# A Review of Transfer Parameters of I, Cs and Pu (2)
## -Concentration Ratios in Freshwater and Marine Fish-

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**Abstract.** In part (1) we reviewed references used for the transfer coefficient from feed to cow's milk of Cs, I and Pu and compared those data with recently published data. In part (2), we have reviewed water-to-fish concentration ratio (CR) data to reconfirm the validity of the original values used in transfer parameter summary reports, such as prepared by the Atomic Energy Society of Japan, and to compare the recommended CR data with newly reported values. We back-traced the references used in the parameter summary reports and found that the CR data did not change much from the values in the 1960's and 1970's for both freshwater and marine fish. We note that due to the difficulty to find the original data in easily accessible digital forms, not all of the original data in the old publications were confirmed, but because experts had judged and provided these recommended data, these data may be assumed to be reliable. Our new data survey showed that not many data were available for I and Pu; for Cs reported values were almost the same as reported in summary reports.

***KEYWORDS: Water-to fish concentration ratio, Environmental dose assessment, Parameter values***

## 1 INTRODUCTION

One of the most important pathways of radionuclides released from nuclear waste disposal sites to humans is via food consumption. Various types of transfer parameters, such as concentration ratios of radionuclides from environmental media to foods are used for radiation dose assessment models. The most reliable parameter values can be provided from laboratory tracer experiments or field observations of the radionuclides or stable elements. Parameter values are also calculated by using metabolic models for the case of farm animals. However, there are still difficult-to-obtain parameters; for such cases, expert judgement is used. Together with the parameter values, their data collection processes are need to be recorded to keep data transparency. With this information, data users can easily understand the quality of the parameter data. In IAEA reports, e.g. TRS 364 [1], 422 [2] and 479 [3], recommended parameter values are published with their original data sources; thus the values provided in such reports are highly reliable, and these values are frequently used in dose assessment models in many countries.

We note that such recommended parameter values consist of old literature values with some updates, if there were any, at the time of the data summary activity. In part (1) included in the Proceedings of IRPA 15 [4], we traced back references used for the transfer coefficient from feeds to cow's milk of Cs, I and Pu in IAEA reports such as TRS 364 [1]. Our traced back results showed that field observation data or tracer experiments in the 1950's to 1970's were the main sources of the parameter data.

In Japan, the parameter values compiled in IAEA reports (TRS 364 [1], 422 [2]) have been used such as JAEA TRU 2 [5] and a review by the Atomic Energy Society of Japan (AESJ) [6]. Although cow's milk is one of the popular food materials in European and North American countries, other countries have different food cultures. It is necessary to consider the food consumption trends in each country for dose assessment by ingestion. For Japan, seafood ingestion pathways are important because Japan is one of the biggest fish consuming countries in the world [7]. Thus, in the dose assessment for nuclear waste management, water-to-fish concentration ratio (CR) is of great concern. In this second review paper, we especially focus on Cs, I and Pu for their importance in environmental dose assessment for nuclear waste disposal.

# 2 MATERIALS AND METHODS

## 2.1 Tracing Original CR Data Used in Parameter Summary Reports for Dose Assessment

The transfer parameter, water-to-fish concentration ratio (CR; L/kg FW) is defined and was calculated as follows:

$$CR = A_{\mathrm{fish}} / A_{\mathrm{water}}$$

where $A_{\mathrm{fish}}$ is activity concentration in fish (Bq/kg FW) and $A_{\mathrm{water}}$ is that in the surrounding water (Bq/L) where the fish was caught.

Transfer parameter values provided in data summary reports such as IAEA reports are generally based on original data sources. In the reports, the CR listing of each radionuclide includes the numbers of original data used for the analysis, geometric mean with geometric standard deviation, arithmetic mean with standard deviation, and minimum and maximum values. The original data sets were usually kept by the researchers who worked on preparation of the summary reports. Therefore, it is difficult to check each datum; what we could do is trace the references used in the summary reports. Here, we have checked details of the references used in the summary report of parameters published in Japan [5].

We also traced reports from Radioactive Waste Management Center [8, 9] and IAEA Technical Report Series No. 422 (TRS 422) [2] and No. 472 [10] for marine fish and freshwater fish.

## 2.2 Survey of Newly Reported Data

According to the published year of the summary reports listed in section 2.1, we assumed that the data collation would be made a year before publication of these reports; thus, we set different periods to survey new literature for marine and freshwater fish in Japan. For CR_Cs, we did not use any observed after 2011 because the Cs long-term effects of the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident to fish would still be occurring [11]. Takata et al. [12] reported that many water-to-biota systems had not reached equilibrium conditions and CRs for many fish species were still higher than CRs before the accident. On the other hand, the effects by Pu released in the FDNPP accident have been small and short-lived iodine ($^{131}$I) was not detected long-term after the accident; therefore, we judged CR data for these elements after 2011 to be acceptable. Considering these conditions, finally, we set the periods of data survey as follows.

***<I and Pu>***
Marine fish: 1992-2019 (in Japanese literature), 2004-2019 (in English literature)
Freshwater fish: 1996-2019 (in Japanese literature), 2010-2019 (in English literature)

***<Cs>***
Marine fish: 1992-2010 (in Japanese literature), 2004-2019 (in English literature, excluding data affected by the FDNPP accident)
Freshwater fish: 1996-2010 (in Japanese literature), 2010-2019 (in English literature, excluding data affected by the FDNPP accident)

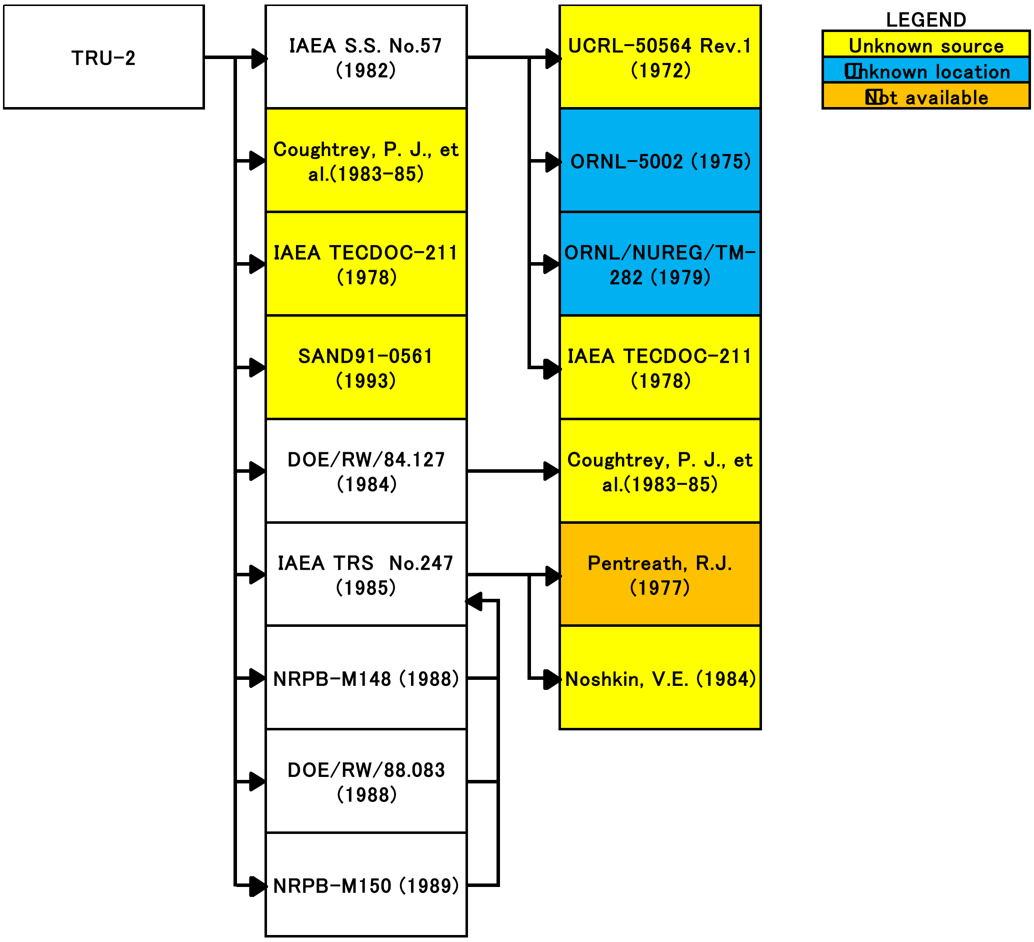
For the new data survey, we used the following three search engines: Google scholar (https://scholar.google.co.jp/) because it covers large areas of scientific publications, Science Direct (https://www.sciencedirect.com/) because it covers Elsevier Publications and provides quick updates of their publications, and J-stage (https://www.jstage.jst.go.jp/browse/-char/ja) because it covers papers in Japanese and English published in Japanese journals. The keywords used were, "iodine (I), cesium (Cs) or plutonium (Pu)", "concentration factor", "radioactivity", "fish", and "freshwater, river, lake, marine, or sea", and we searched the keywords in both English and Japanese.

# 3 RESULTS AND DISCUSSION

## 3.1 Back-tracing of CR Data of Cs, I and Pu in Marine Fish in Summary Reports

The back-tracing results starting from TRU 2 [5] are shown in Figure 1 (note: due to the page limitation of the proceedings, not all the references are in the end list). Most of the water-to-fish CR data were from summary reports by the IAEA, the DOE and the NRPB. We checked further to see whether we could find the original data sets. Coughtrey et al. [13] provided CR data with original sources but many of them were obtained in the 1960's and 1970's and are not available digitally therefore we could not confirm the original data. The most recent case, i.e. SAND91-0561 [14], might include data obtained in the 1980's. Unfortunately, however, these data did not include Japanese literature values because most of the cited literature were published outside Japan. In other summary reports, we could not find the data sources or we even could not find the data location in the reports. Even if the original data were difficult to get, we still trusted the data published in the summary reports because at least their authors used "expert judgement" in providing the CR data.

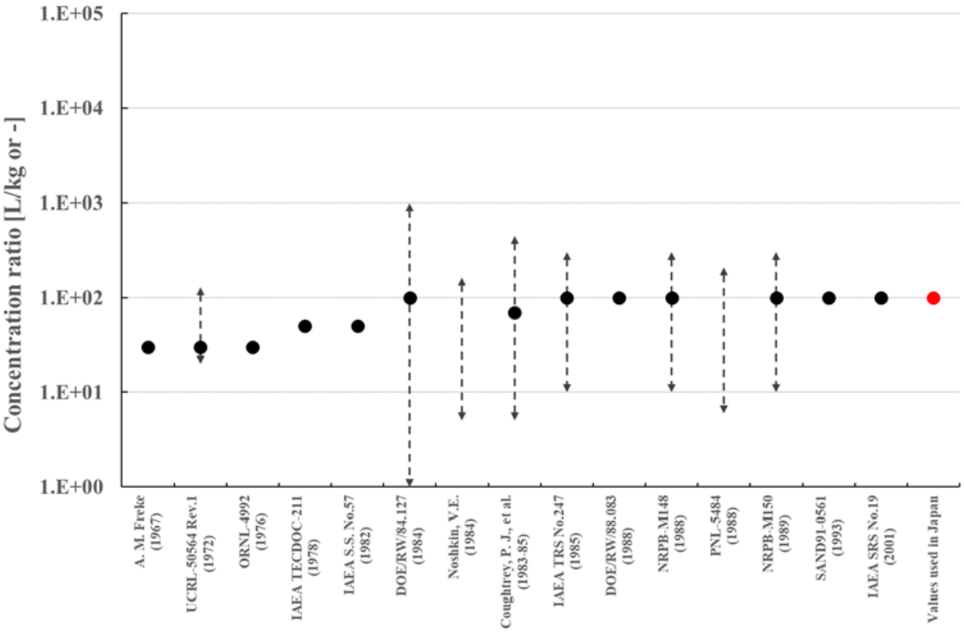
**Figure 1**. Traced-back references in TRU 2 [5] for CR_Cs values for marine fish.



Next we compared the reported CR_Cs data from these publications and the results are plotted in Figure 2. The data reported in the 1960's and 1970's were slightly lower than those used in TRU 2 [5]; however, from these comparisons, we judged the recommended CR_Cs values had not changed much since the 1980's. A similar trend was found for CR_Pu (data not shown). For I, the CR data were almost the same among the surveyed reports (CR=10 L/kg) (data not shown).

Some reports showed large variation of the data, ca. more than two orders of magnitudes was reported in DOE/RW/84.127 [15] as shown in Figure 2; the variation was also large for I and Pu in the same report (data not shown). According to Figure 1, DOE/RW/84.127 [15] referred to Coughtrey et al. [13] which had a narrower distribution of CR_Cs.

**Figure 2:** Reported CR_Cs (L/kg, or unitless (-) when Bq/kg of water was used) in literature values (black) and that used now for dose assessment in Japan (red). Dotted bars show the CR_Cs value ranges.



## 3.2 Comparison of the CR Data in Marine Fish in Summary Reports and in New Reports

The parameter values already used should be reliable ones because almost no changes were found for the last several decades, which means experts at the time of parameter collation selected those CR values. More data are available now with development of measurement methods for low levels of Cs, I and Pu, and it is important to compare old and new data sets to make the parameter values more accurate

CR_Cs values were found in eight recent documents for marine fish (N=57). Data from Japan were found in four papers and limited to the edible part in marine fish, e.g., ref. [16]. Edible part data of these recent values agreed well with TRS 422 as shown in Figure 3. Although conversion factor from muscle to whole body for marine fish is reported as 1.0 in TRS 479 [3], according to the figure, CRs for edible parts (mostly muscles) were higher among recent observation results as well as among reported values.

For I, we found only one paper with one datum that was obtained by measuring I-129, and the value was not reported from Japan. The value (CR=100 L/kg) agreed well with the values recommended by IAEA TRS 422. Probably by using stable iodine data, we could provide more CR data for iodine in marine fish. For marine biota other than fish, such stable iodine base CR measurement results have been reported [17].

For Pu, data for two marine fish species were reported (N=6 in total), and again, no data were provided from Japan. The tissue part for this CR value of Pu were not clear but the TRS 422 value fell within the range of the newly reported values. The value obtained on the Cumbrian coast [18] was 5 L/kg and this was lower than those for the Baltic Sea by two orders of magnitude; CR values for four fish species in the Baltic Sea ranged from 100-1900 L/kg [19]. It was not clear why the differences were large between the two locations, likely factors affecting the CR of Pu would be different Pu forms and fish species. Further observation is needed for a detailed discussion.

**Figure 3:** Summary of newly reported CR values for Sea Water (SW) fish in whole body, edible, other and unknown parts compared to CR values (L/kg, or unitless (-) when Bq/kg of water was used) reported in TRS 422 [5]. Dotted bars show the CR_Cs value ranges.

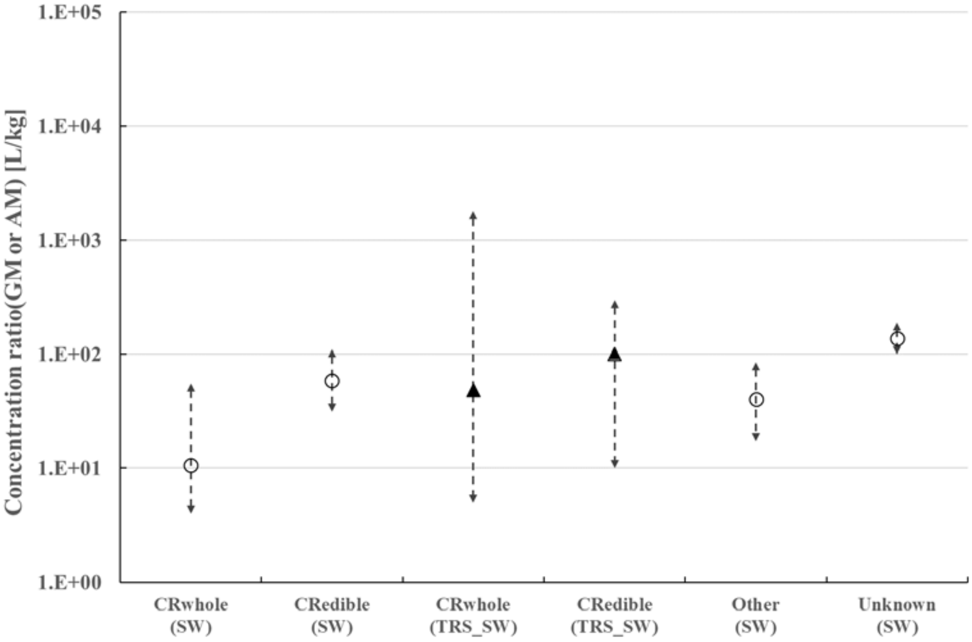
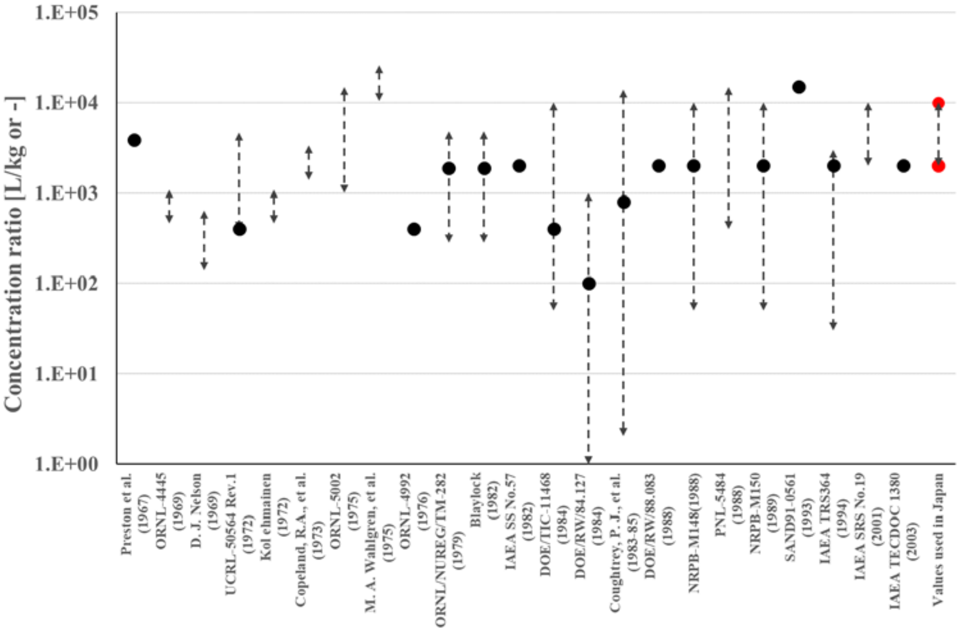


**Figure 4:** Plot of CR values of Cs for fresh water fish (L/kg, or unitless (-) when Bq/kg of water was used) used in the literature for dose assessment. Dotted bars show the CR_Cs value ranges.



## 3.3 Back-tracing of CR Data of Cs, I and Pu in Freshwater Fish in Summary Reports

The CR data used in obtaining the literature survey results are shown in Figure 4. Fresh water fish CR_Cs had two orders of magnitude difference among reports. The DOE/RW/84.127 [15] CR value was the lowest and had a wide range of variation, while the SAND91-0561 [14] value was the highest. For Cs, just as Rowan et al. [20] described, the food chain led to Cs bioaccumulation. Thus the observed differences might be the effect of which trophic level fish species were collected for CR measurements.

The values used in Japanese assessment models [5, 6] were set at a somewhat high level, probably, to avoid underestimation of the dose.

The CR_I results in freshwater fish are shown in Figure 5. The CR values were within the range of 10-70 L/kg, which was slightly higher than marine fish (data not shown). The recommended values showed similar trend for marine fish, that is, the values were almost the same from the 1960's to 2000's. For this element case, too, the values obtained in Japan were not used. But recently, stable iodine measurement results have become available [21], and providing CR data using stable I is the next step to enhance the iodine CR data.

**Figure 5:** Plot of CR values of I for fresh water fish (L/kg, or unitless (-) when Bq/kg of water was used) used in the literature for dose assessment. Dotted bars show the CR_Cs value range.

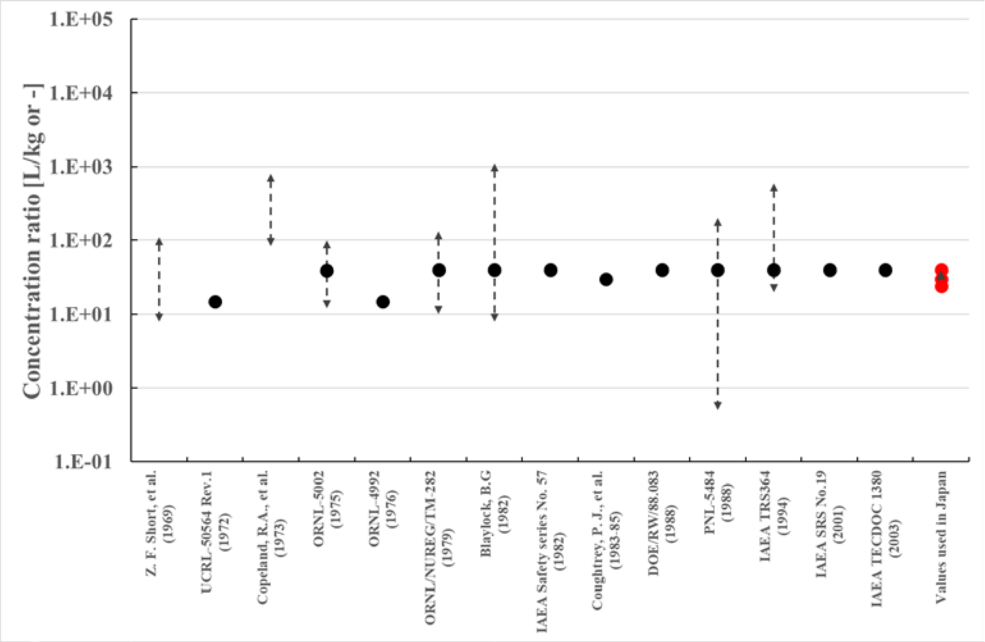
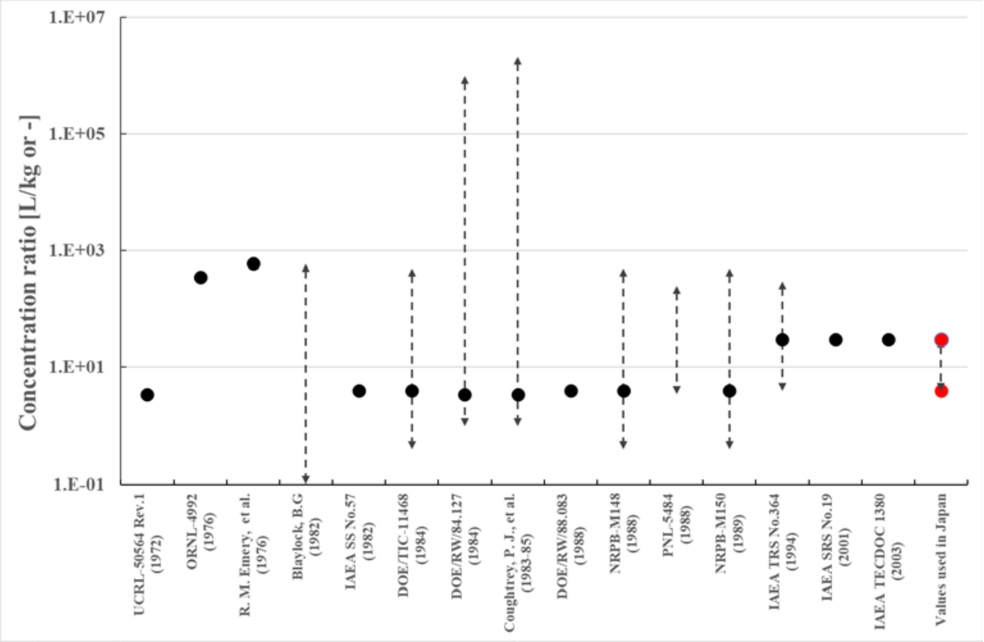


**Figure 6:** Plot of CR values of Pu for fresh water fish (L/kg, or unitless (-) when Bq/kg of water was used) used in the literature for dose assessment. Dotted bars show the CR_Cs value range.



For Pu (Figure 6), two CR data of the 1970’s had a relatively higher value than those reported in the 1980’s by two orders of magnitude. Then in the 1990's, the CR value increased five-fold. The CR_Pu values used in Japanese dose assessment [5, 6] had these ranges. As seen in DOE/RW/84.127 [15] and Coughtrey et al. [13], CR_Pu values ranged over more than five orders magnitude; thus although a narrow range for the recommended value was used in Japan, it is necessary to pay attention to the large CR_Pu variation. The reason for this large variation was unclear and further studies are needed to clarify the major factors affecting the CR_Pu values.

## 3.4 Comparison of the CR Data in Freshwater Fish in Summary Reports and in New Reports

After we checked the publications as shown in section 2.2, we found that there was no CR for Pu, one paper for I [22] and seven papers for freshwater fish for Cs, e.g., ref. [11].

Numbers of CR_Cs values for freshwater fish were 130; and CRs of edible parts were observed to be relatively higher compared to those for whole body or other tissue parts. This means that Cs amounts were relatively higher in fish meat than other parts. The CR values of whole body ($CR_{whole}$) and edible part ($CR_{edible}$) of fish in FW were within the ranges of the TRS series. $CR_{whole}$ values in brackish water fish from the literature were 1-2 orders of magnitude higher than those in the TRS series; however the reason was not clear. Although the site location for high level nuclear waste disposal has not been decided yet in Japan, since coastal areas are potential sites, it may be desirable to supply CR values near coastal areas where brackish environments occur.

For iodine CR, one paper using stable iodine reported 49 data (need the reference name or number here). This recent paper had almost the same CR value as those reported in TRS 472 [10]. Interestingly, whole body CR_I was higher than edible part CR, which is opposite the trend for Cs, thus I would not be concentrated in muscle part.

## 4 CONCLUSIONS

We back-traced the references used in parameter summary reports used in Japan such as TRU 2 [5] and AESJ [6]. These reports referred to internationally available parameter summary reports such as IAEA TRS 422 [2]. Although we carried out a further reference survey but we could not go back to all of the original data because some are not available in digital format. Finally, we found that the back-traced CR data did not change much from the values in the 1960’s and 1970’s for both freshwater and marine fish. We carried out a new data survey to compare their data with those in the parameter summary reports but not many data were available for I and Pu; for Cs reported values were almost the same as in the summary reports.

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