Concentration Factors for $^{137}$Cs in Japanese Coastal Fish (1984—1990)

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(Received, October 25, 1995)
(Revision received, March 12, 1996)
(Accepted, March 12, 1996)

Concentration factors/$^{137}$Cs/Marine fish/Stable Cs/Japanese coastal waters

Concentration factors (CFs; Bq kg$^{-1}$ in wet fish muscle/Bq kg$^{-1}$ in filtered seawater) for $^{137}$Cs were determined in Japanese coastal fish collected from 1984 to 1990. $^{137}$Cs/Cs (stable) atom ratios were also examined to clarify the distribution equilibrium of $^{137}$Cs between marine fish and seawater. The geometric mean of CF in Japanese coastal fish was 52±4 (standard error of the mean), with values ranging from 14 to 133. $^{137}$Cs/Cs atom ratios both in marine fish and seawater indicate that the distribution of $^{137}$Cs was in equilibrium between fish muscle and seawater. Therefore, CF values obtained in the present study can be regarded as equilibrated. Our results show that the CFs for $^{137}$Cs in Japanese coastal fish were within the range of Japanese guidelines, but were below the recommended IAEA value.

INTRODUCTION

Concentration factors (CFs; $^{137}$Cs Bq kg$^{-1}$ in fish/$^{137}$Cs Bq kg$^{-1}$ in filtered seawater) are used as a transfer parameters in assessments of the public dose from radioactivity in the marine environment. To derive a CF value, the concentration of $^{137}$Cs and stable Cs in some marine fish have previously been investigated$^1$. Genellary, $^{137}$Cs concentrations were higher in fish muscle than in viscera$^2$, and gave CFs of 43±12 (arithmetic mean and standard deviation)$^3$. During 1963—1970, $^{137}$Cs atmospheric fallout peaked in 1963 and subsequently decreased. The $^{137}$Cs concentration in Japanese surface seawater also decreased, but the rate of decrease was slower than that of the atmospheric fallout$^3$. During 1963—1970, $^{137}$Cs atmospheric fallout peaked in at 1963 and subsequently decreased. The $^{137}$Cs concentration in Japanese surface seawater also decreased, but the rate of decrease was slower than that of the atmospheric fallout$^3$. The $^{137}$Cs concentration in fish muscle decreased in accordance with the reduction of $^{137}$Cs concentration in surface seawater and fallout$^3$, but the $^{137}$Cs concentration has recently leveled off and has shown no significant regional variations$^4$—$^{10}$. The CF values for $^{137}$Cs, however, exhibited a wide range, from 4.5 to 240$^{1,11}$—$^{19}$. Some of these values were reported under the non-equilibrium conditions, except those reported by Suzuki et al. (1973)$^1$ and Nakahara et al. (1980)$^{18}$.
In this article, we report $^{137}$Cs and stable Cs concentration in 18 species of Japanese coastal fish in order to estimate the recent CF values of $^{137}$Cs in Japanese coastal fish. For seawater, we used the reported data of $^{137}$Cs and stable Cs in surface waters\(^{20}\). To demonstrate the equilibrium state of $^{137}$Cs distribution between fish and seawater, we studied not only the concentration of $^{137}$Cs, but also atom ratios of $^{137}$Cs/Cs (stable). To examine the equilibrium state, the comparison between specific activity ($^{137}$Cs Bq kg\(^{-1}\)/Cs $\mu$g kg\(^{-1}\)) of fish muscle and seawater has been generally used in the past\(^{11}\). However, we used the atom ratios (atoms of $^{137}$Cs/atoms of stable Cs) rather than the specific activity. The reasoning for this has been described elsewhere\(^{20}\). Here we show that the atom ratio is preferable than the specific activity because of the accuracy in expressing the relation between $^{137}$Cs and Cs as a dimensionless unit. In environmental study of $^{129}$I, the atom ratios of $^{129}$I/$^{127}$I is commonly used in expressing the relation of two nuclides\(^{22}\). Therefore, we use atom ratios of $^{137}$Cs/$^{133}$Cs (stable) in this study. We have also compared our derived CF value with those recommended by IAEA\(^{23}\).

**MATERIALS AND METHODS**

From 1984 to 1990, fish samples were collected from selected coastal areas of Japan (Fig. 1).

![Fig. 1. Sampling locations in Japanese coastal waters. ● : Fish sample, □ : Seawater sample.](image)
Thirty two samples of fish, representing 18 species, were collected by set-net fishing. Fish samples were rinsed with clean seawater and muscle tissue was separated from viscera and hard tissues. Fish muscle was oven-dried for 24 hours at 110°C and ashed at 250°C and 450°C, for 24 hours and 48 hours, respectively.

The analytical procedures for $^{137}$Cs and stable Cs has been described elsewhere$^{20,21,24}$. Hence, the procedure is only briefly described here; $^{137}$Cs in the ashed fish muscle (30 g) was separated radiochemically by the ammonium-molybdophosphate method and measured with a low-background gas-flow $\beta$-counter, which was calibrated with $^{137}$Cs standards. For neutron activation analysis of stable Cs in fish samples, ashed samples (from 300 to 500 mg) and reference standard for Cs were irradiated in the JRR-4 nuclear reactor at Japan Atomic Energy Research Institute, and $\gamma$-radioactivity of induced $^{134}$Cs was measured with a $\gamma$-ray spectrometer with Ge (Li) detector.

The concentration factor (CF) for $^{137}$Cs was calculated by dividing the concentration of $^{137}$Cs in wet fish muscle (Bq kg$^{-1}$) by the concentration in filtered seawater (Bq kg$^{-1}$)$^{20,21}$. $^{137}$Cs concentration in seawater was expressed using units of Bq kg$^{-1}$ instead of Bq l$^{-1}$ in order to make the CF value a dimensionless unit$^{23}$.  

RESULTS

Data for $^{137}$Cs and stable Cs concentrations in fish muscle are shown in Table 1 together with $^{137}$Cs/Cs atom ratios and CF values for $^{137}$Cs. The $^{137}$Cs concentrations in fish muscle were ranged from 0.08 to 0.44 Bq kg$^{-1}$-wet. These values are similar to the range reported for Japanese coastal fishes collected during 1987—1990 (0.16—0.47 Bq kg$^{-1}$-wet)$^{27}$. Stable Cs concentrations in fish muscle varied between 4.4 and 23 ng g$^{-1}$-wet, and were comparable to the reported values of 4.4 and 23 ng g$^{-1}$-wet for coastal species$^{11,18,25}$.

The $^{137}$Cs and stable Cs concentrations in coastal waters and the $^{137}$Cs/Cs atom ratios in Japanese coastal waters during 1984—1990 were reported by Tateda and Koyanagi (1994)$^{20}$ (Table 2). The $^{137}$Cs concentrations in filtered seawater were between 3.5 and 5.1 mBq l$^{-1}$ at study sites during 1984—1990. Variations of values between years were not significant. During the 1960s, $^{137}$Cs levels in Japanese coastal waters were within the range of 8.9—28 mBq l$^{-1}$$^{1,26}$. These values are higher than those of today because of the higher atmospheric radioactive fallout during that period. During the 1970s and early 1980s, $^{137}$Cs level in seawater decreased to 3.2—6.3 mBq l$^{-1}$$^{27—29}$ due to the reduction in atmospheric radioactive fallout. Our results from the late 1980s showed lower $^{137}$Cs concentrations than those in the early 1980s. The only exception was the $^{137}$Cs level in seawater in 1986, corresponding to the Chernobyl accident, but the affected period was restricted to a few months$^{5}$. The $^{137}$Cs release from nuclear facilities in Japan has been negligible. Therefore, the $^{137}$Cs concentration in Japanese coastal waters can be regarded as being almost unchanged during the present study period. The mean stable Cs concentrations in Japanese coastal waters was 0.29 $\mu$g l$^{-1}$ with a range of 0.24—0.34 $\mu$g l$^{-1}$, and the geometric mean value. As the reported stable Cs concentrations in seawater were within the range of 0.15—0.55 $\mu$g l$^{-1}$ $^{20,30}$, it is evident that the sites investi-
gated in this work had not been affected by riverine run-off or large inputs of land-derived substances, which can cause a variation of stable Cs concentration in seawater.

Since the $^{137}\text{Cs}$ levels in seawater and biological samples can be regarded as almost constant during 1984—1990\cite{20,24}, we used the data of $^{137}\text{Cs}$ levels in seawater and fish muscle as being representative of those levels during the present study period. By these data, we identify the

| Species               | Location | year | $^{137}\text{Cs}^{a)$ (Bq kg$^{-1}$-wet)} | Cs$^{b)$ (ng g$^{-1}$-wet)} | $^{137}\text{Cs}/\text{Cs}$ atom ratio ($\times 10^{-9}$) | Concentration factor |
|-----------------------|----------|------|---------------------------------|-----------------|---------------------------------|---------------------|
| *Siganus fuscensenns* | Tsuyazaki | 1986 | 0.19 ± 0.01 | 18 ± 1 | 3.2 | 43 |
| *Hexagrammos agurammus* | Tsuyazaki | 1986 | 0.23 ± 0.01 | 13 ± 1 | 5.4 | 52 |
| *Hexagrammos otakii* | Katsuura | 1985 | 0.2 ± 0.01 | 11 ± 1 | 5.5 | 44 |
| *Hexagrammos otakii* | Katsuura | 1986 | 0.29 ± 0.01 | 13 ± 1 | 6.8 | 65 |
| *Hexagrammos otakii* | Hachinohe | 1986 | 0.38 ± 0.01 | 14 ± 1 | 8.2 | 86 |
| *Hexagrammos otakii* | Oshoro | 1989 | 0.11 ± 0.01 | 7.1 ± 0.4 | 4.7 | 30 |
| *Hexagrammos otakii* | Oshoro | 1990 | 0.19 ± 0.01 | 11 ± 1 | 5.2 | 54 |
| *Hexagrammos otakii* | Hachinohe | 1990 | 0.26 ± 0.01 | 13 ± 1 | 6.1 | 73 |
| *Hexagrammos otakii* | Hachinohe | 1987 | 0.44 ± 0.01 | 23 ± 1 | 5.8 | 102 |
| *Pleurogrammus azonus* | Oshoro | 1986 | 0.31 ± 0.01 | 14 ± 1 | 6.7 | 70 |
| *Pleurogrammus azonus* | Hachinohe | 1986 | 0.4 ± 0.07 | 14 ± 1 | 8.7 | 91 |
| *Pleurogrammus azonus* | Hachinohe | 1987 | 0.37 ± 0.01 | 21 ± 1 | 5.3 | 88 |
| *Ditrema temmincki* | Katsuura | 1985 | 0.25 ± 0.01 | 11 ± 1 | 6.9 | 51 |
| *Ditrema temmincki* | Katsuura | 1986 | 0.27 ± 0.01 | 14 ± 1 | 5.8 | 61 |
| *Ditrema temmincki* | Hachinohe | 1986 | 0.27 ± 0.01 | 13 ± 1 | 6.3 | 61 |
| *Ditrema temmincki* | Hachinohe | 1987 | 0.06 ± 0.01 | 4.4 ± 0.2 | 4.1 | 14 |
| *Sebastes inermis* | Katsuura | 1985 | 0.31 ± 0.01 | 17 ± 1 | 5.5 | 69 |
| *Sebastes pachycephalus* | Katsuura | 1985 | 0.19 ± 0.01 | 8.8 ± 0.4 | 6.5 | 41 |
| *Sebastes marmoratus* | Katsuura | 1985 | 0.2 ± 0.01 | 15 ± 1 | 4.0 | 44 |
| *Sebastes baramenueke* | Hachinohe | 1986 | 0.24 ± 0.01 | 13 ± 1 | 5.6 | 54 |
| *Parapristipoma trilineatum* | Katsuura | 1985 | 0.19 ± 0.01 | 10 ± 1 | 5.8 | 41 |
| *Scombropis hoops* | Katsuura | 1986 | 0.59 ± 0.01 | 23 ± 1 | 7.8 | 133 |
| *Onchorhyncus keta* | Hachinohe | 1988 | 0.08 ± 0.01 | 10 ± 1 | 2.4 | 20 |
| *Kareius bicoloratus* | Katsuura | 1986 | 0.13 ± 0.01 | 9.4 ± 0.5 | 4.2 | 30 |
| *Limanda schlencki* | Hachinohe | 1986 | 0.32 ± 0.01 | 17 ± 1 | 5.7 | 72 |
| *Limanda yokohamae* | Hachinohe | 1987 | 0.19 ± 0.01 | 11 ± 1 | 5.2 | 45 |
| *Paralichthys olivaceus* | Hachinohe | 1986 | 0.22 ± 0.01 | 11 ± 1 | 6.1 | 50 |
| *Paralichthys olivaceus* | Katsuura | 1986 | 0.29 ± 0.01 | 15 ± 1 | 5.9 | 65 |
| *Paralichthys olivaceus* | Hachinohe | 1987 | 0.28 ± 0.01 | 16 ± 1 | 5.3 | 66 |
| *Paralichthys olivaceus* | Hachinohe | 1988 | 0.23 ± 0.01 | 12 ± 1 | 5.8 | 57 |
| *Hippoglossoides dubius* | Hachinohe | 1987 | 0.1 ± 0.01 | 12 ± 1 | 2.5 | 23 |
| *Microstomus achen* | Hachinohe | 1986 | 0.22 ± 0.01 | 11 ± 1 | 6.1 | 50 |

$^{a)$ measured value ± counting error
$^{b)$ measured value ± analytical error
The geometric mean and standard error for 137Cs/Cs atom ratios in environmental samples. Because the data distribution was proved to follow statistically the log-normal distribution. The geometric means of 137Cs/Cs atom ratios between seawater and fish showed no significant differences (Kolmogorov-Smirnov test, p<0.05). This result indicates that 137Cs distribution is equilibrated between filtered seawater and fish muscle. Suzuki et al. (1978) showed that 137Cs distribution reached an equilibrium state between seawater and fish muscle during 1963–1970. Our result is compatible with Suzuki et al.’s finding.

### DISCUSSION

The CF values for 137Cs in coastal fish muscle studied here are shown in Table 1. These CF

| Location  | Year | 137Cs (mBq l⁻¹) | Cs (μg l⁻¹) | 137Cs/Cs atom ratio (×10⁻⁹) |
|-----------|------|----------------|------------|-----------------------------|
| Sakata    | 1984 | 5.1±0.2        | 0.29±0.01  | 5.3                         |
| Katsuurra | 1984 | 5.0±0.3        | 0.25±0.01  | 6.9                         |
| Sado      | 1985 | 4.8±0.2        | 0.26±0.02  | 5.8                         |
| Tsuyazaki | 1985 | 4.7±0.2        | 0.34±0.02  | 4.2                         |
| Rokkasho  | 1987 | 5.0±0.2        | 0.33±0.02  | 5.0                         |
| Tsuyazaki | 1987 | 3.7±0.2        | 0.31±0.02  | 3.7                         |
| Oshoro    | 1989 | 4.2±0.2        | 0.30±0.02  | 4.6                         |
| Rokkasho  | 1989 | 3.6±0.2        | 0.33±0.02  | 3.6                         |
| Oshoro    | 1990 | 3.5±0.2        | 0.25±0.01  | 3.5                         |
| Rokkasho  | 1990 | 3.7±0.2        | 0.24±0.01  | 3.8                         |

* a) measured value ± counting error
* b) measured value ± analytical error
values were derived from the seawater and fish data in the same year. However the $^{137}\text{Cs}$ concentrations in seawater in 1986 and 1988 were not obtained. Therefore we assumed, based on the expected exponential decrease of $^{137}\text{Cs}$ in seawaters\textsuperscript{24}, that the $^{137}\text{Cs}$ concentration in seawater would be 4.5 (in 1986) and 4.1 (in 1988) mBq $\ell^{-1}$. The CF values given in Table 1 were calculated by using a density of seawater of 1.025 kg $\ell^{-1}$\textsuperscript{17,23}. The cumulative percent of $^{137}\text{Cs}$ CFs in fish muscle is shown in Fig. 2. The data distribution of $^{137}\text{Cs}$ CFs is close to log-normal. However, the data sets showed three significantly different groups (Kolmogorov-Smirnov test, $p<0.05$). The geometric mean of CFs in the main group was 55 ($\pm 2$: standard errors of mean), and that of CF of the whole data set was 52 ($\pm 4$: standard errors of mean).

![Fig. 2. The cumulative of $^{137}\text{Cs}$ concentration factors in fish muscle.](image)

Many factors are known to affect the $^{137}\text{Cs}$ concentration factor in fish muscle. For example, the $^{137}\text{Cs}$ concentration at steady state is known to vary with body size\textsuperscript{31} and taxa\textsuperscript{14}. Environmental factors also affect the concentration of $^{137}\text{Cs}$ in fish muscle: the brackish water species (e.g. Japanese sea perch \textit{Lateolabrax japonicus}) showed higher $^{137}\text{Cs}$ concentrations than other coastal species\textsuperscript{18}. All the fish samples investigated in this study were adult, and all of the sampling sites were free from the effect of river run off. Therefore the different groups of $^{137}\text{Cs}$ CFs (Fig. 2) may be attributed to the difference in species\textsuperscript{17}. However, the number of data for each species were not sufficient to draw the conclusion. The verification of this difference in CF group should be carefully analysed in future studies.

The $^{137}\text{Cs}$ CF in Japanese coastal edible fish in the late 1960s was 43 $\pm$ 12 (arithmetic mean and standard deviation; on Bq kg$^{-1}$/Bq $\ell^{-1}$)\textsuperscript{11}. This value can be converted to 43 $\pm$ 5 (geometric mean and standard error of mean value, on Bq kg$^{-1}$/Bq kg$^{-1}$ base). Since the ratios of ($^{137}\text{Cs}/\text{Cs}$)$_{\text{fish}}$/$(^{137}\text{Cs}/\text{Cs})_{\text{seawater}}$ were within the range of 0.6 $\sim$ 1.6, we can assume equilibration in distribution of $^{137}\text{Cs}$ between fish muscle and seawater. Statistically, there is a significant difference (Kolmogorov-Smirnov test, $p<0.05$) between the CF data presented here and those of Suzuki \textit{et al.}\textsuperscript{11}. This difference is possibly due to the annual variation of $^{137}\text{Cs}$ fallout in 1960's,
which affected $^{137}$Cs levels in seawater$^1$.

The IAEA$^{23}$ recommended a value of 100 as a generalized CF for $^{137}$Cs in fish. This recommended value was confirmed by Steel$^{17}$, who reported CF of $92 \pm 43$ (cod), $58 \pm 17$ (haddock), $39 \pm 16$ (plaice) and $150 \pm 82$ (whiting) (arithmetic mean and standard deviation) for North Sea fish collected from 1978 to 1985. These arithmetic means can be converted to geometric means $83 \pm 6$, $56 \pm 3$, $36 \pm 2$ and $127 \pm 16$ (standard error of mean value). Our results and Suzuki et al.'s$^1$ findings showed somewhat smaller CF values than those of IAEA and Steel$^{17}$. The most probable reason is the difference in radioecological environments between European coastal waters and Japanese coastal waters. The concentration of $^{137}$Cs in North European coastal waters is influenced by the $^{137}$Cs discharges from the Sellafield reprocessing plant into the Irish Sea$^{32}$. This situation resulted in large annual variations of $^{137}$Cs concentrations in North Sea seawater, and affected the distribution of $^{137}$Cs in the adjacent sea areas$^{33}$. In addition, the $^{137}$Cs in the seabed of Irish Sea is being remobilized and affects the $^{137}$Cs concentration in seawater$^{34}$. For Japanese coastal fish, the recommended CFs value of $^{137}$Cs is 30 under authorization in the public dose assessment, both for the light-water nuclear power reactor (in the guidelines published by Japanese Atomic Energy Commission), and for the Tokai fuel reprocessing plant$^{19}$. The results of our study show larger values than the guideline and authorized CFs. These CFs, however, were estimated under the consumption of “whole” fish by the public. Therefore, it is reasonable to suggest that the authorized CF value would smaller than our result because $^{137}$Cs is less concentrated in the viscera and in the bone of fish.

In conclusion, the $^{137}$Cs levels ($^{137}$Cs/Cs atom ratio) in muscle of Japanese coastal fish almost leveled off during 1984—1990. The distribution of $^{137}$Cs between seawater and fish muscle reached an equilibrium state during the study period. The derived CF values for $^{137}$Cs in Japanese coastal fish showed three different groups, however the reason is unknown. The estimated $^{137}$Cs CFs for muscle of Japanese coastal fish gave the geometric mean value $52 \pm 4$ (standard error of mean value).

ACKNOWLEDGEMENTS

The authors would like to thank Miss Y. Sugita for assisting with sample analysis, the staffs of Hokkaido University, North Eastern Regional Fisheries Research Institute, Marine Ecological Research Institute and Kyushu University for collecting the fish and seawater samples. Dr. P. McDonald, Westlakes Research Institute, U. K. and Dr. M. Carroll IAEA Marine Environmental Laboratory, assisted us with the proofread of the manuscript for English usage. This research was granted by the Central Research Institute of the Electric Power Industry.

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