Distribution, dynamics, and fate of radiocesium derived from FDNPP accident in the ocean

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\textbf{ABSTRACT}

This paper gives an overview of the research results obtained in 10 years after the accident at Fukushima Daiichi Nuclear Power Plant (FDNPP), focusing on the distribution and dynamics of $^{137}$Cs, which is one of major accident-derived radionuclides. Immediately after the FDNPP accident, 8–21 PBq of $^{137}$Cs was transported to the ocean mainly due to direct discharge to the ocean, and deposition in the ocean via the atmosphere. $^{137}$Cs in seawater traveled eastward on the surface of the North Pacific Ocean along the Kuroshio Extension over a period of several years. Some of $^{137}$Cs is also recirculated to the western margin of the subtropical Pacific via the intermediate layer. The concentration of radiocesium in marine organisms also increased immediately after the accident, and then decreased over time. The concentration of radiocesium in brackish and demersal fishes decreased at a slower rate than the concentration of radiocesium in surface-dwelling fish. The amount of $^{137}$Cs accumulated on the seafloor is only about 1\% (0.2PBq) of the amount carried to the ocean, but it remains in the sediments in the coastal area for a long period of time and gradually migrates to the seawater and ecosystems near the seafloor.

\textbf{1. Introduction}

The tsunami with a height of more than 10 m caused by the Great East Japan Earthquake in 2011 caused a loss of power at the Fukushima Daiichi Nuclear Power Plant (FDNPP) operated by Tokyo Electric Power Company Holdings (TEPCO). As a result of extremely rising of the temperature and pressure inside the reactors, a large amount of radioactive material was released into the environment. A major feature of the environmental impact by this accident is that amounts of radionuclides were released directly into the ocean. In addition to monitoring surveys by national and local governments, academic research surveys by many universities/institutes were carried out, highlighting the distribution and the dynamics of radionuclides in the ocean.

In this accident, various radionuclides generated in the reactor during the operation were released to the environment, most of which had a half-life of less than one year. On the other hand, $^{137}$Cs, which remains in the environment for a relatively long time, is regarded as the main radionuclide to be monitored, and many findings have been reported. The accident-derived radionuclides in the ocean are (1) discharged directly into the ocean, (2) deposited in the ocean surface after being released into the atmosphere, and (3) supplied from rivers after being deposited on the land surface. Supply from the processes (1) and (2) are thought to have caused major pollution in the ocean immediately after the accident.

On 13 April 2021, the Japanese Government decided to release ‘treated water,’ from which radionuclides have been removed by a multineuclide removal system such as Advanced Liquid Processing System (ALPS) from the contaminated water generated at the facility, into the ocean. The National Committee of Experts has carefully planned to ensure that the impact of this ocean release on the marine environment and ecosystems is sufficiently small. To predict the fate of the treated waters in the ocean, it would also be helpful to properly review the dynamics of radionuclides released into the ocean for 10 years after the accident.

This review paper outlines the transport of radiocesium (mainly $^{137}$Cs), major accident-derived radionuclides, in two timeframes: immediately after the accident (about one year) and then to the present. Especially in the latter case, the migration of the radionuclides in seafloor and marine organisms where the effects of radionuclides remain in the long term will be explained. Finally, we will summarize the matters that should be noted in understanding the marine environment in future.
2. Radiocesium in the coastal seawater

2.1. Release amount into the ocean

In understanding the marine pollution due to radioisotopes shortly after the FDNPP accident, two major pathways of radionuclides into the ocean should be considered. One was direct discharge of radionuclides from the facility into the ocean. On 2 April 2011, TEPCO reported that water containing a high concentration of radionuclides was leaking from a concrete crack near the intake of FDNPP Unit 2 [1]. TEPCO stopped the visible leakage by injecting water glass (sodium silicate) into a pit near reactor 2 from 6 to 21 April 2011. It was difficult at that time to estimate this release rate, because there was little information on the accident. Under such circumstances, Kawamura et al. [2] estimated the $^{137}$Cs release rate based on the surface $^{137}$Cs concentration data and the report by the Prime Minister of Japan and His Cabinet [3]. The surface $^{137}$Cs concentration data were monitored at the northern and southern discharge channels of FDNPP. TEPCO reported that the total $^{137}$Cs release amount during 120 hours over the period of April 1–6, 2011 was $0.94 \times 10^{15}$ Bq. Kawamura et al. [2] converted the concentration to the release rate by multiplying the constant obtained from the comparison of the concentration with the information by TEPCO. Finally, they extended this estimate over the period of March 21–30 April 2011 and estimated the total $^{137}$Cs release amount in this period to be approximately $4 \times 10^{15}$ Bq. Tsumune et al. [4] also estimated the $^{137}$Cs release rate using a regional ocean model and monitoring data. They suggested by $^{131}$I/$^{137}$Cs activity ratio in monitoring data that direct discharge into the ocean started from 26 March 2011. The total $^{137}$Cs release amount from March 26 to the end of May, 2011 was estimated to be $(3.5 \pm 0.7) \times 10^{15}$ Bq, which corresponds to the estimate by Kawamura et al. [2]. Then, Kawamura et al. [5] revised the $^{137}$Cs release rate, which indicated that the total $^{137}$Cs release amount from March 26 to 30 June 2011 was approximately $3.53 \times 10^{15}$ Bq. They also estimated the amount of atmospheric deposition of $^{137}$Cs to the sea surface, which was another pathway of $^{137}$Cs into the ocean, and it was suggested that the cumulative $^{137}$Cs deposition to the sea surface in the North Pacific from March 11 to 31 May 2011 was approximately $9.89 \times 10^{15}$ Bq. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [6] compiled these estimations with some other estimates and summarized that the amount of $^{137}$Cs released into the marine environment to be $3 \times 10^{15}$–$6 \times 10^{15}$ Bq by direct release from FDNPP and $5 \times 10^{15}$–$1.1 \times 10^{16}$ Bq by the atmospheric deposition. Besides, Aoyama et al. [7] estimated the accumulated $^{137}$Cs deposition amount at the sea surface in the North Pacific to be $1.5 \times 10^{16}$–$1.8 \times 10^{16}$ Bq based on observational data and model simulations. While UNSCEAR estimates only consider estimates from the landside, Aoyama et al. [7] consider the overall mass balance of radiocesium, including the abundance not only on land but also in the ocean. As a result, they reported a higher estimate than UNSCEAR.

By the outflow of contaminated water to the FDNPP’s port, which is thought to have occurred around 26 March 2011, the concentration of $^{137}$Cs in the seawater in the port rose to more than $500,000$ Bq/L, and recorded the highest value on 6 April 2011. As a result of the main outflow being suppressed, the $^{137}$Cs concentration in the coastal ocean near the FDNPP’s port decreased by several orders of magnitude toward the end of April 2011, and it seemed that the outflow of the contaminated water was blocked. However, especially in the port, the subsequent decrease in concentration was extremely slow compared to outside the port, suggesting that the outflow continues. In order to estimate the amount of $^{137}$Cs outflow to the outside of the port, Kanda [8] analyzed data at one representative observation point near the center of the harbor in the FDNPP’s port. By estimating the seawater exchange rate inside and outside the port from the observed decreasing trend in the concentration, the amount of $^{137}$Cs outflow from shortly after the accident to September 2012 was calculated, and concluded that, as of 2012, the outflow of $^{137}$Cs continued while it was reduced to less than one-thousandth of that shortly after the accident.

Machida et al. [9] followed this method and estimated the outflow of $^{137}$Cs to the outside of the port until the June 2018. Figure 1 shows the temporal change in the estimated monthly outflow of $^{137}$Cs from 2011 to 2018 outside the FDNPP port. The $^{137}$Cs outflow showed a declining trend, and the $^{137}$Cs outflow from

![Figure 1. Estimated monthly outflow of $^{137}$Cs from April 2011 to June 2018 outside the FDNPP’s port (Machida et al. [9], with permission). (Main panel) Seven-year temporal variation of monthly estimation of the $^{137}$Cs discharge inventory from the port of FDNPP by using the improved method of Kanda [8] in the period from April 2011 to June 2018. (Inserted panel) Focused variation of the main panel. Color figure available online.](image-url)
Figure 2. (Left) Location of river observatories and $^{137}$Cs deposition density measured by the airborne survey in May 2011 [20]. (Right) Cumulative $^{137}$Cs transport at each observatory based on $^{137}$Cs flux from Taniguchi et al. [19]. Monitoring at Iwanuma and the five other observatory began June 2011 and October 2012, respectively. Color figure available online.

the port, which was $2 \times 10^{15}$ Bq/yr in 2011 (due to lack of monitoring data in the port, this value does not include the leak in March 2011), decreased to 1/10,000 (~2 × 10^{11} Bq/yr) in 2017. Notably, the monthly $^{137}$Cs outflow decreased by an order of magnitude from the beginning of 2014 to the end of 2016. It can be considered that this decrease was due to the coverage of the harbor seabed (Phase 1: July 2014–April 2015, Phase 2: June 2015–December 2016), installation of groundwater impermeable wall (October 2015), removal of highly contaminated retained water (-December 2015) and filling of tunnels and towers (-December 2015). It should be noted that suspended $^{137}$Cs can also contribute to the monitoring data, which is the basis of Figure 1. It is possible that radiocesium-bound particles deposited on the surface and drainage channels flowed out due to rainfall, causing fluctuations in the $^{137}$Cs concentration in the harbor.

Due to the FDNPP accident, radiocesium released into the atmosphere was deposited on the land surface. Radiocesium deposited on the land surface was strongly adsorbed on soil particles, especially fine soil particles such as clay and silt [10,11]. Radiocesium adsorbed on soil particles was washed off from the land surface by rainfall-runoff processes [12,13]. The subsequent washed off radiocesium is transported into the ocean through rivers. Radiocesium in river exists as dissolved form and particulate form. The particulate fractions of radiocesium activity were 27–85% of total radiocesium activity in the normal flow condition [14–17] but were more than 90% in the flooding [14,15]. In the period from 10 August 2011, to 11 May 2012, it was estimated based on observations that 84–92% of the total radiocesium transported in the Abukuma River Basin was carried in particulate form [18]. Considering these results, the suspended particles significantly contribute to the transport of radiocesium through rivers into the ocean. According to continuous monitoring for 6 years, the concentration of particulate $^{137}$Cs in the river declined in two phases [19]. The first phase is between June 2011 and March 2012, the second phase is between April 2012 and August 2015. The average effective half-life of $^{137}$Cs at observatories in the first phase was 0.57 years and that in the second phase was 3.17 years. The concentration of particulate $^{137}$Cs in the river decreased sharply one year after the accident and then decreased more gradually. Taniguchi et al. [19] estimated the $^{137}$Cs flux at each observatory based on the monitoring data.

Figure 2 shows the cumulative $^{137}$Cs transport at representative observatories based on $^{137}$Cs flux estimated by Taniguchi et al. [19]. In this figure, the location of each river and a map of $^{137}$Cs deposition in the surrounding area [20] are also shown. The cumulative $^{137}$Cs discharge from the Abukuma River (observed at Iwanuma) was estimated to be approximately $1.2 \times 10^{13}$ Bq from June 2011 to August 2015 and that from other five rivers were estimated to be $3 \times 10^{10}–9 \times 10^{11}$ Bq from October 2012 to August 2015. Since the FDNPP accident, several numerical studies on the environmental fate of $^{137}$Cs deposited on the land surface have been performed [21–25]. Based on these results of the cumulative radiocesium transport over the period of more than one year, the ratio of that to the amount of initial deposition was estimated to be 0.11–0.68%/yr. These results suggested that radiocesium deposited on the land surface could continue to be a source of supply to the
ocean, although the cumulative radiocesium transport was small compared to the amount of initial deposition. The monitoring and numerical studies suggested that the transport of radiocesium from the land surface to the ocean could persist in the long term.

From the above estimates, $^{137}$Cs were released into the ocean within about three months of the occurrence of the accident in March 2011, and the total amount of $^{137}$Cs released to the North Pacific during this period is estimated to be $8 \times 10^{15} - 2.1 \times 10^{16}$ Bq (3–6 $\times 10^{15}$ Bq of direct discharge, 5–15 $\times 10^{15}$ Bq of atmospheric deposition and $\sim 1.3 \times 10^{13}$ Bq from rivers).

2.2. Initial dispersion of dissolved radiocesium

The mechanisms of oceanic initial dispersal and dilution of dissolved radiocesium that leaked from the FDNPP on and around the continental shelf off Fukushima have been investigated with model assessments by several computational studies [26–28] because it was very difficult to obtain appropriate measurements immediately after the accident. Miyazawa et al. [26] modeled oceanic $^{137}$Cs dispersal released directly from the FDNPP, using a Eulerian passive tracer transport model coupled with a regional circulation model. They conducted a series of numerical experiments to identify the sensitivity of oceanic $^{137}$Cs dispersal to surface winds, tides, and river discharges, and they highlighted that the role played by wind facilitates the meridional extension of the distribution of surface $^{137}$Cs on the shelf, while the tidal effects were minor. Estournel et al. [27] performed a numerical hindcast for dissolved $^{137}$Cs in the ocean, taking into account both direct release and atmospheric deposition. They identified that in addition to the importance of wind, freshwater discharges from rivers enhanced the offshore transport of $^{137}$Cs via surface-concentrated buoyant plumes. Masumoto et al. [28] conducted an inter-model comparison of oceanic $^{137}$Cs dispersal using five different regional oceanic models including the preceding two models. The five models at horizontal resolutions ranging from 600 m to 3.0 km were found to show overall qualitative similarities. They suggested that modeled surface $^{137}$Cs dispersal depends considerably on the lateral grid resolution of the model, where finer-resolution models are more capable of reproducing the prevailing coastal trapped along shelf transport that acts to retard immediate offshore transport and subsequent dilution via the Kuroshio-Oyashio current system (Figure 3 [29]). They also suggested that the warm water eddy pinched off from the Kuroshio Extension Jet in April 2011 was important to the surface $^{137}$Cs dispersal pattern.

Kamidaira et al. [30] also suggested that the high resolution 3-D modeling is important for successful 3-D radionuclide dispersal realization, particularly in coastal shelf regions such as near the FDNPP (Figure 3). They computed oceanic $^{137}$Cs dispersal with the submesoscale eddy-resolving model (horizontal grid resolution: 1 km), and showed that submesoscale dynamics (the approximate scale ranges are 0.1–10.0 km horizontally, 0.01–1.00 km vertically, and hours-days temporally) are important for representation of not only surface $^{137}$Cs

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**Figure 3.** (Left) Modeled $^{137}$Cs concentrations from the Sea Gurn FDM-L2 model at the sea surface in June 2011 (background: Kamidaira et al. [30]) and observed $^{137}$Cs concentrations (circles: Buesseler et al. [86]) using the same color scale. (Right) General sea surface current in June 2011 (arrows) and bottom topography (background). Information on ocean currents was compiled according to the Japan Coast Guard [29]. Color figure available online.
distribution but also vertical $^{137}$Cs transport by eddy processes. It revealed that 42.7% of the FDNPP-derived $^{137}$Cs was transported downward below the mixed layer by eddies with the major contribution being from seasonal ageostrophic secondary circulations induced by submesoscale eddies. Summarizing these studies, five key mechanisms, that could have determined the initial oceanic dispersal of the FDNPP-derived dissolved $^{134}$Cs shortly after the accident occurred, were proposed: (1) coastally trapped along-shelf currents driven by wind; (2) buoyancy inputs due to adjacent riverine freshwater discharges; (3) a mesoscale clockwise lateral circulation that developed southwest of the FDNPP in April 2011 associated with the migration of a warm water eddy pinched off from the Kuroshio Extension Jet, which interacted with (1); (4) the Kuroshio-Oyashio system; (5) submesoscale coherent structures and associated ageostrophic secondary circulations that transported $^{137}$Cs below the mixed layer by eddy processes. After the initial dispersion described above, $^{137}$Cs in the Pacific Ocean had been transported eastward by water currents of the subtropical and subarctic gyres at the surface and westward by the mode water transport via associated deep clockwise-circulating gyres at the subsurface [31]. These widely dispersed oceanic $^{137}$Cs in the meridional direction would decrease to the average pre-accident concentration several years after the accident (Figure 4 [32]).

3. Fate of dissolved radiocesium in the ocean

After the initial dispersion described in Section 2.2, especially the Kuroshio and its Extension Jet was responsible for subsequent radiocesium dispersal and dilution into the Pacific Ocean. These radiocesium have two major fates: one is transported to the east of the Pacific Ocean at the ocean surface by the subtropical and subarctic gyre system currents, and the other is transported southwestward due to formation/subduction of subtropical mode waters.

Tsubono et al. [33] suggested that their simulated surface $^{134}$Cs derived from FDNPP was transported the central part of the North Pacific one year after the Fukushima accident by the Kuroshio and its Extension Jet (Figure 5). This $^{134}$Cs concentration was decay-corrected to 11 March 2011, and considering that the $^{134}$Cs/$^{137}$Cs activity ratio of the accident-derived radiocesium on that day is about 1, the $^{134}$Cs distribution in Figure 5 is equivalent to that of accident-derived $^{137}$Cs. The simulations were consistent with observed $^{137}$Cs distribution on the surface of the North Pacific [32].

Figure 4. (Left) Time-longitude plot of $^{134}$Cs in surface water in the North Pacific Ocean from March 2011 to August 2014. Activity is shown as a color scale and a black dot means that $^{134}$Cs activity was below the detection limit. (Right) Time-longitude plot of $^{137}$Cs surface water in the North Pacific Ocean from March 2011 to August 2014. Activity is shown as a color scale. This figure is reprinted from Aoyama et al. [32] with permission of the publisher. Color figure available online.
Aoyama et al. [32] also showed that surface $^{137}$Cs concentrations in most areas of the North Pacific dropped to pre-accident levels within a few years after the accident (Figure 4). The FDNPP-derived dissolved radioceusium has been measured in seawater samples collected throughout the North Pacific Ocean immediately after the accident [32,34,35]. They found $^{134}$Cs in surface mixing layer in the north of the Kuroshio Front several months after the accident [32], in the Bering Sea in summer 2012 [34], and along the U.S. coastline to California in spring 2016 [35].

Radioceusium arriving by mode water transport had been observed in the Japan Sea and the East China Sea [36]. Surface water transportation...
through subsurface layer due to subduction of the mode waters in the Pacific Ocean has been investigated even before the accident [37]. Thus, it was expected that FDNPP-derived radiocesium in the surface water would be transported due to formation/subduction of mode waters, especially Subtropical Mode Water (STMW) on the southern side of the Kuroshio Extension, after the initial dilution as mentioned in Section 2. STMW, which is defined as a water mass with an isothermal layer of around temperature 16–18°C formed by deep vertical convection in winter [38]. Indeed, significant FDNPP-derived 137Cs were found in core waters of STMW within a year after the accident [31,39]. Inomata et al. [36] investigated 137Cs transport process from the North Pacific Ocean to the Japan Sea through the East China Sea during 2012–2016. They estimated \((0.21 \pm 0.01) \times 10^{15}\) Bq. 5.0% of the estimated total amount of FDNPP-derived 137Cs in the STMW entered into the Japan Sea before 2016 through Tsushima Strait after they entrained into the Kuroshio (Figure 6). As a result, concentration of radiocesium in surface water in the Japan Sea reached a maximum in 2015–2016. Their result suggested that several yearly delayed increase of radiocesium concentration in the Japan Sea were caused by subduction associated with formation of STMW in North Pacific Ocean and meridional transport of subtropical water by the Kuroshio.

4. Sediments

4.1. Accident-derived radionuclides in seabed sediments

In the monitoring survey after the Fukushima Daiichi accident, various radionuclides derived from the accident were also detected in seabed sediments and marine organisms. Within the radionuclides, two isotopes of radiocesium (134Cs and 137Cs) emitted particularly high radioactivity, and the concentration of 137Cs in seabed sediment increased by two orders of higher than the average pre-accident concentration (~2 Bq/kg [40]). Considering that 134Cs, which was not detected before the accident, was also detected from the seafloor, and the 134Cs/137Cs radioactivity ratio (approximately 1, which was attenuated on 11 March 2011) coincides with the estimated value of the inventory ratio in the reactor, it can be concluded that almost all radiocesium measured by the monitoring survey is derived from FDNPP. In addition to radiocesium, other accident-derived radionuclides were also detected in sediments included 131I, 129I, 129mTe, 110mAg, 95Nb, and 125Sb [41]. Since the short-lived radionuclides emit a large amount of radiation in a short period of time, concerns about radiation exposure assessment are often concentrated. Among the radionuclides, the concentration of 131I (half-life 8.02 days), which is particularly highly bioavailable, is recorded up to 28 Bq/kg. By this monitoring survey, 131I was last detected in sediment on 8 June 2011 (1.3 Bq/kg). Radionuclides such as 90Sr

![Figure 7](image_url)  
*Figure 7. Distribution of 137Cs activity concentration in surface sediment (0–3 cm). 137Cs activity is displayed logarithmically. Data are from National Radiation Authority, Japan [41]. Color figure available online.*
4.2. Overall characteristics of horizontal distribution of $^{137}$Cs concentration

Figure 7 shows part of the monitoring results of $^{137}$Cs concentration in sediments observed by the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan and the Nuclear Regulation Authority (NRA), Japan [41]. The result is the $^{137}$Cs activity concentration in 0–3 cm sediment layer. In the sediments after the accident, $^{137}$Cs was detected mainly in the range from 38.7°N to 35.5°N, and its concentration ranged from 2 to 520 Bq/kg. Considering that the concentration in the same sea area in 2010, the year before the accident, was 0.3 to 2 Bq/kg [40], the concentration of $^{137}$Cs in sediments increased over 1–2 orders of magnitude higher than that before the FDNPP accident. A concurrent survey reported that the concentration of $^{137}$Cs in seawater off Fukushima decreased 2–3 orders of magnitude between immediately after the accident and August 2012 (see Figures 1 and 4). On the other hand, the $^{137}$Cs concentration in the sediment shown in Figure 7 has remained its level and distribution characteristics over the two years.

4.3. Vertical distribution and total amount

As described above, the result of NRA’s monitoring survey shown in Figure 7 is the concentration of $^{137}$Cs in the sediment of the surface (0–3 cm) layer. Therefore, the number of data on the distribution of radiocesium to deeper sediments is limited.

As an example of the vertical distribution of radiocesium in the sediment, Figure 8 shows the vertical distribution of $^{137}$Cs in the sediment at five representative stations from the nearshore area near FDNPP (water depth 18 m) to the offshore area (water depth 300 m) reported by Otosaka [48]. In the offshore area where the water depth exceeds 300 m, almost all of the $^{137}$Cs in the sediment existed above the 3 cm layer, which is the reference layer for monitoring survey. On the other hand, the contribution of $^{137}$Cs in the deeper (>3 cm) sediment layers was about 30% at semi-offshore stations (water depth: 75–158 m) and more than 70% at nearshore stations (water depth <50 m).

The thickness of the sediment used for monitoring survey is 3 cm for NRA and 10 cm for TEPCO. These surveys appear to report on different criteria. However, the former mainly covers offshore areas deeper than 100 meters, and the latter covers the nearshore area shallower than 100 meters. As a result, it can be considered that each of these surveys monitors sediment layers appropriate for the target sea area.
Otosaka and Kato [49] estimated the total amount of $^{137}\text{Cs}$ in the sediment to be $0.1 \times 10^{15} – 0.3 \times 10^{15}$ Bq, considering the concentration of radiocesium in the deep sediment. Assuming that the total amount of $^{137}\text{Cs}$ released into the ocean by the accident (the total amount of direct discharge and deposition from the atmosphere) is $8 \times 10^{15} – 2.1 \times 10^{16}$ Bq, as described in Section 2.1, it is 1 to 4% of the supply of $^{137}\text{Cs}$ to the ocean, is presumed to have been deposited in the sediment. In this estimate, they found a horizontal distribution trend in which the $^{137}\text{Cs}$ inventory in sediments increased exponentially with decreasing water depth in areas where accumulation of sedimentary radionuclides was significant (i.e. 38.7°N – 35.5°N) (Figure 9). Based on this distributional pattern, it is estimated that 80% of the sedimentary $^{137}\text{Cs}$ accumulated in the coastal areas shallower than 100 m.

4.4. Factors regulating the distribution of radiocesium in seabed sediments

Radiocesium preferentially adsorbs to fine-grained sediments, mainly with high specific surface area and high clay mineral content [50,51]. Many studies have shown that $^{137}\text{Cs}$ concentrations in seabed sediments obtained after FDNPP show that $^{137}\text{Cs}$ content is 1.5 to 6 times higher in small (<75 μm) sediments than in coarse sediments [52–55]. It has also been pointed out that fine-grained sediments may have high organic matter concentrations, which may result in increased concentrations of radiocesium for organic matter adsorption [56]. In fact, a model of radiocesium transfer from seawater to sediment considering multiple size classes has been shown to better reproduce the concentration of radiocesium in sediments without considering size distribution [57].

As described in Section 4.2, the general distribution pattern of radiocesium in seabed sediments has not changed significantly at the spatial scale of each area (on the order of several km to several tens of km) over the years since the accident. This result indicates that the mobility of radiocesium on the seafloor is relatively low, and that an average value in a certain spatial range can be regarded as a representative level of radiocesium activity concentration in the area. On the other hand, locally (on the order of several meters to several hundred meters), radiocesium in the sediment is heterogeneously distributed. Thoronton et al. [58] investigated detailed horizontal distribution of radionuclides near the surface sediment using a sodium iodide detector that can be towed near the seafloor. They found that the radiocesium concentration tended to be locally high in a small submarine valley depression. The detailed observations in such depressions have shown that low flow velocities promote sedimentation of radiocesium-bound fine particles, and that low shear stress suppresses the remobilization of these particles [59].

4.5. Initial deposition

It has been pointed out that the three major deposition processes to coastal areas, where remarkable accumulation of radiocesium was observed, are (1) transport of radiocesium-bound particles from rivers, (2) biological uptake and sinking to the seafloor, and (3) adsorption of dissolved radiocesium on the surface of suspended particles/sediments.

Otosaka and Kato [49] estimated the amount of $^{137}\text{Cs}$ accumulation per unit area of coastal sediment is estimated to be in the range of $5.2 \times 10^{13}$ and $1.6 \times 10^{14}$ Bq/m², using parameters such as the concentration of dissolved radiocesium near the sea floor, the typical distribution coefficient of Cs between sediment and seawater ($K_d$: 3,500 L/kg), and the thickness of ‘active’ layer of surface sediments (1 cm). By multiplying this amount by area of coastal region (<100 m depth; 7.9 × 10² m²), the amount of dissolved $^{137}\text{Cs}$ adsorbed on the seafloor can be estimated to be $4 \times 10^{13}$ to $1.3 \times 10^{14}$ Bq. This $^{137}\text{Cs}$ amount may be underestimated, but it supports majority of $^{137}\text{Cs}$ inventory in the coastal area, and this process can be concluded as a reasonable process for accumulating radiocesium on the seafloor. The fact that the distribution of the concentration of radiocesium in sediments generally matches the passage history of contaminated plumes [60] also supports this concept. Although the detailed results differ depending on the numerical models, the characteristics of the distribution of radiocesium in sediments can be generally well simulated based on this concept [61–63].

In general, radiocesium in sediment can be categorized into four fractions; exchangeable, organic, and the residues. The ‘residue’ is a fraction that is not eluted by
the extraction treatments, and can be thought to be adsorbed to strong adsorption sites of the layered minerals. Otosaka and Kobayashi [52] found that over 80% of the radionuclides was strongly associated with the sediment (possibly a mineral component) collected from 70 to 100 km south of FDNPP in 2011. On the other hand, surprisingly, the remaining fraction, which is considered relatively labile, also accounts for a certain percentage over the years following the FDNPP accident. This indicates that there is a continuous exchangeable fraction between seawater (or pore water) and sediment. Similar results were obtained from stations closer to FDNPP [56,64].

As pointed out by Murota et al. [65], it is considered that the radiocesium near the surface of the sediment particles is gradually shifting to the irreversible adsorption process while maintaining a constant exchangeable fraction. This concept has been applied, for example, to a two-solid phase migration model for sedimentary radionuclides [66]. If appropriate parameters can be determined, it is expected that this concept will contribute to the prediction of radiocesium distribution in sediments around Fukushima.

Although details will be given in other reviews, highly radioactive particles that appear to have been released into the atmosphere immediately after the accident have also been found in the seabed sediment and suspended particles in seawater [67–69]. This would be an issue to be considered when discussing the heterogeneity of sedimentary radiocesium in and/or the impact on ecosystems in the limited area around the particles.

In addition, even in the area where there is no direct supply of radionuclides from the FDNPP, such as Japan Sea [70] and Tokyo Bay [71,72], radiocesium has been detected from seabed sediments. Continuous surveys of the seafloor in these areas may provide an answer to the behavior of riverine particulate radiocesium in the marine environment.

### 4.6. Temporal changes

Figure 10 shows the temporal change of $^{137}$Cs concentration in the surface sediment (0–10 cm) by TEPCO’s monitoring [73]. Here, the data of 71 stations, mainly from a depth of 50 m or less, are plotted in the figure. The concentration of $^{137}$Cs decreases between 2011 and 2015 at a rate of about 30% per year. Overall, $^{137}$Cs concentrations in sediments show a decreasing trend at a faster rate than the decay of $^{137}$Cs radioactivity (half-life 30.1 years). This reduction rate is consistent with the rate estimated by Otosaka [48] (29%/yr) for 63 stations from the coast of Miyagi to Ibaraki (<100 m), and by Kusakabe et al. [55] (26%/yr) for the area within 30 km of FDNPP. Although the rate of change varies between the stations (~63%/yr to +54%/yr: a negative value indicates decrease) [48], a rate of about ~30% has been obtained at most nearshore stations off Fukushima. The fact that $^{137}$Cs in sediments in the coastal zone is decreasing at a substantially constant rate, regardless of the initial deposition amount, indicates that a common mechanism in the coastal zone controls the decrease in $^{137}$Cs in sediments.

Otosaka [48] described the main processes that influence the temporal change of radiocesium concentration in seabed sediments: (1) vertical mixing of sediments [48,54], (2) resuspension and horizontal export [55,74,75], and (3) desorption from sediments [76–78]. From Figure 10, it is also observed that the $^{137}$Cs concentration in the sediment has been decreasing more slowly after 2016. It should also be considered that the above-mentioned process and its balance may change over time.

Between June 2011 and December 2015, the concentration of $^{137}$Cs in surface sediments decreased to about one fifth. It is estimated that about 60% of the decrease was transported to the deeper sediment layer, and more than 10% was migrated into the seawater column across the seawater-sediment interface and moved horizontally out of the target sea area (and 11% was due to radioactive decay) [48]. Therefore, theoretically, it is estimated that the migration of radiocesium into the deep sediment due to the vertical mixing of sediments was almost terminated, and the decrease rate of $^{137}$Cs concentration in surface sediments actually slowed down. This recent slow decrease is thought to be due to continuous resuspension of surface sediments and desorption of radiocesium.

The deep sediment layers in which radiocesium was buried in the initial stage after the accident becomes the peak layer that records the history of the accident in the long term, while the source that could release radiocesium into the overlying water through pore water [78]. Thus, the process of depletion of radiocesium from seabed sediments may change over time. When estimating changes in radiocesium concentration (abundance) in the seafloor, it should be considered that the controlling processes change over time.

The process of accumulation of radiocesium on the seafloor shown above has been modeled by several numerical simulations [61–63]. These models are expected to be used for predicting long-term changes in the seafloor environment. For more accurate predictions, optimization of various parameters such as kinetics for adsorption/desorption, and thickness of the ‘active’ layer of the seabed sediment is essential. Recent studies have shown that there are regional differences in these parameters [48]. Further observations and laboratory experiments to estimate these parameters will be necessary.

Particularly in coastal areas that receive terrestrial materials, the amount of radiocesium in seabed sediments is balanced by disappearance of initially deposited radiocesium from the seafloor and terrestrial supply. At
the same time, as radiocesium accumulated on the seabed in the early days of the accident has decreased, the relative amount of radiocesium flowing from rivers has increased. Accurate estimations of the riverine input of radiocesium and the detailed understanding of the behavior of radiocesium in riverine particulates in estuaries/coasts are also issues that need to be clarified.

5. Marine biota

The concentrations of accident-derived radionuclides in marine organisms (mainly fish) since the FDNPP accident are continuously monitored by the Fisheries Agency, Japan [79]. From 2011 to 2014, accident-derived radiocesium at levels above the regulation standard of fishery products (100 Bq/kg-wet with sum of $^{134}\text{Cs}$ and $^{137}\text{Cs}$ activities) was frequently detected from fishes. While the concentration of radiocesium in surface-dwelling fishes has quickly decreased until 2013, the concentration of radiocesium in brackish and demersal fishes has been slow to decline, and it is detected very rarely even recently.

A dynamic food chain transfer model suggests that the concentration of radiocesium in demersal fish in the coast of Fukushima cannot be explained by the supply of radiocesium from surface seawater alone [80]. It is also suggested that the additional sources of radiocesium exist near the seafloor [81]. Recent simulations by those authors have demonstrated that the labile (rich in organic matter) fraction in sediments, especially in the early stages of the accident, cannot be ignored as a source of radiocesium to demersal fishes [56]. Although the detailed factors have not been identified at this stage, direct observation of the behavior of radiocesium inside the sediment is expected to reduce the uncertainty of these simulations.

Concentration levels of radionuclides in fishes around Fukushima have fallen sufficiently from a viewpoint of dose assessment [82]. Nevertheless, as described above, a certain percentage of the labile fractions are present in the sediment. There is concern that such ‘feed components’ may be incorporated into the marine ecosystem through benthic food web [83–85].

As a feed component other than sediment, the radiocesium concentration in zooplankton was investigated [86–90], and it was found that radiocesium in zooplankton strongly reflected the concentration of radiocesium in seawater and show a heterogeneous horizontal distribution. Furukawa et al. [91] also experimentally demonstrated that marine fish excrete radiocesium through the potassium channels of the gills. These results indicate that fish take up radiocesium from both food components and seawater.

The measurement data of various nuclides and organisms obtained after the accident were organized as environmental transition parameters applicable to the dose evaluation model, and the latest data collection on ‘concentration factors’ was published [92] and updated continuously [93]. On the other hand, a huge amount of survey data after the accident remains unpublished, and systematic compilation of them will be an issue for the future.

6. Conclusions

From the FDNPP accident to the present, assessment studies on the radiocesium that leaked from the FDNPP in the marine environment had been vigorously conducted both observationally and computationally. Through these studies, the dynamics of radiocesium in the marine environment has been gradually revealed. Modeling studies have played a major role in the analysis of the initial dilution of dissolved radiocesium after the accident and the subsequent dispersion mechanism because it was very difficult to obtain appropriate measurements immediately after the accident. At the same time, observational studies played a major role in the analysis of the medium- to long-term distributions and pathway of radiocesium in the ocean after the accident. In addition, method for estimation of the radionuclide release rate from the nuclear facilities was developed using the ocean model and the monitoring data. This method will have an important role in predicting impact of radionuclides on the marine environmental in the case of future accidents.

These post-FDNPP accident studies have shown that mathematical marine dispersion model of radionuclide migration can be powerful tool in environmental assessment. Based on these experiences, several research institutes have been developing oceanic radionuclide forecasting systems using ocean models in order to prepare for future incidents [94,95]. These systems are expected to provide rapid prediction of the radionuclide concentration in the marine environment including seawater and seabed sediment by executing hypothetical radionuclide release calculations.

The most recent problem to be overcome is the preliminary assessment of pollution impact on the adjacent ocean due to the future release of ALPS water into the ocean, which was decided as part of the decommissioning of FDNPP. Oceanic radionuclide forecasting systems are expected to play an active role in both ex-ante impact assessments through forecast simulation and ex-post analysis through hindcast simulation. For instance, Zhao et al. [96] conducted the numerical simulation to track the oceanic transport and dispersion of hypothetical tritium released from the radioactive water of the FDNPP. They
suggested that the majority of tritium with radioactive water of the FDNPP would be mixed and diluted rapidly in the coastal waters off Japan, and then transported eastward along Kuroshio extension. However, the source term is based on many assumptions, and it will take some more time to assess the validity of the results. In any case, it is necessary to appropriately disclose the source information when the treated water is released into the ocean.

Another issue is the assessment of the impact of radionuclides in the seabed sediments off the Fukushima coast on the marine environment. Since radionuclides in sediments are retained longer than those in seawater, it is desirable to be able to predict long-term changes in the seafloor environment by numerical simulation. In order to ensure the accurate reproducibility of numerical experiment, further investigation and optimization of various parameters for modeling the deposition of radionuclides on seabed sediments. It has been pointed out that the release of treated water into the ocean may lead to the uptake of particle-reactive radionuclides into the seafloor and the transfer to ecosystems [97]. Models that can reproduce the transfer of radionuclides into sediments/ecosystems are also expected to be useful in assessing their effects.

The findings of these ongoing research studies are expected to be extremely valuable information not only for FDNPP accident but also for nuclear incidents that may occur in various areas of the world.

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No potential conflict of interest was reported by the author(s).

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