Distributions of Radiocesium and Plutonium in the Korean Seas and North Pacific after the Fukushima Accident, 2011–2014

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Highlights:
What are the main findings?
• The first radiocesium and Pu isotopes data in Korean seas were present in 2011–2012;
• Higher $^{134}$Cs were observed in Korean seas immediately after the Fukushima accident;
• Fukushima-derived nuclides were introduced to Korean seas by the atmosphere for up to 2 months.

What is the implication of the main finding?
• The residence times of $^{137}$Cs in Korean seas water is estimated to be 8–25 years;
• The dataset could be used to predict the dispersion of anthropogenic pollutant;
• The dataset could also be used to validate the model for future NPP accident around the Korean seas.

Abstract: The distributions of artificial radionuclides, radiocesium ($^{134}$Cs and $^{137}$Cs) and plutonium isotopes ($^{238}$Pu and $^{239+240}$Pu), in the surface water around the Korean seas (East/Japan Sea and Yellow Sea) in 2011–2012 and in three sections in the North Pacific between 2011 and 2014 were examined. The $^{137}$Cs activities in the surface water in the Korean seas in 2011 (immediately after the Fukushima nuclear power plant (NPP) accident on 17 March 2011) were comparable or not significantly different relative to those in 2010 and 2012. However, $^{134}$Cs, which had been not detected in the study area before the Fukushima accident (under the detection limit of 0.1 mBq kg$^{-1}$ level), were detected rapidly in 2011 after the accident (in about 60% of the 72 samples) and gradually disappeared due to their short half-life ($t_{1/2} = 2.06$ years) in 2012 (detected in about 16% of the 24 samples). In addition, the highest activities of radiocesium and Pu isotopes appeared locally in some stations of the Korean Strait region (located between Korea and Japan) within 1–2 months immediately after the accident. This suggests that the radioactive nuclides released immediately after the Fukushima accident were significantly introduced through the atmosphere, based on recent studies conducted in neighboring areas. We also showed that the spatial distribution of radiocesium in the North Pacific moved eastward from 2012 to 2014, and we attempted to quantify the residence time of radiocesium ($^{137}$Cs) in the Korean seas based on the long-term (tens of years scale) temporal trends of $^{137}$Cs activity data, which have been collected since the 1960s and 1970s. The estimated retention time of $^{137}$Cs in the East/Japan Sea and Yellow Sea were $25 \pm 0.6$ and $8.0 \pm 0.1$ years, respectively. These results are expected to be used as a preliminary study for a potential future event of a marine radioactive accident (which, of course, cannot be predicted) and as basic data for predicting the influences of radionuclide releases in the ocean.

Keywords: artificial radionuclides; radiocesium; plutonium; Korean seas; fukushima nuclear power plant (NPP) accident
1. Introduction

Radiocesium (Cs) isotopes $^{137}$Cs ($t_{1/2} = 30.08$ yr) and $^{134}$Cs ($t_{1/2} = 2.06$ yr) are artificial radioactive gamma ($\gamma$)-emitting isotopes released during nuclear weapons tests and from nuclear facility accidents [1]. The largest event of radioactive Cs was the Chernobyl nuclear power plant (NPP) accident in the USSR; it was documented that about $85 \times 10^{15}$ Bq of $^{137}$Cs was directly leaked into the ocean (direct input via Dnipro River) and atmosphere and a still significant amount of radionuclides remain in the sediments [2,3]. These Cs isotopes are very soluble in seawater [4], like other alkali elements. Therefore, Cs isotopes have been used as tracers to better understand processes related to ocean currents and water mass circulations [5–13].

The Plutonium (Pu) isotopes, $^{239}$Pu ($t_{1/2} = 24,110$ y) and $^{240}$Pu ($t_{1/2} = 6,561$ y), are alpha ($\alpha$)-emitting artificial radionuclides mainly released from atmospheric bomb tests during the Cold War (1950s to 1960s) [14–16]. Together with their very long half-lives, the reprocessing of nuclear fuel and nuclear facility accidents (such as Chernobyl) were also sources of $^{239}$Pu and $^{240}$Pu in the marine environment [17–19]. Since Pu isotopes are highly particle-reactive in the marine environment, they can be used as tracers for various oceanic processes, such as water mass transport, particle (export) flux, and sedimentation [20–22].

The Fukushima nuclear accident caused by the Great East Japan Earthquake on 11 March 2011 was designated as the highest grade (seventh) of the International Nuclear Event Scale (INES) established by the International Atomic Energy Agency (IAEA) [23]. After the Fukushima accident, a higher radioactive cesium concentration was observed in the waters close to the marginal seas of the North Pacific [24–27]. As a result, it was estimated that 15.2–18.3 PBq of $^{137}$Cs were released into the North Pacific and, eventually, were transported to North America and the Arctic Ocean along the main ocean current of the North Pacific [28–31]. Recently, some previous studies on Fukushima NPP-derived radionuclides were conducted in the marginal seas of the westernmost part of the North Pacific around the far east of the Asian continent. For example, substantially higher numbers of $^{134}$Cs and $^{137}$Cs than those reported in the past were observed in the East China Sea from 2011 to 2015 [32]. Aoyama et al. [33] suggested that radiocesium from the Fukushima accident could be transported to the waters around the Korean seas through subtropical mode water (STMW) and central mode water (CMW) (Figure 1), but there has been no direct evidence yet, unlike the reported results of the radionuclide activities in the Korean seas.

There was dumping of nuclear waste by Russia into the East/Japan Sea (in Korean seas) in the early 1990s [34]. More recently, besides the ocean dumping and the Fukushima NPP accident, there were several NPPs constructed rapidly in this region (a total of 24 nuclear power reactors are currently in operation), accounting for 26% of the total domestic electricity production [35] (Figure 1). In addition, neighboring countries in the far east of Asia (China, Japan, and Russia) also have more than 80 nuclear reactors in operation; it is a region highly dependent on nuclear power, accounting for the top 15% of the world’s 33 nuclear plants (https://www.iaea.org/pris, accessed on 1 September 2022). This status of radioactive facilities in the far eastern Asian regions suggests that both radioactive monitoring and preliminary case studies are necessary to prepare for potential future accidents in this region, since many anthropogenic activities related to nuclear energy issues are also planned. Therefore, the main objective of this study was to investigate the spatial and temporal distribution of radioactive Cs ($^{134}$Cs and $^{137}$Cs) and Pu isotopes ($^{238}$Pu and $^{239+240}$Pu) around the Korean seas (East/Japan Sea and Yellow Sea) directly after the Fukushima accident (2011–2012) (Figure 1), for the first time. We also investigated the three sectional distributions of $^{134}$Cs and $^{137}$Cs in the North Pacific surface water in 2012–2014 to monitor the long-range transport and influence of these artificial radionuclides. Based on these results, we also attempted to evaluate how the artificial radioactivity input, such as the Fukushima NPP accident, affected the surroundings of the Korean seas. We also expect that measured data in this study could be applied to improve and validate existing simulation models for predicting the dispersion of radioactive plumes into the ocean.
2. Materials and Methods

2.1. Sampling

Seawater sampling was conducted in Korean seas (East/Japan Sea and Yellow Sea, etc.) in 2011–2012 and in the North Pacific from June 2012 to July 2014 on several research cruises (R/V Ieodo and R/V Onnuri of the Korea Institute of Ocean Science and Technology (KIOST) and Icebreaker R/V Araon of Korea Polar Research Institute (KOPRI)) (Figure 1). Samples of water were collected by an on-board pumping system (>80 L per sample), immediately filtered (<1 μm), and acidified to pH < 2 by adding 6 M HCl (~50 mL).

2.2. Analytical Procedure

2.2.1. \(^{134}\)Cs and \(^{137}\)Cs Analysis

The water samples were co-precipitated with Mn through the addition of KMnO\(_4\) (25 mL per 60 L seawater) and 0.4 M MnCl\(_2\) solution (50 mL of 60 L seawater), adjusting the pH to 9–10 (by adding 10 M NaOH), and storing overnight. The settled Mn hydroxide was recovered in a beaker after the supernatant was centrifuged and removed (3500 rpm, 15 min). Then, the dried Mn co-precipitates were dissolved with 8N HNO\(_3\) in a 100 L beaker. The sample was heated to remove the organic matter with H\(_2\)O\(_2\). Then, this preconcentrated sample was co-precipitated with Ammonium Phosphomolybdate Trihydrate (AMP) (>24 h) by adding 13 g of AMP and 20 mg of Cs carrier (Stable Cs) to 13 g of the sample (with vigorous stirring for >1 h) \([12,36–38]\). After discarding the supernatant with a siphon, the precipitate was centrifuged (3500 rpm, 15 min). The remaining AMP co-precipitate was dissolved using 10 M NaOH and heated. Then, the sample was diluted with 500 mL of distilled water (DIW) and adjusted to pH ~2. The sample was co-precipitated again by adding 1 g of AMP (also stirring) for one day. After removing the supernatant by centrifuging, the remaining AMP precipitate was dissolved in 2–3 mL of 10 M NaOH and collected in a Marinelli beaker (10 mL volume with a height of 4 cm) \([12,36–38]\). The
radionuclides, $^{134}$Cs and $^{137}$Cs, were measured (counted) using a gamma ($\gamma$)-spectrometer (Mirion Technologies (Formerly known as Canberra), Meriden, CT, US) equipped with a high-purity Ge (HPGe) detector (Well-TYPE HPGe coaxial detector). The $\gamma$-spectrometer used in this study had a 35.50 mm depth, 59.70 mm diameter, and a 5.25 mm distance from the window, and the relative efficiencies were 25%. Prior to the sample analyses, the $\gamma$-spectrometer was calibrated using a mixed $\gamma$ standard solution (Multi-nuclide Standard, Eckert & Ziegler Isotope Products Laboratories (IPL), Santa Clarita, CA, US) in a sample container that was geometrically the same type as ours (Marinelli beaker). The average counting times were more than two days for all samples, and the average chemical yields were about 70%.

2.2.2. $^{238}$Pu and $^{239+240}$Pu Analysis

$0.2490 \pm 0.0009$ mL of $^{242}$Pu spike was added to the seawater sample immediately after the sample’s pretreatment (filtration and acidification). After waiting for the complete dissolution of the spiked solution (with vigorous stirring for >1 h), the sample was coprecipitated with Mn (the radiocesium analysis and the details of the Mn coprecipitation step were as shown in Section 2.2.1).

Then, this preconcentrated and redissolved (with HNO$_3$ and H$_2$O$_2$, described in Section 2.2.1) sample was extracted by solid-phase extraction (SPE) twice, using an ion exchange column (10–12 cm length of plastic column filled with anion exchange resin, AGI-X8, 100–200 mesh), sealed with glass/quartz wool (2.5 cm length). Briefly, 0.5 g of NaNO$_2$ was added to the samples to prepare the ion exchange. After washing the column with DIW, 50 mL of 8N HNO$_3$ were passed through the column for resin conditioning. Then, the samples were passed through the column at a speed of 1–2 mL min$^{-1}$ using gravity. The column was washed with 80 mL of 10N HCl, and the adsorbed samples were eluted using 100 mL of 0.1N NH$_4$I/9N HCl solution (at a flow rate of 1–2 mL min$^{-1}$. The eluted samples were dissolved in 5–10 mL of concentrative HNO$_3$ (heated), H$_2$O$_2$, and 10 mL of 10 N HCl in order to remove the excess iodine. Then, the fully dried samples were dissolved in 40 mL of 10N HCl repeatedly and purified by one more column extraction with approximately half the amount (~50 mL) of 1N NH$_4$I/9N HCl elution solution. The purified Pu eluted solutions were dried again with (2 mL of concentric) HNO$_3$ and (1 mL of) 0.3 M Na$_2$SO$_4$ (to reduce Pu$^{6+}$ to Pu$^{4+}$ for electroplating by adsorption) and dried with heat.

The samples were transferred to an electrodeposition cell mounted with a stainless-steel disc (16 mm diameter, pre-washed with acetone). To insert the sample disc into the plating cell, we washed the sample with a small amount of DIW, and 4 drops of 0.3% Thymol blue were added to neutralize with concentric ammonia. Then, the sample was adjusted to a pH of 2.2 by adding (1%) sulfuric acid and (1%) ammonia solution. The Pu was separated under 1 A for an hour. Finally, the stainless-steel discs electroplated with Pu were counted using $\alpha$-spectrometry (passivated implanted planar silicon (PIPS) detector with multichannel analyzer (MCA), Mirion Technologies (Formerly Canberra), US). The average counting times were more than five days for all samples. The average chemical yields were approximately 80%. The final activities were calibrated using the chemical yields expected by $^{242}$Pu.

3. Results and Discussion

3.1. Spatiotemporal Distributions of $^{134}$Cs and $^{137}$Cs in the Korean Seas, 2011–2012

The spatial distribution of radioactive Cs isotopes ($^{134}$Cs and $^{137}$Cs) in the Korean seas (East/Japan Sea and Yellow Sea) in 2011 and 2012 is shown in Figure 2. In 2011, the activities of $^{134}$Cs and $^{137}$Cs ranged between 0.11 and 1.10 mBq kg$^{-1}$ (average 0.38 $\pm$ 0.11 mBq kg$^{-1}$, n = 42) and 0.11 and 2.46 mBq kg$^{-1}$ (average 1.41 $\pm$ 0.12 mBq kg$^{-1}$, n = 74), respectively. In 2012, the activities of $^{134}$Cs and $^{137}$Cs ranged between 0.38 and 0.73 mBq kg$^{-1}$ (average 0.53 $\pm$ 0.20 mBq kg$^{-1}$, n = 4) and 0.17 and 1.85 mBq kg$^{-1}$ (average 1.22 $\pm$ 0.11 mBq kg$^{-1}$, n = 25), respectively (Figure 2). The range of $^{137}$Cs activities in 2011 was similar to that in 2012 within the uncertainty range (Figure 2). The $^{134}$Cs activities in 2011 and 2012 also
well with results of Kamenik et al. [41], where an average of 1.73 mBq kg\(^{-1}\) was observed in the East Sea and Yellow Sea between 2005 (first regular annual survey in the Korean seas) and 2010 [40]. Noticeably, detectable \(^{134}\)Cs activities were observed in more than half of the samples (42 of the 72 samples) in 2011. The highest \(^{134}\)Cs was 1.10 mBq kg\(^{-1}\), (the only data with \(>1\) mBq kg\(^{-1}\)) observed on 29 March 2011 in the Korean Strait (Figure 2) (a narrow strait between the Korean Peninsula and Japan as the mouth of the Pacific margin to the East/Japan Sea), just two weeks after the Fukushima NPP accident. This result agreed well with results of Kamenik et al. [41], where an average of 1.73 mBq kg\(^{-1}\) of \(^{134}\)Cs was detected in the North Pacific (between Japan and Hawaii) from June 2011 to September 2012. Lee et al. [42] reported that \(^{34}\)Cs were detected in rainwater (0.01 to 334 ± 74 mBq kg\(^{-1}\)) inland in South Korea, whereas \(^{134}\)Cs were not found in rainwater in the past [43]. Therefore, the substantially higher \(^{134}\)Cs in the Korean seas was likely from atmospheric deposition. \(^{134}\)Cs were not detected in most of the samples in 2012, likely due to the short half-life of \(^{134}\)Cs (2.06 yrs). However, \(^{134}\)Cs > 0.3 mBq kg\(^{-1}\) at the four stations in the East/Japan Sea were still detected (Figure 2), suggesting that artificially originated \(^{134}\)Cs activities measured in this study showed a similar range to those (0.8–1.4 Bq m\(^{-3}\) from April to June 2011) in the East China Sea and South China Sea (adjacent seas to our study area) [39].

![Figure 2](image_url)

**Figure 2.** The spatial distributions of the \(^{134}\)Cs ((a,b), upper panel) and \(^{137}\)Cs ((c,d), lower panel) activities in the Korean seas from 2011 ((a,c)) to 2012 ((b,d)). The red square indicates the region (Korean Strait) where the sample (data) was collected on 29–30 March 2011.
were still steadily being introduced into the Korean sea region more than one year after the accident.

Here, we used the radiocesium data from the annually reported national monitoring result of the Korea Institute of Nuclear Safety (22 stations in the Korean seas from 2005 to 2012) [44,45], because our radiocesium data did not match spatially in 2011 and 2012. The annual mean $^{134}$Cs and $^{137}$Cs activities in the Korean seas from 2006 to 2012 are shown in Figure 3. In terms of the $^{137}$Cs activities in the Korean sea water during the seven years of this period (including before and after the Fukushima accident), there was little difference within the uncertainty range. However, the sudden increase (1–2 orders of magnitude) in $^{134}$Cs activity between 2010 and 2011 that halved between 2011 and 2012 (Figure 3), suggested a direct influence of the input of artificial radionuclides in the Korean seas after the Fukushima NPP accident.

![Figure 3](image-url)

**Figure 3.** The comparisons of the annual mean activities of $^{134}$Cs and $^{137}$Cs in the Korean seas from 2006 to 2012. All the data were applied from the national report of Korea Institute of Nuclear Safety (KINS) [45]. We note that the data from 2011 and 2012 are in the same 22 locations, but the data before 2010 were only for eight stations per year before the expansion of the monitoring region due to the Fukushima accident.

3.2. Spatial Distributions of Pu Isotopes in the Korean Seas in 2011

The spatial distribution of the radioactive Pu isotopes ($^{238}$Pu and $^{239+240}$Pu) in the Korean seas (East/Japan Sea and Yellow Sea) in 2011 is shown in Figure 4. The activities of $^{238}$Pu and $^{239+240}$Pu ranged from 0.07 to 17.99 μBq kg$^{-1}$ (average 0.96 ± 0.16 μBq kg$^{-1}$, n = 44) and 2.47 to 66.58 μBq kg$^{-1}$ (average 9.38 ± 0.48 μBq kg$^{-1}$, n = 54), respectively. These $^{239+240}$Pu activities were about 2–3 times higher than the activity ranges (2.11–18.0 μBq kg$^{-1}$, average 4.91 μBq kg$^{-1}$, n > 100) of the previous five years (2006–2010) in the same area [45]. Regionally, there was not much difference between the Pu activities in the Yellow Sea and the East/Japan Sea (Figure 4).

However, the highest activities of both $^{238}$Pu and $^{239+240}$Pu (about 2–3 times higher than other samples) were observed in the Korean Strait region on 30 March (where the highest Cs activities were also observed in same period, see Section 3.1), just two weeks after the Fukushima NPP accident (Figure 4). Recently, Hirose [46] documented that Fukushima NPP accident-derived nuclides were transported to Fukuoka, Japan (the closest city to our research area, the East/Japan Sea), located right next to the Korean Strait (where the highest Cs and Pu levels were observed in this study, two weeks after the accident) from atmospheric deposition up to two months after the accident. In addition, Kaeriyama et al. [47] reported that the radioactive Cs activities suddenly increased in sinking particles
from sediment trap in the water column of the East/Japan Sea region (spatially close to (tens of kilometers distance) our study area) for one month after the Fukushima NPP accident, indicating the large input of atmospheric dust into the water column. These previous studies suggest that a significant number of radionuclides from the Fukushima NPP may have been introduced from atmospheric depositions into the Korean seas [48–50].

Figure 4. The spatial distributions of: (a) $^{238}$Pu; and (b) $^{239+240}$Pu and activities. The red square indicates the region (Korean Strait) where the samples (data) were collected on 29–30 March 2011 (same region as in Figure 3).

3.3. Sectional Distribution of $^{134}$Cs and $^{137}$Cs in the North Pacific

The spatial distributions of the $^{134}$Cs and $^{137}$Cs in the North Pacific from 2012 to 2014 are shown in Figure 5. Overall, the Cs activity was higher in the northern section (Section 1 in Figure 5) than in the south (Sections 2 and 3 in Figure 5). The activities of $^{134}$Cs and $^{137}$Cs ranged from 0.13 to 6.24 mBq kg$^{-1}$ (average $1.77 \pm 0.31$ mBq kg$^{-1}$, $n = 9$) and 0.73 to 8.95 mBq kg$^{-1}$ (average $2.08 \pm 0.19$ mBq kg$^{-1}$, $n = 26$), respectively, in 2012; 0.36 to 0.97 mBq kg$^{-1}$ (average $0.61 \pm 0.07$ mBq kg$^{-1}$, $n = 9$) and 0.18 to 2.92 mBq kg$^{-1}$ (average $1.44 \pm 0.06$ mBq kg$^{-1}$, $n = 48$), respectively, in 2013; and 0.52 to 0.61 mBq kg$^{-1}$ (average $0.56 \pm 0.04$ mBq kg$^{-1}$, $n = 2$) and 0.95 to 1.97 mBq kg$^{-1}$ (average $1.44 \pm 0.02$ mBq kg$^{-1}$, $n = 13$), respectively, in 2014 (Figure 5). The maximum $^{137}$Cs activity was $8.95 \pm 0.64$ mBq kg$^{-1}$ in 2012 at 49° N and 162° E (Figure 5), and $1.65 \pm 0.08$ mBq kg$^{-1}$ and $1.47 \pm 0.10$ mBq kg$^{-1}$ in 2013 and 2014, respectively (Figure 5). Overall, both the $^{134}$Cs and $^{137}$Cs activities decreased over time. In addition, the location of the highest $^{137}$Cs activity moved eastward, such as 49° N, 162° E (8.95 $\pm$ 0.64 mBq kg$^{-1}$) in 2012; 47° N, 162° E (2.92 $\pm$ 0.12 mBq kg$^{-1}$) in 2013; and 50° N, 166° E (1.97 $\pm$ 0.11 mBq kg$^{-1}$) in 2014 (Figure 5). Previous studies have documented that this radionuclide transport in the North Pacific is a complex influence of the main ocean current and atmospheric input [50,51]. Although less radioactive cesium was observed in the North Pacific in 2014 compared to 2012, Smith et al. [52] observed radioactive cesium from Fukushima off the west coast of Canada in August 2014, indicating that radioactive cesium from Fukushima may have been transported continuously during this period [33,53].

3.4. Isotopic Ratios of Radionuclides in the Korean Seas and the Pacific

Buesseler et al. [54] reported that the $^{134}$Cs/$^{137}$Cs activity ratio was close to 1 after the Fukushima accident, which can be used to identify the source of radioactive Cs and relevant radionuclides. For example, Aoyama et al. [33] reported that the $^{134}$Cs/$^{137}$Cs
activity ratio in Hasaki, Japan was in the range of 0.81 ± 0.07–1.08 ± 0.08 in the surface waters from March 2011 to October 2012 and from June 2014 to June 2016.

Figure 5. The sectional distributions of radiocesium, $^{134}$Cs (upper panel, (a–c)); and $^{137}$Cs (lower panel, (d–f)), from three (#1 to #3) sections in the North Pacific, 2012–2014.

In this study, $^{134}$Cs/$^{137}$Cs was 0.20 ± 0.08 (n > 40) in 2011 and 0.08 ± 0.56 (n = 4) in 2012 in the Korean seas (Figure 6), which was lower than the initial ratio of 1.0. According to previous study [45], the $^{134}$Cs/$^{137}$Cs ratio in the Korean seas (from 30 March to 8 June 2011) was 0.1–0.5, which agreed well with our results. These relatively lower $^{134}$Cs/$^{137}$Cs could be due to the dilution effect in both the ocean and the atmosphere (e.g., the air characteristics of South Korea controlled by the Westerlies) [55]. Immediately after the accident, a large amount of radioactive cesium fallout entered and exited the atmosphere, and the tendency of the observation value to decrease over time was similar to the change in our study results.

Figure 6. The activity ratios: (a) $^{134}$Cs/$^{137}$Cs in the Korean seas (2011); (b) $^{238}$Pu/$^{239+240}$Pu (2011) in the Korean seas; and (c) $^{134}$Cs/$^{137}$Cs in the North Pacific (2012).
The $^{134}$Cs/$^{137}$Cs ratios in the North Pacific section were 0.13–0.80 (average 0.38 ± 0.03, n = 9) in 2012, 0.18–0.51 (average 0.31 ± 0.04, n = 9) in 2013, and 0.38–0.45 (average 0.42 ± 0.005, n = 2) in 2014 (Figure 6). $^{134}$Cs was no longer detected in 2014 indicating the Fukushima NPP-derived $^{134}$Cs decayed due to their short half-life ($t_{1/2} = 2.06$ yr).

The specific difference in $^{238}$Pu/$^{239+240}$Pu appears to be due to the chemical and physical properties of $^{238}$Pu and $^{239+240}$Pu, as $^{238}$Pu is known to have high environmental mobility [56]. Based on this, we also attempted to investigate the $^{238}$Pu/$^{239+240}$Pu ratio to determine the source of the Pu in the Korean sea water (Figure 6). The $^{238}$Pu/$^{239+240}$Pu ratio was 0.27 and 2.20 in March and April 2011, respectively. This result agreed well with previous research, in which the Fukushima nuclear accident-derived $^{238}$Pu/$^{239+240}$Pu was reported to be 2.97 [57]. Therefore, the Pu during this period (March to April 2012) of this study can be regarded as originating from the Fukushima accident. We noted the remarkably higher $^{238}$Pu/$^{239+240}$Pu observed in the Yellow Sea (Figure 6). We believe that this higher $^{238}$Pu/$^{239+240}$Pu was due to the resuspension of Pu (already deposited in the past) in the shallow level of the Yellow Sea (average ~80 m depths) where the suspended sediment concentrations were also remarkably high (>10–100 mg L$^{-1}$) [58], but no direct evidence has been found yet.

The date of the Fukushima nuclear accident (11 March 2011) was set as D-day (day 0), and the $^{239+240}$Pu/$^{137}$Cs ratio sampling over time in all the seawater samples in this study is presented in Figure 7. The $^{239+240}$Pu/$^{137}$Cs ratio gradually decreased with time. A previous study reported that the average $^{239+240}$Pu/$^{137}$Cs ratio between 2006 and 2010 in our study area ranged from 0.002 to 0.006, and that from the global fallout level of nuclear tests (in the 1970s) was 0.011 [45] (Figure 7). This result implies that the radionuclides released into the Korean Sea from Fukushima NPP had a high impact within 20 days after the accident and then returned to the background concentration levels of the past.

3.5. Estimation of the Mean Residence time of $^{137}$Cs in Korean Seas

The effective environmental half-life of $^{137}$Cs is a good indicator to evaluate the actual behavior of $^{137}$Cs in the ocean. In the marine environment, $^{137}$Cs activity in the surface
seawater decreases exponentially with time. The temporal change in the $^{137}$Cs activity in the ocean can be expressed by the following exponential Equation (1) [59]:

$$A_t = A_0 \exp(-at)$$

(1)

where $A_t$ is the $^{137}$Cs activity during year $t$ (mBq kg$^{-1}$), and $A_0$ is the initial $^{137}$Cs activity in $t_0 = 1972$ (Yellow Sea) and 1964 (East/Japan Sea) (mBq kg$^{-1}$). The estimated effective environmental half-life ($T_{EF}$) of $^{137}$Cs in the East/Japan Sea and the Yellow Sea were $11.1 \pm 0.1$ years and $4.7 \pm 0.1$ years (from the coefficients, $\alpha$, from exponential (decay) fitting curve in Figure 8), based on the compiled $^{137}$Cs data in the Korean seas since the 1960s (data from the IAEA MARIS (IAEA’s MARine Information System, https://maris.iaea.org/, accessed on 1 September 2022) web database). Then, the mean residence time $T_m$ of $^{137}$Cs becomes a realistic indicator and can be expressed through the following Formula [60] (2).

$$\frac{1}{T_m} = \frac{\ln(2)}{T_{EF}} - \frac{\ln(2)}{T_R}$$

(2)

where $T_{EF}$ is the effective environmental half-life, and $T_R$ represents the radiological half-life ($T_R = 50.17$ yrs) of $^{137}$Cs. The estimated residence time ($T_m$) of the $^{137}$Cs in the East/Japan Sea and the Yellow Sea were $25 \pm 0.6$ years and $8.0 \pm 0.1$ years, respectively, which is comparable to that in other adjacent marginal seas; the East China Sea ($45.6 \pm 3.8$ years), South China Sea ($36.8 \pm 3.1$ years) and (Chinese part of) Yellow Sea region ($12.0 \pm 1.0$ years) [39] were calculated in the same manner.

![Figure 8](image-url) The long-term trends (1960s to present) of $^{137}$Cs activity in the Korean seas. Past data other than those measured in this study were downloaded and applied from the IAEA MARIS (IAEA’s MARine Information System, https://maris.iaea.org/, accessed on 1 September 2022) web database. The environmental removal constant (exponential decrease) to define the environmental half-life was obtained by the coefficient ($k$) of the best-fitting curve (of exponential decay, $A = A_0 \exp(-kt)$), ($0.146$ yr$^{-1}$ and $0.062$ yr$^{-1}$ in Yellow Sea and East/Japan Sea, respectively) The fitting curve equation is calculated by with Sigma–Plot professional 10.0 software.

4. Conclusions

The distributions of artificial radionuclides, radiocesium ($^{134}$Cs and $^{137}$Cs) and plutonium isotopes ($^{239}$Pu and $^{239+240}$Pu), in the surface water around the Korean seas (East/Japan Sea and Yellow Sea) in 2011 and 2012 and in the three sections of the North Pacific from 2012...
to 2014 were determined. The $^{137}\text{Cs}$ activities in the surface water in 2011 were comparable or not significantly different in the Korean seas (immediately after the Fukushima nuclear power plant (Fukushima NPP) accident on 17 March 2011) relative to those in 2012. $^{134}\text{Cs}$, which was not detected in the study area before the Fukushima accident, was detected throughout 2011, from immediately after the accident; then, it gradually disappeared in 2012 due to its short half-life ($t_{1/2} = 2.06$ years). In addition, the highest activities of radionuclides released immediately after the Fukushima accident were mainly introduced through the atmosphere based on recent previous studies conducted in neighboring areas.

We also showed that the pathway that released radionuclides gradually moved from the North Pacific to the east from 2012 to 2014. We attempted to estimate the residence time of radionuclides ($^{137}\text{Cs}$) in the Korean seas based on the long-term trends of the $^{137}\text{Cs}$ data (from the 1960s). The mathematically calculated residence time of $^{137}\text{Cs}$ in the East/Japan Sea and Yellow Sea were $25 \pm 0.6$ and $8.0 \pm 0.1$ years, respectively. These results are expected to be used as a preliminary study for potential future events of marine radioactive accidents. The dataset in this study would also provide significant information to the national authority for defining the radiological state of the specific marine compartments using existing codes (such as ERICA assessment tool (https://erica-tool.com/, accessed on 1 September 2022)). Furthermore, the dataset could be used to study the dispersion of anthropogenic pollutants in areas close to NPPs [61,62] in the future, and also for the validation of the models (e.g., dispersion and ecosystem). To predict and evaluate the potential influences of radionuclide releases into the ocean more accurately, more extensive studies are necessary, including: (i) the continuous monitoring of radionuclides in the marginal seas around the far eastern Asian region, especially where the major NPPs are concentrated; (ii) the quantitative inventory of the current radionuclide level including the deeper water column (based on activity data along with depth) in this region; (iii) the advancement of marine transport models of radionuclides based on the radionuclide inventory; and (iv) rapid detection technology for extremely low levels of radioactive nuclides in marine samples that require large amounts of sample volume, such as ~100 L of seawater from marine samples.

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