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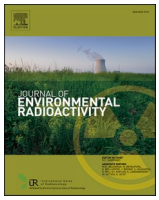

# Transfer of $^{129}$I to freshwater fish species within Fukushima and Chernobyl exclusion zones

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ABSTRACT

Unique data is reported on the transfer of $^{129}$I iodine from freshwaters to fish as well as the internal distribution within fish from the Fukushima and Chernobyl exclusion zones (ChEZ). Samples of water, sediments and fish were collected in the contaminated ponds Inkyozaka and Suzuuchi, and in the less contaminated Abukuma river in Fukushima, as well as in the contaminated Glubokoye lake and in the less contaminated Starukha lake in ChEZ. In water, $^{129}$I was mainly present as low molecular mass (LMM) and negatively charged species, while a minor fraction was associated with colloidal fraction, most probably organic material in water. The sediment-water $^{129}$I apparent distribution coefficients, Kd, ranged from 225 to 329 L/kg, equal that of stable iodine, but did not correlate with $^{129}$I/$^{127}$I ratio or $^{129}$I/$^{137}$Cs ratio as the environmental distribution of radioactive iodine was different from that of stable iodine and radioactive cesium. Concentration ratios (CR) of $^{129}$I in muscle of freshwater fish ranged from 85 to 544 across waterbodies with limited water exchange, similar in Fukushima and Chernobyl, but varied with respect to fish species. Thus, this is the first results on the transfer of $^{129}$I to freshwater fish, showing that the CR for freshwater fish is higher than CR reported for marine fish. Concentrations of $^{129}$I in fish muscle were, however, lower than in the intestinal content, indicating the influence of more contaminated dietary ingredients probably of terrestrial origin based due to $\delta^{13}$C signal on as well as of biodilution. The present results highlighted also that the radiation dose in fish was highly inhomogeneously distributed. Based on the present $^{129}$I/$^{127}$I atomic ratio of $10^{-5}$ in the most contaminated fish in the ponds in Fukushima and Glubokoye lake in Chernobyl, however, a radiation dose of 10 μSv/y would not pose any harm to the fish population.

## 1. Introduction

Iodine is a trace element essential for human and animal health. Marine environments are the major source of stable iodine ($^{127}$I, Muramatsu and Wedepohl, 1998), and iodine levels decrease in terrestrial and freshwater ecosystems with distance from the sea (Schwehr and Santschi 2003; Hou et. al. 2009). Radioactive isotopes of iodine are produced from nuclear fission, where the short-lived $^{131}$I ($t_{1/2} = 8.02$ d) is important for short-term assessments and the long-lived $^{129}$I ($t_{1/2} =$ 15.7 million years) is important for long-term assessments (UNSCEAR, 2017). $^{129}$I in the environment originates mostly from nuclear weapons and the nuclear fuel cycles (Hou et., al. 2009). The total estimated release of $^{129}$I from nuclear weapons tests (NWT, 1945–1975) is about 150 kg (Eisenbud and Gesell, 1997). Thus, the $^{129}$I levels, as well as the $^{129}$I/$^{127}$I atom ratios, increased in the environment due to weapons testing, especially in the northern hemisphere. The $^{129}$I/$^{127}$I ratio in the marine environment ranges from $10^{-11}$ to $10^{-10}$ and in the terrestrial environment from $10^{-11}$ to $10^{-9}$ (Hou et., al. 2009; Snyder and Fehn, 2004). $^{129}$I has also been released from nuclear reprocessing facilities, such as Sellafield in UK, La Hague in France (Raisbeck and Yiou, 1999)

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and Rokkasho in Japan (Udea et al., 2015). Until 2007, La Hague reprocessing plant discharged about 3800 kg $^{129}$I to the English Channel, and Sellafield reprocessing plant discharged about 1400 kg $^{129}$I to the Irish Sea. Releases caused $^{129}$I levels and $^{129}$I/$^{127}$I ratios (from $10^{-8}$ to $10^{-5}$) to increase significantly in the Irish Sea, English Channel, North Sea, and Nordic Seas (Hou et., al. 2009). From 2006 to 2008 Rokkasho reprocessing plant discharged $^{129}$I, $5 \times 10^{8}$ Bq into the Pacific Ocean increasing the $^{129}$I/$^{127}$I ratios in marine biota to $10^{-7}$ (Udea et al., 2015). Sellafield, La Hague and Rokkasho also released $^{129}$I to the atmosphere, and levels of $^{129}$I and $^{129}$I/$^{127}$I ratios increased ($10^{-6}$ to $10^{-4}$) in terrestrial areas close to the plants in Europe. In addition, $^{129}$I was released during nuclear events such as the Chernobyl and Fukushima accidents. Total estimated release of $^{129}$I from Chernobyl was 1.3–6 kg (Hou et., al. 2009), while the estimated release from Fukushima Dai-ichi Nuclear Power Plant (FDNPP) was about 1.2 kg (Hou et al., 2013). Presumably, about 13% of the released $^{129}$I deposited on the land of Japan (Morino et al., 2011), and compared to control areas, the $^{129}$I/$^{127}$I ratio in contaminated soils increased to about $10^{-5}$, three order of magnitude higher than background (Yang et al., 2017). Thus, the release and global impact of $^{129}$I from Chernobyl and Fukushima was lower compared to NWT, but the impact in the specific exclusion zones was strongly higher than NWT (Snyder and Fehn, 2004; Sahoo et al., 2009).

In waters, iodine can be present in different physico-chemical forms (e.g. iodide ($I^-$), iodate ($IO_3^-$), organo-iodine), with iodide predominate in freshwaters (Schwehr and Santschi, 2003). The distribution of iodine species depends on key factors such as pH, redox and dissolved organic carbon (DOC, Santschi and Schwehr, 2004). Fish accumulate iodine from both water and diet (Hunn and Fromm, 1964; Martinez et al., 2014a), and the $^{129}$I/$^{127}$I ratio in fish is expected to reflect the habitat of the fish (Kusuno et al., 2015b). $^{127}$I levels in marine fish species are about 5–10 times higher than in freshwater fish (Eckhoff and Maage, 1997), while the $^{129}$I uptake in freshwater fish is assumed to be higher than in marine fish due to less competition from stable $^{127}$I.

In radioecology, impact assessments associated with human and the environment have traditionally been placed upon the use of transfer factors (e.g., concentration ratios, CRs) whereby, the activity concentrations of radionuclides in organisms are divided by the activity concentrations in their habitat (i.e., surrounding media such as water, IAEA, 2010, 2014). Although available data compilations on such ratios are comprehensive, no $^{129}$I data exist for freshwater fish, even though, freshwater fish, despite being less important than seawater fish, are also assumed to be a significant iodine source for humans (Eckhoff and Mage, 1996). Lacking $^{129}$I data, other extrapolation methods (e.g., using stable analogues) have been used in handbooks (IAEA, 2010). However, the CR values based on stable isotopes can be different because radioactive fallout is a relatively recent addition to the surface of the catchments compared to that of stable isotopes having a different distribution (IAEA, 2010).

To assess long-term consequences of radioactive iodine contamination in the environment, information on the environmental behavior of $^{129}$I species is important (Salbu, 2007). As the information on transfer of $^{129}$I in freshwater is scarce, the present work focuses on the speciation of iodine in the water, quantifying the distribution of $^{129}$I between sediment-water and fish in three waterbodies within the Fukushima area and from two different waterbodies within Chernobyl exclusion zone (CheZ). Based on the distribution data the apparent distribution coefficients (Kd) and concentration ratios (CR) of $^{129}$I were derived and compared with respect to variables such as sampling site, stable iodine and radioactive $^{137}$Cs. The results should make important contributions to the parameterization of freshwater ecosystem transfer models of $^{129}$I and risk assessments associated with the release of radioactive iodine.

## 2. Material and method

### 2.1. Study area

To obtain information on the concentrations of $^{129}$I in contaminated freshwater ecosystems one field sampling campaign was carried out within and outside the exclusion zone of Fukushima in 2016 and two field sampling campaigns in Chernobyl exclusion zone in 2016 and 2017 (Fig. 1, Table1). In Fukushima, sampling took place in spare filled fire extinguishing pond of Inkyozaka (37°25.499′ N 141°01.05′ E) and rice irrigation pond Suzuuchi (37°24.950′ N 140°58.791′ E), situated 0.24 and 3.75 km from the NPP (Konoplev et al., 2018), respectively, in Okuma Town within the 10-km zone around the Fukushima Dai-ichi NPP. In addition, control samples were collected in the Abukuma river (37°47.17.67′ N 141°29 20.75 ′ E) located 63.32 km north-west of the NPP, outside the exclusion zone near Fukushima city. The Inkyozaka and Suzuuchi ponds were quite shallow, with depths of about 2 and 1 m, respectively, and with limited water exchange (Wakiyama et al., 2017) in contrast to the running water system in Abukuma river (about 190 m$^3$/sec) draining a large catchment area (2921 km$^2$, Taniguchi et al., 2019).

In the Chernobyl exclusion zone (ChEZ) sampling took place in contaminated Glubokoye lake (51°26.431′ N 30°03.502′ E, 6.5 km and 338$^0$ north from the Chernobyl NPP) situated in the northwest track of Chernobyl radioactive fallout, and the less contaminated Starukha lake (51°21.294′ N 30°12.266′ E, located 8.1 km and 115$^0$ south-east of the Chernobyl NPP) situated outside the north-west plume of radioactive fallout (Fig. 1, Teien et al., 2021). Glubokoye lake has a limited water exchange with no outlet, but Starukha lake is connected to Prypjat river during flooding events. The sampling program included water,

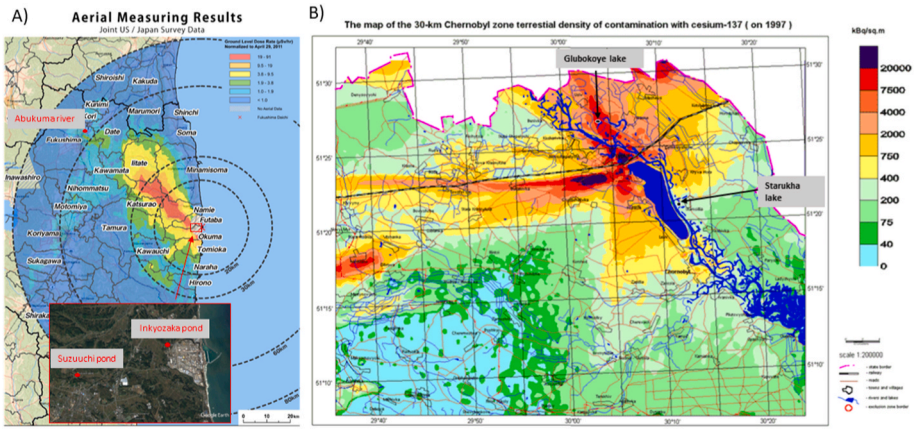


**Fig. 1.** A) Location of Inkyozaka pond, Suzuuchi pond and Abukuma river compared to the Fukushima NPP (Map of detected radioactivity in Fukushima reactor area, en.wikipedia.org) and B) Location of Chernobyl waterbodies Glubokoye lake and Starukha lake within the Chernobyl exclusion zone (UIAR MAP of ChEZ).

**Table 1**
Water characteristics of waterbodies in Fukushima and Chernobyl exclusion zones including activity of $^{137}$Cs, concentration of $^{129}$I and atomic ratios of $^{129}$I with $^{127}$I and $^{137}$Cs.

| Waterbodies | | Fukushima | | | Chernobyl | |
|---|---|---|---|---|---|---|
| | | Inkyozaka pond | Suzuuchi pond | Abukuma river | Glubokoye lake | Starukha lake |
| Coordinates | | 37°25.499′ N 141°01.05′ E | 37°24.950′ N 140°58.791′ E | 37°47.17.67′ N 141°29.20.75′ E | 51°26.431′ N 30°03.502′ E | 51°21.294′ N 30°12.266′ E |
| Date of sampling | | 25.09.2016 | 24.09.2016 | 26.09.2016 | 21.05.2016, 13.06.2017 | 14.06.2017 |
| Temp. (°C) | | 22.1 | 20.4 | 18.1 | 20.3 | 18.5 |
| Cond. (μS/cm)[b] | | 86.9 | 66.4 | 120.4 | 201 ± 10 | 235 ± 10 |
| pH[b] | | 6.81 | 6.35 | 7.08 | 7.4–7.6 | 7.2–7.6 |
| DOC (mg/L) | | 6.03 [3.29] | 5.76 [3.8] | 1.88 | 11.2 [9.9] | 9.8 [5.6] |
| $NO_3^-$ (mg/L) | | 0.09 | 0.03 | 1.2 | 0.09 | 0.5 |
| $Cl^-$ (mg/L) | | 9.35 | 3.84 | 5.89 | 5.2 ± 0.4 | 10.6 ± 1.1 |
| $SO_4^{2-}$ (mg/L) | | 4.49 | 15.2 | 12.1 | 6.8 ± 0.1 | 15.0 |
| Na (mg/L) | | 8.1 ± 0.4 | 3.5 ± 0 | 6.4 ± 0.2 | 4.6 ± 0.3 | 10.4 ± 0.5 |
| Mg (mg/L)[b] | | 1.0 ± 0 | 1.8 ± 0 | 2.6 ± 0 | 4.0 ± 0.3 | 4.3 ± 0.6 |
| K (mg/L) | | 1.9 ± 0.1 | 1.7 ± 0 | 2.3 ± 0 | 1.4 ± 0.4 | 2.9 ± 1.0 |
| Ca (mg/L)[b] | | 4.4 ± 0 | 5 ± 0.2 | 11.6 ± 0.1 | 30 ± 1.4 | 34.7 ± 1.8 |
| $^{133}$Cs (μg/L)[b] | | 0.034 ± 0.003 [0.021 ± 0.003] | 0.006 ± 0.001 [0.005 ± 0.001] | 0.007 ± 0.002 [0.004 ± 0.002] | 0.004 ± 0.0002 [0.004] | 0.004 ± 0.0006 [0.004] |
| $^{134}$Cs (Bq/L) | | 0.64 ± 0.03 [0.49 ± 0.03] | 0.22 ± 0.02 [0.18 ± 0.02] | * | * | * |
| $^{137}$Cs (Bq/L) | | 3.50 ± 0.07 [2.76 ± 0.07] | 1.34 ± 0.04 [1.04 ± 0.03] | 0.006[a] | 3.6 ± 1.0 | 0.023 ± 0.005 |
| $^{127}$I (μg/L)[b] | Unfiltered | 11.3 | 5.8 | 4.8 | 2.34 ± 0.22 | 2.80 ± 0.20 |
| | Particle | <nd | <nd | <nd | <nd | <nd |
| | Colloidal | 1.8 | 0.7 | 0.4 | 0.33 ± 0.56 | <nd |
| | LMM | [9.5] | [5.2] | [4.5] | 2.11 ± 0.52 | 2.80 |
| $^{127}$I/Cl (μg/mg)[b] | | 1.21 | 1.43 | 0.74 | 0.27 ± 0.14 | 0.26 |
| $^{129}$I (x $10^{10}$ at/kg) | Unfiltered | 14.00 ± 0.69 | 1.50 ± 0.02 | 0.09 ± 0.02 | 1.53 ± 0.08 | 0.15 ± 0.00 |
| | Particle | <nd | 0.40 ± 0.02 | <nd | | |
| | Colloidal | 2.00 ± 0.54 | 0.22 ± 0.03 | 0.04 ± 0.02 | 0.10 ± 0.10 | 0.04 ± 0.00 |
| | LMM | [12.00 ± 0.29] [[11.80 ± 0.29]] | [0.88 ± 0.02] [[0.75 ± 0.02]] | [0.05 ± 0.02] [[0.04 ± 0.02]] | [1.68 ± 0.09] [[1.15 ± 0.12]] | [0.11 ± 0.00] * |
| $^{129}$I/$^{127}$I (x $10^{-7}$ at/at) | | 26.2 | 5.74 | 0.43 | 13.8 | 0.72 |
| $^{129}$I/$^{137}$Cs (at/at) | | 29.1 | 5.98 | 0.1 | 3.6 | 47.1 |

* = not analyzed, nd = not detected. Values in single brackets [] are concentration in 10 kDa ultrafiltered (LMM) waterfraction, values in double brackets [[]] are concentration of LMM anionic waterfraction.
[a] Published by Igarashi et al. (2022), collected September 2016.
[b] Significantly different between Fukushima and Chernobyl waterbodies.

sediments (only Fukushima waterbodies) and various fish species.

### 2.2. Sampling of water, sediment and fish

To obtain information about concentrations and speciation of $^{129}I$ and its stable analogue $^{127}I$, water samples were collected from the 5 different waterbodies (three in Fukushima and two in ChEZ). Water samples were filtered (0.45 μm Millipore membrane) and ultrafiltered (10 kDa Pall hollow fibre) *in situ* before analyses of iodine isotopes. Three replicate samples (50 ml tubes) were collected from all fractions to obtain information on particulate, colloidal and low molecular mass (LMM) species of $^{127}I$ and $^{129}I$. Radiocesium ($^{134}Cs$ and/or $^{137}Cs$) and its stable analogue $^{133}Cs$ were also determined in the different water fractions for comparison.

Concentrations of major cations were obtained using ICP-MS (Agilent 8800 ICP Triple Quad) in acidified ($HNO_3$) water samples, concentrations of major anions were obtained by using Lachat IC5000 Ion chromatograph (Zellweger analytics Inc. USA) and dissolved organic carbon (DOC) by using carbon analyzer (Shimadzu TOC5000) in separate 0.45 μm filtered water samples (50 ml) from the waterbodies. Conductivity, temperature and pH were determined *in situ* using an pH meter (WTW 430i with Conductivity probe TertraCon 325 and Sentix 41 pH electrode).

Bottom sediments were only collected from Fukushima waterbodies, in Inkyozaka and Suzuuchi ponds as well as along the waterside edge of the Abukuma river using a core sampler. The cores were sliced into 0–1, 1–2 and 2–3 cm segments, and freeze-dried prior to analysis. The freeze-drying process did not affect the results, as $^{129}I$ analyzed in aliquots of wet sediments showed similar results. Aliquots of the samples were also used to characterize the sediments, such as loss of ignition (organic content) and grain size analysis (Reinoso-Maset et al., 2020).

Fish were caught with gillnets and fishing rods. Three species of fish were collected from Fukushima waterbodies; crucian carp *(Carassius auratus langsdorfii),* common carp (*Cyprinus carpio)* and largemouth bass (*Micropterus salmoides)*. Two different species of fish were collected from the Chernobyl waterbodies; common rudd (*Scardinius erythrophthalmus*) and perch (*Perca fluviatilis*). Dissection of the fish was carried out in accordance with the EMERGE sampling protocol (Rosseland et al., 2001). Fish were sacrificed by a blow on their head, weight and total length were measured, followed dissection and collection of organ samples (i.e., gills, liver, kidney, gonads, muscle, and intestinal content). Muscle tissue was collected without bone after removing the skin. All samples were stored cold in the field, kept in a freezer at −20 °C in the laboratory, and freeze-dried prior to analysis. Fish age was estimated by counting the annuli on their otoliths when found (Wada et al., 2019).

### 2.3. Sample preparation and analysis

#### 2.3.1. Determination of the concentration of $^{129}I$

Aliquots of water fractions, sediment slices and biological material were subjected to further sample preparation prior to analysis. Water sample preparation was based on the method published by Lopez-Gutierrez et al. (2000) with small modifications. About 25 g of water was filtered and 0.4 g KOH and 3 mg of stable iodine (Woodward Iodine) carrier were added. NaClO (10%) was added to release iodine attached to organic matter, and then all iodine was reduced to iodide by $Na_2S_2O_5$. Iodine was then extracted into chloroform after oxidation ($HNO_3$ and $NaNO_2$) and back-extracted into an aqueous $Na_2S_2O_5$ solution. These purification steps were repeated twice. Finally, AgI was precipitated by the addition of $AgNO_3$, the precipitate was mixed with Nb powder and pressed in a copper target for measurements by accelerator mass spectrometry (AMS).

Sediment and fish sample preparation were based on previous published methods applied prior to measurements by AMS (Gómez-Guzmán et al., 2010). About 0.1 g of material were introduced in a microwave oven (Anton Paar Multiwave 3000) together with 7 mg of stable iodine carrier (Woodward iodine) and 10 ml $HNO_3$. The sample were digested at 200 °C for 20 min with continuous control of temperature and pressure in the beakers. Iodine was then extracted from the liquid phase into $CHCl_3$ and back-extracted into $Na_2S_2O_5$ solution as for water samples. Finally, AgI was precipitated by the addition of $AgNO_3$, mixed with Nb powder and pressed for AMS measurement.

$^{129}I$ was determined using Accelerator Mass Spectrometry (AMS). The method is based on the extraction of $I^-$ ions from the sample in a Cs sputter ion source, then injected in a 1 MV Tandetron accelerator (High Voltage Engineering Europe, Netherlands). After passing through a He stripper, $I^{2+}$ was selected in a magnetic deflector. While stable $^{127}I^{2+}$ were detected as a current in a Faraday cup, $^{129}I^{2+}$ ions went through an electrostatic deflector and was detected in an ionization chamber. Standard samples with known $^{129}I/^{127}I$ were continuously measured to correct the isotopic ratio, as well as blank samples (Woodward iodine) for background evaluation. The instrumental background was typically $^{129}I/^{127}I = 1\text{-}2 \times 10^{-13}$, while samples showed ratios in the order of $10^{-12}$ to $10^{-10}$. More details on the AMS facility can be found in Calvo et al. (2015) and Scognamiglio et al. (2016).

#### 2.3.2. QQQ ICP-MS for stable element determination

The concentrations of stable elements including stable iodine ($^{127}I$) and stable cesium ($^{133}Cs$) were determined in aliquots of water fractions, sediment slices and biological material using QQQ ICP-MS. Prior to analysis, aliquots of sediment or biological materials were subjected to microwave oven assisted alkaline digestion for iodine and acid digestion for cesium using an UltraClave at 200 °C. Tetramethylammonium hydroxide (TMAH) solutions were used for alkaline digestion, while ultrapure $HNO_3$ was used for acid digestion. After digestion, the samples were allowed to cool to room temperature and diluted with ultrapure water before measurements. Analyzed certified reference materials (CRM) indicated good accuracy (NCSZC73013: 0.32 ± 0.04 mg I/kg and 0.13 ± 0.01 mg Cs/kg compared to CRM value 0.36 ± 0.12 mg I/kg and 0.13 ± 0.02 mg Cs/kg, respectively. NCS DC 73325: 20 ± 3 mg I/kg compared to CRM value 19 ± 2 mg I/kg and ERM-BD151: 1.6 mg I/kg compared to CRM value 1.78 ± 0.17 mg I/kg). $^{129}I/^{127}I$ ratios in samples were calculated by $^{129}I$ determined by AMS and $^{127}I$ determined by ICP-MS based on atomic ratio.

#### 2.3.3. Gamma spectrometry for radioactive Cs determinations

Aliquots of biological materials and sediment samples were used for quantitative determination of radiocesium ($^{134}Cs$ and $^{137}Cs$). Fukushima samples were assayed out using a NaI detector (PerkinElmer Wizard2 2480 automatic gamma counter) or germanium detector (HPGe, CANBERRA GC3018)(Konoplev et al., 2018; Reinoso-Maset et al., 2020), while samples from Chernobyl were measured using germanium detectors (GEM-30185, EG & G ORTEC, USA) (Teien et al., 2021). The sediment-water-fish distributions of radiocesium in the freshwater bodies in Fukushima were compared with $^{129}I$ to identify similarities of these radionuclides released during the FDNPP accident. $^{129}I/^{137}Cs$ was calculated based on atomic ratio.

#### 2.3.4. Isotope analysis of carbon (δ13C) and nitrogen (δ15N)

Aliquots of fish muscle and stomach content were analyzed with respect to $\delta^{15}N$ to estimate trophic levels, and $\delta^{13}C$ for indication of the food source. Freeze dried homogenates were weighed, wrapped in tin capsules and analyzed using isotope-ratio mass spectrometry (IRMS). Analysis of certified reference material (IAEA-N1 and IAEA-N2 for nitrogen, and IAEA-CH6 for carbon) reflected acceptable accuracy measurements.

### 2.4. Statistical analysis

Results of individual measurements were used in statistical analyses and in calculation of average values with standard deviations. Pairwise (Pearson's) correlations were used to calculate $R^2$ and nonparametric

Mann-Whitney test was used to identify differences among groups or between Fukushima and Chernobyl waterbodies. Statistical analyses were performed using Minitab. P-values $< 0.05$ was interpreted as significant.

## 3. Results and discussion

### 3.1. Water characteristics

The Fukushima waterbodies were characterized as soft waters of pH 6.4 to 7.1, with moderate conductivity ranging from 66 to 120 μS/cm, and with moderate dissolved organic carbon levels (DOC 5.8–6.0 mg/L). The Chernobyl lake waters were characterized as hard with pH 7.2–7.6, conductivity of 201–235 μS/cm and 5.2–10 mg/L of dissolved organic carbon. The pH, ionic strength, as well as concentration of Ca and Mg were higher in the Chernobyl waterbodies than in the Fukushima waterbodies. These variables were also higher in the running Abukuma river compared to the ponds in Fukushima (Table 1).

The water concentrations of Na and Cl decreased from Inkyozaka to Suzuuchi pond, as expected, due to increasing distance from the coast. The concentration of Cl in the Abukuma river was higher than in Suzuuchi pond and agreed with previously reported concentrations (Tagami and Uchida, 2006). Although the Abukuma river is situated farther from the coast, the catchment is large, and it is assumed that the concentration of Cl, and possibly Na, is associated with precipitation being influenced by the marine system (Tagami and Uchida, 2006). The concentration of Na and Cl was, however, within the same range as in Chernobyl lakes, situated far from the coast with limited marine influenced precipitation.

In Fukushima waterbodies the concentrations of dissolved (particle size less than 0.45 μm) stable Cs ranged from 0.006 to 0.034 μg/L and followed a trend similar to that of Na and Cl ($R^2 = 0.98$ and 0.82, respectively), with highest concentrations in Inkyozaka pond. In Chernobyl waterbodies, however, the concentration of stable Cs was $0.004 \pm 0.0006$ μg/L and significantly lower than in the Fukushima waterbodies. Results indicates that the source of stable Cs could be the marine environment, due to the good correlation with NaCl in Fukushima and low correlation in Chernobyl situated far from the marine environment. The activity concentrations of $^{137}$Cs in the Fukushima ponds ranged from 1.3 to 3.5 Bq/L and were higher in Inkyozaka pond than in Suzuuchi pond (Table 1), but similar to ranges previously observed in the Glubokoye lake in Chernobyl (Konoplev et al., 2018; Wakiyama et al., 2017, Baloga et al., 2011). Thus, the activity concentrations of $^{137}$Cs in the waterbodies in Fukushima and Chernobyl exclusion zones were surprisingly similar, while the activity concentration in the Starlukha lake, situated outside the track of Chernobyl radioactive fallout was 100 fold lower.

#### 3.1.1. Concentrations of stable $^{127}I$

In Fukushima waterbodies the concentrations of stable iodine ranged from 4.8 to 11.3 μg/l and decreased from Inkyozaka pond to Suzuuchi pond and to the Abukuma river, i.e., decreased with increasing distance from the coast. In Chernobyl waterbodies the concentration of stable iodine ranged from 2.3 to 2.8 and was significantly lower than in the Fukushima waterbodies. The concentration ratio of $^{127}$I/Cl in Fukushima waterbodies varied between waterbodies and was significantly higher than in Chernobyl waterbodies, while the concentration ratios of $^{133}$Cs/$^{127}$I were more similar in range. This supports that the marine environment, rich in both iodine and cesium, would be an important source in the Fukushima area, located closer to the coast, than Chernobyl. Results supports also earlier findings that the mobility of iodine differs from Cl (Tagami and Uchida, 2006; Sheppard et al., 1993). The main fraction of $^{127}$I (82–93%) in the waters was present as low molecular mass (LMM) inorganic negatively charged species. A minor fraction (7–16%) was, however, present as colloidal species most probably associated with organics.

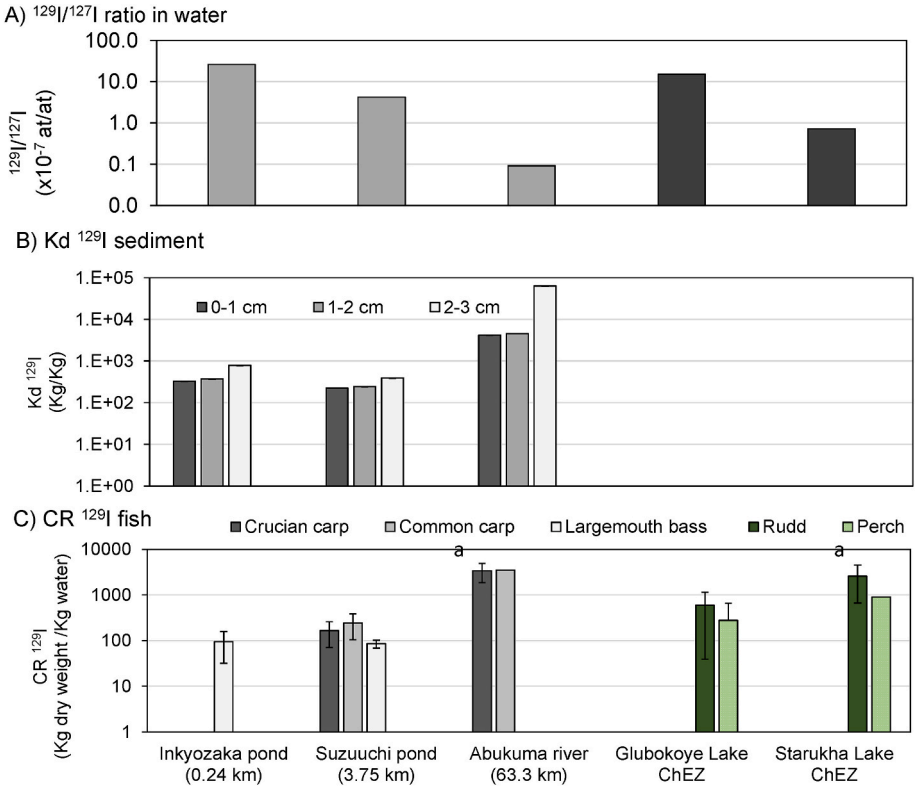


**Fig. 2.** (A) Atomic ratio of $^{129}$I/$^{127}$I in water; (B) Sediment Kd of $^{129}$I; and (C) Concentration ratios (CR) for $^{129}$I in fish in Inkyozaka Pond, Suzuuchi pond and the Abukuma river at increasing dictance from the Fukushima NPP and Glubokoye lake and Starukha lake in chernobyl exlusion zone [a] = Significantly different between reservoirs of similar species. Error bars indicate standard deviation, N = 3–11.

#### 3.1.2. $^{129}I$ in water

The concentrations of dissolved (particle size less than 0.45 μm) $^{129}I$ in the water from Fukushima waterbodies ranged from 0.9 x $10^9$ to 1.4 x $10^{11}$ at/kg and indicated that the concentration of $^{129}I$ decreased with distance from the NPP situated at the coast, as expected. In Chernobyl waterbodies the $^{129}I$ ranged from 0.15 x $10^{10}$ to 1.5 x $10^{10}$ at/kg and was 10-fold higher in the Glubokoye lake, situated within the northwest track of Chernobyl radioactive fallout compared to Starukha lake situated outside the track (Table 1, Fig. 1). Thus, $^{129}I$ level in Chernobyl waterbodies was within the range observed in Fukushima waterbodies. The present concentration level obtained for $^{129}I$ was higher than previously reported for water samples collected from the Fukushima area (3 x $10^6$ to 3 x $10^9$ at/L) as well as from the Abukuma area (Matsuzaki et al., 2013).

The atomic ratios of $^{129}I/^{127}I$ ranged from 2.62 x $10^{-6}$ to 4.35 x $10^{-8}$. In Fukushima waterbodies the ratios were highest in Inkyozaka pond, closest to the NPP, and about 1- and 2-fold lower in Suzuuchi pond and the Abukuma river, respectively (Table 1, Fig. 2). Thus, the $^{129}I/^{127}I$ ratio varied with distance from the Fukushima NPP. In Chernobyl waterbodies, the $^{129}I/^{127}I$ were highest in Glubokoye lake situated within the fallout plume and 2- fold lower in Starukha lake, situated outside the plume. Although the concentration of stable iodine was lower in Chernobyl waterbodies, the $^{129}I/^{127}I$ ratio in Fukushima waterbodies and Chernobyl waterbodies were within the same range.

The atomic ratio of $^{129}I/^{137}Cs$ in Fukushima and Chernobyl waterbodies were also in the same range (from 3.6 to 47.1, Table 1) although the activity concentration of $^{137}Cs$ deposited in the Chernobyl waterbodies had decreased by almost one half-life at the time of sampling. Starukha lake situated outside the track of Chernobyl radioactive fallout had the highest ratio. This suggest that the spatial distribution of $^{129}I$ differ from $^{137}Cs$.

The main fraction of $^{129}I$ (60–94%) in the waterbodies was present as LMM iodine, while 7–50% was present as colloidal species. Thus, the size distribution of $^{129}I$ agreed with the observed size distribution pattern of stable $^{127}I$. The colloidal iodine is probably reflecting iodine associated with organic material since the concentration of colloidal organic material was 2–4 mg/l and sorption of iodine to colloidal organic matter can be significant (Otosaka et al., 2011). A significant part of $^{137}Cs$ and stable Cs (16–42%) were also associated with the colloidal fraction in Fukushima waterbodies (Table 1), and in a greater fraction than iodine. This is in agreement with previous results showing a significant fraction of particulate radiocesium were present in the ponds and that a certain fraction of $^{137}Cs$ has been associated with specific inert micrometer sized $^{137}Cs$ particles released from unit 2 or 3 (Igarashi et al., 2019). Thus, $^{129}I$ and $^{137}Cs$ have different size distribution patterns in water bodies, with $^{129}I$ present mainly in the LMM size fraction, being more mobile than $^{137}Cs$, being more associated with larger sized fractions. The mobility of iodine was also supported by the fact that 40–85% of iodine in the waterbodies was present as LMM inorganic negatively charged species.

**Table 2**
Sediment characteristics from Fukushima waterbodies including activity of $^{137}Cs$ and concentration of $^{129}I$.

| | Depth (cm) | Inkyozaka pond | Suzuuchi pond | Abukuma river |
|---|---|---|---|---|
| Organic matter (%)[a] | 0–3 | 6.5 ± 1.3 | 9.5 ± 0.4 | 9.0 ± 1.9 |
| Sand 63 μm - 2 mm (%)[a] | 0–3 | 87.7 ± 3.1 | 55.3 ± 8.1 | 61.6 ± 10.9 |
| Silt 2–63 μm (%)[a] | 0–3 | 12.3 ± 3.1 | 42.3 ± 7.0 | 38.0 ± 10.8 |
| Clay <2 μm (%)[a] | 0–3 | 0.1 ± 0.1 | 2.5 ± 1.1 | 0.4 ± 0.1 |
| $^{137}Cs$ (kBq/kg) [a] | 0–1 | 93.8 | 149.5 | 0.37 |
| | 1–2 | 90.1 | 164.4 | 0.51 |
| | 2–3 | 66.2 | 172.3 | 0.29 |
| $^{127}I$ (mg/kg) | 0–1 | 2.1 ± 0.1 | 4.0 ± 0.0 | 10.7 ± 1.2 |
| | 1–2 | 2.1 ± 0.2 | 3.3 ± 0.0 | 10.7 ± 0.6 |
| | 2–3 | 1.9 ± 0.1 | 3.4 ± 0.3 | 6.0 ± 0.7 |
| $^{129}I$ (x $10^{12}$ at/kg) | 0–1 | 46.0 ± 2.30 | 2.48 ± 0.21 | 0.80 ± 0.02 |
| | 1–2 | 52.0 ± 2.40 | 2.66 ± 0.29 | 0.87 ± 0.04 |
| | 2–3 | 110.0E ± 2.6 | 4.31 ± 0.39 | 12.0 ± 0.47 |
| $^{127}I/OC$ | 0–1 | 271 | 440 | 1217 |
| (mg/%) | 1–2 | 314 | 334 | 962 |
| | 2–3 | 368 | 348 | 830 |
| $^{129}I/^{127}I$ | 0–1 | 46.2 | 1.31 | 0.16 |
| (x $10^{-7}$ at/at) | 1–2 | 52.2 | 1.70 | 0.17 |
| | 2–3 | 124 | 2.70 | 0.42 |
| $^{129}I/^{137}Cs$ | 0–1 | 3.6 | 0.12 | 1.5 |
| (x $10^{-1}$ at/at) | 1–2 | 4.2 | 0.12 | 1.4 |
| | 2–3 | 12 | 0.18 | 75 |
| Kd $^{127}I$ (x $10^2$ L/kg)[b] | 0–1 | 1.86 | 6.85 | 22.0 |
| Kd $^{129}I$ (x $10^2$ kg/Kg)[b] | 0–1 | 3.29 | 2.25 | 42.1 |

[a] Published by Reinoso-Maset et al. (2020).
[b] Based on 0.45 μm filtered water samples.

### 3.2. Sediments in Fukushima

#### 3.2.1. Sediment characteristics

Sediment characteristics differed among the three sites in Fukushima waterbodies. The organic matter in sediments varied from 6.5 to 9.5% (Reinoso-Maset et al., 2020), lowest on average in Inkyozaka pond and highest in Suzuuchi pond (Table 2). Sediments from Inkyozaka pond was characterized with a very high fraction of sand, with clay being less than 1%. Sediments from Suzuuchi pond and the Abukuma river were characterized by high fractions of sand and silt, with up to 3% clays in Suzuuchi pond and less than 1% clay in the river.

#### 3.2.2. $^{127}I$ in sediments

Concentrations of stable $^{127}I$ in the sediments were in the range 2.1–10.7 mg/Kg, lowest in Inkyozaka pond sediments and highest in the Abukuma river sediments (Table 2). The $^{127}I$ concentrations within the 0–3 cm sediment layers were about constant. Thus, results indicated limited changes of iodine over time within each waterbodies, as expected. However, the concentration of stable $^{127}I$, as well as the relationship between $^{127}I$ and organic matter (%) in the sediment ($^{127}I/OC$ ratio), increased from ponds to the river, i.e., with distance from the coast. Thus, the $^{127}I$ in the sediments did not reflect the concentration in the waters, as the trends of $^{127}I$ in the water were opposite of what was observed in the sediment.

#### 3.2.3. $^{129}I$ in sediments

The concentrations of $^{129}I$ and $^{137}Cs$ in the upper sediment surface layers were in the range of 8.0 x $10^{11}$ to 4.6 x $10^{13}$ at/kg and 0.4–150 kBq/kg, respectively (Table 2). The concentration of $^{129}I$ was highest in Inkyozaka pond sediments, 20-fold lower in Suzuuchi pond and 50-fold lower in sediments from Abukuma river (Table 2). Thus, the concentrations of $^{129}I$ as well as $^{137}Cs$ in sediments from the ponds and the Abukuma river decreased with distance from the NPP, as expected from a point-source contaminant plum. The levels of $^{129}I$ in surface sediments were lower than reported in Fukushima surface soils in 2017 (Yang et al., 2019), while the $^{137}Cs$ activity concentrations in sediments were in agreement with previous reports (Wada et al., 2019). In the present work, the atomic ratio of $^{129}I/^{137}Cs$ in the freshwater sediments varied from 0.01 to 7.5 at/at, lowest in the ponds and highest in the river. These ratios are within the atomic ratio ($^{129}I/^{137}Cs = 0.99$) reported from the marine ecosystem outside the Fukushima NPP which is also affected by the input from the Abukuma river (Otosaka et al., 2018). The $^{129}I$ concentrations in sediment layers increased significantly with sediment depths in the three waterbodies, while the activity concentration of $^{137}Cs$ showed a different trend, being more constant or decreasing with depth. The activity concentration of $^{137}Cs$ in sediments collected in 2011, shortly after the accident, showed highest concentrations in the surface sediments, decreasing with depth (Yoshimura, et al., 2014).

**Table 3**
Fish characteristics Fukushima waterbodies, fish muscle activity of $^{137}$Cs and concentration of $^{129}$I, atomic ratios between $^{129}$I and $^{127}$I and $^{137}$Cs, and CR of $^{137}$Cs and iodine for fish samples collected from Inkyozaka pond, Suzuuchi pond and the Abukuma river. Values are given as average+/-std, in dry weight muscle. Dry weight of fish muscle was in average 21% of the wet weight.

| Waterbody | Inkyozaka | Suzuuchi | | | Abukuma river | |
|---|---|---|---|---|---|---|
| | Largemouth bass (*Micropterus salmoides)* | Crucian carp (*Carassius auratus langsdorfii)* | Common carp (*Cyprinus carpio)* | Largemouth bass (*Micropterus salmoides)* | Crucian carp (*Carassius auratus langsdorfii)* | Common carp (*Cyprinus carpio)* |
| Sample size | 2 | 10 | 3 | 3 | 11 | 1 |
| Age (year) | 0 | 5.7 ± 2 | 0.7 ± 0.6 | 1 | 2.7 ± 2.5 | 0 |
| Weight (g) | 11.5 ± 2.1 | 491 ± 92 | 229 ± 167 | 287 ± 15 | 416 ± 344 | 108 |
| Length (cm) | 10.3 ± 0.3 | 32.4 ± 3.3 | 24.7 ± 8.1 | 26.2 ± 0.6 | 27.2 ± 7.8 | 19 |
| K (g/cm$^3$) | 1.0 ± 0.1 | 1.5 ± 0.2 | 1.3 ± 0.1 | 1.6 ± 0.0 | 1.6 ± 0.2 | 1.6 |
| $\delta^{15}$N | 7.46 ± 0.65 | 13.1 ± 0.5 | 11.4 ± 0.1 | 13.5 ± 0.4 | 13.3 ± 0.4 | 14.2 |
| $\delta^{13}$C | −32.6 ± 0.4[b] | −22.6 ± 0.6 | −22.7 ± 0.7 | −22.4 ± 0.0 | −24.3 ± 0.9 | −22.7 |
| $\delta^{13}$C-intestinal cont. | −30.7 ± 9.3 | −22.8 ± 1.9 | −23.9 ± 2.8 | −26.7 ± 1.8 | −26.8 ± 1.4 | −25.4 |
| $^{137}$Cs (kBq/kg) | 14.9 ± 4.2 | 31.0 ± 4.0 | 15.3 ± 3.9 | 27.7 ± 2.4 | 0.17 ± 0.27 | 0.15 |
| $^{127}$I (mg/kg) | 0.15 ± 0.03[b] | 0.13 ± 0.09 | 0.05 ± 0.02[a] | 0.24 ± 0.03 | 0.21 ± 0.08[b] | 0.23 |
| $^{129}$I (x 10$^{12}$ at/kg) | 13.28 ± 8.78 | 1.81 ± 1.03 | 3.02 ± 1.96 | 0.83 ± 0.08 | 0.64 ± 0.29 | 0.66 |
| $^{129}$I/$^{127}$I (x 10$^{-6}$ at/at) | 20.18 ± 16.14 | 3.71 ± 3.22 | 10.82 ± 5.05[a] | 0.74 ± 0.16 | 0.78 ± 0.45 | 0.61 |
| $^{129}$I/$^{137}$Cs (x 10$^{-1}$ at/at) | 6.11 ± 2.57 | 0.43 ± 0.22 | 1.61 ± 1.35 | 0.22 ± 0.04 | 61.50 ± 66.9 | 32.20 |
| CR $^{137}$Cs (x 10$^4$ L/kg) | 0.43 ± 0.12 | 2.31 ± 0.29 | 1.14 ± 0.29 | 2.07 ± 0.18 | | |
| CR $^{127}$I (x 10$^2$ L/kg) | 0.13 ± 0.02 | 0.22 ± 0.15 | 0.08 ± 0.04 | 0.41 ± 0.05 | 0.44 ± 0.17 | 0.48 |
| CR $^{129}$I (x 10$^2$ kg/kg) | 0.95 ± 0.63 | 1.65 ± 0.94 | 2.43 ± 1.38 | 0.85 ± 0.17 | 33.76 ± 15.33 | 34.70 |

[a] Significantly different between species within one reservoir.
[b] Significantly different between reservoirs of similar species.

Samples collected 5 years later also demonstrated that the activity decreased with depth although the activity was more similar in the top centimeters (Reinoso-Maset et al., 2020; Wakiyama et al., 2019). The atomic ratio of $^{129}$I/$^{137}$Cs in the present work increased, however, by sediment depth, reflecting differences in mobility between the radioisotopes.

The atomic ratio of $^{129}$I/$^{127}$I in sediments was in the range of 1.6 x $10^{-8}$ to 4.6 x $10^{-6}$. The $^{129}$I/$^{127}$I ratio decreased with distance from the NPP with a 10–40 fold difference between each waterbodies, opposite the trend of $^{127}$I that increased with distance from the coast. The $^{127}$I concentrations in the 0–3 cm sediment layers were about constant, while the $^{129}$I concentrations varied by a factor of 2, lowest in the surface layer. Thus, the $^{129}$I/$^{127}$I ratio increased with depth, indicating that sedimentation of contaminated materials with less $^{129}$I amount has occurred during the last years. This is also in accordance with the fact that the $^{129}$I/$^{127}$I ratio in the sediments at 2–3 cm depth was one order of magnitude lower than that reported for surface soil in 2017 (Yang et al., 2019).

#### 3.2.4. Apparent distribution coefficients for $^{129}$I

Apparent distribution coefficients (Kd, L/kg dry weight) for $^{127}$I and $^{129}$I were high and in the range of 200–4000 L/kg, and are within ranges reported for iodine in freshwater ecosystems (IAEA 2010). The Kd for both $^{127}$I and $^{129}$I were lowest in the ponds with minor water replacement and 10-fold higher in Abukuma river with continue water replacement from the catchment (Table 2). Thus, the water- surface sediment interactions were similar for $^{127}$I and $^{129}$I, while Kd increased in the river compared to ponds although the $^{129}$I/$^{127}$I ratio decreased (Fig. 2b). As it was expected that Kd will decrease with decreased $^{129}$I/$^{127}$I ratio as similar species of $^{127}$I and $^{129}$I can compete for sorption sites, results indicate that the water- sediment interaction of iodine is not fast enough to compensate for the continue water replacement in rivers making it difficult to compare apparent Kd in river with waterbodies of limited water replacement.

The $^{127}$I/OC (organic carbon) ratios in the sediments of the two ponds were similar, indicating that the higher level of organic matter in Suzuuchi pond could be one reason of increased concentration of iodine in the sediments of that pond. Sediments of Abukuma river were more enriched in organic matter. Thus, increased level of organic carbon in sediments is probably one factor contributing to the increased transfer of $^{129}$I to sediments, e.g., from Inkyozaka pond, to Suzuuchi pond and to the Abukuma river.

### 3.3. Fish

#### 3.3.1. Fish characteristics

Five different fish species were collected, three in Fukushima waterbodies; crucian carp *(Carassius auratus langsdorfii),* common carp (*Cyprinus carpio)* and largemouth bass (*Micropterus salmoides*) (Table 3), and two fish species in waterbodies within Chernobyl exclusion zone; common rudd (*Scardinius erythrophthalmus*) and perch (*Perca fluviatilis*) (Table 4). The fish collected in both Fukushima and Chernobyl waterbodies had generally very good condition factor, 1.2–1.6 g/cm$^3$, except one small juvenile bass collected in Inkyozaka pond, 1.0 g/cm$^3$.

In Fukushima waterbodies the $\delta^{15}$N signals in fish muscle ranged between 11.4 and 14.2‰ for all fish species (except the juveniles of bass, Table 3) and was 3.1‰, and thus one trophic level higher in fish muscles than in the diet (stomach content with $\delta^{15}$N ranging from 6.5 to 12). In Glubokoye lake in ChEZ the $\delta^{15}$N signals in fish muscle ranged between 7.12 and 8.47‰ for the two species (Table 4). Results demonstrated no significant difference in trophic levels between fish species collected from any of the waterbodies as the difference in $\delta^{15}$N between species was lower than 3‰ (Beaudoin et al., 2001), although the $\delta^{15}$N was significantly higher in perch than in rudd.

**Table 4**
Fish characteristics waterbodies within Chernobyl exclusion zone (ChEZ), fish muscle activity of $^{137}$Cs and concentration of $^{129}$I, atomic ratios between $^{129}$I and $^{127}$I and $^{137}$Cs, and CR of $^{137}$Cs and iodine for fish samples collected from Glubokoye lake and Starukha lake. All values are given as average+/-std, in dry weight muscle. Dry weight of fish muscle was 21% of the wet weight.

| Waterbody | Glubokoye lake | | Starukha lake | |
|---|---|---|---|---|
| | rudd (*Scardinius erythrophthalmus*) | perch (*Perca fluviatilis*) | rudd (*Scardinius erythrophthalmus*) | perch (*Perca fluviatilis*) |
| Sample size | 10 | 3 | 9 | 1 |
| Weight (g) | 11.5 ± 2.1 | 236 ± 74 | 416 ± 344 | 108 |
| Length (cm) | 10.3 ± 0.3 | 26.3 ± 2.8 | 27.2 ± 7.8 | 19 |
| K (g/cm$^3$) | 1.2 ± 0.1 | 1.3 ± 0.2 | 1.2 ± 0.1 | 1.3 |
| $\delta^{15}$N | 7.12 ± 0.26[a] | 8.47 ± 0.27 | | |
| $\delta^{13}$C | −30.0 ± 0.5[a] | −30.9 ± 0.4 | | |
| $^{137}$Cs (kBq/kg)[b] | 18.6 ± 8.5 | 14.2 ± 3.6 | 0.13 ± 0.01 | * |
| $^{127}$I (mg/kg) | 0.21 | 0.29 | 1.05 | 0.30 |
| $^{129}$I (x 10$^{12}$ at/kg)[b] | 9.79 ± 9.95 | 4.97 ± 6.96 | 2.48 ± 1.79 | 0.86 |
| $^{129}$I (x 10$^{12}$ at/kg) intestinal cont. | 35.4 ± 38.6 | | 34.7 ± 53.7 | |
| $^{129}$I/$^{127}$I (x 10$^{-6}$ at/at)[b] | 10.7 | 3.6 | 0.4 | 0.6 |
| $^{129}$I/$^{137}$Cs (x 10$^{-1}$ at/at) | 4.16 | 2.56 | 119 | |
| CR $^{137}$Cs (x 10$^4$ L/kg)[b] | 0.46 ± 0.28 | 0.39 ± 0.10 | 0.059 ± 0.29 | |
| CR $^{127}$I (x 10$^2$ L/kg) | 0.99 | 1.37 | 3.75 | 1.07 |
| CR $^{129}$I (x 10$^2$ kg/kg)[b] | 5.44 ± 5.52 | 2.76 ± 3.87 | 22.95 ± 19.21 | 8.97 |

[a] Significantly different between species within one reservoir.
[b] Significantly different between reservoirs of similar species.

### 3.3.2. *Fish food sources*

In Fukushima waterbodies the $\delta^{13}$C signal in fish muscle ranged from −22.4 to −32.6‰ for all fish species independent of species and water body (Table 3). In Glubokoye lake in ChEZ the $\delta^{13}$C signal in fish muscle ranged from −29.0 to −31.3‰ for the two fish species (Table 4). The $\delta^{13}$C signal in the intestinal content was significantly lower (1.9 in average difference, P = 0.02) compared to the muscle of fish, indicating that intestinal content on the day of sampling was not the major food source as the $\delta^{13}$C signal is not expected to change more than 0.4‰ by trophic fractionation (Nielsen et al., 2018). The results indicated that the food sources could be classified as intermediate between food of terrestrial and aquatic origin (Matsuzaki et al., 2010; Chikaraishi et al., 2011; Solomon et al., 2011). Both Fukushima juvenile bass in Inkyozaka pond and rudd in Glubokoye lake in ChEZ had $\delta^{13}$C signals similar to those reported for terrestrial sources (Chikaraishi et al., 2011; Øvergaard, 2022).

### 3.3.3. *$^{129}$I in fish*

The concentration of $^{129}$I in fish muscle sampled in Fukushima waterbodies ranged from 8.4 x 10$^{11}$ to 13.02 x 10$^{13}$ at/kg and was in the same range as observed in Chernobyl waterbodies (8.6 x 10$^{11}$ to 9.8 x 10$^{12}$ at/kg, Tables 3 and 4). The concentration of $^{129}$I was highest in fish collected from the most contaminated waterbodies in both Fukushima waterbodies and Chernobyl waterbodies, with a factor of 2–16 lower in similar fish species either in less contaminated waterbodies with increasing distance from NPP or outside the plume of fallout.

### 3.3.4. *$^{129}$I concentration ratio for freshwater fish*

The observed concentration ratios (CR) for $^{129}$I (atom/kg dry weight fish per atom/kg water) were in the range 85–3470 based on all fishes sampled (correspond to 18–728 L/Kg wet weight as dry weight muscle was 21% of wet weight), both from Fukushima waterbodies and Chernobyl waterbodies (Tables 3 and 4). There were no differences in the CR range observed in fish from Fukushima waterbodies compared to fish from Chernobyl waterbodies. The concentrations of $^{129}$I were thereby higher in the fish muscle than in the water from all waterbodies. This is supported by the observed CR for $^{127}$I, that was in range 7.8–41 for fish where $^{127}$I were measured (1.6–8.2 L/kg wet weight). Thus, the trend is different from that observed in the marine environment. In the marine environment, the CR of $^{127}$I in fish is about 1 L/kg, and the $^{129}$I/$^{127}$I ratio in fish is reported to be similar to that of seawater (Kusuno et al., 2015) or brackish water (Udea et al., 2015). Higher CRs of iodine in freshwater fish than in marine species have previously been reported (Eckhoff and Maage 1997). However, this is the first results on the transfer of $^{129}$I to fish, showing that the CR of $^{129}$I for freshwater fish is also higher than CR reported for marine fish.

It could be argued that higher concentrations of stable iodine in marine environment than in freshwater could decrease the uptake of $^{129}$I in marine fish due competing effects of stable I. The effect of stable iodine on the uptake of $^{129}$I was not possible to identify in the present work as the isotopic ratios of $^{129}$I/$^{127}$I in waterbodies with limited water exchange were within the same range for both Fukushima waterbodies and Chernobyl waterbodies. The observed atomic ratio of $^{129}$I/$^{127}$I in fish were in the range of 0.6–20 x 10$^{-6}$ and about a factor of 10 higher compared to observed atomic ratios in the corresponding waterbodies. Thus, the $^{129}$I/$^{127}$I ratio in fish was different compared to the water, unlike observations in the marine and coastal environment (Kusuno et al., 2015; Udea et al., 2015).

Higher $^{129}$I CR was observed in waterbodies with running water (897–3470 dry weight) than in ponds and lakes with limited water exchange (85–544 dry weight) across Fukushima and Chernobyl areas. Observations from differences in $^{129}$I Kd, sediment-water interactions from the same waterbodies (Fukushima) showed also lower concentration in water compared to sediments in running water from Abukuma river than in the ponds, supporting trends for CR in fish. This could be attributed to different water composition at time of sampling in rivers compared to normal situations or to migrations of fish, making the observed CR in running waterbodies less credible than in waterbodies with limited water exchange. However, such differences between waterbodies were not observed for $^{137}$Cs CR, indicating different sources or different body accumulation pathways for the two radionuclides. The observed atomic ratio of $^{129}$I/$^{137}$Cs in fish muscles were, however, in the same range for water and samples of fish muscles (Tables 3 and 4).

Observed $^{129}$I CR indicate differences between fish species within the same water body, although the number of fish was too low to conclude. The $^{129}$I CR in Largemouth bass samples was significantly lower than for carps in Fukushima waterbodies. Differences were also observed for body concentrations of stable iodine, where the concentration of $^{127}$I in muscle of common carp was lower than that of bass. Differences in the iodine concentrations in fish muscles between marine fish species are also well known due to uptake of iodine from both water and diet (Eckhoff and Maage 1997). The difference in $^{129}$I/$^{127}$I between water and fish and the change in CR indicated different feed sources than waterborne $^{129}$I in fish. This is in agreement with the assumption that fish accumulate iodine via the diet (Kusuno et al., 2015; Martinez et al., 2014b).

### 3.3.5. *Tissue distribution of $^{129}$I*

The concentration of $^{129}$I and $^{127}$I varied significantly among various tissues and organs, as shown for crucian carp from the Suzuuchi pond in Fukushima (Fig. 3). The concentration of $^{129}$I was lowest in the gills,

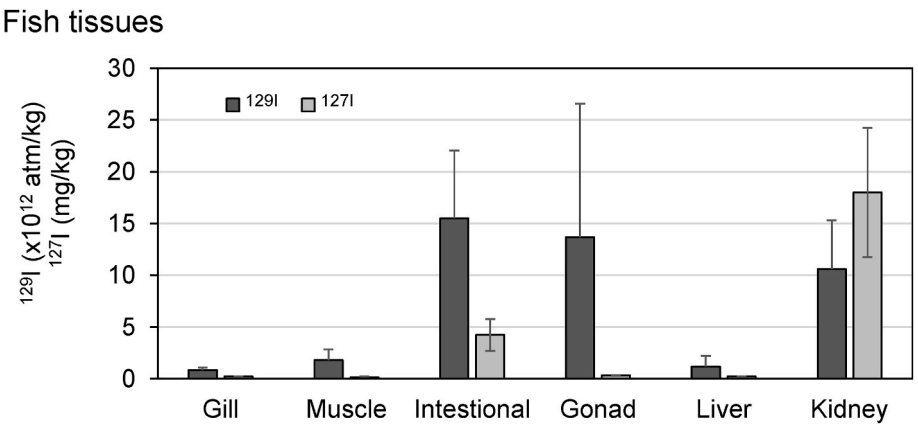


**Fig. 3.** Concentration of $^{129}$I and $^{127}$I in different fish tissue of crucian carp in Suzuuchi pond (n = 5).

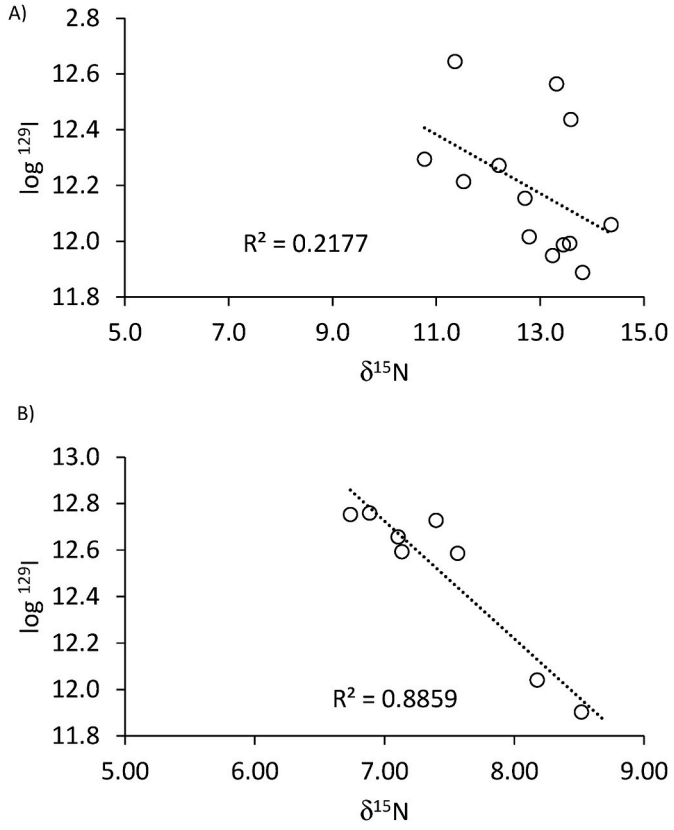


**Fig. 4.** Correlation between $\delta^{15}$N and log $^{129}$I in fish from A) Suzuuchi pond in Fukushima exclusion zone and B) Glubokoye lake in Chernobyl exclusion zone.

liver and muscle, while highest in kidney and gonad. These trends were not observed for stable iodine showing different distributions with lowest concentrations in gonad. The $^{129}$I/$^{127}$I atom ratio was thus highest in gonad and 10-fold lower in other tissues (Table S1). This indicates different tissue distributions for $^{129}$I and $^{127}$I. For comparison, strong correlation existed for stable analogues of other radionuclides, e.g., as observed between $^{133}$Cs and $^{137}$Cs ($R^2$ = 0.96), showing similar trends. The $^{129}$I concentration was among the lowest in the muscle tissue compared to other tissues, while the activity concentrations of $^{137}$Cs was highest in muscle tissues. The results highlighted that the dose distribution in fish is highly inhomogeneous, and it is essential to compare similar tissues when activity concentrations of radionuclides between fishes are compared.

### 3.3.6. *Dietary uptake of $^{129}$I in fish*

The concentrations of $^{129}$I in the diet (intestinal content) were higher than in the fish muscle at the time of sampling (Fig. S1), and the calculated CR in fish from waterbodies with limited water exchange was in the range 18–114 (85–544 dry weight, Tables 3 and 4). The trophic magnification factor (TMF, kg/kg) of $^{129}$I was 0.12 ± 0.7 and 0.21 ± 0.17 (muscle/intestinal content) in Fukushima waterbodies and Chernobyl waterbodies, respectively, (based on fish sampled for intestinal content). Uptake of waterborne trace elements through the gills is an important exposure pathway. Our results indicated, however, minor influence of the water borne iodine on uptake in fish as the observed CR was high and varied between similar fish species within a waterbody. In addition, the fact that the $^{129}$I/$^{127}$I ratio in water and fish were different, supported the assumption that the $^{129}$I was also transferred to fish by diet. The negative correlation between Log$^{129}$I in fish muscle and $\delta^{15}$N ($P < 0.001$) within Chernobyl waterbodies ($R^2 = 0.89$, Fig. 4) indicated that biodilution would affect the $^{129}$I through the food web. This is supported by the fact that TMF was lower than 1 for $^{129}$I. More data are, however, needed to conclude as the trend was not significant in Fukushima ponds, as the intestinal content $\delta^{13}$C signal at the time of sampling did not reflect the main food source. Fish food items varies and $\delta^{13}$C as well as trace elements such as iodine among food items (Nielsen et al., 2018).

The diet of common carp (*Cyprinus carpiocarassius*) has also been reported to be dominated by benthic and planktonic invertebrates (Matsuzaki et al., 2010), while terrestrial prey items represent a significant part of the diet, especially in juvenile largemouth bass (Hodgson and Hansen, 2005; Sutton and Ney, 2002), and rudd (Revnne and JeuErl, 1991). The $\delta^{13}$C values for carp species in this study varied from −25.8 to −22.2‰, suggesting that the fish utilized the littoral zone originated food source as feed more frequently than the juvenile bass, having lower $\delta^{13}$C values in stomach content, similar to ranges previously reported for terrestrial invertebrates in Japan (Chikaraishi et al., 2011). Thus, dietary ingredients of $^{129}$I transfer to fish could also include foods with terrestrial $^{129}$I/$^{127}$I ratio source signals different from those of the waterbodies.

### 3.4. *Dose contribution*

Dose contribution of $^{129}$I has been calculated (Hou et al., 2009) to be about 1 μSv/y for organisms with $^{129}$I/$^{127}$I atomic ratio of $10^{-6}$ and about 0.1 mSv/y for organisms with $^{129}$I/$^{127}$I atomic ratio of $10^{-4}$. The present $^{129}$I/$^{127}$I atomic ratio in the range of $10^{-5}$ in the most contaminated fish in this study, i.e., in the ponds in Fukushima and in Glubokoye lake in Chernobyl, would result in a radiation dose of 10 μSv/y, significantly lower than 1 mSv/y for human. Compared to the total internal dose contributions from $^{137}$Cs and $^{90}$Sr in fish from Glubokoye lake (8.2–9.8 μGy/h, Lerebours et al., 2018), the radiotoxicity of $^{129}$I in

fish is not expected to cause any harm to the fish or to fish eating predators.

## 4. Conclusion

Information on $^{129}I$ in freshwater, sediment and fish have for the first time been documented, based on sampling campaigns in three waterbodies in Fukushima exclusion zone and two waterbodies in Chernobyl exclusion zone. Results demonstrated that waterborne $^{129}I$ was mainly present as LMM and negatively charged species, but also that a minor fraction was associated with colloidal sizes, most probably organic material. The water-sediment apperent distribution coefficient (Kd) for $^{129}I$ was observed to be in the range 225–329 L/kg in waterbodies with limited water exchange in Fukushima and similar to stable iodine. However, $^{129}I$ seemed more mobile in sediments than $^{137}Cs$, being retained in the upper sections. The observed concentration ratios (CR) for $^{129}I$ in fish muscle tissue to water were in the range 85–544 (atom/kg dry weight fish per atom/kg water) across waterbodies with limited water exchange in Fukushima and Chernobyl. Thus, the CR of $^{129}I$ for freshwater fish is higher than CR reported for marine fish. As the $^{129}I/^{127}I$ ratio in fish was different than in water and the $^{129}I$ concentration level in fish was lower than in the intestinal content, results indicate that $^{129}I$ in fish could be attributed to dietary uptake of contaminated feed, probably of terrestrial origin, and biodiluted. As the source and then the environmental distribution of radioactive iodine is different from that of stable iodine, data of $^{129}I$ is important to assess consequences of radioactive iodine contamination in the environment. The current data is the first results on the transfer of $^{129}I$ to freshwater fish and show that the transfer in Fukushima waterbodies were similar to the Chernobyl waterbodies having similar $^{129}I/^{127}I$ ratio.

## 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.

## Data availability

Data will be made available on request.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2023.107269.

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