

Some Considerations on Water-to-Fish Transfer Data Collected in Japan for Radionuclides and Stable Elements -11252

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ABSTRACT

The water-to-fish concentration factor (CF) is defined as the ratio of concentration of a radionuclide in the fish part (Bq/kg-fresh) to that in water (Bq/L, unless otherwise specified). CFs have been used in mathematical models for environmental safety assessment of radioactive waste disposal. In the present paper, published global fallout Cs-137 and Sr-90 concentration data for fish living in freshwater or seawater areas in Japan were collected to calculate CFs. Among the collected data, suitable data to obtain CF values for Cs-137 (CF-Cs) and Sr-90 (CF-Sr) were identified. They included Cs data for three species of freshwater fish, and the CF-Cs in muscle for the three species ranged from 31 to 855. Suitable data were identified for 14 seawater fish species, and the CF-Cs in muscle for 12 of them ranged from 15 to 481. For Sr-90, no data sets were available for any freshwater fish. Data were identified for eight species of seawater fish, and the CF-Sr in muscle for seven of them ranged from 0.4 to 7.9. Geometric mean values of CF-Cs and CF-Sr were almost the same among the seawater fish species. Stable element data were also collected to check whether or not the same group elements had similar CF values. The CFs for the Cs and K pair and the Sr and Ca pair were close, but a statistical difference was observed. Thus it is difficult to use data of K and Ca for calculating CF-Cs or CF-Sr. When CF-Cs and CF-stable Cs (calculated from literature values) were compared, no statistical difference was observed possibly because physico-chemical forms of stable Cs and Cs-137 are almost the same. Thus stable Cs can be used as an analogue of Cs-137 for CF determination. The applicability hypothesis is also valid for other radionuclide-stable element combinations where they are in physico-chemical equilibrium.

INTRODUCTION

In radiation dose assessment for humans from radioactive nuclear waste disposal sites, food consumption is one of the important pathways; thus, it is necessary to know the transfer of radionuclides from environmental media to food materials. Recently, eating habits are changing and consumption of seafood is increasing worldwide so that more data on water-to-fish concentration factors (CFs) are needed. This CF (L/kg, unless otherwise specified) value is defined as the ratio of concentration of a radionuclide in fish part (Bq/kg-fresh) to that in water (Bq/L). The International Atomic Energy Agency (IAEA) recently published the Technical Report Series No. 472 (TRS-472) [1] to update parameter values from the previous version TRS-364 [2]. The update included addition of freshwater fish data. For seawater fish, data compiled in the IAEA's TECDOC-211 [3] values were revised in its TRS-422 [4]. Some of the fish concentration factors compiled in these four reports are listed in Table I.

In these reports, fish species and also CF ranges were not listed. According to TRS-422, because the range of minimum and maximum CFs was one order of magnitude (or less) from the recommended values in a reliable database, the CF ranges were not included in its tables. However TRS-422 also noted in footnotes the few elements for which reliable data indicated that greater variability was apparent for a given CF.

Although the CF ranges are narrow, it is desirable to know whether or not there are differences among fish species to give better data to the people who do radiation dose assessment. It is also useful to compare the data obtained worldwide with domestic Japan data to identify area differences. In this report, therefore,

radionuclide transfer data for fish living in freshwater or seawater areas in Japan were surveyed and those data were compared with data of related stable elements, such as potassium for radiocesium.

Table I. Recommended concentration factors for edible portions of freshwater and seawater fish (L/kg).

Element	Freshwater fish		Seawater fish	
	TRS-364[2]	TRS-472[1]	TECDOC-211[4]	TRS-422[3]
Na	20	76	0.1	1
K	-	3200	-	-
Ca	-	12	1	2
Mn	400	240	500	1000
Fe	200	170	1000	30000
Cu	200	230	-	-
Zn	1000	3400	2000	1000
Sr	60	2.9	1	3
Cs	2000	2500	50	100

Note: '-' denotes no data.

DATA ANALYSIS

Extensive global fallout concentration data in fish and waters of Japan are available in environmental radioactivity survey reports and studies [5]. From [5], about 100 reports written from 1960-1980 were surveyed for the present paper. Among their data, suitable data to obtain CF values for Cs-137 (CF-Cs) and Sr-90 (CF-Sr) were found. Radioactivity concentration data in fish tissues (muscle, internal organ, or whole body, in fresh weight) were divided by the concentration data in water collected from almost the same place and the year of the fish sampling to obtain CFs.

The distribution pattern of calculated CF values for each fish species was a log-normal one so that the geometric mean (GM) could be calculated if the number of samples was more than three. Some results are shown in Fig.1. Arithmetic mean (AM) was also provided if the number of samples was more than two. Single value (SV) was used if there was only one available datum for the fish.

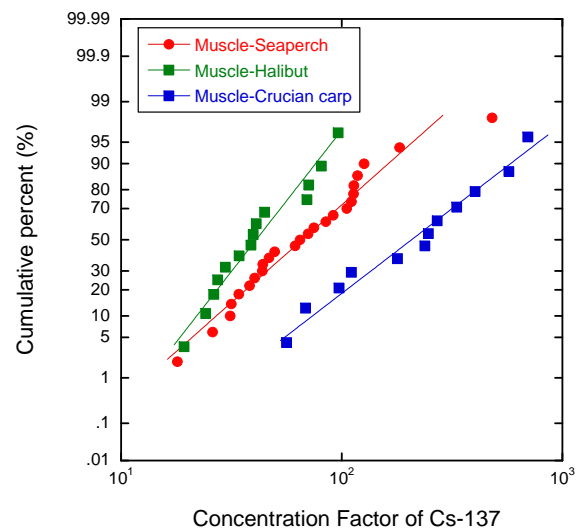


Fig.1. Probability distributions of Cs-137 concentration factors for three fish species collected in Japan.

RESULTS AND DISCUSSION

Concentration Factors for Freshwater Fish

After surveying the reports selected from [5], CF-Cs for three freshwater fish species were obtained: carp (wild and cultured), crucian carp (wild), and Japanese bitterling (wild). The CFs obtained are listed in Table II. For muscle, CF-Cs values for all three species ranged from 31 to 840. The GM value for wild carp was 241 (n=5) and that for cultivated carp was 237 (n=5) so that there was no clear difference between wild and cultivated carp. For crucian carp and Japanese bitterling, the GM values were 207 (n=12) and 78 (n=3), respectively. Because bitterling have a shorter lifetime than carp and crucian carp, the bitterling CF values were likely to be affected. Data comparison of CF-Cs for muscle and internal organ is made in Fig.2, and there was a high correlation by the t-test ($R=0.88$, $p<0.001$). The slope of the best fit line was almost 1 and no statistical difference was observed. Thus, Cs-137 distributed uniformly in muscle and internal organ for freshwater fish. The GM of CF-Cs for freshwater fish (edible part) compiled in TRS-472 [1] was 2500 (range: 140-15000), which was one order of magnitude larger than the GM of CF-Cs obtained from the Japanese data reviewed.

Table II. Concentration factors of Cs-137 for freshwater fish tissues (L/kg).

Common name	Latin name	Collection type	Tissue type	N ^{*1}	GM ^{*2}	AM/SV ^{*3}	Min.	Max.
Crucian carp	<i>Carassius langsdorfi</i>	Field	Muscle	12	207	272	56	697
Crucian carp	<i>Carassius langsdorfi</i>	Field	Internal organ	12	252	308	91	887
Carp	<i>Cyprinus carpio</i>	Field	Muscle	5	241	378	31	840
Carp	<i>Cyprinus carpio</i>	Field	Internal organ	5	255	472	19	943
Carp	<i>Cyprinus carpio</i>	Cultured	Muscle	5	237	317	96	855
Carp	<i>Cyprinus carpio</i>	Cultured	Internal organ	5	217	262	103	610
Carp	<i>Cyprinus carpio</i>	Cultured	Whole	1		331		
Japanese bitterling	<i>Acheilognathus melanogaster</i>	Field	Muscle	3	78	83	59	125
Japanese bitterling	<i>Acheilognathus melanogaster</i>	Field	Internal organ	3	29	38	11	69

*1: Numbers of CFs calculated, *2: Geometric mean, and *3: Arithmetic mean or single value.

The CF-Cs data obtained in this study were compared with CF of potassium (CF-K) because Cs and K are both alkali metals. The CF-K values as well as CFs of other stable elements were calculated for carp and crucian carp using stable element concentration in edible part [6] and GM data from a survey of Japanese rivers water samples [7]. The CFs obtained are shown in Table III. The CF-K values for both carp and crucian carp were 2860 that were close to the recommended CFs-K 3200 (range: 570-15000) [1]. On the other hand, compared to K, all CF-Cs values were smaller than recommended CF, possibly because K is selectively taken up by freshwater fish.

When CFs of other stable elements, i.e. Na, K, Ca, Mg, Mn, Fe, Cu and Zn, for muscle and internal organ of cultured carp were compared, differences of five times or more were found for Mn, Fe, Cu, and Zn, although the differences were small (less than 2-fold) for Na, K, Mg, and Ca. These data in Table III were compared with those listed in TRS-472. The tendency was almost the same; however, CFs of Ca, Mg, Fe, and Cu were higher than the recommended values. The reasons were not clear, but usually essential element concentrations in freshwater fish organs are almost the same [6], thus other factors in Japanese rivers such as elemental concentration, pH, and electrical conductivity, must have affected the CF of the stable elements. The fate of other elements and radionuclides is also likely to be affected by these factors so that it is necessary to collect other element concentration data in fish for comparison.

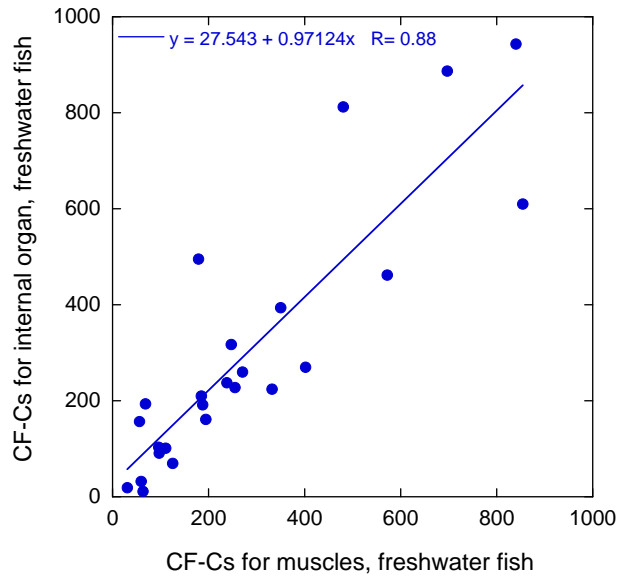


Fig.2. Correlation between CF-Cs for muscles and internal organ samples in freshwater fish.

Table III. Concentration factors of eight stable elements for freshwater fish tissues (L/kg).

Common name	Latin name	Collection type	Tissue type	Na	K	Ca	Mg	Mn	Fe	Cu	Zn
Crucian carp	<i>Carassius langsdorfi</i>	Field	Muscle	59	2860	100	120	42	612	727	4750
Carp	<i>Cyprinus carpio</i>	Cultured	Muscle	96	2860	9	115	21	204	910	3000
Carp	<i>Cyprinus carpio</i>	Cultured	Internal organ	187	2020	9	99	208	1270	5640	17500

Concentration Factors for Seawater Fish

For seawater fish, CF-Cs values could be calculated for 14 fish species using data found in the reports selected from [5] and they are given in Table IV. There are three tissue types, muscle (edible part), internal organ and whole, and some fish species have CF-Cs values for all three. The numbers of calculated CF data

ranged from 1 to 20 for each species or tissue types. All CF-Cs values in muscle ranged from 15 to 481. The GMs of CF-Cs of nine species were in the narrow range of 27-66, which was lower than for freshwater fish and almost the same as the recommended value in TRS-422 for seawater fish. Because the collected CF-Cs values for nine fish species were almost the same, one recommended CF-Cs for seawater fish might be acceptable. A high correlation between CF-Cs and fish weight has been reported [8]; however, according to this report, CF-Cs values were still in a narrow range of about 10-180 for 10-10000 g weight fish and the change of CF-Cs with the weight was small. There might be a fish weight effect on CF-Cs, but it should not be significant.

For internal organ samples, the CF-Cs values ranged from 22 to 404, and the GMs of the CF-Cs for five species ranged from 35 to 93, which was also a narrow range. There was no significant difference between CF-Cs values of muscle and internal organ as was found for freshwater fish. Although freshwater fish had a high correlation between CF-Cs values of muscle and internal organ, only a weak correlation ($R=0.58$, $p<0.001$) was found as shown in Fig.3. It is probable that Cs behavior in a seawater fish body is different from that in a freshwater fish body. The ANOVA test of GMs of CF-Cs values for muscle, internal organ and whole for each fish species did not indicate any statistical difference, although there were slightly lower values for the whole compared to the other two organs, possibly because Cs-137 concentrations in other parts, such as bone, fin, and skin are lower than those in muscle and internal organ [9].

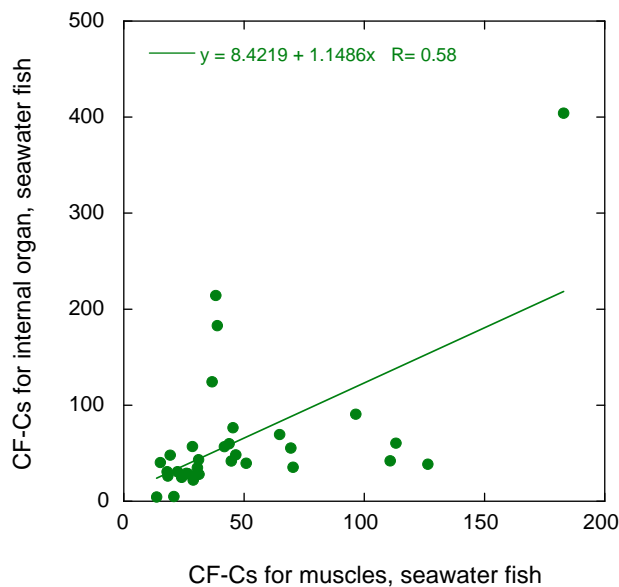


Fig.3. Correlation between CF-Cs for muscles and internal organ samples in seawater fish.

Then CF-K values in muscle samples were compared with CF-Cs values. From the survey of [5], potassium concentration in seawater was 0.4g/L on average and that in muscle of sea fish was 0.4-6.4 g/kg with GM of 3.0 g/kg ($n=20$) so that the CF-K for seawater fish ranged from 1 to 16.1 with GM of 7.5. The CF-Cs values for the same samples ranged from 21-64 with GM of 34, and a significant difference was observed (t-test, $p<0.001$). There was no correlation between TF-K and TF-Cs for fish species so that K and Cs may be behaving differently in a seawater fish body. Compared to freshwater fish, CF-K values were lower in seawater fish. This was possibly due to the difference of metabolism between freshwater and seawater fish which could affect CF values.

Table IV. Concentration factors of Cs-137 for sea fish tissues (L/kg).

Common name	Latin name	Tissue type	N ^{*1}	GM ^{*2}	AM/SV ^{*3}	Min.	Max.
Horse mackerel	<i>Trachurus japonicus</i>	Muscle	3	40	42	29	51
		Internal organ	3	56	58	40	77
		Whole	7	33	36	14	55
Mackerel	<i>Scomber japonicus</i>	Muscle	2		52	47	58
		Internal organ	1		48		
		Whole	2		19	4	33
Brown sole	<i>Pleuronectes herzensteini</i>	Muscle	9	48	50	30	87
		Internal organ	1		27		
		Whole	10	33	35	15	54
Japanese Seaperch	<i>Lateolabrax japonicus</i>	Muscle	25	66	88	18	481
		Internal organ	10	58	94	26	404
		Whole	10	27	32	4	52
Black rockfish	<i>Sebastes ventricosus</i>	Muscle	6	27	29	15	45
		Internal organ	6	35	36	22	43
		Whole	7	40	40	30	48
Fat geenling	<i>Hexagrammos otakii</i>	Muscle	2		41	30	53
		Whole	1		32		
Yellow tail	<i>Seriola quinqueradiata</i>	Muscle	8	42	46	21	69
		Internal organ	3	43	46	25	57
		Whole	3	32	35	17	46
Japanese whiting	<i>Sillago japonica</i>	Whole	2		33	22	43
Bastard halibut	<i>Paralichthys olivaceus</i>	Muscle	14	41	46	19	96
		Internal organ	3	93	107	48	183
		Whole	1		28		
Sea bream	<i>Pagrus major</i>	Muscle	3	37	38	26	50
		Whole	1		8		
Rockfish	<i>Sebastes schlegelii</i>	Muscle	4	38	38	34	44
		Internal organ	1		124		
White croaker	<i>Genyonemus lineatus</i>	Muscle	6	48	50	31	72
Gurnard	<i>Chelidonichthys spinosus</i>	Muscle	2		55	39	70
Whitebait		Whole	20	27	30	13	63

*1: Numbers of CFs calculated, *2: Geometric mean, and *3: Arithmetic mean or single value.

There were also CF-stable Cs values for muscle samples of some fish species and the Cs-137 concentration range was 14-24 with a GM of 17.1 $\mu\text{g}/\text{kg}$ -fresh. Since stable Cs concentration in seawater is 0.5 $\mu\text{g}/\text{L}$ seawater [10], the GM of CF-stable Cs was calculated to be 34 (range: 28-48, n=7), which was almost the same as listed in Table IV. CF-Cs in the same muscle samples with one whitebait (whole) sample are plotted against CF-stable Cs in Fig.4. The values agreed well with a correlation factor of 0.88 ($p=0.004$) by the t-test, but the range for CF-stable Cs was narrower. Probably, physico-chemical forms of stable Cs and radioactive Cs were not the same at the sample collection sites; however, their physico-chemical forms were almost the same because their CF values were almost the same. From the results, it was concluded stable Cs may be used as an analogue for Cs-137 in CF determination if Cs-137 concentration in fish is too low to measure and if stable Cs and Cs-137 are in physico-chemical equilibrium.

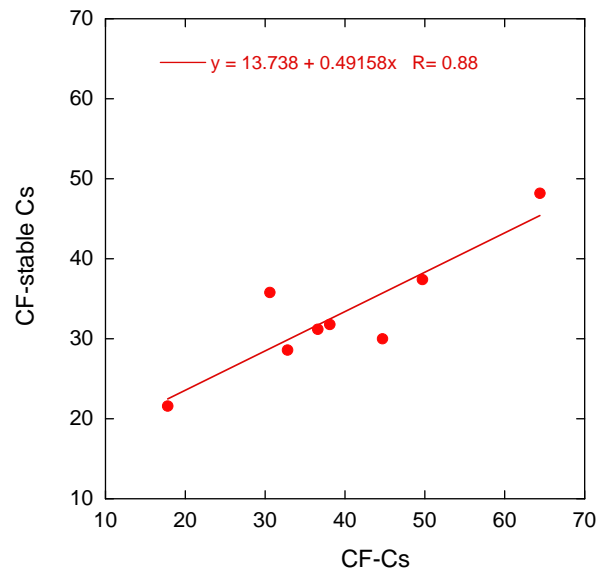


Fig.4. Correlations between CF-Cs and CF of stable Cs for seawater fish samples.

The CF-Sr data are listed in Table V for muscle (seven species) and for whole (two species). CF-Sr values in muscle of all species were in a narrow range, from 0.4 to 7.9. The GMs of seven fish species ranged from 1.6-3.1, which was almost the same as reported in TRS-422 [1]. Thus for Sr-90, it would not be necessary to provide CF values for each fish species.

Calcium concentration data were also found for some muscle samples. The Ca concentration range was 0.05-0.57 g/kg-fresh with a GM of 0.12 g/kg-fresh, while the concentration in seawater is 0.412 g/L. Thus CF-Ca ranged from 0.12-1.4 with a GM of 0.3, which was one order of magnitude lower than for CF-Sr. Indeed, significant difference was observed by the t-test ($p<0.001$) between CF-Ca and CF-Sr values. No correlation was found between CF-Ca and CF-Sr, thus, the distribution pattern of Ca and Sr should be different in seawater fish.

CF values of eight stable elements (Na, K, Ca, Ng, Mn, Fe, Cu and Zn) were also calculated using literature values and they are given in Table VI. Element concentration data in 97 species commonly found in Japan [6] were used for this calculation and the open ocean sea water concentrations of these elements were also used. The TF-Ca values were almost the same as mentioned above, and the CF levels were almost the same as those published in TECDOC-211 and TRS-422, although CF of Fe in TRS-422 is slightly higher than the present result.

Table V. Concentration factors of Sr-90 for sea fish tissues (L/kg).

Common name	Latin name	Tissue type	N ^{*1}	GM ^{*2}	SV ^{*3}	Min.	Max.
Brown sole	<i>Pleuronectes herzensteini</i>	Muscle	7	2.9	3.7	0.7	6.8
Japanese seaperch	<i>Lateolabrax japonicus</i>	Muscle	15	2.5	2.8	1.4	6.1
Yellow tail	<i>Seriola quinqueradiata</i>	Muscle	5	2.0	2.7	0.4	6.2
Bastard halibut	<i>Paralichthys olivaceus</i>	Muscle	11	2.8	3.7	0.4	7.8
Sea bream	<i>Pagrus major</i>	Muscle	3	1.9	2.0	1.4	3.1
Sea bream	<i>Pagrus major</i>	Whole	1		1.0		
Rockfish	<i>Sebastes schlegelii</i>	Muscle	4	3.1	3.7	1.9	7.9
Whitebait		Whole	20	2.6	3.2	0.7	6.3
White croaker	<i>Genyonemus lineatus</i>	Muscle	6	1.6	2.0	0.7	3.4

*1: Numbers of CFs calculated, *2: Geometric mean, and *3: Single value.

Table VI. Concentration factors of eight stable elements for sea fish tissues (L/kg).

Sea fish (97 species)	Na	K	Ca	Mg	Mn	Fe	Cu	Zn
GM	0.08	9	0.49	0.24	720	2200	1600	1200
Min.	0.03	3	0.07	0.14	500	500	400	600
Max.	0.25	13	2.1	0.67	10500	13000	6800	3900

CONCLUSION

About 100 reports on global fallout concentration data in Japanese environmental samples were selected from environmental radioactivity survey reports and studies from 1960 to 1980 published by the Science and Technology Agency [5]. Their data were used to calculate water-to-fish concentration factors (CFs). The calculated CF-Cs values for freshwater and seawater fish were almost the same as those of recommended values compiled by IAEA. All Japanese freshwater and seawater fish species showed similar values with a narrow range of CF-Cs. The same results were obtained for Sr-90 for seawater fish. Thus, it is not necessary to collect CF values for Sr-90 and Cs-137 for various kinds of species. However, the distributions of Sr and Cs in fish tissues are different; for example, Sr concentration is higher in bone than other tissues, and Cs concentrations in muscle and internal organ are higher than in bone, thus, tissue-dependent data are needed.

It was also found possible to use stable elements as analogues of radionuclides to obtain CF values. The same group elements, e.g., CF-Cs and CF-K, and CF-Sr and CF-Ca did not agree with each other so that it

is difficult to use other elements as analogues; however, the stable element and the radionuclide agreed, i.e. CF-Cs and CF-stable Cs. This approach can be used for other stable element – radionuclide sets to provide more CF values for environmental dose assessment.

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