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Transfer of elements relevant to radioactive waste into chironomids and fish in boreal freshwater bodies

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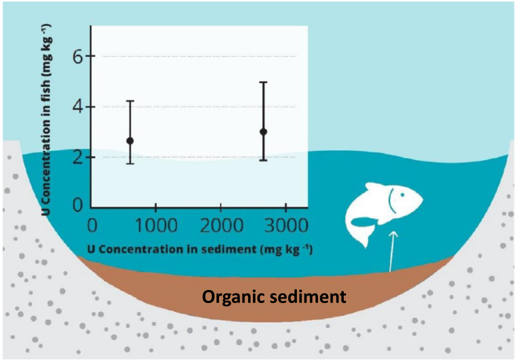
HIGHLIGHTS

- Transfer of elements into chironomids and fish was investigated in freshwater ponds.
- Sediment is likely to be the main source of most elements.
- Concentration in fish is not adequately predicted by constant concentration ratio.
- The data are needed for the development of radioecological models.

GRAPHICAL ABSTRACT



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ABSTRACT

Information on transfer of elements and their radionuclides is essential for radioecological modeling. In the present study, we investigated the transfer of Cl, Co, Mo, Ni, Se, Sr, U and Zn in a boreal freshwater food chain. These elements were selected on the basis that they have important radionuclides that might be released into the biosphere from various stages of the nuclear fuel cycle. Water, sediment, chironomid larvae (*Chironomus sp.*), roach (*Rutilus rutilus*) and perch (*Perca fluviatilis*) were sampled from two ponds near a former uranium mine and one reference pond located further away from the mining area. Concentrations measured in water, sediment and the three animal species indicated the importance of sediment as a source of uptake for most of the elements (but not Cl). This should be considered in radioecological models, which conventionally predict concentration in aquatic organisms from concentration in water. The results also show that the assumption of linear transfer (constant concentration ratio) may not be valid for elements into fish. The results of this study show that further basic research is needed to understand the fundamental processes involved in transfer of elements into freshwater organisms in order to develop radioecological models.

1. Introduction

Radioecological modeling is an essential tool for predicting transfer of radionuclides in ecosystems, estimating doses, and assessing possible adverse effects on human health and ecosystems (Hilton, 1997; Avila

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et al., 2003; Shaw, 2005; Caffrey et al., 2014; Haanes et al., 2020). The use of the models requires quantitative data on transfer of elements and their radionuclides in the environment. Only limited amount of such data is available on boreal freshwater bodies.

The current radioecological models for predicting steady-state concentrations in biota are generally based on the use of concentration ratios (CR). Concentration ratio is defined as the ratio of radionuclide concentration in biota to the corresponding concentration in soil, sediment, or water (IAEA, 2010; Copplestone et al., 2013; IAEA, 2014; Brown et al., 2016). The biota/medium concentration ratios for other contaminants than radionuclides are commonly described by terms such as bioconcentration factor, bioaccumulation factor, and biota sediment accumulation factor (McGeer et al., 2003; Rowan et al., 2014). These ratios are simple to use but have limitations. Many physical, chemical, and biological processes occurring in nature are aggregated into one parameter, which increases the uncertainty of model predictions (Hinton et al., 2013). For example, many of the models describing the transfer in aquatic environment do not consider the exposure of benthic organisms via sediment or suspended particles (Konovalenko et al., 2017). One of the current challenges in radioecology is to identify the key processes affecting the transfer and develop modeling to be process-based (Hinton et al., 2013; Konovalenko et al., 2017).

One basic assumption related to CRs is that the uptake of elements into living organisms is linear, i.e., that the CR for an element is constant regardless of its concentration in the medium. However, this assumption may not be correct in all ecosystems and for all elements. Studies in boreal forest species have revealed clear deviation from the linearity assumption (Tuovinen et al., 2011, 2016a, 2016b), and non-linear uptake of elements and their radionuclides has also been observed in other studies in e.g. plants (Timperley et al., 1970; Sheppard and Sheppard, 1985; Palm, 1994; Krauss et al., 2002; Han et al., 2006), fish (Pyle and Clulow, 1997) and earthworms (Keum et al., 2013). In the present study, we evaluated the validity of this assumption in boreal aquatic ecosystems.

To fill the knowledge gap on the transfer of elements in boreal aquatic food chains, samples of water, sediment, larvae of chironomid midges (*Chironomus sp.*), roach *(Rutilus rutilus)* and perch *(Perca fluviatilis)* were collected from three ponds in Eastern Finland. Roach and perch are common fish species in Finnish freshwaters, and the diet of both species includes benthic organisms. Moreover, several benthic organisms such as chironomid larvae consume and recycle autochthonous and allochthonous organic matter (Jones and Grey, 2004), which may result in uptake of contaminants bound to organic matter.

Like in our previous studies (Roivainen et al., 2011a, 2011b, 2012; Tuovinen et al., 2011, 2016a, 2016b), total element concentration was measured rather than specific radionuclides. This approach is based on the assumption that, in equilibrium, radioactive and stable isotopes of the same element behave similarly in ecosystems, and data on total element concentration can thus be utilized in models predicting transfer of radionuclides in case of long-term radioactive contamination such as failure of a spent nuclear fuel repository (IAEA, 2010).

The elements studied were chlorine (Cl), cobalt (Co), molybdenum (Mo), nickel (Ni), selenium (Se), strontium (Sr), uranium (U) and zinc (Zn). These elements were selected on the basis that they have important radionuclides that might be released into the nature from various stages of the nuclear fuel cycle from mining to final disposal of spent nuclear fuel (OECD, 2006; Posiva, 2012).

2. Materials and methods

2.1. Sampling sites

All the samples were collected from three ponds near the Paukkajanvaara former uranium mine in Eastern Finland, approximately 40 km NE of the city of Joensuu (N 6981372, E 653304, ETRS-TM35FIN), (Fig. 1). The mine was used for uranium extraction from 1959 to 1961 and closed because it proved to be uneconomic (Colpaert, 2006). After 30 years the mine was rehabilitated with clay and till and planted with pine trees (Colpaert, 2006). Of the three ponds, Iso Hiislampi (N 6981416, E 653352, ETRS-TM35FIN) is located closest to the abandoned mine. Pieni Hiislampi (N 6981778, E 653324, ETRS-TM35FIN) is downstream of Iso Hiislampi. Ruosmanlampi (N 6982224, E 654851, ETRS-TM35FIN) is located nearby but in a different watershed and used as a reference pond in this study. The maximum depths of the ponds were 5 m (reference pond), 10 m (Pieni Hiislampi) and 14 m (Iso Hiislampi), (Metsähallitus, 2020). The surface areas of the ponds were 6.20 ha, 1.23 ha and 2.53 ha for reference pond, Pieni Hiislampi and Iso Hiislampi, respectively. Samples were collected in March 2016 (from Pieni Hiislampi and Iso Hiislampi), June 2017 (from all three ponds), and November 2017 (from all three ponds).

2.2. Sediment samples

In November 2017, sediment samples were collected at six randomly selected spots from each pond, aiming at a sampling that represents the whole pond. The six samples per pond were pooled for analysis. The samples were collected with Ekman sampler (17 × 17 cm). The samples were transported to the laboratory and stored at −20 °C.

2.3. Water samples

Surface water samples were collected from each pond with buckets at two randomly selected spots in June 2017. The water pH and water conductivity were also measured (YSI Professional Plus, YSI Incorporated, Yellow Springs, Ohio, USA) from surface of the ponds. In the laboratory, 100 ml of water from each sample was placed into a beaker and evaporated for 40 h at 50 °C to reach 20 ml. The samples were then stored at 4 °C before analysis.

2.4. Sampling of chironomid larvae

Sampling was conducted in March 2016 at Pieni Hiislampi and Iso Hiislampi, and November 2017 at the reference pond. The chironomid larvae were collected with the same method used for sediment samples at the depths of approximately 3 to 6 m from the pond surface. One of the samples in Pieni Hiislampi was collected at the deeper part of the pond (approximately 8 m) near a water stream flowing from Iso Hiislampi. Samples were sieved from the sediment in the field. We continued sampling of chironomid larvae at different times of the year in the reference pond, but no chironomids were found. Thus, two samples per pond (each consisting of approximately 40 chironomid larvae) from two ponds were collected and transported to the laboratory and stored at −20 °C. Before freezing the samples, gut contents of chironomid larvae were purged for about 1 h in clean water.

2.5. Fish samples

Fish samples were collected with fishing rods in June 2017 from all ponds. Ten roach samples and 10 perch samples per pond were collected from Iso Hiislampi and Pieni Hiislampi. No roaches were caught (despite later attempts during the year) in the reference pond, but six perches were collected. The samples were transported to the laboratory and stored at −20 °C. The guts were removed with plastic knife after freezing. The average perch dry weight (g), was 7.47 (SD 3.24) in Iso Hiislampi, 15.8 (SD 8.36) in Pieni Hiislampi and 4.98 (SD 1.55) in the reference pond. The roach dry weights (g), were 9.23 (SD 4.78) and 6.74 (SD 2.62) in Iso Hiislampi and Pieni Hiislampi, respectively.

2.6. Analysis of the samples

Organic matter content of sediment samples was determined by measuring loss on ignition. Sediments were oven-dried at 65 °C overnight and were then ground and homogenized. Empty crucibles

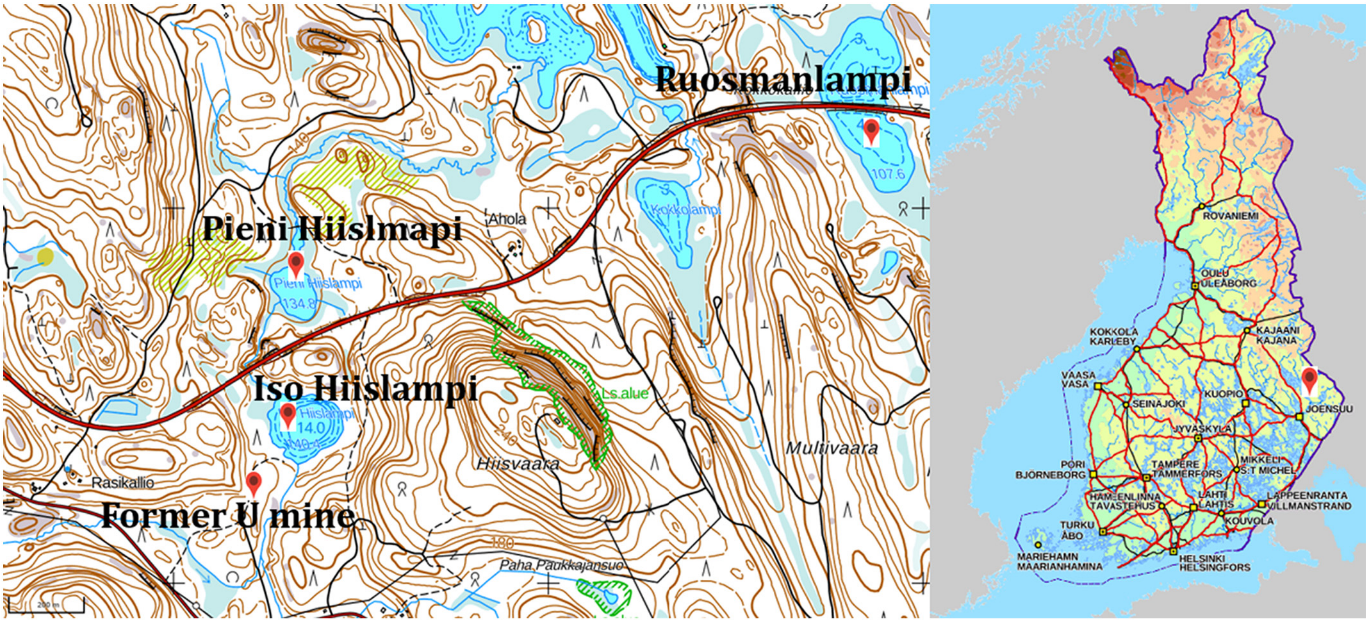

Fig. 1. The locations of two ponds (Iso Hiislampi and Pieni Hiislampi) near Paukkajanvaara former uranium mine and a reference pond (Ruosmanlampi) in Eastern Finland. Iso Hiislampi is located close to the former mine. The map is taken from National Land Survey of Finland, license CC 4.0 (Topographic map raster 1:50000; 05/2021).

were pre-heated in oven at 550 °C for 1 h and weighed. They were then filled with 1 g of sediment (three replicates for each sample were analyzed) and weights of crucibles with dried sediments were determined. The samples were then placed in the oven at 550 °C for 2 h and the weight of crucibles with ashes was measured. The % weight reduction (sediment dry weight minus ash weight; loss on ignition), was used as a measure of the organic matter content. Carbon and nitrogen concentrations of the sediments were analyzed with elemental analyzer (Thermo Finnigan, FLASH EA 1112, Milan, Italy) in order to calculate the C/N ratio. 2–3 mg of each dried sediment was added to tin cups for analysis.

Multi-elements analyses were carried out in a commercial laboratory in Kuopio, Finland (Labtium Ltd., accredited according to FINAS T025 EN ISO IEC 17025). All the analyses included blanks and every 20th sample was measured twice. Tomato leaves (SRM 1773a) and lake sediment (NW-WQB-1) were used as certified reference materials. Inductively coupled plasma-mass spectroscopy (ICP-MS, Thermo iCap Q, Thermo Fisher Scientific, Waltham, Massachusetts, USA) and inductively coupled plasma optical emission spectrometer (ICP-OES, Thermo iCap 6500 duo, Thermo Fisher Scientific, Waltham, Massachusetts, USA) were used to analyze the concentrations of metals and selenium after nitric acid (HNO_3) digestion (following the standard procedure US-EPA 3051). Before the analyses, sediment, fish, and chironomid samples were freeze-dried with freeze dryer (Zirbus lyophilizator, Zirbus technology GmbH, Harz, Germany) under a vacuum. Fish and chironomid samples were then homogenized, using a cutting mill in Teflon container equipped with metal blades. Sediment samples were sieved to <2 mm. 0.5 g of each sample (sediment, fish, and chironomid) was then weighed into a Teflon high temperature digestion vessel. 10 ml of concentrated HNO_3 was added, and samples were digested with CEM MARS microwave oven (8.5 bar, 30 min). After cooling, the volume was made up to 50 ml with deionized water. Water samples were filtered to 0.45 μm, acidified with 10 ml of HNO_3 and 50 ml of volume was made by adding deionized water before analysis by ICP-MS and ICP-OES. Analyses of Cl in sediment and fish samples followed the standard method (SFS-EN 15408). One gram of each sample was pressed to a pellet and combusted in an oxygen atmosphere in a closed combustion vessel. The acidic gas was absorbed to a carbonate/bicarbonate solution. The solution was then filtered. Finally, the prepared solution (of sediment and fish samples) and the filtered water (as described above) were analyzed with Thermo Electron Dionex ion chromatography (ICS 3000, Thermo Fisher Scientific, Massachusetts USA) following the standard method (SFS-EN ISO 10304-1). Cl concentrations in chironomids were not analyzed because of small amount of sample material. Concentrations in sediment, chironomids and fish are given per dry weight (DW), and DW-based concentration ratios are reported. For comparison (in the discussion section) to published values based on fresh weight, the dry/fresh weight values given in Tables 1 and 2 were used for conversion.

2.7. Statistical analyses

To compare the perch concentrations and perch-to-sediment/water CR values between three ponds One-Way ANOVA with Tukey's test for pairwise comparison was used. To compare the roach concentrations and roach-to-sediment/water CR values between two ponds independent-samples *t*-test was used, not assuming equal variances. Because of log-normal distributions, data were log-transformed before analyses. The chironomid data were not tested statistically, as only two chironomid samples per pond were collected. All statistical analyses were performed using SPSS 25 for Windows (SPSS Inc., an IBM Company). Differences were considered statistically significant when $P \leq 0.05$.

3. Results

All sediments were highly organic, the total organic matter content being the highest in Iso Hiislampi (91%), followed by Pieni Hiislampi (74%) and the reference pond (40%). The C/N ratios were 33, 29 and 15, respectively. Water pH was circumneutral in all ponds (approximately 6.5–6.7) and water conductivity was 4.8 mSm^{-1} in the reference pond, 5.2 mSm^{-1} in Pieni Hiislampi and 5.4 mSm^{-1} in Iso Hiislampi.

The element concentrations in sediment, except for Sr, were highest in Iso Hiislampi, which is the pond closest to the former mine (Table 1). The sediment concentrations of the elements were generally the lowest in the reference pond. The concentrations in surface water were below the detection limit for many of the elements studied (Table 1). In general, the concentrations were orders of magnitude higher in sediment than in water. However, Cl concentrations were much lower in

Table 1
Element concentrations in water (mg L^{-1}) and sediment (mg kg^{-1} DW) in two ponds near a former uranium mine (Pieni Hiislampi and Iso Hiislampi) and in a reference pond. Arithmetic mean of two samples per pond is given for water. For sediment samples one pooled sample per pond was analyzed.

	Co	Mo	Ni	Se	Sr	U	Zn	Cl
Water								
Reference	<0.001*	<0.002*	<0.002*	<0.001*	0.02	<0.001	<0.02	1.10
Pieni Hiislampi	<0.0002	<0.001	0.0006	<0.0006	0.01	0.003	<0.02	0.80
Iso Hiislampi	<0.0002	<0.001	0.001	<0.0006	0.01	0.006	<0.02	0.55
Sediment								
Reference	8.55	1.80	23.2	2.60	22.4	5.14	95.1	0.02
Pieni Hiislampi	27.8	2.91	27.0	2.21	36.2	441	86.5	0.03
Iso Hiislampi	58.2	17.7	89.5	6.07	30.6	2685	165	0.02

The arithmetic mean of dry/fresh weight ratios in sediments was 0.06 in the reference pond, 0.04 in Pieni Hiislampi and 0.07 in Iso Hiislampi. *The water samples from the reference pond were analyzed later than the other water samples. Because of the separate analyses, the detection limits for Co, Mo, Ni and Se were not identical.

sediment than in water. Among the animal species studied, the highest element concentrations were found in chironomids except for Sr and Zn (Table 2). All concentrations measured are reported in Supplementary Table S2.

Because of the lower concentrations in water compared to sediment, the calculated animal-to-water CR values were much higher than CRs derived from sediment for other elements than Cl. As concentrations of the elements in animal and water samples were below detection limits for many elements, animal-to-water CR values could only be reliably calculated for Cl, Sr and U (Table 3, see animal-to-water CRs for other elements in Supplementary Table S1). The animal-to-sediment CR values were higher in chironomids than in fish, except for Sr and Zn (Table 4). The CRs of most elements tended to be higher in roach than in perch; only the CRs for Se and Cl were higher in perch than in roach. For perch, the data suggested an inverse relationship between CR and concentration in sediment for most elements: the highest CR values were observed in the pond with the lowest concentrations in sediment, and lowest CR values in the pond with highest concentrations. A similar trend was observed for roach, although roach samples from the reference pond were not available. The fish-to-water CR for Cl showed a similar pattern as the fish-to-sediment CRs for the other elements: there was an inverse relationship between CR and concentration in water (note that Cl concentration was the highest in the reference pond, Table 1). The chironomid-to-sediment CR values did not show substantial or systematic differences between the two ponds; the maximum differences were less than 3-fold, and the direction of the difference varied.

To further illustrate the uptake of Co and U (for which there were clear differences in sediment concentrations) into fish and chironomids, their concentrations and CR values are plotted against sediment concentration. The concentrations of Co and U in roach did not increase with increasing concentration in sediment, and the CR values decreased with increasing sediment concentration (Fig. 2). A different pattern was seen in chironomids: concentrations of Co and U increased with increasing concentration in sediment, and the CR values were approximately constant (Fig. 3).

4. Discussion

This study included determination of element concentrations in both water and sediment, as well as in two fish species and in a sediment-dwelling benthic organism contributing to the same food web. Samples from three ponds formed a gradient of environmental element concentrations.

Overall, the concentrations in both water and sediment agree with previous data from Iso Hiislampi, indicating that one-time sampling was representative. According to Tuovinen et al. (2016c), the peak sediment concentration of U in Iso Hiislampi at depths of 6–8 cm was approximately 27 Bq g^{-1} (2186 mg kg^{-1}), which is consistent with the sediment concentration in this study (2685 mg U kg^{-1}). Concentrations of elements in water were also comparable to previous data showing concentration of U in stream water samples from 0.002 to 0.012 mg L^{-1} (Tuovinen et al., 2016c) and concentrations of Co, Ni and Zn of 0.0001 mg L^{-1}, 0.0011 mg L^{-1} and 0.006 mg L^{-1} in Iso Hiislampi, respectively (Tuovinen et al., 2019).

The element concentrations and CR values were generally higher in chironomids than in fish. This is not surprising, as chironomid larvae are sediment-dwelling organisms and ingest sediment particles, in which concentrations are high. The results of a recent study are broadly consistent with our findings. Väänänen et al. (2019) reported that the

Table 2
Geometric means of element concentrations (mg kg^{-1} DW) in chironomid, roach and perch samples in two ponds near a former uranium mine (Pieni Hiislampi and Iso Hiislampi) and in a reference pond. Values in parentheses indicate range for chironomids and geometric standard deviation for fish samples.

	Co	Mo	Ni	Se	Sr	U	Zn	Cl
Chironomids								
Pieni Hiislampi ($n = 2$)	0.47 (0.44–0.50)	0.45 (0.43–0.49)	0.69 (0.60–0.80)	1.54 (1.41–1.71)	0.72 (0.57–0.91)	4.78 (3.81–6.00)	91.2 (88.3–94.2)	n.a.
Iso Hiislampi (n = 2)	2.02 (1.91–2.13)	1.27 (1.15–1.41)	2.52 (2.00–3.20)	6.98 (6.60–7.40)	1.46 (1.37–1.56)	35.5 (22.8–55.2)	92.0 (75.6–112)	n.a.
Roach								
Pieni Hiislampi ($n = 10$)	0.11 (1.09)	0.07 (1.34)	< 0.2	1.32 (1.12)	77.8 (1.14)	2.60 (1.62)	165 (1.19)	0.27 (1.05)
Iso Hiislampi (n = 10)	0.10 (1.12)	0.06 (1.16)	< 0.2	1.78 (1.17)	64.9 (1.14)	3.00 (1.62)	144 (1.18)	0.27 (1.09)
P	0.79	0.52		0.02	0.10	0.52	0.11	1
Perch								
Reference ($n = 6$)	< 0.1	< 0.04	0.23 (2.22)	1.58 (1.03) a	34.5 (1.11) a	< 0.02	103 (1.14) a	0.26 (1.07) a
Pieni Hiislampi (n = 10)	< 0.1	< 0.04	< 0.2	1.83 (1.14) a	42.4 (1.16) b	0.99 (1.41)	67.6 (1.22) b	0.32 (1.06) b
Iso Hiislampi (n = 10)	< 0.1	< 0.04	< 0.2	2.36 (1.21) b	40.1 (1.13) b	1.49 (1.62)	73.3 (1.21) b	0.32 (1.03) b
P				0.002	0.02	0.03*	0.002	0.005

The arithmetic mean of dry/fresh weight ratio in fish was 0.27 (range from 0.25 to 0.29). In chironomids, the dry/fresh weight ratio is approximately 0.16 (Armitage et al., 2012).
t-test was used for comparisons when there were samples from two ponds, and ANOVA with Tukey's test for comparisons when there were samples from three ponds. Different letters (a and b) indicate significant difference between the ponds in Tukey's test ($P < 0.05$). **t*-test was used for U as the concentrations in the reference pond were below detection limit and not included in the analysis.
n.a. = data is not available.

Table 3
Geometric mean and geometric standard deviation (range for chironomids) of animal-to-water CR (L kg^{-1}) in two ponds near a former uranium mine (Pieni Hiislampi and Iso Hiislampi) and in a reference pond.

	Sr	U	Cl
Chironomids			
Pieni Hiislampi	44.9 (35.9–56.2)	1594 (1270–2000)	n.a.
Iso Hiislampi	97.5 (91.3–104)	5904 (3800–9200)	n.a.
Roach			
Pieni Hiislampi	4864 (1.15)	867 (1.62)	0.33 (1.05)
Iso Hiislampi	4326 (1.15)	500 (1.59)	0.48 (1.08)
P	0.07	0.02	0.001
Perch			
Reference	1728 (1.11) a	n.a.	0.24 (1.08) a
Pieni Hiislampi	2654 (1.16) b	331 (1.42)	0.39 (1.05) b
Iso Hiislampi	2671 (1.13) b	249 (1.62)	0.58 (1.04) c
P	<0.001	*0.28	<0.001

n.a. = data is not available. *t*-test was used for Pieni Hiislampi and Iso Hiislampi. **t*-test was used for U as there were two ponds. Different letters (a, b and c) indicate statistically significant difference in Tukey's pairwise test ($P < 0.05$).

concentrations of Cu, Ni and As were higher in chironomids than in perch and pike (*Esox lucius*). However, Sr concentration was much higher in the two fish species than in chironomids. The high uptake of Sr into fish is most likely related to the fact that it is a calcium analogue and accumulates to bones (IAEA, 2010; Chowdhury and Blust, 2011; Salem et al., 2014). The concentrations of most elements were clearly lower in the animals than in sediment (CR values much less than one), indicating either low bioavailability in the sediments or active regulation to keep the concentration of toxic elements at a sufficiently low level, and that of essential elements (such as Co and Mo) at an optimal level. Concentrations of all other elements than Cl were much higher in animal tissues than in water (the DW-based CRs were from 45 to more than 11,600 in chironomids and from more than 61 to over 16,500 in fish). Furthermore, animal-to-water CR values are substantially higher than animal-to-sediment CRs (e.g., up to 500,000-fold higher for U in chironomids). These findings are consistent with results showing that sediment rather than water is the main source of uptake in deposit-feeding organisms (Rowan et al., 2014). Consistently with our results, Väänänen et al. (2019) reported that the concentrations of As, Cu, Ni and Zn in four Finnish lakes were much higher in sediment than in water.

Transfer of elements from sediments to chironomids and fish has been addressed in only a few previous studies, and only two of them (Draves and Fox, 1998; Crawford and Liber, 2016) have addressed boreal ecosystems. Reinhold et al. (1999) reported that the DW-based CR between chironomid larvae and sediment was 0.03 for Ni, while the CR for Zn was approximately 0.3. These values were measured for only one sediment, so possible variation with sediment concentration could not be assessed. Turceková et al. (2002) reported that the concentrations of Cu, Zn, Cd, As and Pb were 1 to 3 orders of magnitude lower in perch liver than in the sediment (and concentrations were lower in perch muscle than in liver). Draves and Fox (1998) investigated concentration of Zn in yellow perch (*Perca flavescens*) and several invertebrate taxa (including Chironomidae) that serve as prey for yellow perch. While Zn concentration in chironomids were generally higher in the contaminated section of the river than in the reference section, no differences between the sections were observed in concentrations in perch. The results of this study are consistent with our findings that showed constant concentration in fish but increased concentration in chironomids with increases in environmental concentrations. Crawford and Liber (2016) investigated the bioaccumulation of U in *Chironomus dilutus* by exposing the larvae to 50 and 500 mg U kg^{-1} DW in spiked sediment. Overall, the results showed lower concentration of U (mg kg^{-1} DW) in the larvae than sediment (5–69 mg kg^{-1} DW at 50 mg U kg^{-1} DW and 20–452 mg kg^{-1} DW at 500 mg U kg^{-1} DW).

All the derived CR values from sediment to biota other than Sr for fish are comparable to the broad ranges reported by IAEA (2010) for freshwater fish (0.0007–29.0 for Co, 0.003–2.90 for Ni, 0.09–40.0 for Se and 0.009–130 for Zn) and invertebrates (0.004–0.20 for Co, 0.03–0.19 for Mo, 0.002–1600 for Ni, 0.06–6.00 for Se, 0.002–0.62 for Sr, 0.003–0.06 for U and 0.001–23.0 for Zn). In the present study, Sr was the only element, which showed higher CR values (1.17–2.15) in the whole fish compared to values reported by IAEA (2010), (0.0002–0.95). The FW-based water-to-fish CR values for Sr (from 717 to 1313) and U (from 67 to 234) are in general, consistent with the available literature data (22–710 for Sr in freshwater fish; IAEA, 2010 and 3.8–48,000 for Sr and 0.6–760 for U in benthic feeding fish; IAEA, 2014) and the values for benthic fish, derived from ERICA Assessment Tool (version 1.2), (860 and 72 for Sr and U respectively). Therefore, the values recommended in models are in agreement with the CRs determined in this study. For Cl, on the other hand, the CR values are much lower than published data (0.24–0.58 in this study compared to 25–230 reported by IAEA, 2010 and 1250 from ERICA Tool). Regarding chironomids, water-to-chironomid CRs for U (255–945; FW-based CRs) are in the same range as reported by IAEA (2010) for water invertebrates, (3.6–60,000) and ERICA Tool for insect larvae (200), while much lower CR values were observed for Sr (7–16; FW-based CRs) compared to published data (77–1300; IAEA, 2010) and ERICA Tool for insect larvae (3700).

The CR values reported in the literature vary widely, and it is not surprising that the CR values observed in this study were mostly consistent with the very broad ranges reported. The wide variation of published CRs is largely due to differences between species (generic values, such as CR for “freshwater fish”, are often used in radioecological models)

Table 4
Geometric mean and geometric standard deviation (range for chironomids) of animal-to-sediment CR in two ponds near a former uranium mine (Pieni Hiislampi and Iso Hiislampi) and in a reference pond.

	Co	Mo	Ni	Se	Sr	U	Zn	Cl
Chironomids								
Pieni Hiislampi	0.017 (0.016–0.018)	0.16 (0.15–0.17)	0.025 (0.02–0.03)	0.69 (0.63–0.76)	0.02 (0.015–0.025)	0.011 (0.008–0.013)	1.05 (1.02–1.08)	n.a.
Iso Hiislampi	0.034 (0.032–0.036)	0.07 (0.06–0.08)	0.028 (0.02–0.035)	1.15 (1.08–1.21)	0.048 (0.045–0.051)	0.013 (0.008–0.02)	0.56 (0.46–0.68)	n.a.
Roach								
Pieni Hiislampi	0.004 (1.09)	0.02 (1.33)	< 0.007	0.59 (1.12)	2.15 (1.44)	0.006 (1.62)	1.91 (1.19)	8.24 (1.05)
Iso Hiislampi	0.002 (1.12)	0.003 (1.17)	< 0.002	0.29 (1.17)	2.12 (1.15)	0.001 (1.59)	0.87 (1.18)	11.1 (1.09)
P	0.33	<0.001		0.10	0.83	0.02	0.01	0.01
Perch								
Reference	< 0.11	< 0.02	0.01 (2.22)	0.61 (1.04) a	1.53 (1.11) a	< 0.004	1.08 (1.41) a	11.4 (1.07) a
Pieni Hiislampi	< 0.003	< 0.01	< 0.007	0.83 (1.14) a	1.17 (1.16) b	0.002 (1.41)	0.78 (1.22) b	9.77 (1.06) b
Iso Hiislampi	< 0.002	< 0.002	< 0.002	0.39 (1.21) b	1.31 (1.12) b	0.0006 (1.62)	0.45 (1.21) c	13.5 (1.03) c
P				0.01	0.01	* < 0.001	<0.001	<0.001

n.a. = data is not available. *t*-test was used for Pieni Hiislampi and Iso Hiislampi. **t*-test was used for U as there were two ponds. Different letters (a, b and c) indicate statistically significant difference in Tukey's pairwise test ($P < 0.05$).

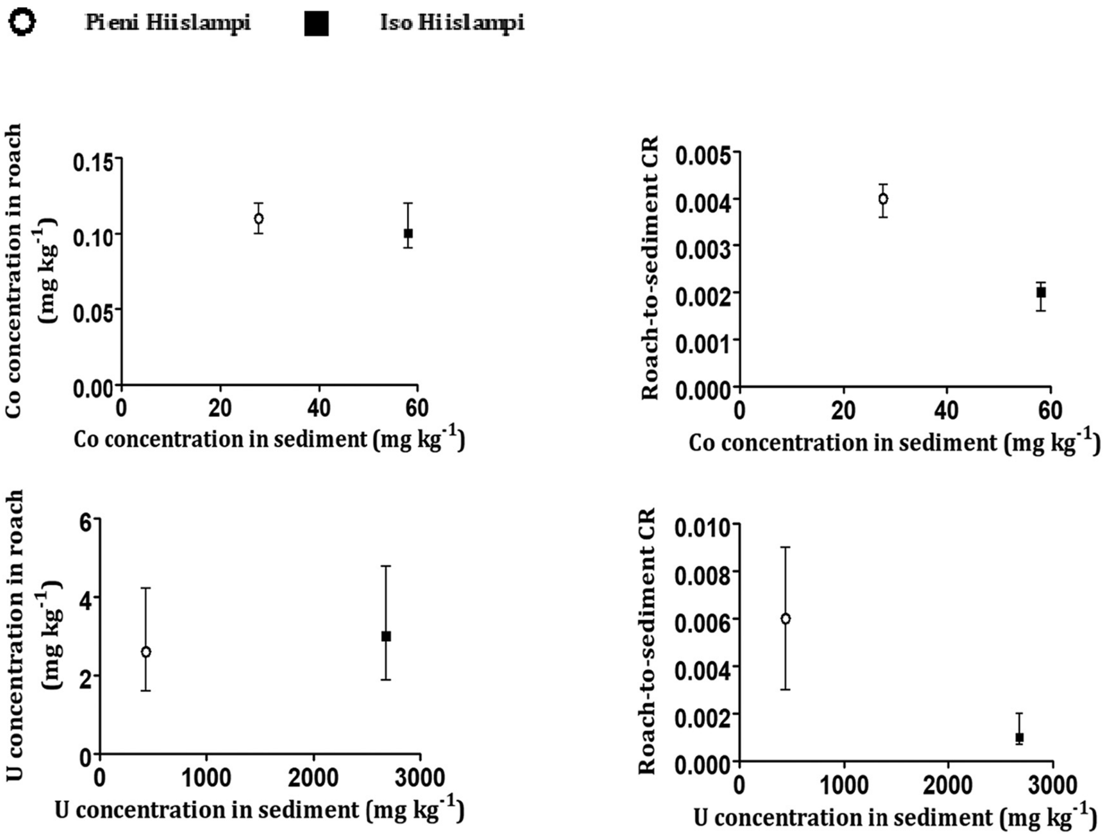

Fig. 2. Cobalt and uranium concentration in roach and roach-to-sediment CR values as a function of sediment concentration in Pieni Hiislampi and Iso Hiislampi. Geometric means are given, and error bars indicate geometric standard deviation.

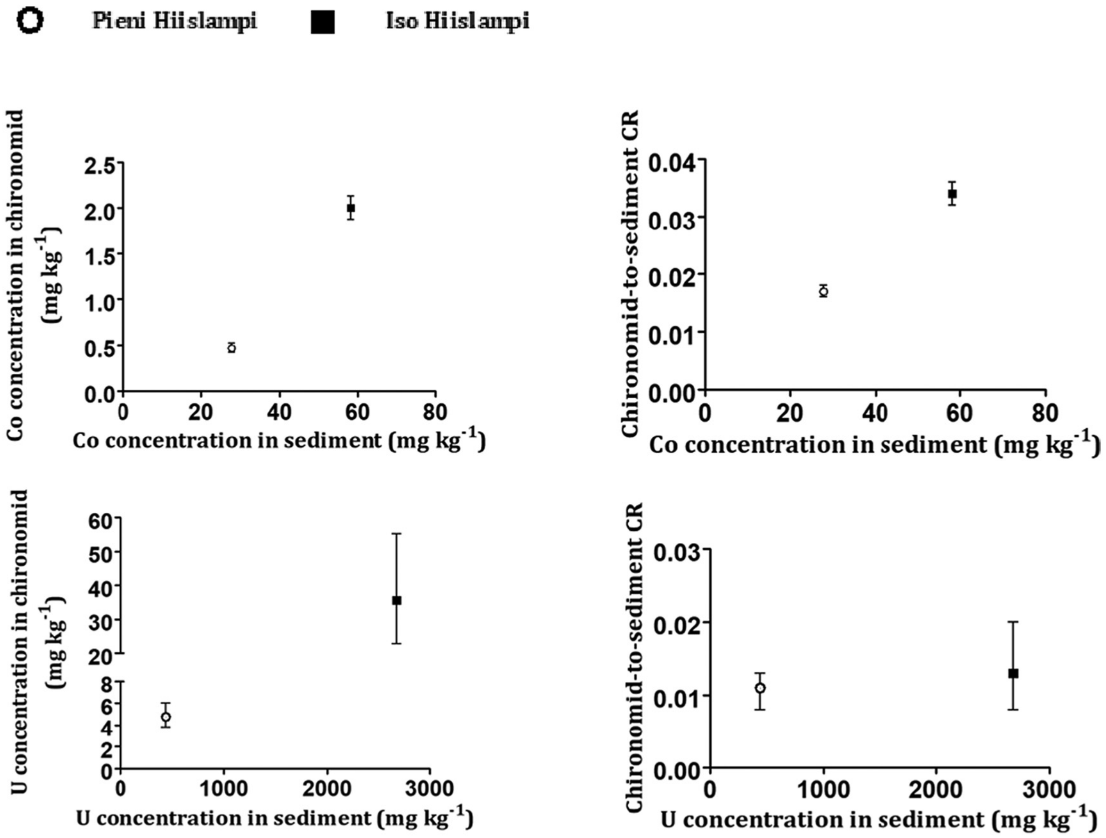

Fig. 3. Cobalt and uranium concentration in chironomids and chironomid-to-sediment CR values as a function of sediment concentration in Pieni Hiislampi and Iso Hiislampi. Geometric means are given, and error bars indicate min/max values.

and environments in which the measurements have been performed. More species- and ecosystem-specific studies are needed to reduce uncertainties in radioecological modeling. The results of the present study add significantly to the data available on boreal freshwater bodies.

In CR-based radioecological models, concentration in aquatic species is generally predicted from concentration in water (Hosseini et al., 2008; Copplestone et al., 2013; Howard et al., 2013; Brown et al., 2016; Hirth et al., 2017) and distribution coefficient (K_d), (IAEA, 2004; Kumblad and Bradshaw, 2008; Howard et al., 2013; Brown et al., 2016).The finding that element concentrations were much higher in sediment than in water (except for Cl), suggests that sediment might be an important source of elements taken up by the organisms studied. According to the classical view, the matter and energy of aquatic ecosystems are mostly derived from photosynthesizing organisms (e.g., phytoplankton), and are subsequently transferred to higher trophic levels in the food chain. However, it is increasingly evident that, in addition to this “green” food chain, an important role is played by the “brown” food chain, based on dissolved and particulate organic matter. An important part of the brown food web may be methane oxidizing bacteria, which incorporate biogenic methane and serve as food for zooplankton and macro invertebrates, thereby supporting the whole aquatic ecosystem (Jones and Grey, 2004; Sanseverino et al., 2012). The brown food web is likely to be of particularly great importance in humus-rich boreal lakes and ponds with a thick layer of organic sediment, such as the ponds that were sampled in this study. In studies that have considered both food ingestion and direct uptake from water, sediment ingestion has been identified as the primary source of metals to deposit-feeding invertebrates (Wang and Fisher, 1999; Rowan et al., 2014). This is highly relevant to the food chain addressed in the present study, comprising sediment-feeding invertebrates and small fish feeding on them. Several studies have shown importance of the transfer of contaminants from the bottom sediments to benthic organisms through the food web also in marine environment (Shigenobu et al., 2015; Bezhenar et al., 2016; Wang et al., 2016). Further research is needed in different types of water bodies for better understanding of the relative contributions of water and sediment.

Aquatic and terrestrial ecosystems are interconnected, and a considerable fraction of substances (including radionuclides) in aquatic sediment can originate from the surrounding terrestrial environment (Reynolds, 2008; Soininen et al., 2015; Evans-White and Halvorson, 2017). Terrestrial origin of the organic matter in our study ponds is supported by the high C/N ratios of the sediments. Organic matter from terrestrial sources (from vascular plants) tends to have C/N ratios of 20 or higher, while C/N ratios between 4 and 10 indicate aquatic (algal) origin (Emerson and Hedges, 1988; Meyers, 1994). The C/N ratios in Iso Hiislampi and Pieni Hiislampi were well above 20, while the reference pond had a C/N ratio of 15, indicating significant contribution from terrestrial sources even in the reference pond.

The element concentrations observed in fish were approximately constant and did not increase with increasing concentration in sediment. Consequently, the CRs (for most elements) decreased with increasing concentration in sediment. This observation could indicate either that uptake form sediment is nonlinear or that water rather than sediment drives the uptake into fish. Examining the relationship between water and fish concentrations was possible only for U and Cl, as Sr concentrations in water were similar in the ponds, and concentrations of the other elements in water were below the detection limit. The U and Cl data were more consistent with nonlinear than with linear transfer also when water was considered as the medium (approximately constant concentration in fish, decrease of CR with increasing concentration in water). Thus, the data do not clearly support the assumption that transfer of elements into fish is linear (can be described with a constant CR), being more consistent with previous results suggesting non-constant CRs in terrestrial plants and animals (Tuovinen et al., 2011, 2016b). The inadequacy of approaches based on concentration ratios has also been shown in studies on the uptake of environmental metals into aquatic biota (McGeer et al., 2003). In contrast to the fish data, concentrations of most elements in chironomids showed the expected increase as a function of concentration in sediment; this was not true for Sr and Zn, but the small differences in sediment concentrations (less than 2-fold) limit the conclusions that can be drawn concerning these two elements. Consistently with linear transfer of elements into chironomids, the CRs were approximately constant (no substantial differences between the ponds). The results are also consistent with the data reported by Crawford and Liber (2016), showing increase of U concentration in *Chironomus dilutus* by increase of U concentration in spiked sediments. In spite of the limitations of the data (low number of samples), it is of interest to speculate possible reasons for the difference between chironomids and fish. The approximately constant element concentrations (non-constant CRs) in fish can be assumed to result from physiological mechanisms that optimize the concentrations of essential elements and keep concentration of toxic elements at a sufficiently low level. As sediment-dwelling and sediment-ingesting organisms, chironomids may have developed mechanisms to tolerate high tissue concentrations, and there is thus less need to control uptake of potentially harmful elements.

The limitations of this study included the fact that no chironomid larvae or roach were found in the reference pond. Perch samples were caught from all ponds, but the concentrations of many elements were unfortunately below the detection limits. Furthermore, most of the element concentrations in water were below the detection limits. However, useful data were obtained for evaluating transfer of several elements (Cl, Co, Mo, Ni, Se, Sr, U, Zn) in a boreal food web for which few previous results are available. Moreover, the concentration gradients between the ponds allowed for the evaluation of the assumption that the transfer of elements into two fish species and chironomids can be described using a constant CR. Another limitation of this study was that possible seasonal variation was not studied. Uptake of elements in aquatic organisms is very complicated and attributed to several parameters (e.g. water salinity, pH, oxygen level and temperature), and, e.g., oxygen concentration and temperature show marked seasonal variation in Finnish conditions (Mäkinen and Lerssi, 2007). This aspect together with sediment characteristics, water physicochemical properties and element speciation should be considered in future studies. Furthermore, data would be needed also on organisms that do not feed on sediment (or on deposit-feeding animals), as the radionuclide concentrations of such organisms are likely to be related to concentrations in water rather than concentrations in sediment (Rowan et al., 2014). In order to validate the findings of this study, it would be helpful to include measurements of radionuclides in future studies, as several studies have suggested that radioactive isotopes may behave differently than stable isotopes of the same element (Wood et al., 2013; Brown et al., 2019; Beresford et al., 2020).

5. Conclusion

The findings of this study indicate that sediment may be more important source for uptake of many elements into aquatic animals in boreal lakes and ponds rich in organic matter. This should be taken into account in radioecological models, which conventionally predict concentration in aquatic organisms from concentration in water. The results also show that the assumption of linear transfer (constant CR value) may not be valid for uptake of all elements into fish but appears to be more suitable for predicting uptake into chironomids. Further basic research is needed to understand the fundamental processes involved in transfer of elements into freshwater organisms in order to develop radioecological models.

CRediT authorship contribution statement

Soroush Majlesi: Conceptualization, Methodology, Investigation, Formal analysis, Writing-Original Draft

Jarkko Akkanen: Conceptualization, Methodology, Validation, Writing, Reviewing and Editing, Supervision

Päivi Roivainen: Methodology, Validation, Writing, Reviewing and Editing, Supervision

Tiina S. Tuovinen: Methodology, Investigation, Data Curation

Jouni Sorvari: Methodology, Investigation, Validation, Writing, Reviewing and Editing

Jonne Naarala: Writing, Reviewing and Editing

Jukka Juutilainen: Conceptualization, Methodology, Validation, formal analysis, Writing, Reviewing and Editing, Supervision, Project administration

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.2021.148218.

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