DETERMINATION OF CONCENTRATION RATIOS OF STABLE SR AND CS FOR FISH, AND ANALYSIS OF FACTORS INFLUENCING THEIR RANGE OF VARIATION

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Water chemistry was found to influence uptake of contaminants by aquatic organisms. Empirical inverse relationships were derived between CRs of fish for stable Sr (CR^{Sr}) and Cs (CR^{Cs}) and water concentrations of their biochemical analogues Ca and K, respectively, illustrating how such relationships could be used in the prediction of more site-specific CR^{Cs} and CR^{Sr} in fish simply from water chemistry measurements. The determination of (CR) for fish is important, because it is used to define concentration of radionuclide in biota and posterior estimation of radiation dose to organism.

I. INTRODUCTION

The radioisotopes Cs-137 and Sr-90 are two of the most important radionuclide contaminants in the aquatic environment, originating mainly from nuclear accidents (e.g. Chernobyl, 1986 and Fukushima, 2011) and in fallout from nuclear weapons tests, and present a significant radiological risk (Ref. 1). The biota-to-water concentration ratio (CR) is usually used to define the concentration of radionuclides in biota and subsequent estimation of radiation dose to organisms. For aquatic organisms it is usually defined as the ratio between the radionuclide concentration in the whole organism and the concentration in the surrounding water.
For fish:

CR_{fish} (L/kg ww) = C_{fish}/C_w= (kg/kg ww)/(kg/L), where C_{fish} is the radionuclide concentration per unit mass of muscle tissue and C_w is the dissolved radionuclide concentration in water (kg/L). The CR definition assumes that the radionuclide in the organism is in equilibrium with its surrounding media, though in reality concentrations of radionuclides in the environment and in organisms usually vary in a dynamic way. Risk assessors often have access to only limited measurements from their site of interest and often need to use literature data or extrapolation methods to obtain CRs. Despite the large number of Cs and Sr studies, temporal variation is rarely measured or taken into account, so its importance as a source of variability in CR estimates and for risk assessment is poorly understood.

Stable elements are increasingly used for the prediction of radionuclide transfer in environmental safety assessment models (Refs. 2, 3 and 4) since the behaviour of stable elements and their corresponding radionuclides are assumed to be similar in the environment if they are in the same physico-chemical forms. Uptake to organisms, including potential biomagnification is also assumed to be similar. However, these assumptions have rarely been tested.

Another way to calculate site-specific Cs-137 and Sr-90 CRs in fish more precisely is using empirical equations together with measurements of K and Ca concentrations in water. Calcium and K are essential biological macroelements and behave similarly in biological tissues to Sr and Cs, respectively. Thus, in cells Cs follows the K pathway and Sr follows the Ca pathway (Refs. 5, 6 and 7), the pairs of ions competing for uptake sites on biological membranes (Ref. 8).

This study used a previously unanalysed dataset for stable Sr and Cs in order to better understand the behaviour of these elements and their radioisotopes in freshwater and brackish water ecosystems, with the overall aim of improving the use of such data in radioecological risk assessments.

II. MATERIALS AND METHODS

II.A. Study Sites

Data were obtained from four lakes (Bolundsfjärden, Eckarfjärden, Fiskarfjärden and Frisksjön) and three coastal areas in the central-east and south-east part of Sweden. The sites have been intensively studied as part of the investigations and risk assessments by the Swedish Nuclear Fuel and Waste Management Co. (SKB) at Forsmark and Laxemar-Simpevarp area during the selection of a site for a future radioactive waste repository for spent nuclear fuel (Ref. 9).

II.B. Water And Fish Samples

Water and fish samples were collected during 2002-2008 at 7 sites. The data was extracted from the SKB 'SICADA' database and subsequent SKB reports (Refs. 10, 11, 12 and 13). For each site, the fish species were categorized as piscivorous or non-piscivorous, depending on the available fish species at each site. Three fish species were considered as piscivorous: pike, perch and smelt and 7 as non-piscivorous: roach, tench, ruffe, bleak and herring, bream and flounder. For each of the seven investigated sites the arithmetical mean (AM), standard deviation (SD), minimum and maximum observed concentrations of Ca, K, Sr, Cs and values pH and ectrical conductivity (EC) of the water were calculated. Electrical conductivity (EC) is related to the Ca and K concentrations in water, as it estimates the amount of total dissolved salts, which in lake water are mainly Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^- $SO4^-$ (Ref. 14).

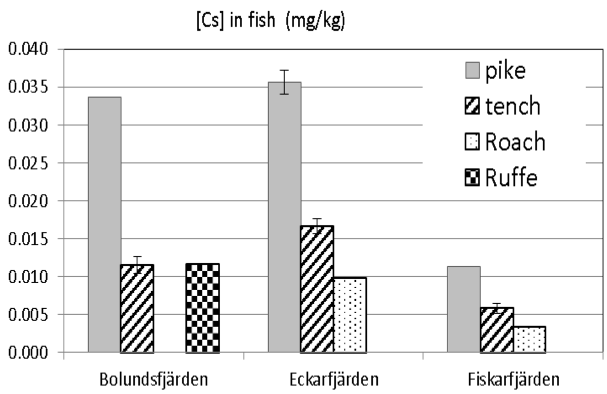

Fig. 1. The arithmetical mean (AM) and standard deviation (SD) of the Cs concentration in four different fish species (pike, tench, roach, ruffe) in the three lakes Bolundfsfjärden, Eckarfjärden and Fiskarfjärden of the Forsmark area, Sweden.

III. RESULTS

III.A Concentration Ratios Of Fish

The fish-water concentration ratios, CR_{fish}, were calculated from fish and water element concentration data that were from the same site and preferably from the same year. Where data of fish and water concentrations from the same year were not available, an average water concentration, with standard deviation (SD), were calculated from data within 1-4 years of the date when the fish was caught. CRs for piscivorous and non-piscivorous fish were calculated separately for each site, and their averages were statistically compared. For each of the seven investigated sites and every type of fish the AM and SD of CR^{Sr} and CR^{Cs} were calculated and presented in Table I. The average Cs concentration in pike was a factor of 3 higher than in roach and approximately factor of 2 higher than in ruffe and tench samples from three study lakes of Forsmark (Fig. 1).

III.B Biomagnification Factor (BMF)

In order to determine if there was biomagnification of Cs or Sr in piscivorous fish, the biomagnification factor (BMF) for each element was calculated for each site as the ratio of the element concentration (μg/kg ww) in muscles of piscivorous fish to the element concentration (μg/kg ww) in muscles of non-piscivorous fish that were considered as a potential food source at each site. Of the species for which data was available, pike was assumed to eat ruffe, roach and tench, and perch assumed to eat roach, bleak and herring. There was no consistent pattern for Sr BMFs (Table I). Only in Bolundsfjärden was there any indication of biomagnification of Sr (BMF for Sr between pike and ruffe of 4.5), but this estimate must be used with caution since it is based on limited data. For Cs, BMFs were in the range 2 – 5.6. For coastal fish, there was no biomagnification for Sr (BMF less than 1) and Cs BMFs of up to 3 were estimated.

III.C. Concentration Ratio And The Effects Of Water Chemistry

III.C.1. CR^{Sr} of fish vs. [Ca] water

There was an inverse relationship between the mean values of CR^{Sr} (all fish species and all sites considered) and the mean Ca concentrations in water (mg/L) (Fig. 2a). The best fit relationship was the exponential function

$$CR([Ca]) = A_2 e^{b_2 \cdot [Ca]}, \qquad (1)$$

ln-transformed to:

$$\ln(CR^{Sr}) = a_2 - b_2[Ca]. \qquad (2)$$

Table II shows the values of the coefficients a_2 and b_2, derived from the linear regression analysis, and the coefficient A_2 and correlation coefficient r.

III.C.2. CR^{Cs} of fish vs. [K] water

There was an inverse relationship between the mean values of CR^{Cs} (all fish species and all sites considered) and the mean K concentrations in water (mg/L) (Fig. 2b). The best fit relationship was the power function:

$$CR^{Cs} = A_1[K]^{b_1}, \qquad (3)$$

log-transformed to:

$$\log(CR^{Cs}) = a_1 - b_1 \cdot \log([K]). \qquad (4)$$

TABLE I. The Cs and Sr concentration in fish muscle (µg/kgww), biomagnification factor (BMF) and concentration ratio (CR) for fish muscle (l/kg ww). N - number of fish samples, AM - arithmetical mean, SV - single value, SD - standard deviation. Piscivorous fish are marked in bold font.

Site	Common name	N	Cs	Sr	Ref	CR Cs (l/kg ww)		CR Sr (l/kg ww)	
			BMF	BMF		AM/SV	SD	AM/SV	SD
Lake									
Bolundsfjärden	**Pike**	1			A	4390.4	231.4	2.41	1.0
	Ruffe	6*	2.88	4.54	A	1523.5	80.3	0.53	0.2
	Tench	3	2.91	1.19	A	1509.1	167.7	2.02	0.9
Eckarfjärden	**Pike**	2			A	4967.8	741.6	4.94	1.1
	Roach	18*	3.60	1.74	A	1378.1	196.4	2.84	0.4
	Tench	3	2.14	0.98	A	2325.2	357.2	5.05	3.9
Fiskarfjärden	**Pike**	5*			A	759.78		3.64	1.0
	Roach	16*	3.33	0.40	A	228.2		9.09	2.6
	Tench	3	1.95	1.87	A	388.98	45.9	1.95	1.6
Frisksjön	**Perch**	3			B	6474.1	990.6	8.96	8.4
	Roach	3	5.57	0.66	B	1162.7	275.1	13.60	1.6
Marine									
Simpevarp area -Borholmsfjärden Bay	Bleak	3	2.47	0.03	B	44	34.4	2.61	1.2
	Bream	2			B	107.48	112.2	1.39	0.4
	Perch	3			B	108.82	87.1	0.08	0.0
Simpevarp coast	Flounder	3			B			0.18	0.1
	Herring	3	3.03	0.32	B			0.21	0.1
Forsmark coast	Ruffe**	3			C	210.36	102.8		
	Roach**	3			C	119.4	52.9		
	Smelt**	3			C	183.47	81.6		

A - (Hannu and Karlsson, 2006); B - (Engdahl, 2006); C - (Kumblad and Bradshaw, 2008)
* in order to gain enough sample matter the individuals were pooled together as one sample
** concentration of whole fish were analysed, while all other samples of fish muscle were analysed

Table III shows the values of the coefficients a_1 and b_1, derived from the linear regression analysis for set of 17 CR^{Cs} values for non-piscivorous and piscivorous fish.

III.C.3. Dependence of CR^{Sr} and CR^{Cs} on EC and pH

In this study, a strong correlation was determined between EC and [K] (r=0.87) and between EC and [Ca] (r=0.99). There was an inverse relationship between CR^{Sr} and EC (all fish at all sites considered; Fig. 3, a) as well as between CR^{Cs} and EC (Fig. 3, b). The best fit equation was the power function

$$CR^{Cs\ or\ Sr} = A_1 \cdot EC^{b_1}, \quad (5)$$

expressed linearly after log-transformation as:

$$\log(CR^{Cs\ or\ Sr}) = a_1 - b_1 \cdot \log(EC). \quad (6)$$

Table IV shows the values of the coefficients a_1, b_1 and A_1, derived from the linear regression analysis. The inverse correlation of CR^{Sr} with pH was moderate (r=-0.6) and for CR^{Cs} with pH it was low (r=-0.2).

IV. CONCLUSIONS

Seasonal variation in concentrations of Sr, Ca, Cs, K in water is large and CRs based on single measurements of water concentrations may therefore lead to erroneous estimation of CR values. The determined empirical inverse relationships between CR^{Sr} for fish and [Ca] in water, as well between CR^{Cs} for fish and [K] in water can be used for the estimation of site-specific CRs based on measured Ca and K water concentrations. EC of water substituted in the derived empirical equations can also be used to estimate site-specific CR^{Sr} and CR^{Cs} for fish where data of Ca and K water concentrations are lacking. Piscivorous lake fish biomagnify Cs but not Sr.

The CR for fish defined for stable Sr and Cs can be used as proxies for their radioisotopes.

TABLE II. The lower confidence limit (LCL) and upper confidence limit (UCL) of 95% CIs and means of the coefficients a_2, b_2 and A_2, and correlation coefficients r, derived from the linear regression analysis for CR^{Sr} for all fish types and [Ca] in water.

	a_2			b_2			A_2			r
	LCL	mean	UCL	LCL	mean	UCL	LCL	mean	UCL	
Sr	1.87	3.05	4.2	-0.065	-0.045	-0.025	6.5	21	68	-0.8

TABLE III. The lower confidence limit (LCL) and upper confidence limit (UCL) of 95% CIs and means of the coefficients a_1, b_1 and A_1, and correlation coefficients r, derived from the linear regression analysis for CR^{Cs} for all fish types and [K] in water.

	a_1			b_1			A_1			r
	LCL	mean	UCL	LCL	mean	UCL	LCL	mean	UCL	
Cs	3.27	3.58	3.89	-1.17	-0.89	-0.61	1881	3839	7838	-0.87

TABLE IV. The lower confidence limit (LCL) and upper confidence limit (UCL) of 95% CIs and means of coefficients a_1, b_1 and A_1, and correlation coefficients r, derived from the linear regression analysis for CR^{Cs} or CR^{Sr} and EC for all fish types combined.

	a_1			b_1			A_1			r
	LCL	mean	UCL	LCL	mean	UCL	LCL	mean	UCL	
Sr	0.99	1.6	2.3	-0.99	-0.69	-0.4	10	43	189	-0.79
Cs	3.7	4.3	4.9	-1	-0.8	-0.5	5270	20915	82988	-0.83

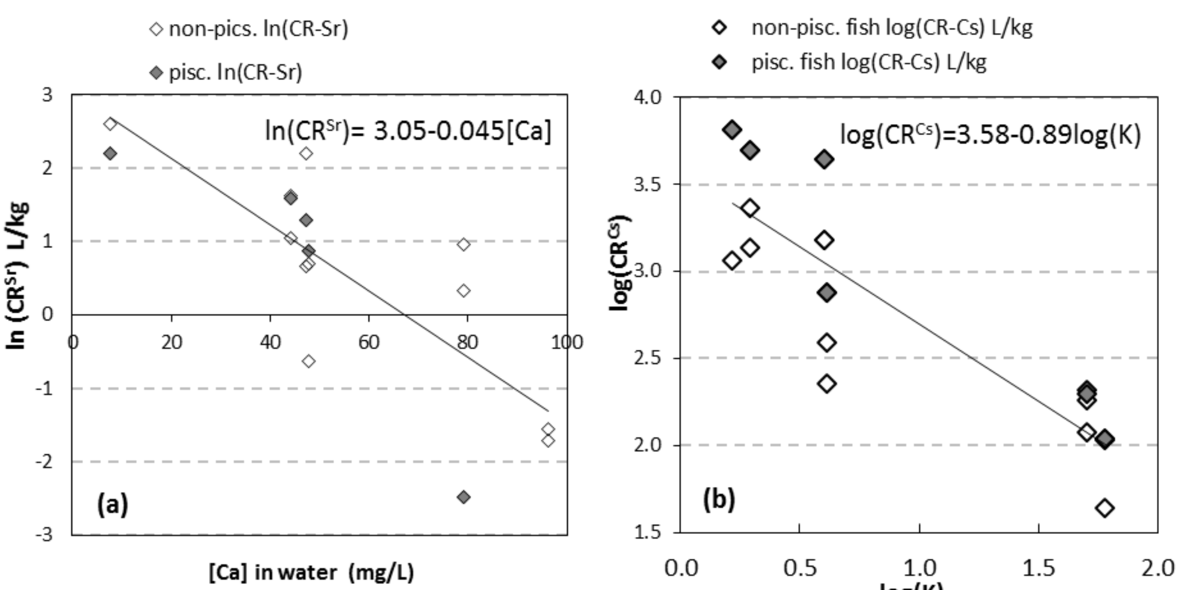

Fig. 2. Arithmetical mean (AM) of CR^{Sr} (a) and CR^{Cs} (b) for piscivorous and non-piscivorous fish observed in this study versus AM of K concentration in the water samples The solid line represents the statistical approximation of observed data for both piscivorous and non-piscivorous fish.

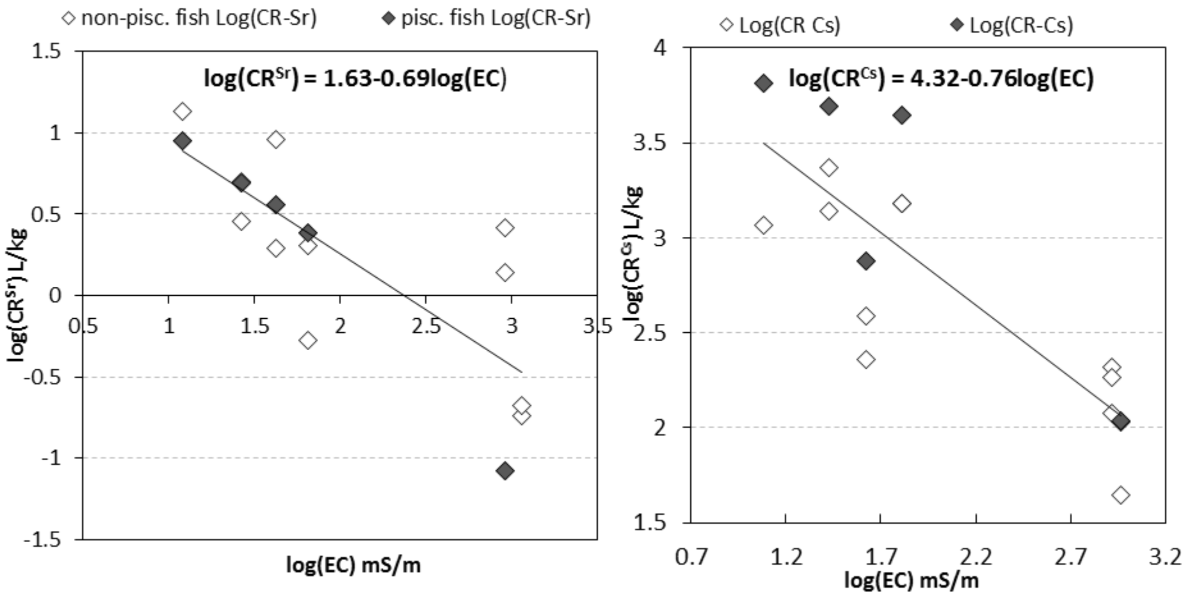

Fig. 3. Relations between water EC and CR in piscivorous and non-piscivorous fish muscle for Sr (a) and Cs (b).

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