



Seasonal changes in uptake and depuration of ^{137}Cs and ^{90}Sr in silver Prussian carp (*Carassius gibelio*) and common rudd (*Scardinius erythrophthalmus*)

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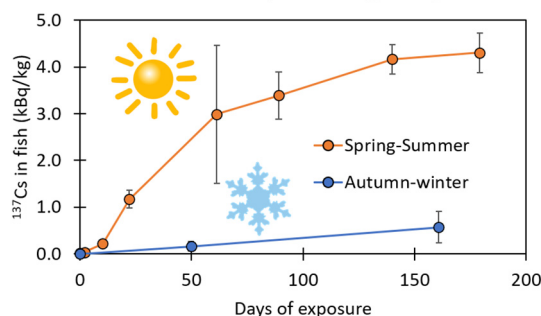
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HIGHLIGHTS

- Transplant field experiments in lakes have provided new information on transfer of radionuclides to fish
- Transfer of radionuclides to fish depends upon season
- Uptake rates for ^{137}Cs and ^{90}Sr in fish during summer are significantly higher than during winter season
- Biological half-lives of ^{137}Cs in fish muscle in winter are significantly higher than in summer

GRAPHICAL ABSTRACT

Accumulation of ^{137}Cs in fish (*Carassius gibelio*)



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ABSTRACT

Dynamic transfer of radionuclides to fish was studied in a series of experiments under field condition in two lakes within the Chernobyl exclusion zone during 2016–2020. “Clean” common rudd (*Scardinius erythrophthalmus*) and silver Prussian carp (*Carassius gibelio*) were transported to the contaminated Glubokoye Lake and kept in cages during several months of exposure, while contaminated Glubokoye fish were kept in cages in the “clean” Starukha Lake. Radiocaesium (^{137}Cs) and radiostrontium (^{90}Sr) were determined in intestine contents, muscle and bone tissues based on repeated samples during several months of exposure.

During summer, the activity concentrations of ^{137}Cs and ^{90}Sr increased with time of exposure in clean fish caged in the contaminated lake. During autumn and winter, however, minor changes in fish uptake occurred during several weeks of exposure to the contaminated water. Furthermore, depuration in the contaminated fish was significant during summer, while insignificant during winter when exposed in the «clean» water. The rate constant of ^{137}Cs uptake in muscle was between 8.0 and 22 day^{-1} during summer, while 0.2 to 1.0 day^{-1} during autumn-winter. Similarly, the rate constant of ^{90}Sr uptake in bone was between 1.4 and 1.6 day^{-1} , while 0.08–0.52 day^{-1} during autumn-winter. Biological half-lives of ^{137}Cs in fish muscle tissue in summer were 77 ± 10 days, while exceeded 230 days during seasons at low water temperature.

The results demonstrated that the transfer of ^{137}Cs and ^{90}Sr to fish was highly dependent upon seasons, in particular the water temperature. The transfer data obtained during low water temperature seasons deviated significantly from transfer data in literature and handbooks. Thus, seasonal changes in radionuclide transfer to fish should be taken into account when radiological impact to fish is assessed.

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1. Introduction

Following the Chernobyl and the Fukushima Daiichi nuclear accidents the activity concentration of radionuclides such as radiocaesium (^{137}Cs) in freshwater fish collected from contaminated water systems reached hundreds of kBq kg^{-1} (IAEA, 2006; Kryshev, 1995; Wada et al., 2016). Still 30 years after the accident in Chernobyl and 5 years after the Fukushima Daiichi accident, the activity concentrations of ^{137}Cs in freshwater fish species situated in closed lakes within the exclusion zones (ChEZ in Chernobyl and FEZ in Fukushima) were hundred times higher than permissible levels for food (Balonov et al., 2018; Gudkov, 2008; Kaglyan et al., 2012; Lerebours et al., 2018; Wada et al., 2019). A similar trend is also reported for radiostrontium (^{90}Sr) in ChEZ (Balonov et al., 2018; Gudkov, 2008; Kaglyan et al., 2012; Lerebours et al., 2018). Due to the high activity levels of ^{137}Cs and ^{90}Sr , the ChEZ represents a unique observatory for field experiments.

Uptake of radionuclides in fish depends on several abiotic factors such as the radionuclide and the speciation, competing ions, robustness of the ecosystem and on biotic factors such as food supply, fish species and life stages (Beresford et al., 2013; Chowdhury and Blust, 2001; Giblin et al., 2015; Pinder III et al., 2009). Knowledge of factors controlling the bioavailability and uptake of radionuclides is critical for understanding bioaccumulation and related impact for organisms as well as for humans (Metian et al., 2019).

Within radioecology and radiological protection, robust models are required to predict the partitioning of radionuclides between environmental media compartments and their transfer through food-chains as well as predicting biological responses within reasonable uncertainties. Comprehensive databases and handbooks are available internationally (IAEA, 2010) and are commonly used although information on underlying processes influencing uptake and early responses is scarce. Current models applied for estimating radionuclide transfer through aquatic food-chains rely primarily on concentration ratios (CR) or bioaccumulation factors (BCF) calculated by simply dividing the activity concentration of a radionuclide in the aquatic organism (Bq kg^{-1} ww) by the activity concentration in water (Bq kg^{-1} or Bq L^{-1}), assuming that equilibrium conditions are attained (Brown et al., 2008). The alternative approach is to apply biokinetic models, which account for time dependent uptake of radionuclides to aquatic organism via various pathways such as absorption directly from the water column via gills and assimilation through the gut epithelium following ingestion of contaminated food, as well as time dependent depuration.

The main route of ^{137}Cs uptake in freshwater fish is via diet, as the uptake through the gills is relatively low (Haque et al., 2017; Hewett and Jefferies, 1976; Smith, 2006). Thus, information about the contamination level in the diet and the amount consumed can be applied to estimate the uptake of ^{137}Cs in fish. The highest activity concentration of ^{137}Cs is generally reported in predatory fish (Smith et al., 2000; Lerebours et al., 2018). However, during the first years after the Chernobyl accident the highest activity concentration of ^{137}Cs was reported in the non-predatory fish species silver Prussian carp (*Carassius gibelio*) and the predatory species were the second most contaminated (Ryabov et al., 1998). In contrast, there is no consensus about the uptake route of ^{90}Sr in fish (Chowdhury and Blust, 2001; Kryshev, 2003; Ophel and Judd, 1962; Ophel and Judd, 1967; Smith, 2006). Additionally, the activity concentration of ^{90}Sr in non-predatory fish species is reported to be higher than in predatory fish species (Lerebours et al., 2018; Kaglyan et al., 2019). Furthermore, there seems to be limited available information on comparative experiments and associated uptake rate constants of ^{137}Cs and ^{90}Sr in fish either via waterborne exposure or via diet.

Bioaccumulated radionuclides are not homogeneously distributed between tissue and organs, and the tissue distribution depends upon

the chemical property of the radionuclide. In teleost fish, ^{137}Cs accumulates in soft tissues such as muscle (Poston and Klopfer, 1986; Suzuki et al., 1979), while ^{90}Sr accumulates predominately in skeletal tissues and scales (Boroughs et al., 1956). Muscle tissues typically contain 55–67% of ^{137}Cs and only 2–4% of ^{90}Sr in fish, while skeletal tissues contain typically 91–97% of ^{90}Sr (Gudkov, 2008; Yankovich et al., 2010). Furthermore, stable analogues can compete with radionuclides in transfer to biota (e.g., potassium, K can compete with ^{137}Cs and calcium, Ca can compete with ^{90}Sr) and thereby reduce the uptake (Chowdhury and Blust, 2001; Smith et al., 2000; Wada et al., 2019). Thus, the fish-to-water concentration ratios (CR) for ^{137}Cs and ^{90}Sr are reported to be inversely related to K and Ca in the surrounding water (Rowan and Rasmussen, 1994; Smith et al., 2000). Therefore, the concentrations of K and Ca have been incorporated in tabulated calculations of CRs for ^{137}Cs and ^{90}Sr , respectively (IAEA, 2010).

Although much attention has been given to determine the transfer of the most important fission yield radionuclides ^{137}Cs and ^{90}Sr from water to fish, information on key influencing factors under natural conditions is still scarce. There is also a conceptual problem when results from laboratory experiments are extrapolated to natural ecosystems such as lakes. In the field, food web structures are complex and the interaction between predators and prey are dynamic, varying with time and space. Following uptake of radionuclides via food, the CR of ^{137}Cs under natural conditions is usually higher than in the laboratory experiment (Chebotina et al., 1992). Furthermore, the effective half-life of ^{137}Cs in fish has also been reported to be highly variable between species (Smith et al., 2002), as well as within the same species (e.g., from 107 days to 385 days for *Cyprinus carpio* (Garnier-Laplace et al., 1997; Kryshev, 2002)). Similarly, the effective half-life of radiostrontium in the body of fish has also been reported to vary significantly, from 38 to 500 days (Kryshev, 2006; Shekhanova, 1983; Tjahaja et al., 2012). Hence, predicting transfer of pollutants under natural conditions especially from laboratory experiment will suffer from very large uncertainties (De Ruiter et al., 2005). It is therefore hypothesized that natural conditions, in particular the seasonality, would influence the metabolism of fish and thereby key variables such as uptake, accumulation and depuration. The objective of the present work was therefore to characterize the uptake and depuration rates of ^{137}Cs and ^{90}Sr in two non-predatory fish species during spring-summer and autumn-winter field conditions in order to establish CR and biological half-lives and to identify if the field observations would deviate from literature data.

2. Material and methods

Reciprocal transplant experiments with fish were performed in freshwater lakes within the Chernobyl exclusion zone (ChEZ) to determine the dynamic transfer of ^{137}Cs and ^{90}Sr to fish under field conditions. Fish with low radionuclide concentrations was transferred into a Chernobyl contaminated lake and fish from the contaminated lake was transferred into a low radionuclide concentration lake. Uptake and depuration were monitored during several months of exposure.

2.1. Study sites

The Glubokoye Lake (N 51.444796°, E 30.063938°) is one of the most radioactive contaminated lakes within the ChEZ and was used as experimental site. Glubokoye Lake is situated in the northwest track of the Chernobyl radioactive fallout plume at a distance of 6.5 km (338°) from the Chernobyl Nuclear Power Plant (Gudkov, 2008). The lake has a length of 1.2 km with a maximum width of 250 m and a depth of 6–7 m in the central part. Over the past 15 years, the activity concentrations of the ^{137}Cs and ^{90}Sr have been

about 2–10 Bq L⁻¹ and 100 Bq L⁻¹, respectively, and seem to remain quite stable (Baloga et al., 2011).

The Starukha Lake (N 51.358174°, E 30.207388°) with low activity concentration of radionuclides was used as a control lake. The lake is situated 8.1 km south-east of the Chernobyl NPP (115°) and outside of the north-west plume of radioactive fallout (Fig. 1).

2.2. Fish species and experimental design

Nineteen separate cage experiments were performed with two domestic and robust non-predatory fish species (Common rudd (*Scardinius erythrophthalmus*) and silver Prussian carp (*Carassius gibelio*)) caged in the ChEZ lakes Glubokoye Lake and Starukha Lake to study the uptake and depuration of radionuclides. Rudd was easy to catch in the lakes and typically feeding from the water column and the surface. The silver Prussian carp is known to be quite insensitive to handling and has also been used in many experimental studies within radioecology (Chowdhury and Blust, 2001; Garnier-Laplace et al., 1997; Ophel and Judd, 1962). The carp is typically feeding from the sediment surface and the water column. The exposure varied between 2 and 6 months during spring-summer and autumn-winter seasons 2016–2020.

Uptake dynamics and accumulation of the radionuclides were studied in fish containing low initial concentrations of radionuclides. The initial concentration served as background and starting point of the uptake model:

- “Clean” common rudd (aged 1+ to 2+ years old, body weight of 22 ± 16 g) were caught in the Starukha Lake. Initial background activity

concentrations: 50 ± 10 Bq ¹³⁷Cs kg⁻¹ in muscle tissue and 400 ± 100 Bq ⁹⁰Sr kg⁻¹ in bones.

- “Clean” silver Prussian carp (aged 1+ to 2+ years old, body weight of 15 ± 10 g) were caught in a floodplain lake in a suburb of Kiev (N 50.663383°, E 30.722267°). Initial background activity concentrations: <10 Bq ¹³⁷Cs kg⁻¹ and <10 Bq ⁹⁰Sr kg⁻¹ in muscle and bone tissue, respectively.

The “clean” fish were transferred to Glubokoye Lake, kept in cages and exposed to the contaminated water to follow the uptake dynamics.

The depuration dynamics of radionuclides was studied in the same fish species contaminated in Glubokoye Lake and transferred to “clean” water in Starukha Lake for several months. The contamination levels served as background and starting point for the depuration model:

- Contaminated rudd (aged 1+ to 2+ years old, body weight of 18 ± 15 g) were caught by rod fishing in Glubokoye Lake. Initial background activity concentrations: 11 ± 5 kBq ¹³⁷Cs kg⁻¹ in muscle tissue and 95 ± 11 kBq ⁹⁰Sr kg⁻¹ in bones.
- Contaminated silver Prussian carp: As small wild silver Prussian carp were difficult to catch in Glubokoye Lake, “Clean” silver Prussian carp exposed for several month (June–November) in Glubokoye Lake were used to study depuration. Initial background activity concentrations: 2.5 ± 0.4 kBq ¹³⁷Cs kg⁻¹ in muscle tissue and 7 ± 2 kBq ⁹⁰Sr kg⁻¹ in the bones (aged 1+ to 2+ years old, body weight of 28 ± 6 g).

To study the effects of seasonality, fish were typically transferred to the lakes during spring (May–June) with high water temperature as

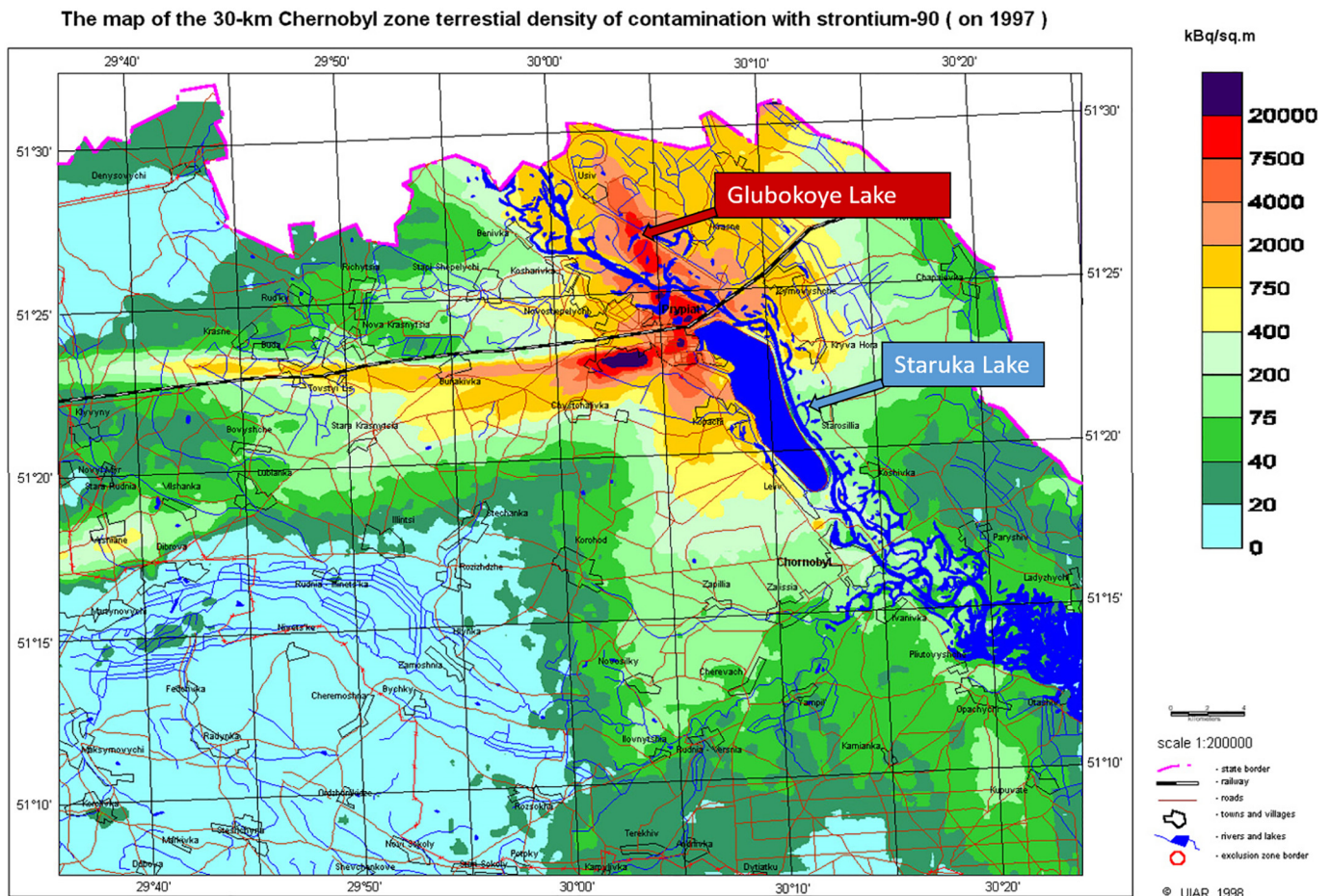


Fig. 1. Location of Glubokoye Lake and Starukha Lake within the Chernobyl exclusion zone (UIAR MAP of ChEZ).

well as during late autumn (October–November) with low water temperature.

Fish were kept in cages of $1 \times 1 \times 1$ m in size, drawn with a plastic mesh of 1 cm and lid at the top. During summer, the cages were placed at the bottom of the lake at a depth of 0.8–1.2 m with the upper part 10–20 cm above the water surface. During winter, cages were placed at a depth of 1.2–1.5 m to ensure that the upper part of the cage was below the ice cover. The species were kept in separate cages with about 40–60 fish in each. The fish could feed on natural food that entered the cage from sediments, water and deposition. Thus, the fish were exposed to the contaminated ecosystem in Glubokoye Lake during a period of 2–11 months with no additional food supply, while fish in Starukha Lake were exposed to “clean” water and natural occurring food organism during the same periods. Control cages with, “clean” rudd from Starukha Lake and contaminated rudd from Glubokoye Lake were kept in their original lakes as controls to elucidate any caging and handling effects.

The number of fish in the cages decreased gradually by increasing time of exposure due to sampling, to 7 fish at the end of each exposure period. To study the transfer of ^{137}Cs at individual level with increasing time of exposure, fish were tagged with pit tag before transferred to separate cages during different seasons in 2018, 2019 and 2020. Seven tagged fish were kept in each cage (4 cages for uptake and 2 cages for depuration).

In total, 13 separate cages were included to study the uptake (3 cages with rudd and 10 cages with carp) and 6 cages were included to study the depuration of radionuclides in fish (4 and 2 cages with rudd and carp, respectively). The field experiment received an ethics approval from the NUBiP commission of Ukraine.

2.3. Water sampling

Water samples were collected from each lake regularly during each experimental period. To obtain information about activity concentration of radionuclides in the lake water, 1 L was collected from Glubokoye Lake and 20 L from Starukha Lake. Water was filtered (0.45 μm or 1 μm) directly *at site*, or after acidification (0.1% HNO_3) in the laboratory.

To obtain information of general water quality parameters, three replicates of 50 mL samples of unfiltered and *at site* filtered water were collected. Information of major anions were obtained by using Lachat IC5000 Ion chromatograph (Zellweger analytics Inc. USA) and dissolved organic carbon (DOC) by using carbon analyser (Shimadzu TOC5000) in separate 0.45 μm filtered water samples (50 mL). Temperature, pH and conductivity were regularly determined *in situ* (WTW). In addition, water temperature was logged continually using Onset HOBO UA-001-64 Waterproof Pendant 64 K Temperature Data Loggers (Onset Computer Corporation, USA) located in the middle of the cage.

2.4. Fish sampling

Fish samples were collected before caging and at different time intervals during exposure, typically after 2 days, 1, 2, 4, 8 and 12 weeks. At each time point, samples were collected from 7 fish, randomly selected from each cage and sacrificed by a blow on their head. Weight and body length were measured, followed by blood sampling via a syringe, dissection and collection of muscle, intestine content and bone samples according to the EMERGE sampling protocol (Rosseland et al., 2001). Muscle tissue was collected after removing the skin. Small tissues were transferred to 5 mL plastic tubes (Sarstedt), while larger tissues were placed in plastic bags. All samples were stored cold before kept in freezer at -20°C until analysis. The weight of live tagged fish was determined regularly *at site* during exposure until the end of each set of experiments.

2.5. Determination of the activity concentration of ^{137}Cs and ^{90}Sr

Determination of ^{137}Cs activity concentration in water and fish samples (muscle and intestine content) was carried out using defined containers (Marinelli vessels with 1 L volume for water and vials of 5 cm^3 or 20 cm^3 for fish samples). Gamma measurements were performed using a low-background γ -spectrometric complex with a multi-channel analyser ASPEC-927 (software GammaVision 32) and high-purity Ge detector (GEM-30185, EG & G ORTEC, USA) with energy resolution of 1.78 keV at the ^{60}Co 1.33 MeV line in low background passive lead protection. The wet weight of samples was recorded using a scale (for large samples KERN pfb, accuracy of 0.01 g and for small samples AXIS AD200, accuracy of 0.001 g).

To determine whole body ^{137}Cs activity in tagged fish, 7 live fish from each cage were placed in a Marinelli vessel containing water, with a total mass of 1000 g. Gamma measurements (count rate) using the photopeak at 661.6 keV (counting time 600–1000 s) were carried out in the field using a scintillation gamma spectrometer SEG-05 (AKP, Ukraine) with lead shielding. Repeated ^{137}Cs activity measurements showed that although the fish were moving within the Marinelli vessel during the measurements, the scatter effects did not exceed 20%. After measurements, the fish were released into to the cages. Thus, whole body measurements allowed the ^{137}Cs activity level in the same group of fish to be followed during exposure in a series of experiments. At the end of each series of experiments, the ^{137}Cs activity from laboratory measurements of muscle tissues was compared with *at site* measurements. The obtained ratio was 1.5 ± 0.3 , and in agreement with results obtained for larger fish (Gudkov et al., 2008).

The ^{90}Sr activity in fish bone tissues was determined after ashing the samples in a muffle furnace at a temperature of 550°C , and measurements direct on a SEB-01-70 beta spectrometer (AKP, Ukraine). The ash content of silver Prussian carp bones and rudd was $20 \pm 3\%$ ($N = 35$) and $19 \pm 2\%$ ($N = 28$), respectively. The activity of ^{90}Sr in water samples as well as in bone and muscle tissues of fish with activity levels less than 1 Bq was determined using a standard radiochemical procedure (Pavlockaya, 1997). Based on house standards and certified reference materials, the accuracy of both ^{137}Cs and ^{90}Sr was judged to be good (JSAC 0785: 447 ± 25 Bq ^{137}Cs kg^{-1} and 12.4 ± 2.1 Bq ^{90}Sr kg^{-1} compared to CRM value 445 ± 12 Bq ^{137}Cs kg^{-1} and 11.5 ± 2.1 Bq ^{90}Sr kg^{-1} , respectively).

2.6. Stable element determination

The concentrations of stable elements such as ^{133}Cs and ^{88}Sr were determined in aliquots of water fractions and fish tissues using ICP-MS (Agilent 8800). Water samples (50 mL) were acidified (5% V/V HNO_3) prior to analysis, while aliquots of fish tissues were subjected to microwave oven assisted acid digestion using an UltraClave (Milestone) at 260°C . Ultrapure HNO_3 were used for all samples. After digestion, the samples were diluted with ultrapure water (18.2 M Ω cm^{-1}) before measurement. Analysed certified reference materials (CRM) indicated good accuracy (NCSZC73013: 0.13 ± 0.01 mg Cs kg^{-1} and 82 ± 2.8 mg Sr kg^{-1} compared to CRM values 0.13 ± 0.02 mg Cs kg^{-1} and 87 ± 5 mg Sr kg^{-1} , respectively, and DOLT-5: 3.63 ± 0.15 mg Sr kg^{-1} compared to CRM value 3.73 ± 0.26 mg Sr kg^{-1}).

2.7. Data analysis

The time dependent changes of the activity concentration of radionuclides in the body of fish $C_f(t)$ could be described by the linear differential Eq. (1) (Smith, 2006; Smith et al., 2002):

$$\frac{dC_f}{dt} = (k_f + k_w)C_w - (k_b + \lambda)C_f, \quad (1)$$

where $C_w(t)$ and $C_f(t)$ are activity concentrations of the radionuclides in water and fish (Bq kg^{-1}), respectively, at time t (days); k_f and k_w are the

rate of uptake of the radionuclides in fish by diet and water (day^{-1}), respectively; k_b is the rate of depuration of radionuclides from fish (day^{-1}); λ is decay constant $6.6 \cdot 10^{-5} \text{ day}^{-1}$ for ^{90}Sr and $6.3 \cdot 10^{-5} \text{ day}^{-1}$ for ^{137}Cs .

In the case of transferring “clean” fish ($C_f(0) = 0 \text{ Bq kg}^{-1}$) into a radioactive contaminated lake with water activity concentration (C_w , Bq kg^{-1}), the solution of Eq. (1) has the form of Eq. (2):

$$C_f(t) = \frac{(k_f + k_w) \cdot C_w}{(k_b + \lambda)} (1 - \exp(-(k_b + \lambda)t)) \quad (2)$$

Initially at small values, $((k_b + \lambda) \cdot t) < 0.5$, Eq. (2) can be approximated to a linear dependency shown in Eq. (3):

$$\begin{aligned} C_f(t) &\approx (k_f + k_w) \cdot C_w \cdot t \\ C_f(t)/C_w &\approx (k_f + k_w) \cdot t \end{aligned} \quad (3)$$

Using measured concentrations of radionuclides in fish ($C_f(t)$ from the first 100 days of exposure) and water (C_w), Eq. (3) was used to determine the uptake rates of radionuclides in fish for each cage during different seasons.

In case of transferring radioactive contaminated fish into a “clean” lake ($C_w = 0$), the rate of uptake ^{137}Cs in fish via diet and from water was insignificant ($k_f = 0$ and $k_w = 0$). In this case, the solution of Eq. (1) follows an exponential decline described in Eq. (4):

$$\begin{aligned} C_f(t) &= C_f(0) \cdot \exp(-(k_b + \lambda)t) \\ C_f(t)/C_f(0) &= \exp(-(k_b + \lambda)t) \end{aligned} \quad (4)$$

Eq. (4) was used to determine the rate of depuration of radionuclides from fish for each cage during different seasons. Thus, the biological half-life of the radionuclide determined from tissues or from the whole fish was $T_{1/2} = \ln(2)/k_b$, and the effective half-life was: $T_e = \ln(2) / (k_b + \lambda)$.

In situations where the activity concentration of radionuclides in fish is in equilibrium with the concentration in water, no changes in the activity concentration in fish would occur with time ($\frac{dC_f}{dt} \approx 0$). Consequently, using Eq. (1), fish - water bioconcentration factor (BCF) or concentration ratio (CR) of ^{137}Cs and ^{90}Sr can be described as shown in Eq. (5):

$$CR = \frac{C_f}{C_w} \approx (k_f + k_w) / (k_b + \lambda) \quad (5)$$

Concentration ratio (CR) of ^{137}Cs and ^{90}Sr in fish relative to water was calculated using Eq. (5), Bq kg^{-1} wet weight per Bq kg^{-1} water.

2.8. Statistical analysis

The uptake data was fitted to linear, polynomial and sigmoid curves, and best fit with the lowest Akaike and Bayesian information criterion (AIC/BIC) values was obtained by a linear model. Best fit for the depuration data was also obtained by a linear model after log transformation. Thus, the effect of season on time dependent changes in accumulation and depuration of ^{137}Cs in the intestine contents, of ^{137}Cs in muscle and ^{90}Sr in bone tissues were analysed using a linear mixed model (extension of linear regression models) with cage as experimental unit (random effect) using residual variance structure. The fixed effect was Season with number of Days exposed within each season as covariate.

Raw data such as activity concentration of fish (tissue or whole body measurements) was either collected from individuals or as an average of 7 fish at the day of sampling. When only average measurements were available, each fish was given an individual random value from the normal distribution defined by the average activity concentration

of the fish that date, with measurement uncertainties as the standard deviation.

To identify changes in activity concentration in fish tissues by increased time of exposure, the activity level was set to zero at the first date of each season regardless of previous exposure. Subsequent values within the same season and cage were calculated as changes from this initial value for the specific season. Season was generally defined by water temperature above and below 7°C . Day after start of exposure in different seasons was the linear regressor nested within season. Pairwise (Pearson's) correlations were used to calculate R^2 from all measured values for the variables evaluated. Statistical analyses were performed using JMP Pro v15.2.1 (SAS institute, Cary, NC, USA). The mixed model diagnostics indicated no major deviations from normally distributed error terms. The significance of average changes in activity concentration in fish tissues by increasing time of exposure (slope) and in mean activity between the seasons are reported for each outcome (Table S1). The uptake and depuration were somewhat different between each cage of fish and this explained 22.5–97.8% of the total variance. P-values < 0.05 was interpreted as significant.

3. Results and discussion

3.1. General characteristic and activity concentration of radionuclides in lake water

The Glubokoye Lake was characterized as hard water with pH 7.4–7.6, Ca concentration $30 \pm 2 \text{ mg L}^{-1}$, K concentration $1.2 \pm 0.1 \text{ mg L}^{-1}$, conductivity of $201 \pm 10 \mu\text{S cm}^{-1}$ and moderate concentration of dissolved organic carbon (DOC $12 \pm 2 \text{ mg L}^{-1}$). The concentration of stable Sr and Cs were $106 \pm 2 \mu\text{g L}^{-1}$ and $4.9 \pm 1.6 \text{ ng L}^{-1}$, respectively. The water quality in Starukha Lake was quite similar to Glubokoye Lake, with pH 7.2–7.6, conductivity $235 \pm 10 \mu\text{S cm}^{-1}$ and moderate dissolved organic carbon levels (DOC $9 \pm 4 \text{ mg L}^{-1}$). The concentrations of Sr and Cs were $145 \pm 5 \mu\text{g L}^{-1}$ and $4.4 \pm 1.5 \mu\text{g L}^{-1}$, respectively. Thus, the water quality and the concentration of stable analogues of ^{137}Cs and ^{90}Sr were relatively similar in both lakes although the K concentration was about a factor of 2 higher in Starukha Lake compared to Glubokoye Lake. The water temperature varied from 2°C in winter to 28°C in summer in both lakes (Fig. 2a) and was typically below 7°C from October to March, and above 19°C from May–September. The seasonal variations in water temperature were similar in both lakes.

The average activity concentration of ^{137}Cs and ^{90}Sr in Glubokoye water were $3.6 \pm 1.0 \text{ Bq L}^{-1}$ and $100 \pm 11 \text{ Bq L}^{-1}$, respectively, and did not vary significantly during the 4 years of study. This agrees with earlier reported activity levels in Glubokoye Lake (Baloga et al., 2011). The activity concentration of ^{90}Sr was more than a factor of 20 higher than ^{137}Cs , and the concentration ratio $^{90}\text{Sr}/^{88}\text{Sr}$ was $1.87 \cdot 10^{-7}$ (in μg) and a factor of 1000 higher than the concentration ratio of $^{137}\text{Cs}/^{133}\text{Cs}$ (in μg). The activity concentrations of ^{137}Cs and ^{90}Sr in Starukha water were $0.023 \pm 0.005 \text{ Bq L}^{-1}$ and $0.12 \pm 0.02 \text{ Bq L}^{-1}$, respectively, and more than two and three orders of magnitude lower, respectively, than the activity concentration of these radionuclides in Glubokoye water.

3.2. Fish

3.2.1. Condition of fish

The weight of rudd kept in cages decreased from May to September by 20–30% and was constant from September to November for both lakes. The decrease in weight could be attributed to limited access to food or caging effects of the wild fish. Control fish, such as “contaminated” rudd from Glubokoye Lake kept in cages remaining in Glubokoye Lake, showed similar activity concentration compared to wild native fish from the lake, $16.8 \pm 7.5 \text{ kBq kg}^{-1}$ and $12.8 \pm 4.6 \text{ kBq kg}^{-1}$, respectively. The caging effect was therefore judged to be of minor

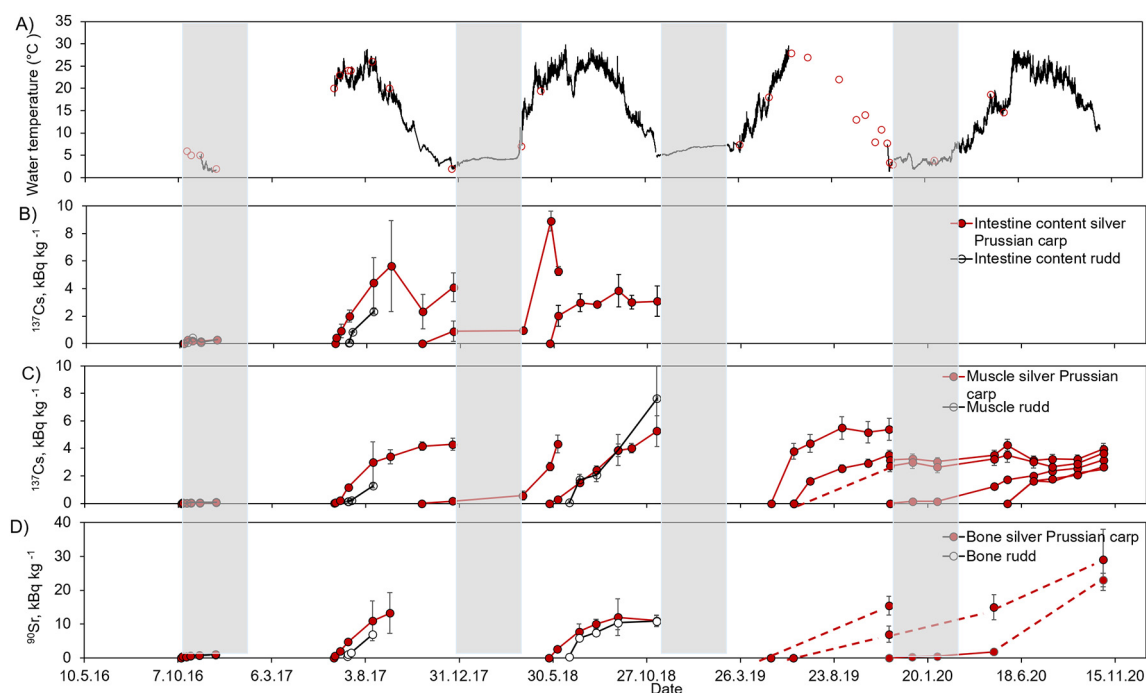


Fig. 2. Variation in water temperature (A), ^{137}Cs activity concentration in intestine contents (B), ^{137}Cs activity concentration in muscle tissue (C) and ^{90}Sr activity concentration in bone tissue (D) of rudd and silver Prussian carp ($N = 7$). Grey area indicates season with low water temperature.

importance. Minimal caging stress was also supported by the measured blood glucose level in caged rudd that were in the normal range (3.7 ± 1.9 mM) and well below levels associated with stressed fish (>7 mM, Kroglund et al., 2001).

The average weight of caged silver Prussian carp in Glubokoye Lake and in Starukha Lake increased from May to November with a factor up to 1.8 (Fig. S1). In contrast, from November 2019 to February 2020, at water temperatures below 7°C , the carp did not increase in weight. Lowered metabolism, reduced food access and feeding activity at lower temperatures was expected, and also reported by others, to result in reduced growth (Handeland et al., 2008; De Giosa et al., 2014).

3.2.2. Activity concentration of ^{137}Cs in intestine content

The activity concentration of ^{137}Cs in contents collected from the intestines of rudd and silver Prussian carp exposed in Glubokoye Lake varied during the year and reached 3 kBq kg^{-1} and 9 kBq kg^{-1} , respectively (Fig. 2b). The ^{137}Cs activity concentration in the intestine contents increased during spring-summer ($P < 0.0001$) and reached significantly higher levels in spring-summer season than in autumn-winter season ($P < 0.0001$), and highest in May. The low activity measured in the intestine contents during the autumn-winter season can most likely be attributed to reduced feeding (Handeland et al., 2008) and reduced access to contaminated feed organisms. However, reduced density of food supply as well as changes in diet composition with altered level of contamination during winter compared to spring and summer cannot be ignored.

3.2.3. Uptake of ^{137}Cs

During caging in May–June and exposure during summer, the ^{137}Cs activity concentration in the “clean” fish increased significantly in the contaminated Glubokoye Lake ($P < 0.0001$), following a first order exponential growth to a maximum (Fig. 2c). When caging fish during October–November, however, the ^{137}Cs activity concentration in fish did not change during autumn-winter. In comparison, the activity concentration of ^{137}Cs in fish exposed for a similar time period of 40 days during summer was more than a factor of 3 higher than during autumn

and winter (Fig. 3, Table S1). As observed for the intestine content, the uptake of ^{137}Cs in fish was high during summer with high activity concentration in the intestine contents, while low during winter with low activity concentration in the intestine contents and reduced fish metabolism due to the low water temperature (De Giosa et al., 2014; Handeland et al., 2008). Correlation between ^{137}Cs in intestine contents and muscle ($R^2 = 0.59$, $P < 0.001$) also supports that the ^{137}Cs uptake in fish tissues was linked to the feed. Although the activity concentration of ^{137}Cs was high in Glubokoye water, the waterborne uptake of ^{137}Cs seemed to be low in fish at seasons with low water temperature. This is also in agreement with previous findings showing that the waterborne uptake of ^{137}Cs in fish is limited (Haque et al., 2017; Hewett and Jefferies, 1976; Smith, 2006).

3.2.4. Depuration of ^{137}Cs

During caging in May–June and exposure during summer, the activity concentration of ^{137}Cs in contaminated fish decreased with time in the “clean” Starukha Lake (Fig. 4). Following 40–50 days of exposure, the ^{137}Cs activity concentration in muscle of rudd decreased with a factor of 2 and continued to decrease following a first order kinetics (Fig. 4, Table S1). When caging fish in “clean” water during autumn, however, no changes in the activity concentration of ^{137}Cs in fish could be observed after 2–3 months of exposure (Fig. 4, right panels). However, the activity concentration of ^{137}Cs decreased significantly in fish that continued to stay in the cage during the next spring and summer. Thus, the ^{137}Cs was still high in fish several weeks after transfer to “clean” water during the winter season, while the level decreased significantly during the summer season. The retention time of ^{137}Cs in fish was therefore highly dependent upon season and the water temperature.

3.2.5. Uptake of ^{90}Sr

During caging in May–June and exposure throughout the summer, the activity concentration of ^{90}Sr increased significantly in fish with increasing time of exposure in the contaminated Glubokoye Lake as observed for ^{137}Cs (Fig. 2d). However, when fish were transferred to the

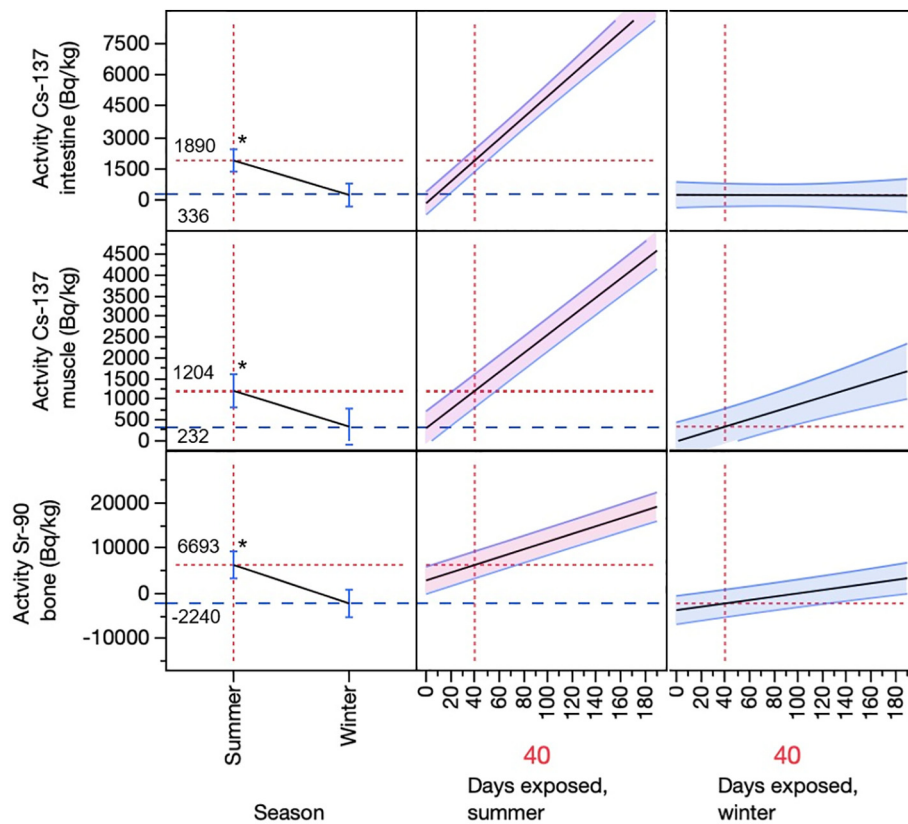


Fig. 3. Marginal model profiles present change in activity concentration of ^{137}Cs in intestine contents, ^{137}Cs in muscle and ^{90}Sr in bone as a function of days after start of exposure to the contaminated Glubokoye Lake at different seasons. Effect of season after 40 days of exposure are indicated with 0.95% confidence bars and * indicate significantly different between seasons ($P < 0.05$). Effects of days within season are illustrated with 0.95% confidence bands on the regression line. $N = 13$ cages.

cage in October–November the activity concentration of ^{90}Sr in bone tissues increased only slightly, as also observed for ^{137}Cs in muscle tissues. For long term caging (11 months, October–September), the ^{90}Sr activity in bone of silver Prussian carp stayed about constant during winter until spring when the activity increased significantly. As shown for ^{137}Cs , the ^{90}Sr uptake in fish was also significantly lower during the season with low water temperature compared to the spring and summer seasons (Fig. 3, Table S1), although the uptake mechanisms were most likely different. The low accumulation of ^{90}Sr during winter is probably due to minimal growth of fish because no changes in the weight of silver Prussian carp were observed during winter, while a 35% increase in weight was observed during summer. Minimal growth of fish during winter with low water temperature is well known from aquaculture in temperate regions even after more than sufficient food supply and high growth rate during summer at high water temperature (Handeland et al., 2008). When the age of fish is determined from skeletal structure such as scales and otoliths, the accumulation in bone structure is also shown to depend upon season. Although the uptake of ^{90}Sr can occur directly from water (Chowdhury and Blust, 2001; Ophel and Judd, 1962; Tjahaja et al., 2012) and the ^{90}Sr water activity concentration was about constant during the year, the ^{90}Sr accumulation in fish bone occurred mainly during seasons with high water temperature stimulating skeletal growth. The exponential rise in plankton and the increased abundance of food during spring seems to be of major importance because the ^{90}Sr activity concentration was higher in fish exposed from May than in fish exposed from June (Ophel and Judd, 1967) although the exposure times were similar. This is supported by the findings that the ^{90}Sr level increased in bone of fish with high growth rate due to addition of clean feed compared to fish with limited growth not receiving additional food (Kashparova et al., 2021).

3.2.6. Depuration of ^{90}Sr

The activity concentration of ^{90}Sr did not decrease in fish within the experimental time periods, up to 80 days, neither during summer nor during winter in the “clean” Starukha Lake (Fig. 4, Table S1). After eleven months of caging there were no significant changes in the ^{90}Sr activity concentration in the bone tissues. Thus, changes in the ^{90}Sr body retention time were too slow to be observed within the experimental period. Body retention times of radiostrontium ($T_{1/2}$) have been reported from 30 to 500 days (Kryshev, 2006; Tjahaja et al., 2012). No changes in the ^{90}Sr levels were observed during the 50–242 days field experimental period, indicating that the biological half-life of ^{90}Sr in fish could be significantly longer than previously reported.

3.3. Rates constants for uptake and depuration of ^{137}Cs and ^{90}Sr

The uptake rates of ^{137}Cs and ^{90}Sr in fish for each cage were determined using Eq. (3) and the least squares method based on the initial linear part of the exposure (Fig. S2). Results demonstrated that the uptake rate of ^{137}Cs in muscle tissue of rudd and silver Prussian carp ($k_f + k_w$, day^{-1}) was significant both during summer and autumn-winter (Table 1) and in the range 8.0–22 day^{-1} during summer at a water temperature above 19 °C, while significantly lower (0.3–0.9 day^{-1}) during seasons with water temperature less than 7 °C. Thus, the uptake rates were more than a factor of 10–50 higher during summer than during winter. The predicted uptake rates at low water temperature are in agreement with uptake rates (0.14–0.22) reported for salmonids in water temperatures between 1.4 and 10 °C (Yamamoto et al., 2015). However, uptake rates observed during summer significantly exceeded reported uptake rates of waterborne ^{137}Cs : $0.2 \pm 0.01 \text{ day}^{-1}$ at 20 °C

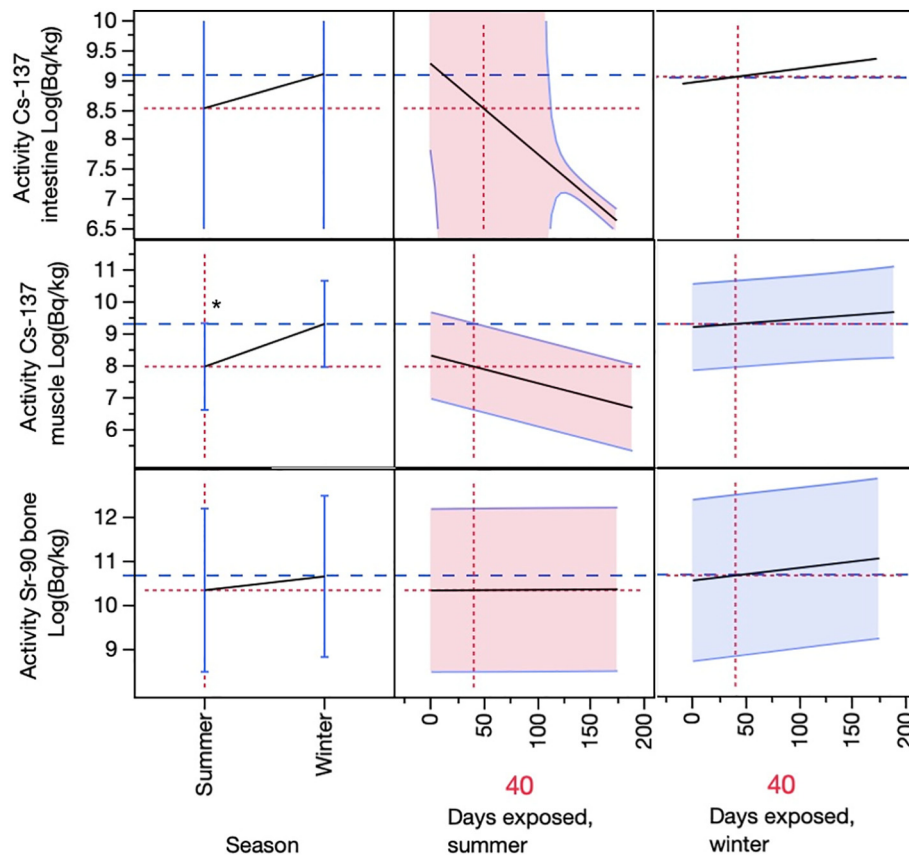


Fig. 4. Marginal model profiles present change in activity concentration of ¹³⁷Cs in intestine contents, ¹³⁷Cs in muscle and ⁹⁰Sr in bone as a function of days after introducing contaminated fish into the clean Starukha Lake at different seasons. Effect of season after 40 days of exposure are indicated with 0.95% confidence bars (* indicate significantly different between seasons (P < 0.05)), while effects of days within each season are illustrated with 0.95% confidence bands on the regression line. N = 6 cages.

(Garnier-Laplace et al., 1997) and 0.24 day⁻¹ at 20 °C (Lebedeva, 1966). Diet seemed to be a more important source of ¹³⁷Cs than water (Pan and Wang, 2016) which explains that the observed field uptake rates during summer exceeded reported laboratory uptake rates for waterborne ¹³⁷Cs. However, the main uptake during winter could be waterborne ¹³⁷Cs because the observed rates were similar to reported uptake rates for waterborne ¹³⁷Cs.

The uptake rates of ⁹⁰Sr in bone tissues of rudd and silver Prussian carp were significant both during summer and winter season, and in the range 1.4–1.6 day⁻¹ during seasons with water temperature above 19 °C, and only 0.08–0.5 day⁻¹ during seasons with water temperature less than 7 °C (Table 1). Thus, the uptake rates in water with temperature average of 23.5 ± 1.5 °C were more than a factor of 3–20 higher than in water with average 3.3 ± 1.7 °C. This variation exceeded

Table 1

Water temperature and uptake rate (k_f + k_w) of ¹³⁷Cs in muscle tissue and ⁹⁰Sr in bone tissues of fish collected from cages at different seasons. Data are from Glubokoye Lake based on several experiments during 2016 to 2020. The rates reflect waterborne and dietary uptake. R² is given for the correlation between observed uptake and days of exposure. Season spring-summer and autumn-winter is defined by water temperature above and below 7 °C, respectively.

Season	Species	Start exposure	Temperature, °C		¹³⁷ Cs muscle			⁹⁰ Sr bone		
			Average ± SD	Min-max	K _f + k _w , day ⁻¹	R ²	P - value	K _f + k _w , day ⁻¹	R ²	P - value
Spring-summer	Carp ^b	11.04.2018	19.8 ± 3.9	10.6–26.9	17 ± 4	0.94	P < 0.0001	–	–	P < 0.0001
	Carp ^b	24.05.2018	24.3 ± 2.2	17.9–29.8	9 ± 3	1.00		1.4 ± 0.3	0.99	
	Carp ^b	14.05.2019	20.3 ± 3.9	14–29.6	22 ± 5	0.96		>0.8	–	
	Carp ^b	13.06.2017	23.2 ± 1.8	18.2–28.8	13 ± 4	0.99		1.6 ± 0.4	0.99	
	Carp ^b	19.06.2019	21.9 ± 2.2	19.5–29.6	>5	–		–	–	
	Carp ^b	19.06.2019	21.9 ± 2.2	19.5–29.6	17 ± 5	1.00		>0.5	–	
	Carp ^b	26.05.2020	23.3 ± 4.4	15.0–28.6	10 ± 3	1.00		>0.7	–	
	Rudd ^a	25.06.2018	24.1 ± 2.4	17.9–29.1	15 ± 4	0.89		1.5 ± 0.4	0.81	
	Rudd ^a	05.07.2017	23.4 ± 2.0	19.6–28.8	8 ± 1	1.00		1.6 ± 0.1	1.00	
Autumn-winter	Carp ^b	12.10.2016	3.3 ± 1.7	1.1–6.2	0.3 ± 0.1	0.95	P < 0.0001	0.2 ± 0.1	0.86	P < 0.0001
	Rudd ^a	13.10.2016	3.3 ± 1.7	1.1–6.2	0.2 ± 0.1	0.45		0.13 ± 0.03	0.98	
	Carp ^b	31.10.2017	4.2 ± 1.1	1.9–11.2	1.0 ± 0.3	1.00		0.5 ± 0.1	0.86	
	Carp ^b	20.11.2019	3.7 ± 0.9	1.3–7.4	0.7 ± 0.2	0.89		0.08 ± 0.02	0.97	

“–” not determined due to too few sample points or not measured.

^a Rudd (*Scardinius erythrophthalmus*).

^b Silver Prussian carp (*Carassius gibelio*).

previously reported differences in uptake rates for ^{90}Sr in fish at comparable temperatures ($0.015\text{--}0.7\text{ day}^{-1}$ in bone at $18\text{--}20\text{ }^{\circ}\text{C}$ (Lebedeva, 1962), $0.2\text{--}0.8\text{ day}^{-1}$ at $15\text{--}20\text{ }^{\circ}\text{C}$ (Ophel and Judd, 1967)).

The results showed that season and associated water temperature had a large impact on the accumulation of radionuclides in fish. This is in contrast to previous findings by Pan and Wang (2016) reporting that temperature had only minor effects on the overall accumulation of ^{137}Cs in fish from estuarine water, although that study did not include fish at temperature below $16\text{ }^{\circ}\text{C}$. Comparing uptake rates of ^{137}Cs and ^{90}Sr , the rate of ^{137}Cs was a factor of 10 higher than for ^{90}Sr . Taking into account conversion factors of 1.1 and 0.14 for ^{137}Cs from muscle to whole body and ^{90}Sr from bone to whole fish body, respectively (Yankovich et al., 2010), the whole body uptake rates corresponded to $9.0\text{--}24\text{ day}^{-1}$ and $0.20\text{--}0.22\text{ day}^{-1}$ for ^{137}Cs and ^{90}Sr , respectively, during summer. Thus, the whole-body uptake rate of ^{137}Cs was about a factor of 40–60 higher than for ^{90}Sr during summer.

The depuration of ^{137}Cs in contaminated fish followed a first order kinetic decrease during summer for both rudd and silver Prussian carp (Fig. 4, Fig. S3), but no significant decrease was observed during winter (Table 2). The correlations between observed and first order kinetic predicted depuration rates ($k_b + \lambda$) are given in Table 2 for each experiment. The average depuration rate for ^{137}Cs from muscle tissue was $k_b = 0.009 \pm 0.001\text{ day}^{-1}$ during summer with water temperatures above $19\text{ }^{\circ}\text{C}$ (Table S1), corresponding to a biological half-life of 77 ± 10 days for ^{137}Cs in rudd and silver Prussian carp. The predicted half-life of ^{137}Cs in fish was in line with earlier reported values; 75 ± 35 days (Kryshev, 2003), 106 ± 5 days (Garnier-Laplace et al., 1997) and $84\text{--}100$ days (Kryshev, 2002). However, the depuration rates were significantly lower during winter ($P < 0.001$) and lower than 0.003 day^{-1} corresponding to a biological half-life of more than 235 days. The biological half-life increased with decreasing water temperature which also is in agreement with previous findings (Kryshev, 2002). Thus, the body retention of ^{137}Cs in fish seemed to be controlled by depuration during summer until the water temperature decreased to levels where metabolism was significantly affected and depuration more or less ceased.

During the experiments, no decrease in the ^{90}Sr levels in the fish bone tissue could be observed (Fig. 4). Thus, it was impossible to properly estimate the half-life, although it would probably exceed 500 days. The findings are in contrast to previous reports, where the ^{85}Sr depuration rate constant was 0.018 day^{-1} and Sr was eliminated from the bone tissues with effective half-life of about 30 days (Tjahaja et al., 2012), or with biological half-life of 77 days (Shekhanova, 1983). The lack of depuration during 342 days of exposure supports that the biological half-life would exceed $560 \pm 270\text{ day}^{-1}$ that was reported by (Kryshev, 2003). Results demonstrated also that long term chronic experiments are required to identify the biological half-lives of ^{90}Sr in fish under field conditions.

3.4. Concentration ratio (CR)

The concentration ratio of ^{137}Cs was in the range 1870 to 5042 based on the activity concentrations in muscle tissue of native rudd and silver Prussian carp caught in Glubokoye Lake and the activity concentration in water (C_f/C_w). CRs of caesium-isotopes have been reported for muscle tissue of freshwater fish to be in range 2400 to 15,000 depending upon the potassium (K) concentration in water (IAEA, 2010). For non-predatory fish, the CR was estimated to be 2742, based on IAEA 2010 (corr. factor $3290 \times K^{-1}$) and taking into account the K concentration of 1.2 mg L^{-1} observed in Glubokoye Lake. This corresponded very well with the obtained CR of ^{137}Cs (2583 ± 1663) for native rudd in Glubokoye and was similar to the CR (range: 1850–2450) reported for freshwater fish in Fukushima water with $1\text{--}1.7\text{ mg K L}^{-1}$ (Wada et al., 2019). The CRs for rudd and silver Prussian Carp based on kinetic parameters ($k_f + k_w = 8\text{--}22\text{ day}^{-1}$ and $k_b = 0.009 \pm 0.001\text{ day}^{-1}$) and Eq. (2) were calculated to be in the range 900–2400 during summer. Utilizing the kinetic parameters ($k_f + k_w = 0.2\text{--}1.0\text{ day}^{-1}$ and $k_b = 0.002\text{--}0.003\text{ day}^{-1}$) during seasons with low water temperature, however, the CRs were significantly lower and in the range 100–500. Thus, uptake during low temperature seasons would be much lower and the fish would be less contaminated than predicted using the recommended CRs (IAEA, 2010). During summer, however, the activity concentration in fish introduced to the contaminated water increased to about 5.8 kBq kg^{-1} , i.e., similar to levels observed in several years old native silver Prussian carp ($6.7 \pm 1.2\text{ kBq kg}^{-1}$). Increased CR of ^{137}Cs due to increased water temperature was also pointed out in the review by Metian et al. (2019), also referring to conflicting reported laboratory results. However, this 2019 review concluded that the temperature did not influence the uptake rate and the CR.

The concentration ratio of ^{90}Sr was 997 ± 61 and 630 ± 16 based on activity concentration in bone tissue of native rudd and silver Prussian carp, respectively, caught in the Glubokoye Lake (C_f/C_w). Utilizing the stable strontium concentration in fishbone ($86 \pm 6\text{ mg kg}^{-1}$) and in water, the CR (bone) was 840 ± 9 and 733 ± 7 of native rudd and silver Prussian carp, respectively, caught in the Glubokoye Lake, which corresponded well to the CR for ^{90}Sr . For freshwater fish the CR for ^{90}Sr in bone was estimated to be 275 based on IAEA 2010 (corr. factor exp. $(9.7\text{--}1.2 \ln(\text{Ca}))$) and taking into account the Ca concentration of 30 mg L^{-1} as observed in Glubokoye Lake. The CR in native fish was, however, a factor of more than 2 higher than predicted based on the equation for non-predatory fish (IAEA, 2010). Since the ^{90}Sr uptake rate decreased during seasons with low water temperature, however, it was expected that the CRs also would change, but to a lesser degree than for ^{137}Cs as the differences in rate constants were much larger. Results indicated, however, that the activity concentration in fish increased to $29 \pm 9\text{ kBq kg}^{-1}$ after about 500 days exposure to the contaminated water and to a significantly lower

Table 2

Depuration rate ($k_b + \lambda$) and effective half-life ($T_{1/2}$) of ^{137}Cs in muscle tissue of contaminated rudd and silver Prussian carp at different seasons from 2016 to 2020 in Starukha Lake. R^2 is given for the correlation between observed depuration and days of exposure. Season spring-summer and autumn-winter is defined by water temperature above and below $7\text{ }^{\circ}\text{C}$, respectively.

Season	Species	Experimental period	Temperature, $^{\circ}\text{C}$		^{137}Cs			
			Average \pm SD	Min–max	$k_b + \lambda$, day^{-1}	$T_{1/2}$, day	P-value	R^2
Spring-summer	Rudd ^a	14.05–19.11.2019	20.3 \pm 3.9	14.0–29.6	0.006 \pm 0.002	116 \pm 39	P < 0.0001	1.00
	Carp ^b	05.05–27.10.2020 ^c	20.0 \pm 4.4	10.4–28.9	0.009 \pm 0.001	77 \pm 9		0.93
	Carp ^b	05.05–27.10.2020	20.0 \pm 4.4	10.4–28.9	0.012 \pm 0.002	58 \pm 10		0.93
	Rudd ^a	13.06–31.10.2017	19.8 \pm 4.7	5.9–28.8	0.0093 \pm 0.0003	75 \pm 2		0.86
	Rudd ^a	25.06–11.09.2018	24 \pm 2.2	17.9–9.1	0.0102 \pm 0.0004	70 \pm 3		0.86
Autumn-winter	Rudd ^a	14.10–06.12.2016	3.3 \pm 1.7	1.1–6.2	<0.0002	>3500	P = 0.29	0.36
	Carp ^b	20.11.19–04.02.2020 ^c	3.7 \pm 1.3	0.5–6.9	0.003 \pm 0.001	231 \pm 77		0.96
	Carp ^b	20.11.19–04.02.2020	3.7 \pm 1.3	0.5–6.9	0.0006 \pm 0.0004	1155 \pm 770		0.71

^a Rudd (*Scardinius erythrophthalmus*).

^b Silver Prussian carp (*Carassius gibelio*), pit tagged and depuration were followed during both winter and summer season.

^c The average weight of silver Prussian carp in this cage was in range 40–60 g and factor of 2 higher than other groups.

level than observed in 1–2 years old native fish (95 ± 11 kBq kg⁻¹). The difference in activity concentration can be attributed to the presence of uncontaminated bone tissue in experimental fish prior to caging in the contaminated water. In such situations, where the native fish have been raised in uncontaminated lake water, the fish would be significantly less contaminated after nuclear fallout than predicted using the recommended CRs (IAEA, 2010).

4. Conclusion

Transplant field experiments with fish contained in cages within lakes in the Chernobyl exclusion zone (ChEZ) during several years have demonstrated that the transfer of ¹³⁷Cs and ⁹⁰Sr to fish are dynamic and that seasonality, especially the associated water temperature, has a significant influence on the accumulation of radionuclides.

Uptake rates of ¹³⁷Cs in muscle tissue and of ⁹⁰Sr uptake in bone tissue of common rudd (*Scardinius erythrophthalmus*) and silver Prussian carp (*Carassius gibelio*) were in range 8–22 day⁻¹ and 1.4–1.6 day⁻¹, respectively, during summer at a water temperature above 19 °C, while only 0.2–1.0 day⁻¹ and 0.08–0.5 day⁻¹, respectively, during seasons with water temperature less than 7 °C. The uptake rates of ¹³⁷Cs in muscle tissues and ⁹⁰Sr uptake in bone tissues were significantly lower during winter than during summer. Overall, the uptake rates for ⁹⁰Sr were significantly lower than for ¹³⁷Cs. Depuration rates of ¹³⁷Cs in muscle tissue corresponded to biological half-lives of 77 ± 10 days during summer and more than 235 days during winter. Although the most extensive experimental period lasted up to 1 year, no significant decrease in the ⁹⁰Sr levels in the fish bone tissue could be observed, hence a biological half-life of more than 500 days was observed.

The CRs for ¹³⁷Cs based on field experiments during summer and associated kinetic parameters were similar to CRs for native fish (chronic exposed whole life) and those reported in the IAEA handbook. However, significantly lower CRs were observed during autumn–winter compared to spring – summer seasons (CR of ¹³⁷Cs was 100–450 for winter and 900–2400 for summer), and the CRs for the winter season were significantly lower than those in IAEA handbook. In case of a new nuclear event, fallout during spring–summer would therefore be much more severe for freshwater fish than deposition during autumn and winter. The findings also underline the fact that laboratory results cannot directly be extrapolated to the field and that field experiments are needed to provide information on seasonal changes in radionuclide transfer to fish that is needed in assessment models. Then, the use of transplant field experiments in natural ecosystems has proved to be a useful tool and should be utilized more often.

CRediT authorship contribution statement

Hans-Christian Teien: Conceptualization, Methodology, Investigation, Writing – original draft. **Olena Kashparova:** Investigation, Formal analysis, Writing – original draft. **Brit Salbu:** Conceptualization, Writing – review & editing. **Sviatoslav Levchuk:** Investigation, Formal analysis. **Valentyn Protsak:** Methodology, Investigation, Formal analysis. **Dag Markus Eide:** Formal analysis, Writing – original draft. **Karl Andreas Jensen:** Formal analysis. **Valery Kashparov:** Conceptualization, Methodology, Investigation, Writing – original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.147280>.

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