the Figure, one of the samples marked with white color is an outlier. It was collected at RT1 in October 2012 and probably had been drifted from a more heavily contaminated area. This sample was not accounted for in the regression analysis. For ^{137}Cs, the intercept of the regression line almost coincides with zero. The slope is equal to $1.2*10^4 \pm 0.2*10^4$ l kg^{-1} (dry weight). Taking into account that dry weight of gastropods amounts to approximately 30% of its fresh weight, the CR^{Cs-137} was estimated as $3.6*10^3 \pm 0.6*10^3$ l kg^{-1}. This value is in good agreement with the default one for freshwater gastropods (in the ERICA Tool it is equal to $2.8*10^3$ l kg^{-1}). The regression obtained for ^{90}Sr in the Techa River was not reliable. Individual ratios were found to be within the range from 200 to 4000 l kg^{-1} (dry weight); the average value was 900±200 l kg^{-1} (dry weight). The slope in Fig 5b is about 140±90 l kg^{-1} (dry weight) which is even smaller than the minimal individual value. In other words, the dependence of ^{90}Sr concentration in the mollusk's body on the water concentration is weak, and other factors may influence ^{90}Sr bioaccumulation.

The Pearson correlation of ^{90}Sr concentration in the mollusk's body and in the bottom sediments was better (r=0.493, p= 0.053), but still not reliable. Thus, the description of ^{90}Sr body

burden for gastropod mollusks cannot be simplified by water-to-body or sediment-to-body conversion. For example, the CR^{Sr-90}, which were evaluated based on the averaging of the individual data for the Techa River and the Miass River, were equal to 270±60 l kg^{-1}, and 2100± 600 l kg^{-1}, respectively.

5.2. Filamentous algae

For measurements of ^{137}Cs and ^{90}Sr concentrations, only 5 and 9 samples, respectively, of the Techa River water were available. Additionally, one sample was collected in the Miass River tested for both ^{137}Cs and ^{90}Sr concentrations. The concentration ratios calculated individually (taking into account that the dry weight of algae makes up approximately 10% of its wet weight) for each of the samples differed significantly for the Techa River and the Miass River for both radionuclides. The CR^{Cs-137} for the Miass River was by 2 orders of magnitude lower than those for the Techa River, and equaled to 80 l kg^{-1} and 1700±500 l kg^{-1}, respectively. By contrast, CR^{Sr-90} for the Miass River was by order of magnitude higher than those for the Techa River, and equaled 300 l kg^{-1} and 10±4 l kg^{-1}, respectively. Such difference can be caused by the difference in anion concentrations in the water of the rivers; however, conclusions can only be made after the statistics is improved. Algae are widely used for radionuclide monitoring in aquatic environments (Carlson and Snoelis, 1994). According to Harney (1969), the mean concentration ratios of ^{137}Cs for different sorts of freshwater algae from different medias (in terms of water mineral composition) are in the range from 80 to 1450 l kg^{-1}. This is in good agreement with the results of the current study. However, the concentration ratios of strontium isotopes (^{89}Sr) published by Harney (1969) ranged from 600 to 2300 l kg^{-1}, which is much higher than the results observed for the Techa River and the Miass River.

5.3. Fish

5.3.1. CR^{Cs-137}

Individual concentration ratios for each species were calculated. The distribution of *CR*s within the groups of the same species caught simultaneously was asymmetrical. There was no significant difference between *CR*s of the same species taken from different sampling sites, including both the Techa River and the Miass River. However, for perches and pikes, the difference between the groups conditionally subdivided into carnivores and phytophagous was significant. The average values (± error of mean) and distribution parameters are presented in Table 7. The default values in the Erica Tool for benthic and pelagic fishes are 6300 and 7100 l kg^{-1}, respectively. These are by order of magnitude higher than CR^{Cs-137} calculated for fishes in the Techa River and the Miass River.

*CR*s of phytophagous perches and roaches demonstrate that the difference in the median values is not high enough to exclude the possibility that the difference is due to random sampling variability (p=0.087). The difference between CRs of all carnivores was also insignificant (p = 0.8). Therefore, the data can be grouped into the carnivores and phytophagous clusters independent of the sampling site and fish species.

Since the fishes were caught in different seasons and because of unfeasibility to exclude the factor of migrations along the non-uniformly contaminated course of the river, the individual concentration ratios expected to be described by the Shifted Log-Logistic distribution, the so-called Generalized Logistic Distribution (GLD). The Log-Logistic distribution is widely used in the hydrological studies (Strupezewski et.al., 2001, Svanidze and Grigolia, 1973) for analysis of

non-stationary objects. The expressions of probability density function (*pdf*) and cumulative distribution function (*cdf*) of GLD are presented by Eqns 1 and 2, respectively:

$$pdf(x) = \frac{(1+kz)^{-1-1/k}}{\sigma(1+(1+kz)^{-1/k})^2}, \qquad (1)$$

$$cdf(x) = \frac{1}{1+(1+kz)^{-1/k}}, \qquad (2)$$

where $z \equiv (x-\mu)/\sigma$; μ is the location parameter equal to median value, σ>0 the scale parameter and *k* the shape parameter.

The comparison of the fitting of pooled data by GLD to generalized extreme value and three-parameter lognormal demonstrated the GLD distribution performed very well compared with these other distributions. Figure 6 presents the cumulative distributions of the experimental results for carnivores (a) and phytophagous (b) of different species and fitting curves of GLD functions obtained with pooled data. Fitting parameters for GLD functions are shown in Table 8.

Solid gray line corresponds to the fitting curves of generalized logistic distribution function obtained with pooled data.

The GLD distribution parameters obtained can be used in the probabilistic approaches for radioecological risk assessments.

5.3.2. $CR^{Sr\text{-}90}$

Being a metabolic analogue of calcium, ^{90}Sr accumulates predominantly in the calcified tissues with mean $T_{1/2}$ of biological elimination rudely equal to 1.5 years (Kulikov and Chebotina, 1988). Therefore, the seasonal differences in water contamination, which are shown in Fig. 2b, do not affect the body burden of ^{90}Sr, but lead to the seasonal differences in the measured body-to water concentration ratios. Figure 7 demonstrates an example of this difference for perches in the Techa River. As can be seen in Figure 7, the autumnal concentration ratios are not dependent on sampling site. In contrast, the concentration ratios calculated for the spring time are site-dependent which reflects the difference in dilution of ^{90}Sr concentrations in the water, which has been shown in Fig. 2b.

Because *CR* is defined as an equilibrium radionuclide activity concentration ratio, the results of the spring fieldtrips cannot be assumed as equilibrium data. The data from the fall fieldtrips to the Techa River sampling sites show that the *CR*s were not site-specific, but the average values were significantly different for perch, roach and pike (Table 9). The default value for CR^{Sr-90} in the Erica Tool is 17 l kg^{-1}. This value is comparable with the Techa River data for perch and pike, but to a lesser degree than that for the Techa River roach and all fishes from the Miass River.

According to Table A1.1, the concentration of Ca^+ in the Miass River was 2 times lower than that in the Techa River, which has resulted in the levels of CR^{Sr-90} in the Miass River being about 2 times higher (Table 9). The maximal accumulation of radionuclides is observed in roaches, the minimal one is observed in pikes.

Assuming the inverse proportionality of CR^{Sr-90} and concentration of Ca^{+} (Vanderploeg et al., 1975), these two parameters were multiplied to obtain a universal characteristic allowing to combine the Techa River and the Miass River data (Eqn. 3).

$$U = CR^{Sr-90} * A_{Ca+}, \quad (3)$$

where A_{Ca+} is concentration of Ca^{+} expressed in g l^{-1}; U is a universal characteristic of bioaccumulation of ^{90}Sr expressed in g kg^{-1}.

The distributions of U were analyzed for each of the species collected in fall season for which concentration of Ca^{+} were taken from the Table A1.1 (Annex 1). It was found that U is asymmetrically distributed parameter. The GLD function was found to be the best fitting for all tree species comparing with the 3-parameter lognormal and generalized extreme functions. Table 10 presents the parameters of generalized logistic distribution functions for universal parameter of ^{90}Sr bioaccumulation U for perch, roach and pike.

The distribution parameters obtained can be used in the probabilistic approaches to risk assessment. Figure 8 presents the comparison of three PDF functions. As it can be seen, pikes are characterized both by lower values and by the narrowest distribution of U. Roaches demonstrate a larger bioaccumulation as well as a larger individual variability.

Conclusions

Concentration of ^{137}Cs in the water and bottom sediments of the Miss River corresponded to background levels was found to be in the range of 0.01-0.05Bq/l and 40-190 Bq/kg respectively. Background levels of 90Sr concentrations in water and bottom sediments were in the range of 0.004-0.04 Bq/l and 20-70 Bq/kg respectively. Significant difference of radionuclide concentrations in the Techa River was found depend on the distance from the historical site of releases. This difference for ^{137}Cs in the water of upper and lower stream was about order of magnitude and in low stream its concentration was equal to background levels. Cesium-137 contaminations of bottom sediments of upper and low the Techa River stream are differ by 2 orders of magnitude and the low Techa River even in the low stream its concentration was 5 times exceeding of the background levels.

Significant spring-tofall difference between radionuclide concentrations in water was observed. Maximal seasonal difference for ^{137}Cs was at RT2 (3.5 times). In spring, the increases in the water level and flow rate (at PT2 the water level rises by 30 cm, and the flow rate increases 4-fold) lead to slowdowns in cesium sorption in the surface waters. Accordingly, the ^{137}Cs concentrations in the water along the river decrease slower than it can be observed in the fall. For ^{90}Sr the seasonal difference in concentrations is determined mainly by dilution in spring.

The mass dependence of ^{137}Cs concentrations in the bodies of perches and roaches were found. The difference in ^{137}Cs accumulation in the fishes with masses below and above 80g and 30g for perches and roaches, respectively, determined by age-related changes from one type of nutrition to another. Pike, that has no mass (age) dependent food preferences, does not demonstrate mass dependence on radionuclide concentrations.

For the Techa River, *Kd* values (for both ^{137}Cs and ^{90}Sr) show the trend to decrease with distance downstream. This can be due to the fact that insoluble fraction of the radionuclide in

bottom sediments is practically not interrelated with the soluble one which is circulating in water and the biotic components of the ecosystem.

The default values of the concentration ratio for ^{137}Cs in the Erica Tool for benthic and pelagic fishes are 6300 and 7100 l kg^{-1}, respectively. These are by order of magnitude higher than CR^{Cs-137} calculated for fishes in the Techa River and the Miass River.

For ^{137}Cs *CR*s of predominant phytophagous (small perches and roaches) demonstrate no difference. The difference between CRs of all predominant carnivores (large perches and roaches as well as pikes) was also insignificant. The distributions of concentration ratios can be described for both phytophagous and carnivores by Generalized Logistic Distributions (GDL) with parameters shown in the table 6.

The default value for CR^{Sr-90} in the Erica Tool is comparable with the Techa River data for perch and pike, but different than that for the Techa River roach and all fishes from the Miass River. This parameter is strongly depended on Ca^{+} concentration in water. The universal characteristic allowing to combine the data of fish caught in the water with different mineralization was proposed by multiplying CR^{Sr-90} and concentration of Ca^{+}. The GLD function was found to be the best fitting of individual universal parameter for all species. The GDL can be used for probabilistic modeling of the concentration ratios in fishes.

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Figure legends

Figure 1. The map of the river basin area demonstrating the locations of sampling sites on the Techa River and the Miass River

Figure 2. Seasonal variation of concentrations of ^{137}Cs (a) and ^{90}Sr (b) in the water of the Techa River at different sampling sites in comparison with the background levels (shaded box).

Figure 3. Comparison of fractions of ^{137}Cs sorbed on particles (with size above 0.45 microns) in the Techa River water in the fall and spring.

Figure 4. Average concentrations of ^{137}Cs in fish with different body mass: (a) perch; (b) roach. Solid gray lines indicate average concentrations above and below mass threshold (dashed line).

Figure 5. Comparison of ^{137}Cs (a) and ^{90}Sr (б) concentrations in water and benthic gastropods.

Figure 6. Cumulative distributions of the observed CR^{Cs-137} for carnivores (a) and phytophagous (b).

Figure 7. Seasonal difference in body-to-water concentration ratios of ^{90}Sr for perches in the Techa River. The error bars correspond to errors of means.

Figure 8. Comparison of probability density functions of the universal parameter (U) of ^{90}Sr bioaccumulation for different species.

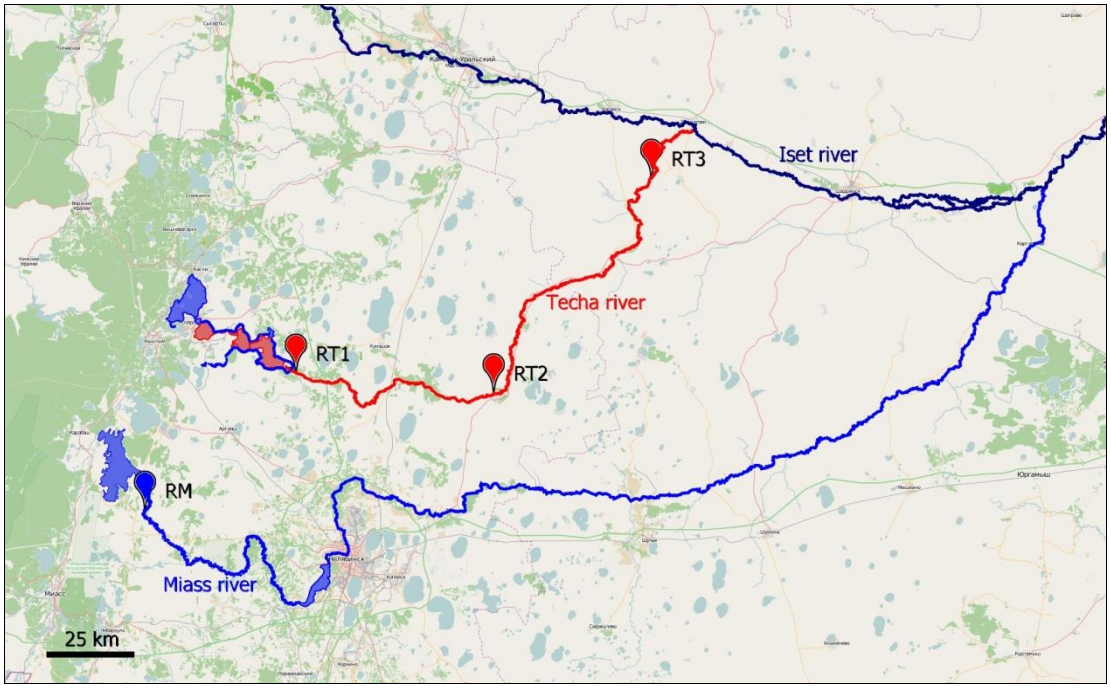


Fig. 1

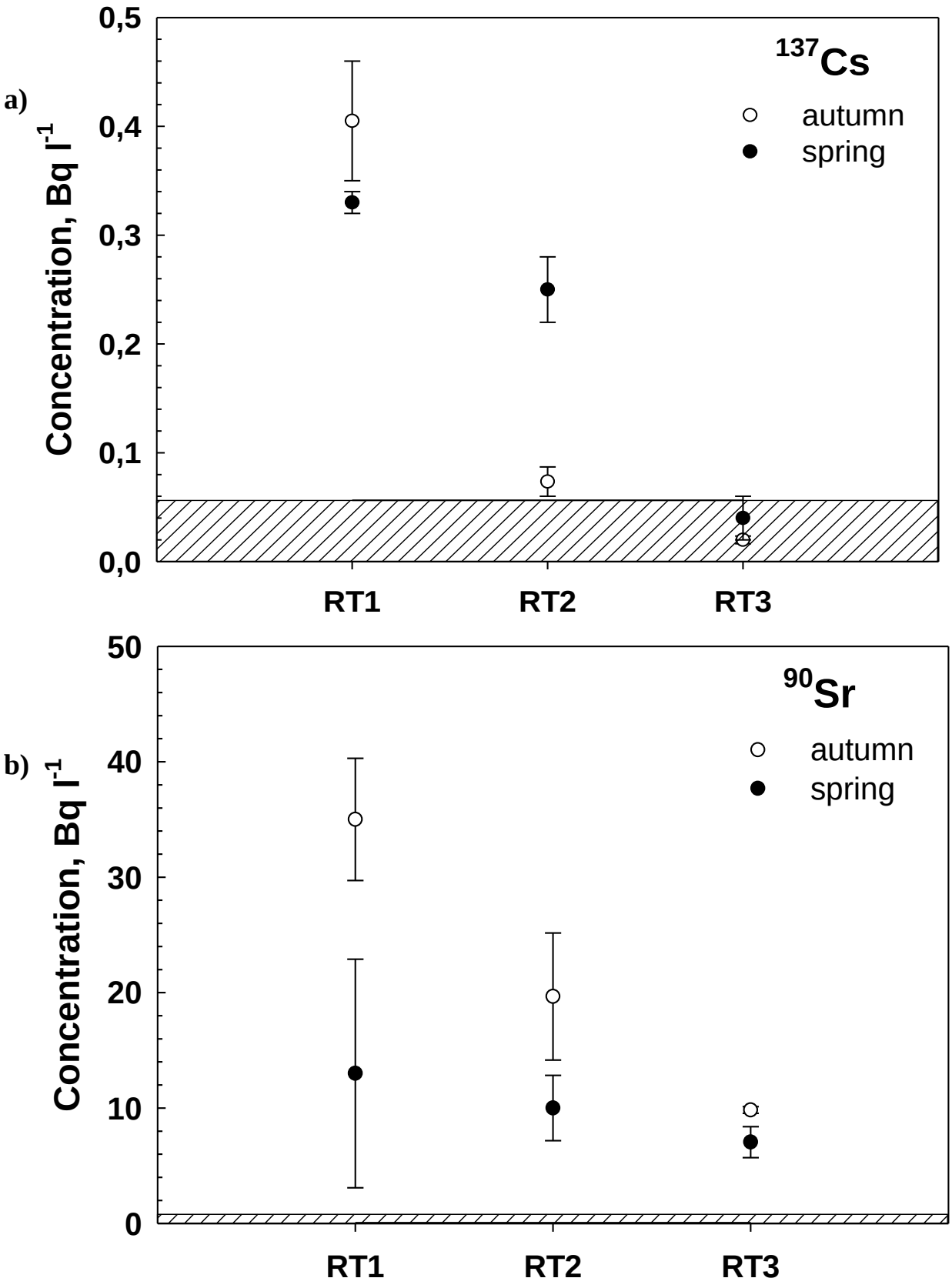

a)
Concentration, Bq l-1
0,5
0,4
0,3
0,2
0,1
0,0
137Cs
autumn
spring
RT1
RT2
RT3
b)
Concentration, Bq l-1
50
40
30
20
10
0
90Sr
autumn
spring
RT1
RT2
RT3

Fig 2.

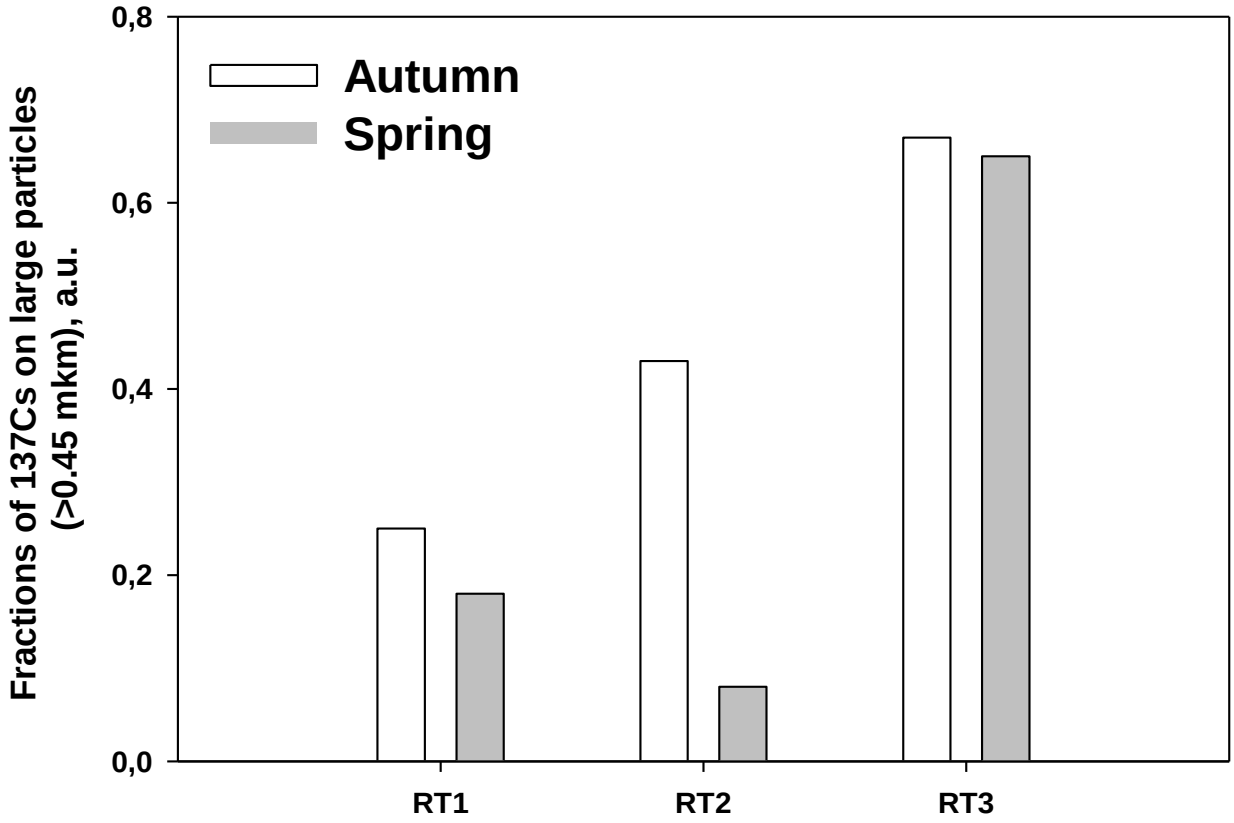

Fractions of 137Cs on large particles
(>0.45 mkm), a.u.
0,8
0,6
0,4
0,2
0,0
Autumn
Spring
RT1
RT2
RT3

Fig. 3

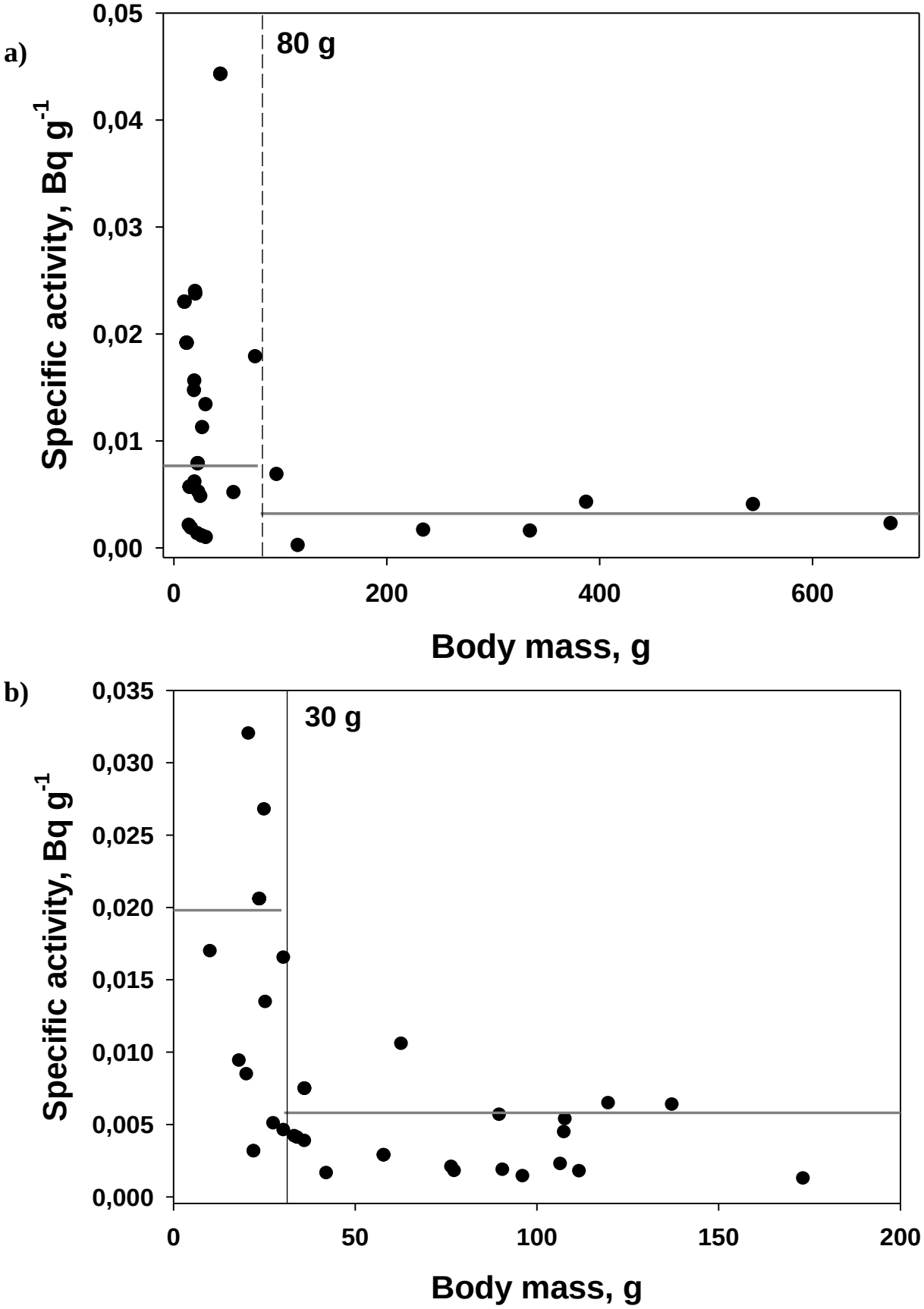

a)
80 g
Specific activity, Bq g-1
0,05
0,04
0,03
0,02
0,01
0,00
0
200
400
600
Body mass, g
b)
30 g
Specific activity, Bq g-1
0,035
0,030
0,025
0,020
0,015
0,010
0,005
0,000
0
50
100
150
200
Body mass, g

Fig. 4.

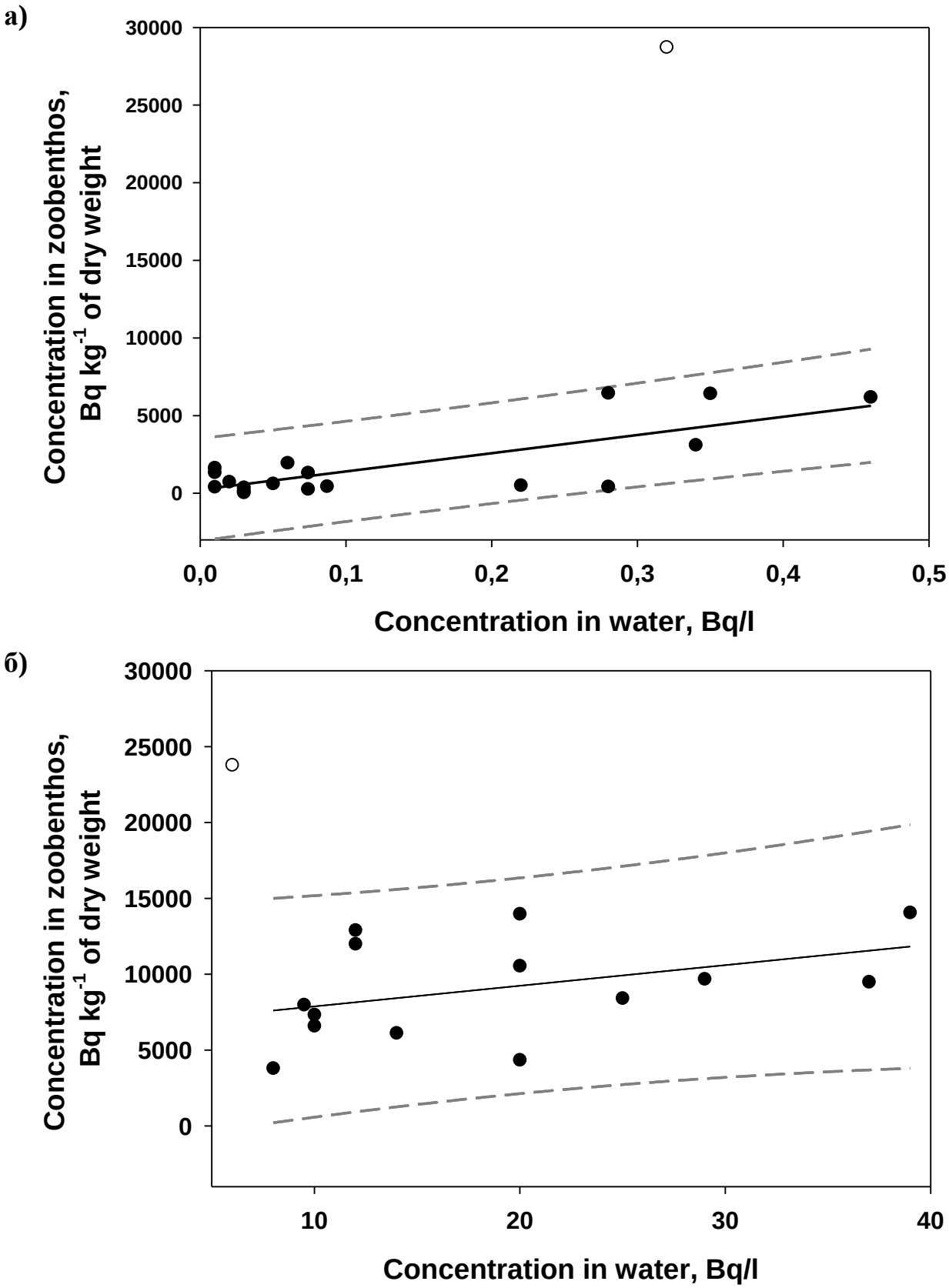
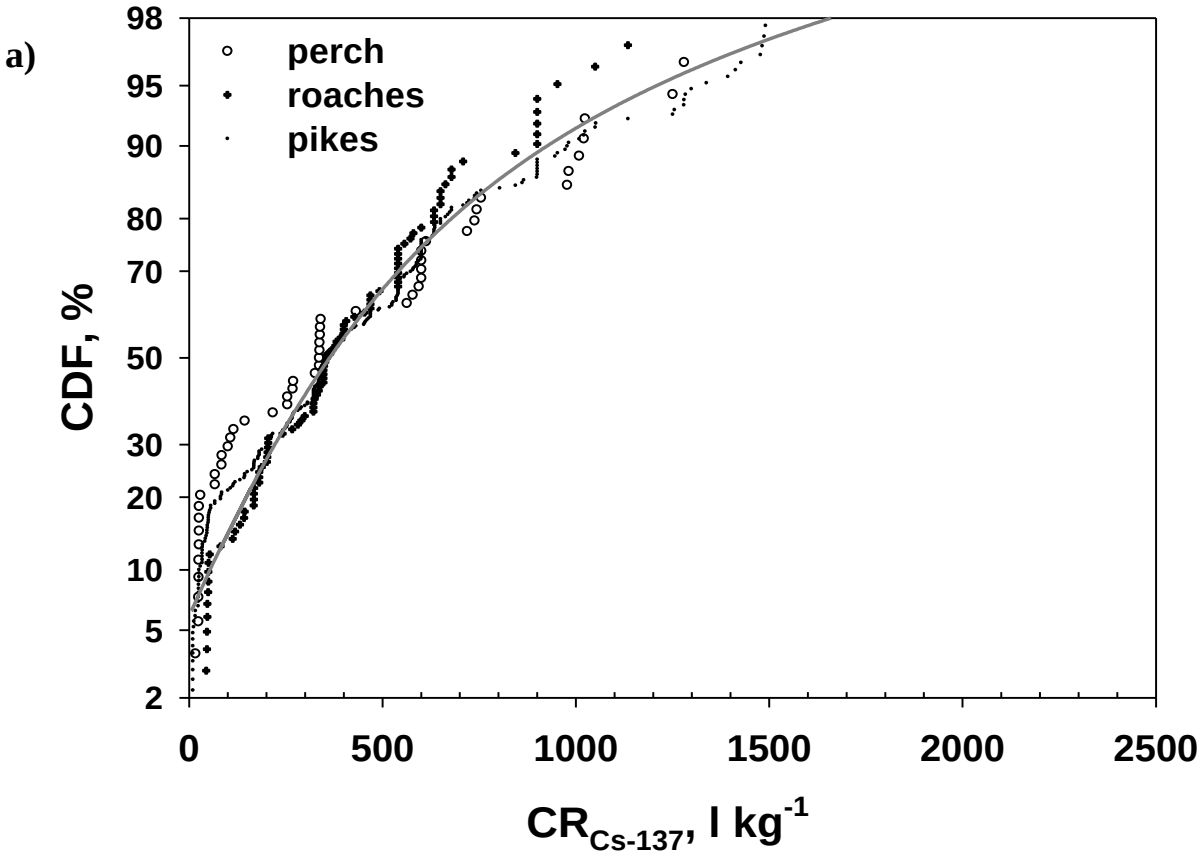

а)
30000
25000
20000
15000
10000
5000
0
Concentration in zoobenthos, Bq kg-1 of dry weight
0,0
0,1
0,2
0,3
0,4
0,5
Concentration in water, Bq/l
б)
30000
25000
20000
15000
10000
5000
0
Concentration in zoobenthos, Bq kg-1 of dry weight
10
20
30
40
Concentration in water, Bq/l

Fig. 5.


a)
perch
roaches
pikes
CDF, %
98
95
90
80
70
50
30
20
10
5
2
0
500
1000
1500
2000
2500
CR Cs-137, l kg-1

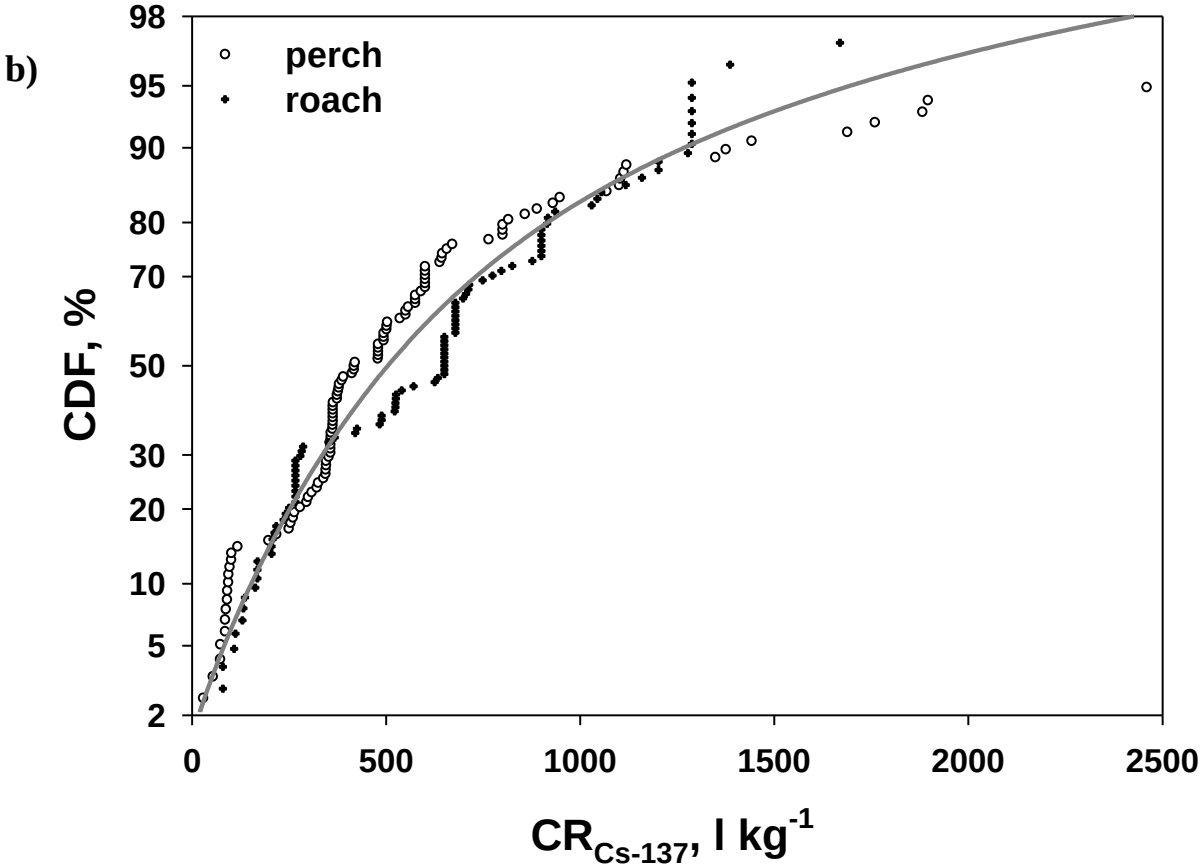

b)
perch
roach
CDF, %
98
95
90
80
70
50
30
20
10
5
2
0
500
1000
1500
2000
2500
CR Cs-137, l kg-1

Fig 6

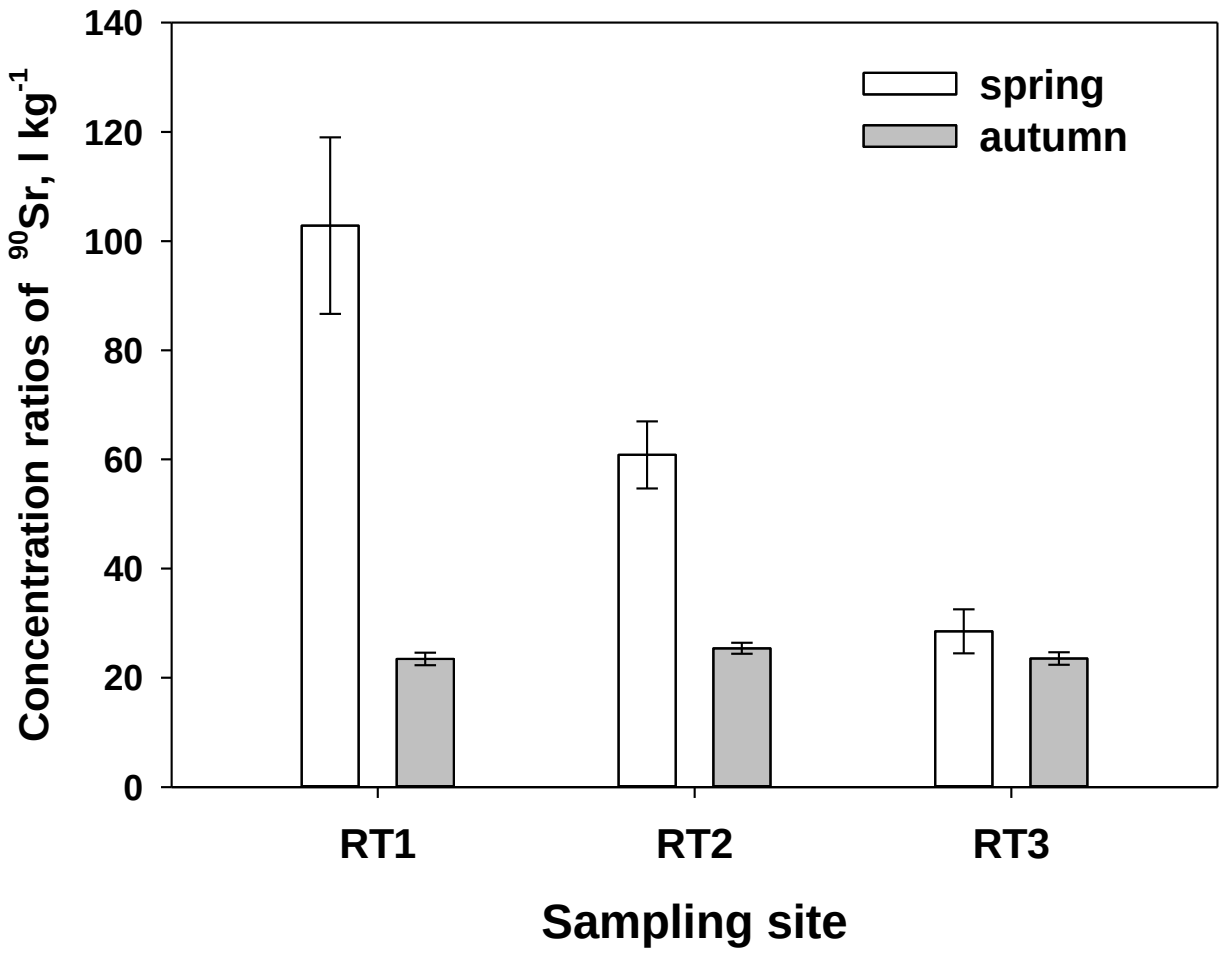

140
120
100
80
60
40
20
0
Concentration ratios of ^{90}Sr, l kg^{-1}
spring
autumn
RT1
RT2
RT3
Sampling site

Fig. 7

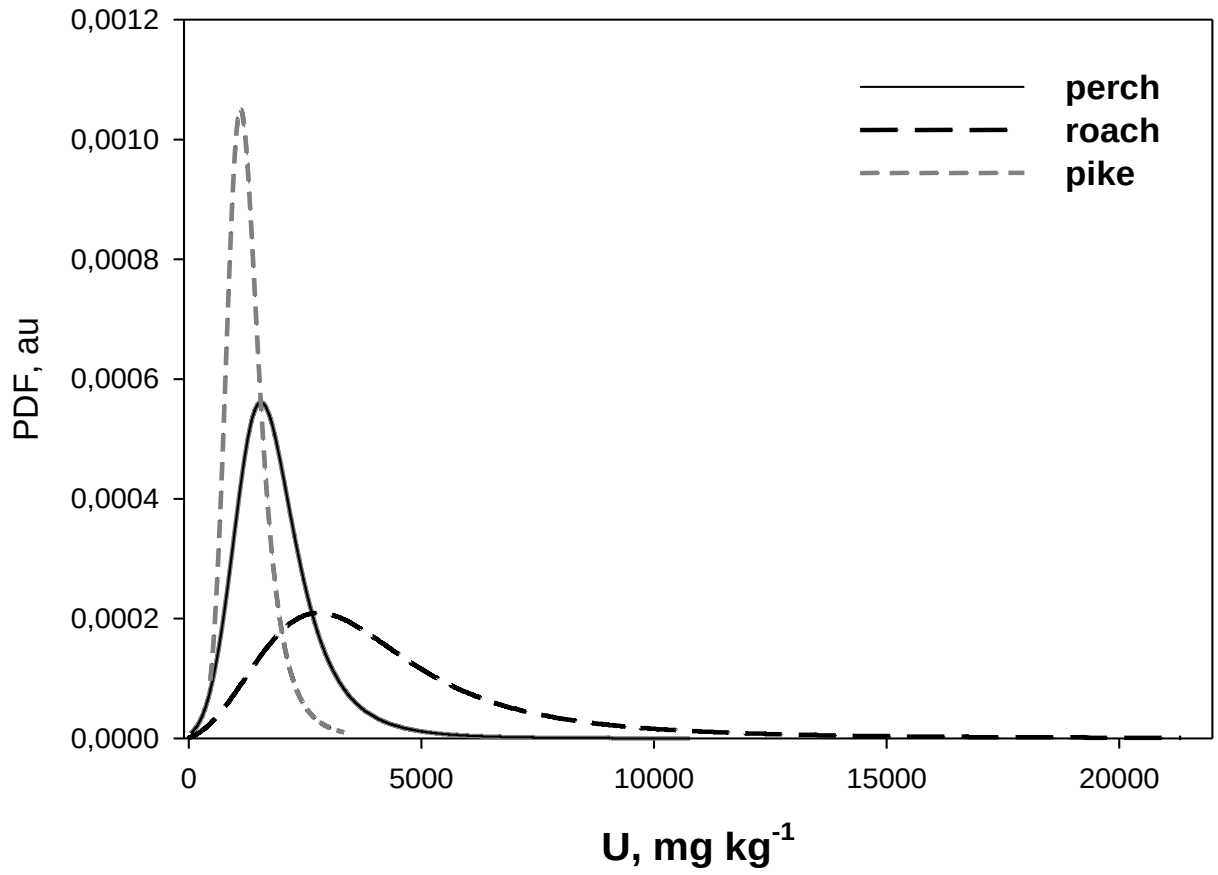

0,0012
0,0010
0,0008
0,0006
0,0004
0,0002
0,0000
PDF, au
0
5000
10000
15000
20000
U, mg kg-1
perch
roach
pike

Fig. 8

Table 1 – Description of fishes under study; N – number of individuals in the group; M: body mass, st.dev.: standard deviation.

Sampling site	Roach		Perch		Pike	
	N	M (st.dev.)	N	M (st.dev.)	N	M (st.dev.)
September 2011						
RT1	10	30 (10)	11	120 (50)	4	1300 (200)
RT2	11	200 (100)	1	220	5	360 (60)
RT3	10	20 (10)	1	420	1	1360
RM	6	20 (10)	10	15 (6)	1	380
April-May 2012						
RT1	22	50 (80)	23	140 (160)	3	550 (500)
RT2	20	30 (20)	8	90 (80)	4	1100 (500)
RT3	18	30 (20)	6	50 (50)	2	700 (900)
RM	11	30 (10)	11	23 (4)	4	220 (110)
August-September 2012						
RT1	10	200 (300)	10	90 (20)	10	1000 (600)
RT2	10	100 (100)	11	150 (50)	10	500 (300)
RT3	11	140 (200)	10	200 (100)	6	800 (400)
RM	4	70 (30)	10	170 (190)	4	800 (500)
April-May 2013						
RT1	9	50 (20)	13	150 (190)	11	400 (500)
RT2	12	300 (300)	13	100 (100)	14	600 (700)
RT3	10	100 (300)	14	50 (90)	8	1100 (900)
RM	8	110 (40)	5	40 (30)	8	300 (300)
August-September 2013						
RT1	10	70 (50)	11	90 (60)	11	300 (200)
RT2	12	50 (40)	13	130 (70)	-	
RT3	12	50 (50)	23	70 (70)	-	
RM	7	120 (40)	9	200 (200)	12	300 (200)
Totally						
The Techa River	187		168		89	
The Miass River	36		45		29	

Table 2 – Performance parameters of radionuclide measurement systems

Facility	Radionuclide	NMU, Bq	DL, Bq
UMF 2000	^{90}Y	0.05	0.17
UMF 1500	^{137}Cs	0.09	0.13

Table 3. Concentrations of man-made radionuclides in bottom sediments of the Techa River and the Miass River. The errors indicated in the Table correspond to the standard deviations of 5 repeated measurements (for values > DL) or standard uncertainties of estimates (for BDL results).

Sampling site	^{137}Cs, Bq kg^{-1} dry weight					^{90}Sr, Bq kg^{-1} dry weight		
	09.2011	04-05. 2012	08-09. 2012	04-05. 2013	08-09. 2013	09.2011	04-05. 2012	08-09. 2012
RM	90±90*	40±40*	40±40*	190±50	-	70±70*	20±20*	20±20*
RT1	16700±800	7500±400	44000±2000	220000±20000	78800±4000	1700±100	920±50	4400±200
RT2	2800±100	4100±20	11600±500	900±100	620±70	800±100	430±30	1080±50
RT3	790±50	340±40	1390±80	180±40	130±20	220±50	39±7*	140±10*

* BDL results

Table 4. Specific activity of ^{137}Cs and ^{90}Sr in zoobenthos and filamentous algae samples (mean±error of mean).

Sampling site	^{137}Cs		^{90}Sr	
	Number of samples	Bq/kg dry weight	Number of samples	Bq/kg dry weight
Gastropods				
RM	5	500±200	6	150±90
RT1	4	14000±7000	5	11000±1000
RT2	7	1900±700	7	9000±1000
RT3	3	800±400	4	6000±1000
filamentous algae				
RM	1	40±10	1	40±10
RT1	2	8000±4000	4	2500±300
RT2	3	1300±300	2	2400±400
RT3	4	180±20	3	1120±70

Table 5. Body-averaged concentrations of ^{137}Cs and ^{90}Sr for fishes caught at different sample sites. Results represented the average values and error of mean in the units of Bq/kg.

Sampling site	^{137}Cs										^{90}Sr					
	Pearch				Roach				Pike		Pearch		Roach		Pike	
	phytophagous		carnivores		phytophagous		carnivores									
	N	A±err	N	A±err	N	A±err	N	A±err	N	A±err	N	A±err	N	A±err	N	A±err
RM	38	13±2	7	3±0.8	31	10±2	10	3.7±0. 7	29	7±2	42	1.2±0. 2	41	1.6±0.5	29	1.7±0. 4
RT1	45	260±30	14	210±30	40	200±20	15	140±10	38	180±10	59	960±60	55	1310±80	38	480±30
RT2	18	150±40	25	60±10					31	60±20	43	550±30			31	350±20
spring					22	190±30	9	60±10					31	640±70		
fall					12	80±40	21	50±5					33	1100±100		
RT3	41	30±10	14	8.4±0.7	30	20±30	26	30±20	15	4.7±0. 7	55	220±10	56	580±40	15	150±20

Table 6. Distribution coefficients of ^{90}Sr and ^{137}Cs in comparison to default values used in ERICA Tool and RESRAD BIOTA

Sampling site	^{137}Cs		^{90}Sr	
	Kd $*10^5$, l kg^{-1}	ERICA Tool/ RESRAD BIOTA *Kd* $*10^5$, l kg^{-1}	*Kd*, l kg^{-1}	ERICA Tool/ RESRAD BIOTA *Kd*, l kg^{-1}
RT1	2.04 (0.16-6.88)	1.37 / 0.005	140 (24-730)	2000 / 30
RT2			57 (17-135)	
spring	0,16 (0.02-0.53)			
fall	0,56 (0.07-1.93)			
RT3	0,28 (0.02-1.4)		16 (4-36)	
RM	0.057 (0.008-0.19)		5400 (500-17500)	

Table 7. ^{137}Cs concentration ratios for perches, roaches and pikes.

Species	N	Mean CR^{Cs-137} ± error of mean, l kg^{-1}	median (25%-75%), l kg^{-1}
Perch			
phytophagous	118	660±60	420 (340-700)
carnivores	55	450±60	340 (80-610)
Roach			
phytophagous	104	660±50	600 (300-900)
carnivores	103	440±30	360 (190-570)
Pike	112	500±50	370 (120 – 600)

Table 8. Fitting parameters of generalized logistic distribution functions of CR^{Cs-137} for carnivores and phytophagous fishes

Parameter of GLD distribution	Carnivores	phytophagous
k	0.273	0.353
σ	187	230
μ	363	504
Min-Max*, l kg^{-1}	25-8730	15-1770

*Empirical results

Table 9. CR^{Sr-90} for perches, roaches and pikes caught in the Techa River and the Miass River

Species	The Techa River		The Miass River	
	N	CR^{Sr-90} ±error of mean, l kg^{-1}	N	CR^{Sr-90} ±error of mean, l kg^{-1}
Perch	76	24±1	24	50±10
Roach	96	58±4	18	130±10
Pike	45	15±1	13	40±6

Table 10. Fitting parameters of generalized logistic distribution functions for the universal parameter of ^{90}Sr bioaccumulation U (g kg^{-1}) for perch, roach and pike. N is number of samples used for the distribution fitting.

Parameter of GLD distribution	Perch (N= 112)	Roach (N=114, 110)	Pike (N=59)
K	0.274	0.405	0.290
σ	0.507	1.240	0.267
μ	1.790	3.780	1.210
Min-Max*, g kg^{-1}	0.047-6.95	0.015-21	0.463-43.4

*Empirical results

Annex 1.

Hydrochemical indexes of water

Table A1.1. Hydrochemical indexes of water at sampling station, Summer 2012.

N	Indexes	Sampling site			
		RM 1	RT 1	RT 2	RT 3
1	pH	8.3 ± 0.1	7.7 ± 0.2	7.7 ± 0.2	7.2 ± 0.2
2	Oxygen concentration, mg/L	11.06 ± 0.03	6.7 ± 0.1		
3	Hardness of water, mmol/L	4 ±1	3.3 ± 0.5	3.3 ± 0.5	3.4 ± 0.5
4	Mineralization (dry residue), g/L	0.3	0.5	0.5	0.5
5	Total alkalinity, mmol/L	2.4 ± 0.2	5.1 ± 0.4	5.1 ± 0.4	5.4 ± 0.4
6	COD, mg/L	4.44	6.32	4.56	3.6
7	K^+, mg/L	4 ± 2	7.5 ± 0.8	7.4 ± 0.7	6.8 ± 0.7
8	Ca^{2+}, mg/L	40 ± 20	85 ± 9	71 ± 7	80 ± 8
9	Mg^{2+}, mg/L	23 ± 5	29 ± 3	38 ± 4	33 ± 3
10	Na^+, mg/L	30 ± 20	61 ± 9	55 ± 8	70 ± 10
11	NH^{4+}, mg/L	0.3 ± 0.1	0.7 ± 0.3	0.5 ± 0.2	0.6 ± 0.3
12	HCO_3^{2-}, mg/L	150 ± 10	310 ± 20	310 ± 30	330 ± 30
13	SO_4^{2-}, mg/L	100 ±308	130 ± 10	120 ± 10	160 ± 20
14	Cl^-, mg/L	16 ± 9	57 ± 6	59 ± 6	40 ± 4
15	NO^{3-}, mg/L	2 ± 1	0.45 ± 0.09	2.5 ± 0.5	1.3 ± 0.3
16	NO^{2-}, mg/L	2 ± 2	4.6 ± 0.9	3.2 ± 0.6	5.3 ± 1.1
17	P, mg/L	0.6 ± 0.5	0.88 ± 0.09	0.10 ± 0.01	1.5 ± 0.2
18	Fe, mg/L	0.03 ± 0.01	0.040 ± 0.008	0.028 ± 0.006	0.05 ± 0.01
19	Cd, mg/L	0.002 ± 0.001	0.005 ± 0.001	0.002 ± 0.001	0.002 ± 0.001
20	Co, mg/L	<0.009	<0.009	<0.009	<0.009
21	Mn, mg/L	<0.003	<0.003	<0.003	<0.003
22	Cu, mg/L	0.009 ± 0.001	0.008 ± 0.002	0.010 ± 0.002	0.008 ± 0.002
23	Ni, mg/L	<0.007	<0.007	<0.007	<0.007
24	Pb, mg/L	<0.005	<0.005	<0.005	<0.005
25	Sr, mg/L	0.1 ± 0.1	0.08 ±	0.14 ±	0.11 ±

			0.02	0.03	0.02
26	Cr, mg/L	<0.018	<0.018	<0.018	<0.018
27	Zn, mg/L	0.028 ± 0.003	0.030 ± 0.008	0.025 ± 0.006	0.04 ± 0.01

The data presented according the annual report Russian-Norwegian project “The Characterization of the Current Status of Ichthyofauna in the Techa River”, 2013.

Annex 2. The deal with measurements, which are wholly or partially below the detection limit

Example 1. ^{90}Sr and ^{137}Cs contamination of the Miass River

In the example the radionuclide detection in the water of the Miass River (contaminated only due to global fallouts) sampled in September 2012 is represented. The measurements of ^{90}Sr were found to be below the DL but above the NMU. Therefore they cannot be considered as an analyte free and the repeated measurements (N=15) of water sample were compared with the repeated measurements of the blank (N=15). In the figure A2.1a the comparison of distributions of measurement results is shown. As it can be seen the histograms are not coincided. Both are well fitted by normal distributions ($\alpha<0.05$). The widths of distributions do not differ significantly; but the mean values are greater than would be expected by chance. This means that there is a statistically significant difference between the blank and the water measurements ($t = -3.170$ with 28 degrees of freedom; $P = 0.004$). The difference of the mean values corresponds to 0.07 Bq; 90% confidence interval obtained with Monte Carlo simulations is found as 0.04-0.1 Bq. In terms of specific activity the ^{90}Sr contamination of the Miass River is about 0.0035 Bq/l (0.002-0.005 Bq/l).

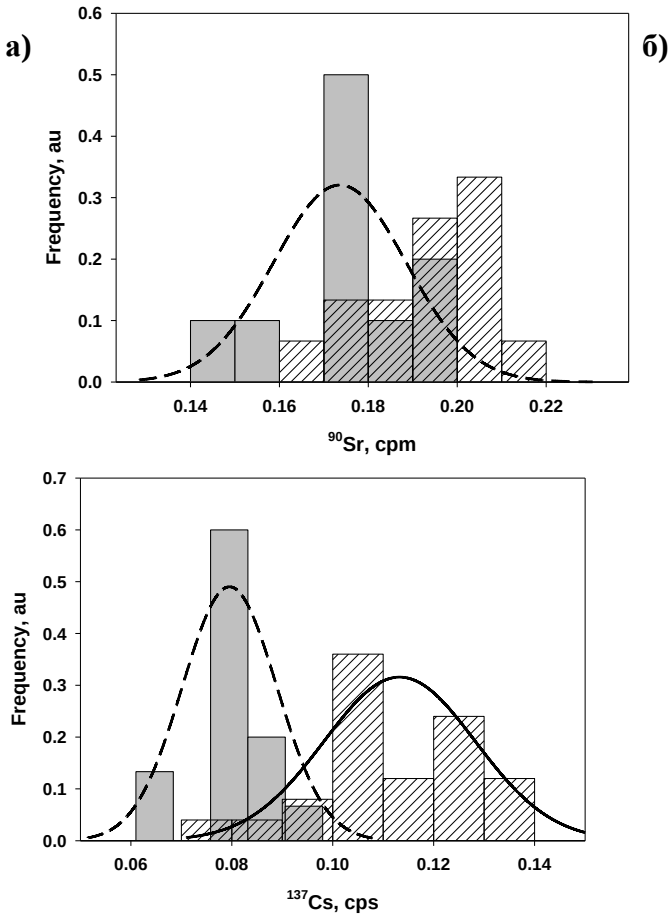


Fig. A2.1 - Comparison of blank and water sample measurements for the Miass River: a) ^{90}Sr; b) ^{137}Cs

About 20% of the ^{137}Cs measurements were BDL. Therefore the similar comparison of the measurement results of the blank (N=15) and water samples (N=25) was performed (Fig. A2.1b). The difference of the mean values corresponded to 0.26 Bq (90 % confidence interval obtained with Monte Carlo simulations is from 0.17 to 0.30 Bq), which corresponds to 0.012 Bq/l (0.009-0.015 Bq/l).

The measurements of the water and bottom sediments collected at different sites and demonstrated relatively low levels of contamination were treated similarly to the method described above.

Example 2. The assignment of an activity value to BDL measurement of ^{137}Cs contamination of perch body

Specimens captured in April of 2012 had the average weight 23 g, which is in 7 times lower than that of perch captured in summer. However no reliable mass dependence of radionuclide concentration was observed and for all capture times the BDL measurements amounted 75%. Therefore the measurement results were pooled together. The measurements of the samples with different weight was adjusted for the same (minimal) sample mass and compared with blank measurements. Figure A2.2 represents the comparison assuming mean blank as zero. A best fit of the distribution shapes were found. In this example there were normal and lognormal (with 3 parameters) distributions (Eqn A2.1 and A2.2).

$$blank \in b(x) = \frac{e^{-0.5(x-\mu)^2/\sigma^2}}{\sigma\sqrt{2\pi}}, where\ \mu = 0; \sigma = 0.012 \quad \text{A2.1}$$

$$Cs \in c(x) = \frac{e^{-0.5(\ln(x-\gamma)-\mu)^2/\sigma^2}}{(x-\gamma)\sigma\sqrt{2\pi}}, where\ \mu = -2.46; \sigma = 0.175; \gamma = -0.065 \quad \text{A2.2}$$

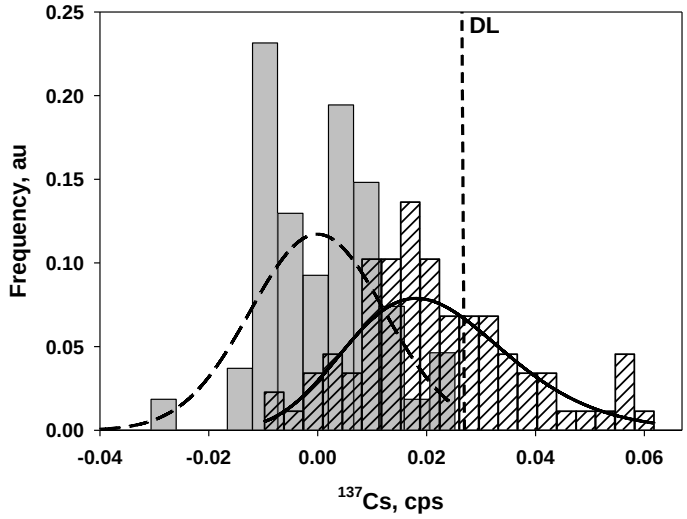


Figure A2.2. Comparison of blank (gray bars) and perch (cross-hatched bars) measurements. Long-dashed line is fitting of the blank data by normal distribution; sold line is fitting of ^{137}Cs measurements by lognormal distribution. The threshold indicated by short-dashed line corresponds to the detection limit (DL).

Random sampling of 1000 value pairs ***C*** and ***B*** taken from distributions (A2) and (A1) correspondingly was performed. The difference ***C-B*** reflects the expected excess of a sample response over the blank response. Two distribution of ***C-B*** were found, viz., (1) for all value pairs and (2) only for the cases with B<DL. Figure C represents the distributions. The first distribution corresponds to group average activity of ^{137}Cs in fish body. The second one allows interpreting the BDL values. Both data sets are distributed symmetrical, however they are not fitted well by normal distribution (according Shapiro-Wilk test P=0.011 and 0.043 respectively).

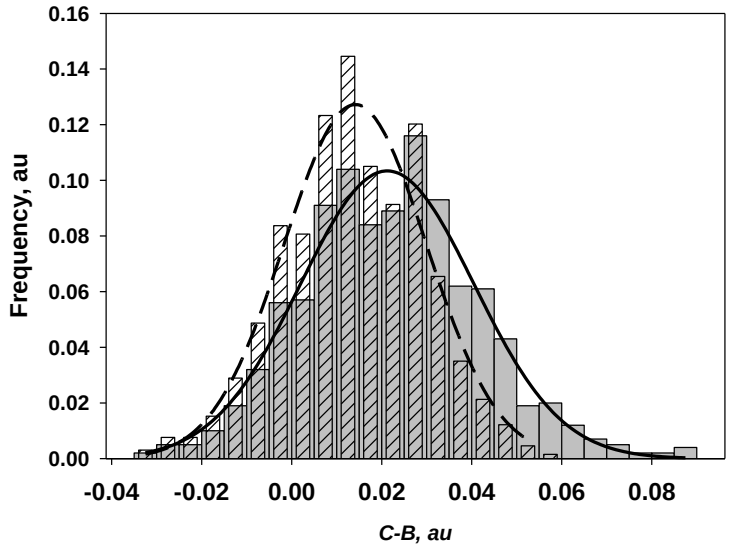


Figure A2.3. Distributions of simulated difference between ^{137}Cs and blank detection. Gray bars represents the difference for all simulated values. The curves are normal distribution fits. Cross-hatched bars corresponds to the selection of the data with B<DL.

As a result the average activity of ^{137}Cs in the body of perch caught in the Miass River has calculated as about 0.2±0.2 Bq. The expected value for the BDL results has been found as equal to 0.13±0.13 Bq. Uncertainty of the estimates has considered both the width of a distribution (Fig. A2.3) and a sample mass variation (the count to dose conversion is depending on sample mass). This value can be assigned as a surrogate ^{137}Cs activity for the Miass River perches with BDL measurements of ^{137}Cs.

The measurement results of ^{90}Sr and ^{137}Cs for different species caught at different sites were treated similarly according the method described above.