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Accumulations of artificial radionuclides ^{137}Cs and $^{239+240}$Pu in anchovy from the Korean seas

Huisu Lee and Intae Kim*

Marine Environmental Research Center, Korea Institute of Ocean Science and Technology (KIOST), Busan 49111, South Korea.

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Abstract – We investigated the accumulation patterns of the artificial radionuclides ^{137}Cs and $^{239+240}$Pu in anchovy – categorized into four groups by size– from Korean seas. The activities of ^{137}Cs and $^{239+240}$Pu were in the ranges of 74–137 mBq kg^{-1} and 0.27–3.21 mBq kg^{-1}, respectively. They generally increased with increasing size (except for $^{239+240}$Pu in large anchovy), indicating radionuclide accumulation by seawater uptake through respiratory and/or higher feed capacity as a manifestation of growth. However, the activity of $^{239+240}$Pu decreased sharply in large anchovy. The calculated concentration factors (CFs) of ^{137}Cs in anchovy were 10.9–20.2, which are an order of magnitude lower than those in other marine organisms. However, the CFs of $^{239+240}$Pu in adult anchovy were 14.0–162.8, which are significantly higher than those in other fishes and comparable (or even higher) to the IAEA recommendation values. The annual effective dose (AED) rates of ^{137}Cs and $^{239+240}$Pu for anchovy consumption (per person) in South Korea were estimated to be 3.7×10^{-6}–6.9×10^{-6} mSv yr^{-1} and 0.26×10^{-6}–3.10×10^{-6} mSv yr^{-1}, respectively. The AED from anchovy consumption is insignificant relative to that of natural radionuclides.

Keywords: artificial radionuclides / ^{137}Cs / $^{239+240}$Pu / anchovy / radionuclide accumulation

1 Introduction

^{137}Cs, an isotope of Caesium (Cs), is produced by gamma (γ)-emission from artificial radiation resulting from nuclear weapons tests or nuclear facility accidents (UNSCEAR, 2000). ^{137}Cs can be harmful to humans based on assessments of bodily exposure because it has a relatively long half-life (30.03 yr) and releases very strong gamma rays (662 keV) (Oh and Ryu, 2006). Furthermore, it is estimated that 15.2 ± 18.3 PBq ^{137}Cs was introduced into the northern Pacific Ocean and deposited in the atmosphere by the recent Fukushima Nuclear Power Plant (NPP) accident in 2011 (Aoyama, 2018). This artificial Cs isotope has chemical characteristics similar to potassium (K), a significant nutrient present in nature, in that both Cs and K are alkali elements. Consequently, ^{137}Cs released in the ocean is soluble in water and is therefore easily taken up by living organisms; furthermore, it known to affect the muscles and stomach (Koide *et al.*, 1982).

$^{239+240}$Pu is the alpha (α)-emitting isotope of Plutonium (Pu) that was released primarily during the 1950s and early 1960s by the atmospheric bomb tests of the Cold War period (Peirson *et al.*, 1982; Clark and Smith, 1988; Donaldson *et al.*, 1997). The half-lives of ^{239}Pu and ^{240}Pu are 24,100 and 6,561 years, respectively – much longer than other artificial radionuclides. $^{239+240}$Pu absorbed by marine organisms is known to pose significant health risks to humans *via* seafood consumption (Kim *et al.*, 2020). Moreover, Pu is known to affect human liver and bone tissues, potentially causing cancer (Nielsen *et al.*, 2012).

As of June 2020, the number of NPPs in South Korea has increased by 80.7% since the 1980s, with the amount of power generated by NPPs having increased to 25%–18% higher than the Organization for Economic Co-operation and Development (OECD) average (Korea Institute of Nuclear Safety, 2019). Hence, research on the potential impact of the release of artificial nuclides such as ^{137}Cs and $^{239+240}$Pu on seawater surrounding the Korean Peninsula from NPP accidents, is needed.

However, studies on the effects of artificial radionuclide accumulation in marine products are rare, other than those conducted immediately following the Fukushima accident (Schiermeier, 2011; Kryshev *et al.*, 2012; Fisher *et al.*, 2013). More recently, Kim *et al.* (2019) reported that ^{137}Cs in some fishes increased with increasing size (weight). However, this study categorized fishes into only two growth stages, limiting the evaluation of artificial nuclide accumulation patterns based on a lack of detail at various growth stages.

*Corresponding author: ikim@kiost.ac.kr

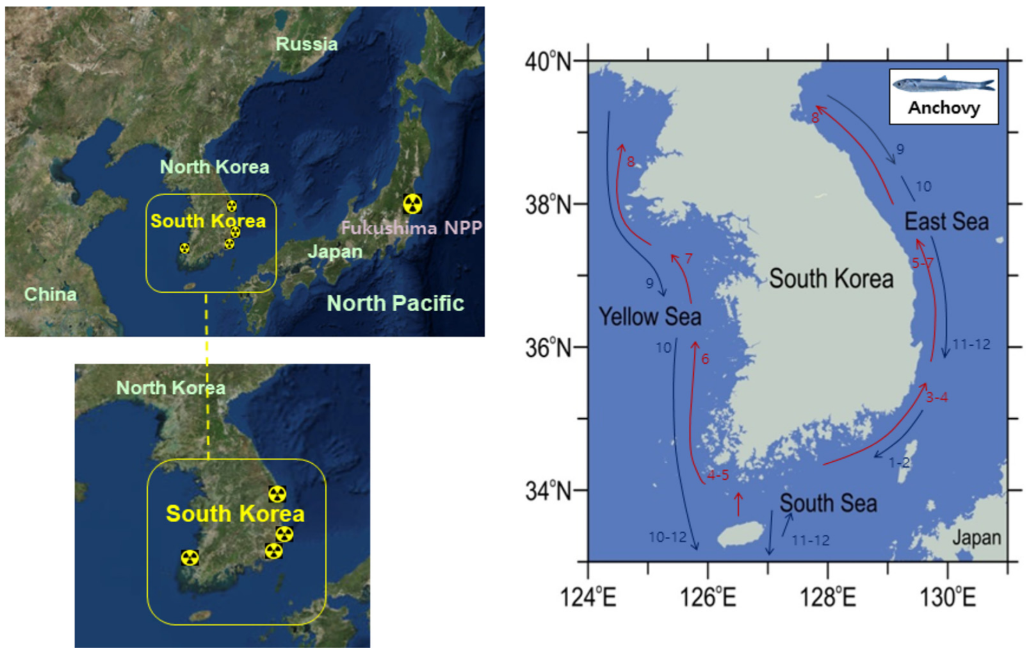

Fig. 1. The Korean seas, sampling region (yellow box), and locations of nuclear power plant facilities in South Korea (left). The migratory map of anchovy (right) around the Korean Peninsula. Arrows denote the direction of migration and the numbers denote the date (month) corresponding to the migration route of anchovy.

Anchovy (*Engraulis japonicus*) is a coastal migratory fish present along the entire coast of South Korea during the summer (Fig. 1) (Cha *et al.*, 1990). Anchovy fisheries dominate the South Korean fishing industry, manifesting very high economic value because anchovies are commercially useful from the fry stage onward (Kim and Lo, 2001; Oh, 2018). Given that all body parts are edible and readily consumed (*e.g.*, for broth in most Korean soups) in Korean food culture, investigation of the accumulation of artificial radionuclides in anchovies in the Korean seas is critical. Moreover, anchovies captured in the coastal waters of the Korean Peninsula are classified into four to six groups (within the same species) according to their growth stages, thereby rendering them a useful fish species for research on the accumulation of artificial radionuclides.

The purpose of this study was to determine the level of artificial radionuclides ^{137}Cs and $^{239+240}$Pu in anchovies from Korean seas and to evaluate the accumulation patterns of these nuclides in anchovy – the fish most widely consumed by the people of South Korea in particular and East Asian countries in general.

2 Methods

2.1 Sample collection and pretreatment

The dried anchovy samples were collected from seafood markets along the southeastern coast of South Korea in 2018. Each box of samples weighed approximately 5 kg. The samples were classified into smallest (1.6–3 cm, XS), small (4–5 cm, S), medium (5–6 cm, M), and large (6–7 cm, L) categories. Before analysis, the dried anchovies were further dried in a drying oven (60 °C) for more than 24 h, after which the dry weight was recorded. The samples were then homogenized and stored in a desiccator. The entire body of each anchovy was used. Here, we noted that the anchovies caught off the coast of Korea vary in size, but only one species (*Engraulis japonicus*) lives and is used as a food ingredient for Koreans. Thus, our study valid for the whole country (around the Korean Peninsula), not for a specific part of Korea.

2.2 Analytical procedure

2.2.1 ^{137}Cs measurements

Four to five hundred grams of anchovy dry-weight was placed in a γ-counter container (Marinelli beaker, Eckert & Ziegler, Germany) and gamma nuclide analysis was performed using a high-purity Ge (HPGe) detector (P-TYPE coaxial detector; Mirion Technologies (Canberra BNLS) NV, San Ramon, CA, USA). The γ-spectrometer used in this study was 39.7 mm in depth and 61.5 mm in diameter and the distance from the window was 5.51 mm. The relative efficiencies of the two detectors were 30% and > 50%, respectively. Prior to conducting sample measurements, the γ-spectrometer was calibrated using a mixed γ standard solution [Multinuclide Standard, Isotope Products Laboratories (IPL), Burbank, CA, USA] that was geometrically the same type as ours (Marinelli beaker). Samples were counted for more than 3 days and the average chemical yields were approximately 70%.

2.2.2 Pu isotope ($^{239+240}$Pu) analyses

The homogenized samples (200–300 g) were incinerated in an electric furnace for 24 h at 600 °C. The temperature of the electric furnace was increased by 100 °C per hour for complete incineration, preventing carbonization. Next, the samples were spiked with the Pu tracer (^{242}Pu; 4 mBq per sample) and acidified with a 1:3 mixture of nitric acid (HNO_3) and

Table 1. Certified and measured values (Bq kg^{-1} based on dry mass) of radionuclide certified reference materials (CRMs).

Radionuclide	CRM	Certified value	Confidence interval ($\alpha = 0.05$)	Measured value ($n = 3$)
^{137}Cs	IAEA-414	5.18	5.12–5.22	5.18 ± 0.38
$^{239+240}Pu$	IAEA-414	0.12	0.116–0.123	0.116 ± 0.013

hydrochloric acid (HCl), then heated at 80 °C to decompose the organic matter. If a white residue remained, concentrated HNO_3 was added to complete the decomposition. After complete decomposition was verified visually, the samples were collected and evaporated to dryness.

The dried samples were dissolved with 50 mL of 8 N HNO_3 and extracted upon passage through an anion-exchange resin column (AGI-X8, 100–200 mesh; Bio-Rad Laboratories, Hercules, CA, USA). Next, Pu isotopes were eluted using 0.1 N NH_4I/9N HCl. The eluted samples were fully dried and subjected to one additional column extraction. The purified Pu-elution solutions were again dried with HNO_3 and Na_2SO_4 to reduce Pu(VI) to Pu(IV) for electroplating by adsorption, then adjusted to a pH of 2.1–2.4 by adding sulfuric acid. The samples were transferred to an electrodeposition cell mounted with a stainless steel disc (16-mm diameter) and the Pu was separated under 1 Å for 1 h. Finally, the stainless steel discs electroplated with Pu were counted using α-spectrometry (PIPS detector with MCA, Mirion Technologies). Samples were counted for more than 5 days and the average chemical yields were approximately 80%. The measurement data were calibrated based on the chemical yields of spiked ^{242}Pu.

2.2.3 Method validation

To validate our analysis methods, radionuclide certified reference material (CRM) IAEA-414 (mixed fish from the Irish and North Seas; IAEA, 2013) from the International Atomic Energy Agency (IAEA) was analyzed together with the samples. The analytical results of the CRM were in good agreement with the certified values for ^{137}Cs and $^{239+240}Pu$ (Tab. 1).

3 Results and discussion.

3.1 Accumulation of ^{137}Cs in anchovies

The ^{137}Cs activity measured in each anchovy sample is listed in Table 2. The average ^{137}Cs activity in the anchovies was 74 ± 11 mBq kg^{-1}, 121 ± 16 mBq kg^{-1}, 132 ± 16 mBq kg^{-1}, and 137 ± 19 mBq kg^{-1}, respectively, in the smallest (XS), small (S), medium (M), and large (L) anchovy. Overall, the ^{137}Cs activity increased with increasing body size (length) ($p < 0.05$) (Fig. 2a). According to Kim *et al.* (2019), it was found that ^{137}Cs accumulates as fish grow (for Spanish mackerel, mullet, and pollack, etc.) in the Korean seas. The higher ^{137}Cs concentration in larger anchovies in this study also seems to be related to artificial radioactivity such as ^{137}Cs accumulation in anchovy according to growth stage.

The ^{137}Cs activity in anchovy was compared with that of other marine organisms collected in recent years (2015–2017) (Fig. 3a). The average ^{137}Cs activity in anchovy was similar to that in other fishes. However, the average ^{137}Cs activity was 1–2 times lower than that of microalgae and 2–3 times higher than that of crustaceans and mollusks (Kim *et al.*, 2019) (Fig. 3a). Since ^{137}Cs in seawater is soluble, it is easily taken up by living marine organisms. In this study, we did not determine the exact mechanism by which ^{137}Cs accumulates in anchovy during growth, but we think it involves both: i) the uptake of seawater containing artificial nuclides from current NPP operations as well as past NPP accidents; and ii) elevated levels due to bioaccumulation by grazing.

3.2 Accumulation of $^{239+240}Pu$ in anchovy

The $^{239+240}Pu$ activity measured in each anchovy sample is listed in Table 2. The mean $^{239+240}Pu$ activity in anchovy was 0.27 ± 0.10 mBq kg^{-1}, 1.58 ± 0.26 mBq kg^{-1}, 3.21 ± 0.37 mBq kg^{-1}, and 1.19 ± 0.23 mBq kg^{-1}, respectively, in smallest (XS), small (S), medium (M), and large (L) anchovy. Overall, $^{239+240}Pu$ activity increased with increasing body size, yet activity decreased noticeably in the large anchovy (Fig. 2b). The $^{239+240}Pu$ activity in anchovy in this study was compared with that of other marine organisms collected in recent years (2015–2017) (Fig. 3b) in the same manner as ^{137}Cs. The $^{239+240}Pu$ activity in anchovy was 1–2 orders of magnitude higher than that in all other marine organisms observed, including fish, crustaceans, and mollusks (Kim *et al.*, 2019) (Fig. 3b). This significantly higher $^{238+239}Pu$ in anchovy is likely to be attributed to the anchovy's unique migratory pattern that circulates the entire coastal area around the Korean Peninsula where several domestic NPPs are located (Fig. 1).

Pu is known to pose a significant health risk to humans *via* exposure through food or respiration. A previous study suggested that soluble Pu is associated with certain diseases, including tumors in bones, the liver, and other organs (Beyea and von Hippel, 2019).

In this study, improved food selectivity in anchovy following growth may be a more important mechanism of $^{239+240}Pu$ accumulation (Fig. 2b) than seawater uptake because Pu(IV) (*i.e.*, PuO_2) is relatively insoluble in seawater, in contrast to ^{137}Cs (Lindahl *et al.*, 2010). In general, as an anchovy grows, feeding rate increases due to the increase in: i) mouth size, ii) swimming ability, and, above all, iii) food search ability. As a strategy to increase the survival rate, it has been known that the improvements of food selectivity of adult anchovy especially could lower the rate of empty stomach (Kim *et al.*, 2013; Yoo and Jeong, 2016).

In contrast to ^{137}Cs, $^{239+240}Pu$ activity in adult (large, L) anchovies decreased 2–3-fold in this study, suggesting that certain *in vivo* mechanisms for reducing (artificial) radionuclides are at work in anchovies. Yue *et al.* (2018) reported that complexes of uranium (U), an actinide element similar to

Table 2. The body length of each anchovy sample group, activities of artificial radionuclides, ^{137}Cs and $^{239+240}$Pu, and calculated concentration factors (CFs) of these nuclides in this study. All uncertainties in this study are denoted as standard deviations for the measured values.

Species (Scientific name)	Body length (cm)	Activity (mBq/kg)		Concentration factor (CFs)	
		^{137}Cs	$^{239+240}$Pu	^{137}Cs	$^{239+240}$Pu
Engraulis japonicus	1.6 ~ 3 (smallest)	74 ± 11	0.27 ± 0.10	10.9 ± 2.2	14.0 ± 6.1
	4 ~ 5 (small)	121 ± 16	1.58 ± 0.26	17.8 ± 3.3	80.4 ± 19.1
	5 ~ 6 (medium)	132 ± 16	3.21 ± 0.37	19.4 ± 3.4	162.8 ± 30.8
	7 ~ 8 (large)	137 ± 19	1.19 ± 0.23	20.2 ± 3.4	60.7 ± 16.1

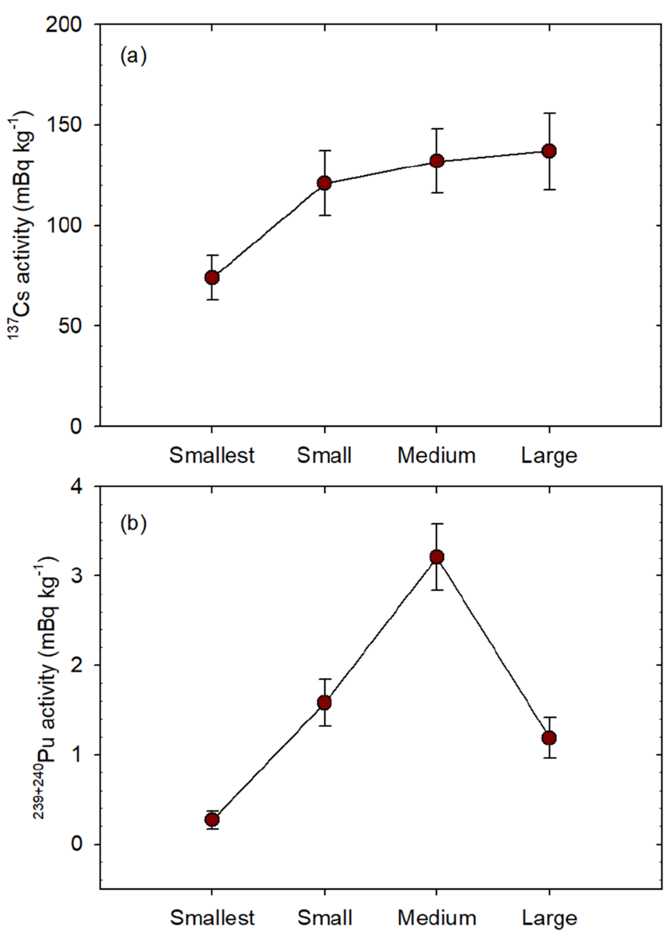

Fig. 2. Activities of (a) ^{137}Cs and (b) ^{239,240}Pu in anchovy according to their size.

$^{239+240}$Pu, with chelating ligands constitute a common mechanism of U detoxification. In particular, metallothionein (synthesized in the liver and kidney) is often considered a significant biochemical antidote, complexing with metals and limiting the availability of toxic chemical substituents (*e.g.*, metals or metalloids) in the cells and tissues of fish such as anchovy (Shi, 1990; King *et al.*, 1997; Kang, 2006; Thompson and Bannigan, 2008; Annabi *et al.*, 2013). Moreover, a recent study found that this low-molecular-weight protein shows efficient detoxifying effects based on acute depletion of U in land animals (Jiong *et al.*, 2010). Therefore, we think that $^{239+240}$Pu accumulated in marine organisms can be sufficiently removed as a result of the metallothionein effect. Furthermore, a more recent study reported that the accumulation of $^{239+240}$Pu was more concentrated in the internal organs than in muscle or skin (or exoskeleton), unlike the accumulation ^{137}Cs (mainly accumulated in muscle) (Kim *et al.*, 2020). This implies that $^{239+240}$Pu can be more efficiently detoxified than ^{137}Cs (as revealed in the present study) in adult fish because metallothionein metabolism mainly occurs in internal organs (*e.g.*, the liver).

3.3 Accumulation rate of ^{137}Cs in anchovies compared to other fishes

In order to compare and evaluate the enrichment pattern of the artificial radionuclide ^{137}Cs according to the growth stage in anchovies and other fish species (Kim *et al.*, 2019), ^{137}Cs activity was plotted against the mean age of fish estimated from body length (data applied from the Korean Fisheries Life Resources Information Center, https://www.nifs.go.kr/frcenter/) (Fig. 4). The activity of ^{137}Cs in other fish samples (anchovy, Spanish mackerel, pollack, mullet, and flounder; data from Kim *et al.* (2019)) increases as fish size increases. The activity of ^{137}Cs in anchovy was lower than that in other fish species, except flounder. However, considering the accumulation rate and the coefficient of exponential growth (fitting) curve of anchovy (1.36) (Fig. 4), the accumulation rate of ^{137}Cs in anchovy was significantly higher than that of Spanish mackerel (0.16), pollack (0.049), and mullet (0.0042) (Fig. 4). This relatively higher ^{137}Cs accumulation rate considering the ages of each fish species seems to be due to the anchovy's unique migratory pattern, circulating the entire coastal area around the Korean Peninsula where several domestic NPPs are located (Fig. 1).

3.4 Concentration factors of artificial nuclides in anchovy

The level of radioactive contamination in anchovy can be evaluated by quantifying the concentration factors (CFs) of

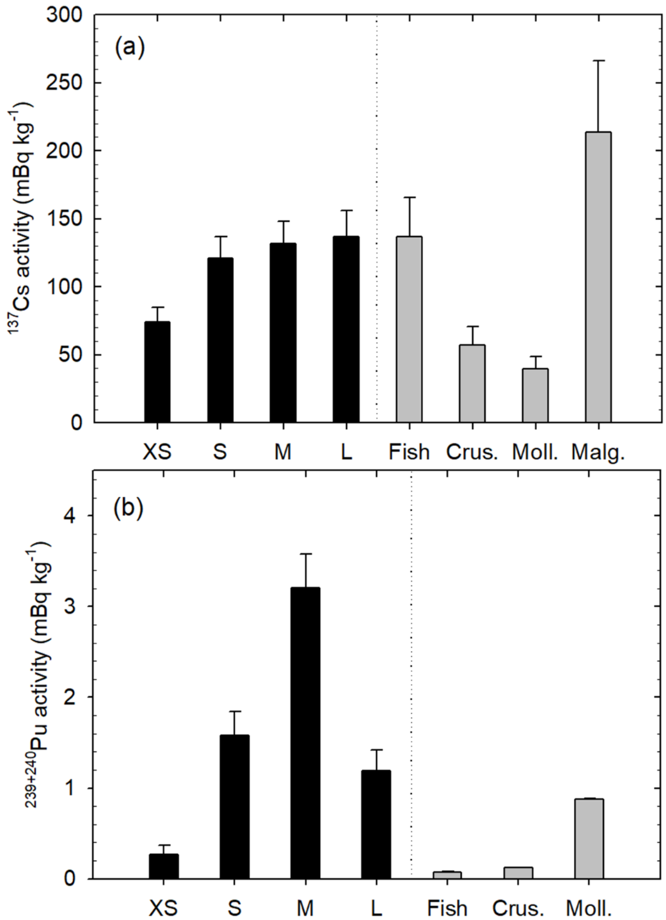

Fig. 3. Comparisons of activities of (a) ^{137}Cs and (b) ^{239,240}Pu in anchovy (black bars) according to size with other groups of marine organisms such as other fish, crustaceans (Crus.), and mollusks (Moll.) (grey bars). The smallest-, small-, medium-, and large-anchovies are expressed as XS, S, M, and L, respectively, for convenience in visualization.

nuclides in marine organisms based on mean activities in seawater. The CFs of the radioactive nuclides were determined as follow (IAEA, 2004):

$$\mathrm{CF} = \frac{\text{Concentration per unit mass of biota}\,(\mathrm{Bq\,kg^{-1}\,wet\ weight})}{\text{Concentration per unit mass of seawater}\,(\mathrm{Bq\,kg^{-1}})}.$$

The calculated CFs for ^{137}Cs and $^{239+240}$Pu in anchovy used in this study are presented in Table 2. In this calculation, the average activities of ^{137}Cs (1.64 ± 0.09 mBq kg^{-1}) and $^{239+240}$Pu (4.78 ± 0.36 μBq kg^{-1}) in the surface water of Korean sea (from the 5 stations in the East/Japan Sea) were measured in February 2018–the same period during which samples were collected for this study. In general, the CFs in the organisms have a wet weight-based value. However, the anchovy samples used in this study were dried; thus, we noted that our CF results were corrected by multiplying 0.2389 to convert to (estimated) wet weight, applying a previously reported value by Kim *et al.* (2017).

The quantified CFs of ^{137}Cs and $^{239+240}$Pu by anchovy consumption are presented in Table 2. The calculated CFs of

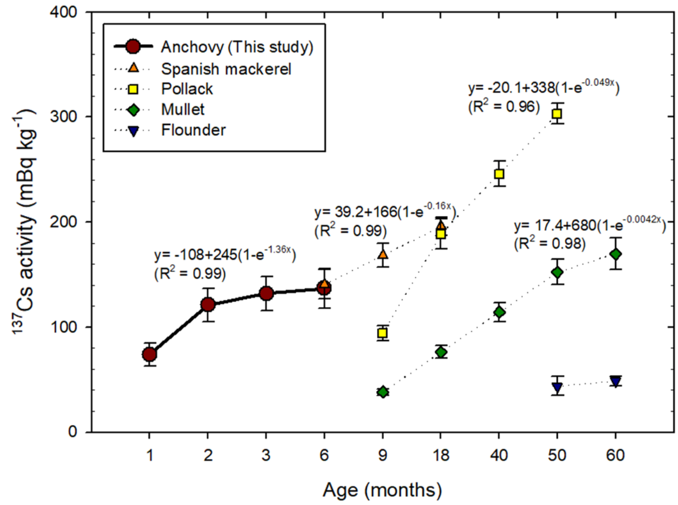

Fig. 4. The plot of mean ^{137}Cs activities in anchovy against their age (months) and comparisons with other fish species. Note that the x-axis does not represent a linear scale but is instead an arbitrary scale.

^{137}Cs were lowest in the smallest anchovy (10.9 ± 2.2) and highest in large anchovy (20.2 ± 3.9). The calculated CFs of $^{239+240}$Pu were also lowest in the smallest anchovy (14.0 ± 6.1) and highest in medium anchovy (162.8 ± 30.1) (Fig. 5). In order to evaluate the accumulation levels of ^{137}Cs and $^{239+240}$Pu in anchovies, these results were compared with those of other marine products and their recommended CF values (IAEA, 2004) (Fig. 5). The calculated CFs of ^{137}Cs in anchovies in this study were generally lower than those of other marine products (other fishes, crustaceans, mollusks, and algae) collected in 2015–2017 (Kim *et al.*, 2019). The mean CF of ^{137}Cs (16.82 ± 1.33) in anchovies were also an order of magnitude lower than the IAEA recommendations (100 for ^{137}Cs in fish). On the other hand, the calculated CFs of $^{239+240}$Pu in anchovy is significantly higher than those in other fishes. In addition, the mean CFs of $^{239+240}$Pu in anchovies were comparable (61 and 80 in small- and large anchovy, respectively) or even higher (163 in medium anchovy) to the IAEA recommendations (100 for $^{239+240}$Pu in fish).

3.5 Quantification of the effective dose rate from anchovy

In this study, we tried to assess the potential risk of radionuclides to humans by quantifying the annual effective dose rate (AED) (Sv yr^{-1}) as follows:

$$AED = A_{AR} \times e(T) \times I_{yr},$$

where A_{AR} is the average activity of an artificial radionuclide in anchovy (Bq kg^{-1}) in this study, $e(T)$ is an effective dose coefficient (Sv Bq^{-1}) (1.30×10^{-8} and 2.50×10^{-7} Sv Bq^{-1} for ^{137}Cs and $^{239+240}$Pu, respectively) (ICRP, 2012), and I_{yr} is the amount of intake per year for a person in South Korea (kg yr^{-1}). The 2017 Food Balance Sheet from the Korea Rural Economic Institute (KREI) was applied to determine the amount of anchovy intake I_{yr} (assuming that anchovies at each growth stage were consumed equally).

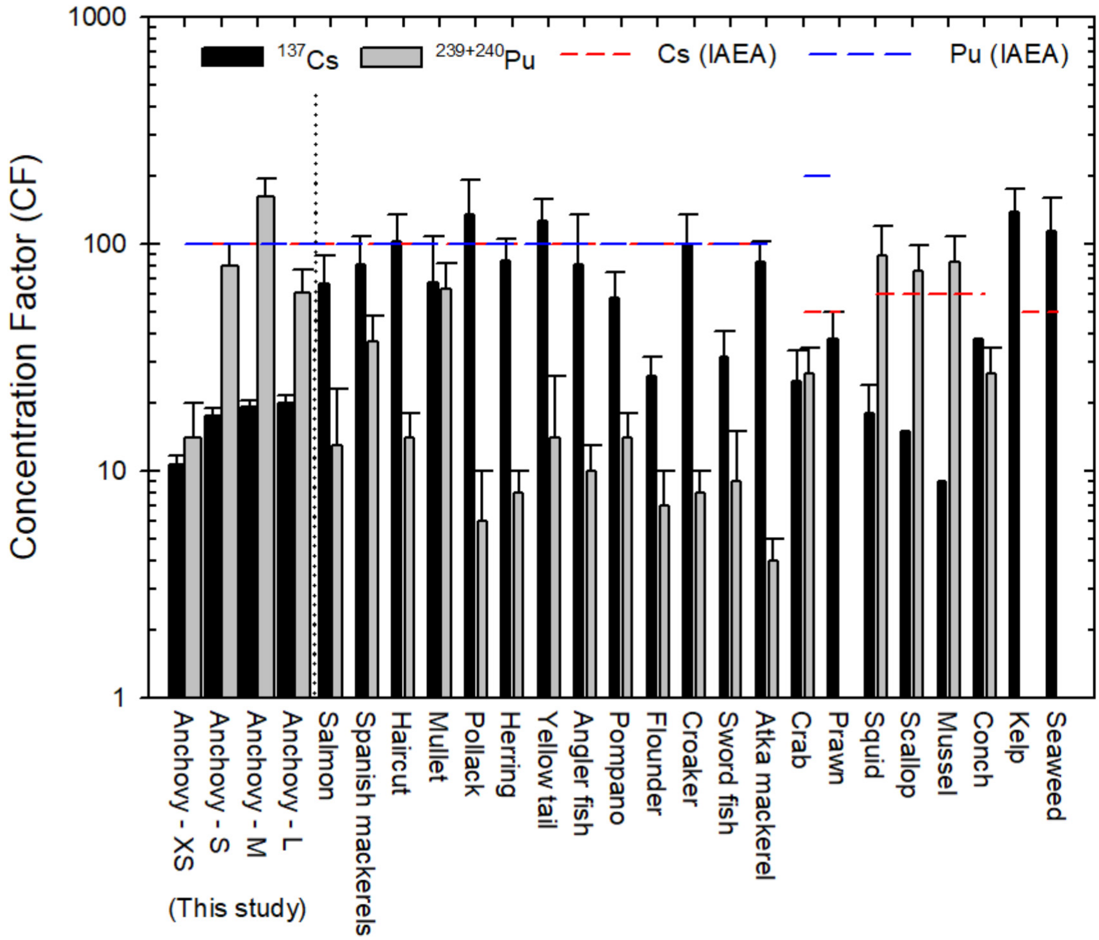

Fig. 5. Comparisons of concentration factors (CFs) of ^{137}Cs (black bar) and $^{239+240}$Pu (grey bar) in anchovy sample in this study and other marine organisms with IAEA-recommended CF values (red for ^{137}Cs and blue for $^{239+240}$Pu). The smallest-, small-, medium-, and large-anchovies are expressed as XS, S, M, and L, respectively, for convenience in visualization.

Table 3. The estimated annual effective dose (AED) rate ($\times 10^{-6}$ mSv yr^{-1}) of artificial radionuclides, ^{37}Cs and $^{239+240}$Pu, for a person in South Korea by anchovy consumption.

Body length (cm)	Annual intake per a person[a] (kg y^{-1})	AED of ^{137}Cs	AED of $^{239+240}$Pu
1.6–3	3.86	3.71 ± 0.55	0.26 ± 0.09
4–5		6.07 ± 0.80	1.52 ± 0.25
5–6		6.62 ± 0.80	3.10 ± 0.36
7–8		6.87 ± 0.95	1.15 ± 0.22

[a] 2017 Food Balance Sheet from Korea Rural Economic Institute (KREI, 2019).

The estimated AEDs of ^{137}Cs and $^{239+240}$Pu by anchovy consumption are presented in Table 3. The estimated AED of ^{137}Cs in anchovy was lowest in the smallest anchovy [$(3.71 \pm 0.55) \times 10^{-6}$ mSv yr^{-1}] and highest in large anchovy [$(6.87 \pm 0.95) \times 10^{-6}$ mSv yr^{-1}], showing an upward trend from smallest to largest anchovy (Tab. 3). The AED of $^{239+240}$Pu, estimated in the same manner as ^{137}Cs, was lowest [$(0.26 \pm 0.09) \times 10^{-6}$ mSv yr^{-1}] in the smallest anchovy and highest [$(3.10 \pm 0.36) \times 10^{-6}$ mSv yr^{-1}] in the medium anchovy. Although other artificial radionuclides such as ^{90}Sr were investigated in this study, the sum of the estimated AEDs of ^{137}Cs and $^{239+240}$Pu by anchovy consumption (approximately 30×10^{-6} mSv yr^{-1}) is significantly lower than that of ^{210}Po (9.4×10^{-2} mSv yr^{-1}) – a natural radionuclide – by entire seafood consumption for a person in South Korea. The total AED value by anchovy consumption in this study was also insignificant compared to the recommended AED limit of 1 mSv yr^{-1} suggested by the International Commission on Radiological Protection (ICRP, 2007).

4 Conclusion

We investigated the distribution and accumulation patterns of the artificial radionuclides ^{137}Cs and $^{239+240}Pu$ in anchovy, a major marine product in the Korean seas and the most popular fishery resource in Korean food culture. The concentration of ^{137}Cs in anchovy increased with increasing body length, indicating that ^{137}Cs accumulates in anchovy based on growth stages. The concentration of $^{239+240}Pu$ in anchovy also increased with growth, but sharply decreased in the adult stage. It is not entirely clear why $^{239+240}Pu$ decreased within a specific growth level, but we hypothesized that adult fish may have developed detoxification abilities against certain harmful radionuclides. The noticeably higher accumulation rate of ^{137}Cs in anchovy was likely due to the relatively short migratory period of anchovies in coastal regions. The concentration factors (CFs) of ^{137}Cs in anchovy were an order of magnitude lower than those in other marine organisms. However, the CFs of $^{239+240}Pu$ in adult anchovy were significantly higher than those in other fishes and comparable (or even higher) to the IAEA recommendations. However, we do not yet understand the exact mechanisms involved in $^{239+240}Pu$ depletion in adult anchovy; therefore, further research is needed. Moreover, further studies on the accumulation rate and detoxification of radionuclides in fishes are also necessary in the future to ensure that safe levels of harmful radionuclides are not exceeded in the typical Korean diet.

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