

Special Report: “Radioactivity in the Marine Environment and in Fisheries Products during the Five Years after the Fukushima Dai-ichi Nuclear Power Plant Accident”

## Distributions of radionuclides in the ocean and their temporal changes

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**Abstract:** There are two types of radionuclides in the ocean environment: natural radionuclides and artificial radionuclides. The former have existed since Earth was formed; the latter have appeared in the oceans since 1945. Since then, the sources of artificial radionuclides have been atmospheric nuclear testing (the largest source), accidents at nuclear power-related facilities, and planned releases from such facilities. The present study describes the levels of natural and artificial radionuclides in the oceans and the spatiotemporal changes in the artificial radionuclides.

**Keywords:** natural radionuclides, artificial radionuclides, ocean

### 1. Introduction

There are many kinds of radionuclides in the environment. They can be divided into the natural radionuclides that have existed since before human beings appeared on Earth, and the artificial radionuclides that have been rapidly increasing in abundance since the end of World War II. The latter arise from causes such as atmospheric nuclear tests and emissions from nuclear power plants and reprocessing facilities. The present study describes the distributions of both types of radionuclides in the ocean environment and their temporal changes up to the time of the accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) in Fukushima Prefecture, Japan.

### 2. Natural radionuclides

There are two main origins for the various types of natural radionuclides on Earth. The primordial radionuclides have existed in Earth’s crust since it was formed; other natural nuclides are generated by cosmic rays. Some primordial radionuclides have a decay series, whereas some do not. Therefore, the natural radionuclides in the ocean can be placed into three categories:

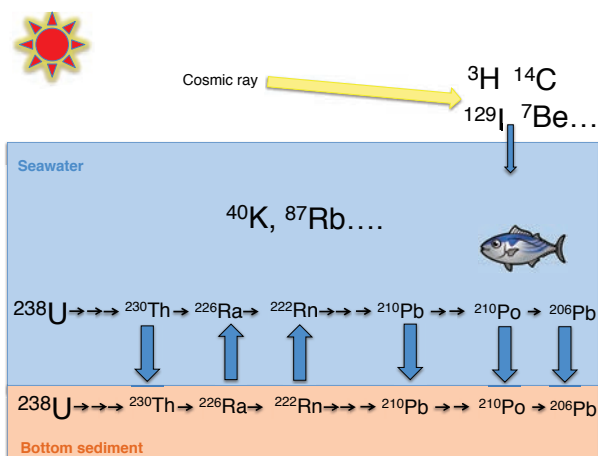
- Nuclides that have a decay series
- Nuclides that do not have a decay series
- Nuclides generated by cosmic rays

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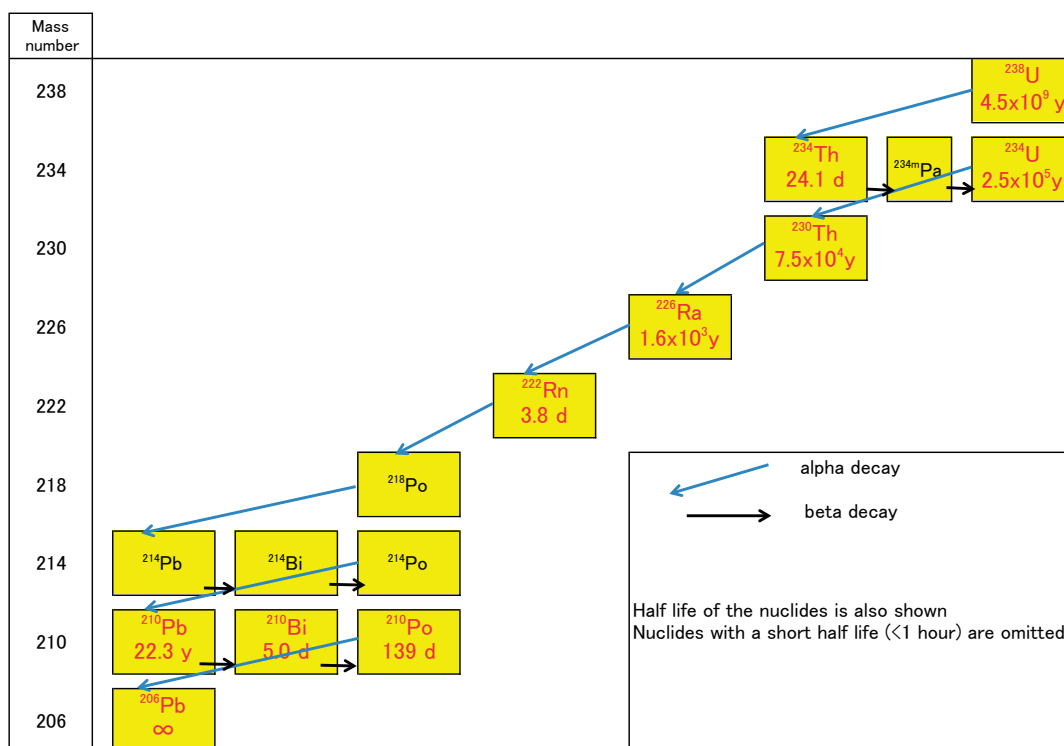
**Fig.1** Schematic of the distribution of natural radionuclides in the oceans. The major nuclides are shown. In the figure, the thick upward arrows show the tendencies for nuclides to dissolve out of marine sediments, and the downward arrows show the tendencies for them to be eliminated from sea water.

**2.1 Nuclides that have a decay series**

In seawater, uranium exists mainly as the uranyl carbonate ion  $[UO_2(CO_3)_3]^{4-}$ , which dissolves very easily in seawater and is distributed almost uniformly throughout all oceans. There are three isotopes of uranium ( $^{234}U$ ,  $^{235}U$ , and  $^{238}U$ ), and the concentration of  $^{238}U$  (half-life  $[t_{1/2}] = 4.5 \times 10^9$  years) is the highest: approximately 3  $\mu\text{g/L}$  (0.04 Bq/L). The concentrations of  $^{235}U$  and  $^{234}U$  are 0.002 and 0.04 Bq/L, respectively.

$^{238}U$  decays into  $^{234}Th$ , which then decays through other nuclides to finally become  $^{206}Pb$ , which is stable; this is called the uranium series (Fig. 2). Nuclides that arise through decay are called descendant nuclides, and nuclides prior to decay are called parent nuclides. In seawater, there is also the actinium series that starts with the decay of  $^{235}U$  ( $t_{1/2} = 7.0 \times 10^8$  years) and the thorium series that starts with  $^{232}Th$  ( $t_{1/2} = 1.4 \times 10^{10}$  years). The total amount of radioactivity in seawater from nuclides of the uranium series is an order of magnitude greater than that of the other two series.

If nuclides belonging to the same series had the same chemical properties, and sufficient time had passed, they would have the same radioactivity (this is called radioactive equilibrium). However, in fact, the nuclides that are formed in a decay series have a variety of chemical properties and show various behaviors in the oceans, so their levels of radioactivity are different. For example, thorium (Th), lead (Pb), and polonium (Po) are relatively insoluble in seawater, so they are easily eliminated from seawater as particles, and their concentrations are therefore less than those of their parent nuclides (see Fig. 1). Conversely, uranium and radium (Ra) are soluble in seawater, so they remain in seawater for a long time. At the same time, they dissolve out of marine sediments, so they show higher



**Fig.2** The uranium series. The main nuclides included in the uranium series. Those with small generation rates from their parent nuclides have been omitted.

radioactivity in seawater than do their parent nuclides. Furthermore, radon (Rn), which is a gas, partially escapes to the atmosphere from seawater. Moreover,  $^{222}\text{Rn}$  that has come from the land via the air decays to generate  $^{210}\text{Pb}$ ,  $^{210}\text{Bi}$ , and  $^{210}\text{Po}$ , which enter the ocean environment.

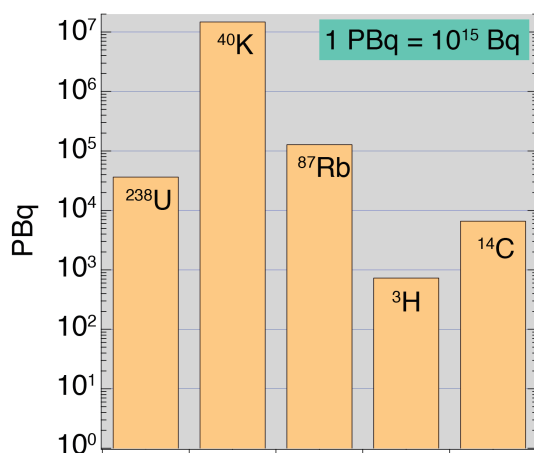
## 2.2 Nuclides that do not have a decay series

One of the major nuclides without a decay series is potassium-40 ( $^{40}\text{K}$ ,  $t_{1/2} = 1.3 \times 10^9$  years).  $^{40}\text{K}$  decays into one of the stable nuclides  $^{40}\text{Ar}$  or  $^{40}\text{Ca}$ . It has the highest radioactivity (11–12 Bq/L) among the natural radionuclides in seawater. There is also rubidium-87 ( $^{87}\text{Rb}$ ,  $t_{1/2} = 4.75 \times 10^{10}$  years), but its concentration (0.11 Bq/L) is only about 1/100th that of  $^{40}\text{K}$ . In the periodic table, potassium and rubidium belong to the alkali metals, along with sodium (Na), so they dissolve extremely easily in seawater. They therefore have the same distribution as salinity, being distributed approximately evenly throughout all of the oceans. There are a few other nuclides that do not have a decay series, but their concentrations are extremely low.

## 2.3 Nuclides generated by cosmic rays

Cosmic rays, which come from outside Earth, cause nuclear reactions in elements in Earth's upper atmosphere, creating many radionuclides. These radionuclides are continuously raining down onto the surface of Earth. The main ones are the following:

- Tritium ( $^3\text{H}$ ,  $t_{1/2} = 12.3$  years)
- Beryllium-7 ( $^7\text{Be}$ ,  $t_{1/2} = 53$  days)
- Carbon-14 ( $^{14}\text{C}$ ,  $t_{1/2} = 5730$  years)
- Iodine-129 ( $^{129}\text{I}$ ,  $t_{1/2} = 1.6 \times 10^6$  years)



**Fig.3** Amounts of the main natural radionuclides existing in the oceans.

Some of the nuclides shown above are readily incorporated into the biogeochemical cycle of the corresponding stable elements. For example, in the atmosphere,  $^{14}\text{C}$  becomes  $^{14}\text{CO}_2$  (carbon dioxide) and dissolves in seawater. Some  $^{14}\text{CO}_2$  is incorporated into organic compounds through photosynthesis, whereas most of the rest circulates with the ocean currents in the form of  $\text{H}^{14}\text{CO}_3^-$ . This is why it is possible to quantitatively describe the movement of seawater by studying the distribution of  $^{14}\text{C}$ .

Figure 3 shows the total amounts of the main natural radionuclides in the oceans. As explained above,  $^{40}\text{K}$  is the most abundant, exceeding  $10^7$  PBq (1 PBq =  $10^{15}$  Bq).  $^{87}\text{Rb}$  is also present in substantial amounts.  $^{238}\text{U}$  is the third most abundant, and the uranium-series nuclides that follow from it (not shown in the figure) are also important.

These various natural radionuclides circulate in the ocean with their own biological and chemical

**Table 1**  $^{40}\text{K}$  concentration in foods

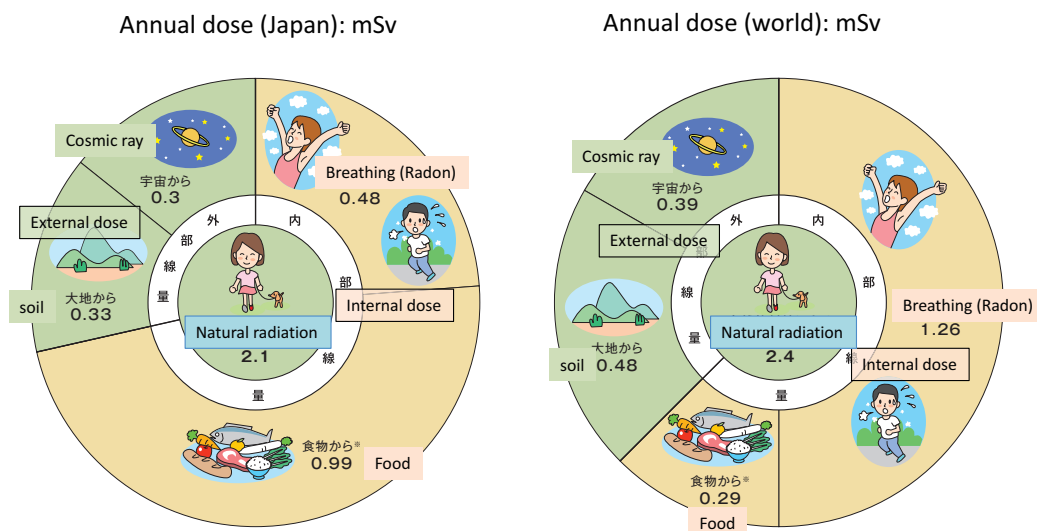
food	$^{40}\text{K}$ (Bq/kg)
dried kelp (kombu)	2,000
dried mushroom (shiitake)	700
tea	600
dry milk	200
brown seaweed	200
spinach	200
beef	100
fish	100
milk	50
rice	30
bread	30
wine	30
beer	10
sake	1

Imai *et al.* (1999)

**Table 2** Reference activity concentrations of natural radionuclides in food

	Activity concentration (mBq/kg)					
	$^{226}\text{Ra}$	$^{210}\text{Pb}$	$^{210}\text{Po}$	$^{232}\text{Th}$	$^{228}\text{Ra}$	$^{228}\text{Th}$
Milk products	5	40	60	0.3	5	0.3
Meat products	15	80	60	1	10	1
Grain products	80	100	100	3	60	3
Leafy vegetable	50	30	30	15	40	15
Roots and fruits	30	25	30	0.5	20	0.5
Fish products	100	200	2,000	-	-	-

Adapted from UNSCEAR (1993)



**Fig.4** Radiation doses received from natural radionuclides. From: Federation of Electric Power Companies, *Graphical flip-charts of nuclear- and energy-related topics*, 2015.

properties and are partially incorporated into marine organisms. Meanwhile, the natural radionuclides on the land and in the air are incorporated into agricultural products and ultimately arrive on our dinner tables. Tables 1 and 2 show the major natural radionuclides contained in our food. The concentration of  $^{40}\text{K}$ , several tens to several hundreds of becquerels per kilogram, is overwhelmingly high compared to other nuclides. Among the nuclides shown in Table 2, the three on the left are the uranium-series nuclides, and the three on the right are from the thorium series. Note that the concentration of  $^{210}\text{Po}$  in fish (2000 mBq/kg) is particularly high. This is because Po has biological and chemical properties that allow it to easily remain in the body. The ratio of the concentration of an element in fish to that in seawater is called the concentration factor: the recommended value for the concentration factor of Po is  $2 \times 10^3$  L/kg, which is in the relatively high group (International Atomic Energy Agency [IAEA], 2004).

Although somewhat beyond the scope of this document, it is worth looking at the radiation doses that we receive from these natural radionuclides (Fig. 4). If we compare the global averages and the averages for Japan, two differences are evident. The exposure of Japanese people to radon is low, whereas the exposure through food is high. Radon ( $^{222}\text{Rn}$ ,  $t_{1/2} = 3.8$  days) is a member of the uranium-series nuclides, generated by  $\alpha$  decay from its parent nuclide radium ( $^{226}\text{Ra}$ ,  $t_{1/2} = 1602$  years). It is one of the chemically stable noble gases, similar to others such as helium and neon. In addition,

the radon isotope  $^{220}\text{Rn}$  (so-called thoron,  $t_{1/2} = 54.5$  seconds) is generated from radium ( $^{228}\text{Ra}$  and  $^{224}\text{Ra}$ ) in the thorium series. Radon and thoron decay through the decay series, until finally becoming the stable  $^{206}\text{Pb}$  and  $^{208}\text{Pb}$ , respectively. If radon or thoron is inhaled as a gas, the descendant nuclides that result from  $\alpha$  decay with a short lifetime are deposited in the organs and tissues that form the respiratory tract, conferring high-energy radiation in these regions, with biological consequences. The differences in the effects of radon shown in Fig. 4 are thought to result from differences in living environments. Specifically, wooden buildings that do not contain much  $^{238}\text{U}$  do not generate radon, and furthermore the good ventilation within such a building makes it difficult for radon gas to accumulate. The relatively large radiation exposure from food comes from  $^{210}\text{Po}$ , which is contained in fish (Table 2). This is because  $^{210}\text{Po}$ , which emits  $\alpha$  radiation, is a large source of radiation in the gastrointestinal tract, especially the mucosa of the small intestine, and the intake radiation conversion coefficient is  $1.2 \times 10^{-6}$  Sv/Bq, about a hundred times that of  $^{137}\text{Cs}$ , which is  $1.3 \times 10^{-8}$ .

### 3. Artificial radionuclides

The three sources of artificial radionuclides in the oceans are as follows:

- Atmospheric nuclear tests
- Accidents at nuclear power-related facilities

**Table 3** Amounts of radionuclides released to the environment

nuclides	half life	PBq*		
		nuclear test	Chernobyl accident	FDNPP accident
<sup>89</sup> Sr	50.5 d	117,000	~115	-
<sup>90</sup> Sr	28.8 yr	622	~10	0.09~0.9 (ocean)
<sup>131</sup> I	8.02 d	675,000	~1,760	200 (atmosphere) 11.1 (ocean)
<sup>134</sup> Cs	2.06 yr	~0	~54	15~20 (atmosphere) 3.6 (ocean)
<sup>137</sup> Cs	30.2 yr	948	~85	15~20 (atmosphere) 3.6 (ocean)
<sup>239</sup> Pu	24.1 yr	6.52	0.03	~0
<sup>240</sup> Pu	6.56 yr	4.35	0.042	~0
<sup>241</sup> Pu	14.3 yr	142	~6	~0

Adopted from UNSCEAR (2000), Casacuberta *et al.* (2013), Kobayashi *et al.* (2013), Tsumune *et al.* (2013), Aoyama *et al.* (2016), Bu *et al.* (2014)

\* 1PBq=10<sup>15</sup>Bq

- Emissions from nuclear power-related facilities

The following subsections provide an overview of each of these.

### 3.1 Artificial radionuclides from atmospheric nuclear tests

From 1945 to 1963, there were more than 500 nuclear

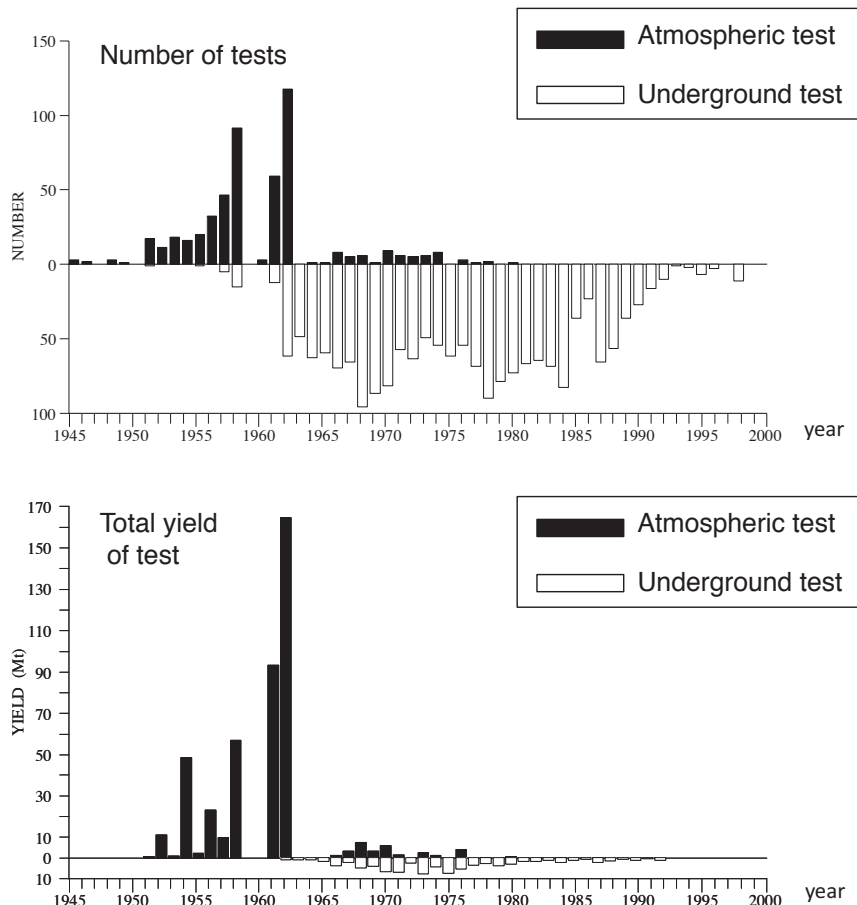
**Table 4** Release of <sup>137</sup>Cs to the ocean by nuclear tests

	Arctic Ocean	Atlantic Ocean	Indian Ocean	Pacific Ocean	sum
northern hemisphere	7	157	21	222	407
southern hemisphere	0	44	63	89	196
sum	7	201	84	311	603

Adapted from IAEA (2005)

unit : PBq

tests in the atmosphere. The total amount of artificial radionuclides emitted into the oceans from these tests is the largest of the three sources listed above. For example, the amount of <sup>137</sup>Cs due to nuclear testing is more than ten times that from the Chernobyl accident, and more than 40 times that from the Fukushima Daiichi reactor accident (Table 3). Atmospheric nuclear



**Fig.5** History of nuclear testing. The information is to be added to the UNSCEAR 2000 report.

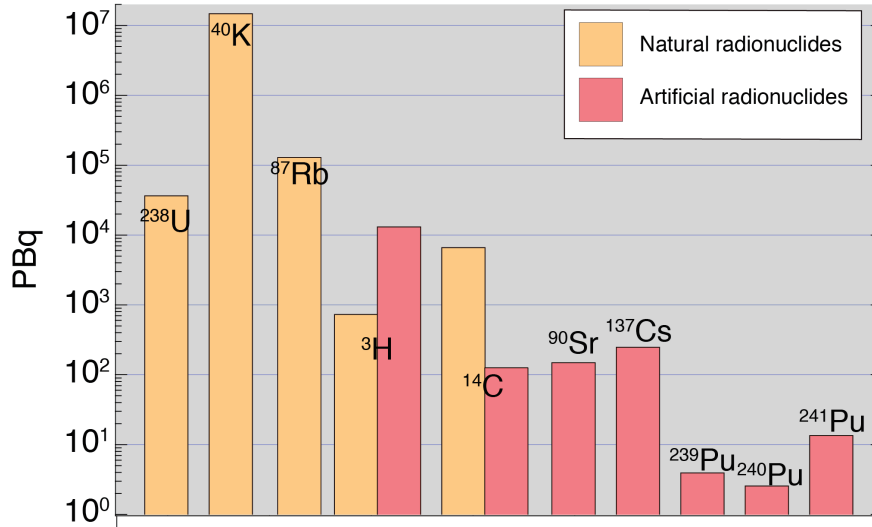


Fig.6 Comparison of the amounts of the main artificial radionuclides and natural radionuclides in the oceans in 2000. Adapted from Figure 3 and Aarkrog (2003).

tests continued until the signing of the Partial Test Ban Treaty in 1963. Some countries did not sign the treaty and continued testing, but the number of atmospheric tests after the signing was much lower (Fig. 5), and the radionuclide emissions have been greatly reduced. Therefore, the pollution due to artificial radionuclides in the oceans was the most serious in the early 1960s. Using  $^{137}\text{Cs}$  as an example, the region with the most pronounced pollution from nuclear testing was the Pacific Ocean, which accounted for about half of what was emitted into the oceans (Table 4). Since 1963, the nuclides in the oceans derived from nuclear tests have

been decreasing as they undergo radioactive decay, but they persist. Figure 6 presents the amounts of the main artificial radionuclides in the oceans as of 2000, along with the natural radionuclides. Out of approximately 600 PBq of  $^{137}\text{Cs}$  emitted, levels had declined to about 1/3 of this by 2000. At the same time, even in 2000, almost 40 years after the end of atmospheric nuclear tests, artificial tritium ( $t_{1/2} = 12$  years) still exceeded the pre-testing natural amount.

The concentration of  $^{137}\text{Cs}$  in the surface layers of the ocean due to nuclear testing is decreasing owing to radioactive decay and mixing with seawater from other

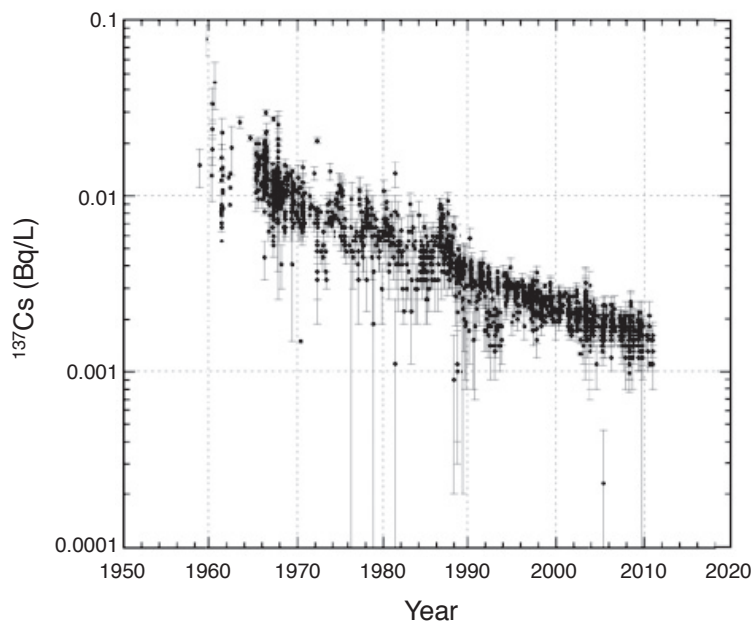
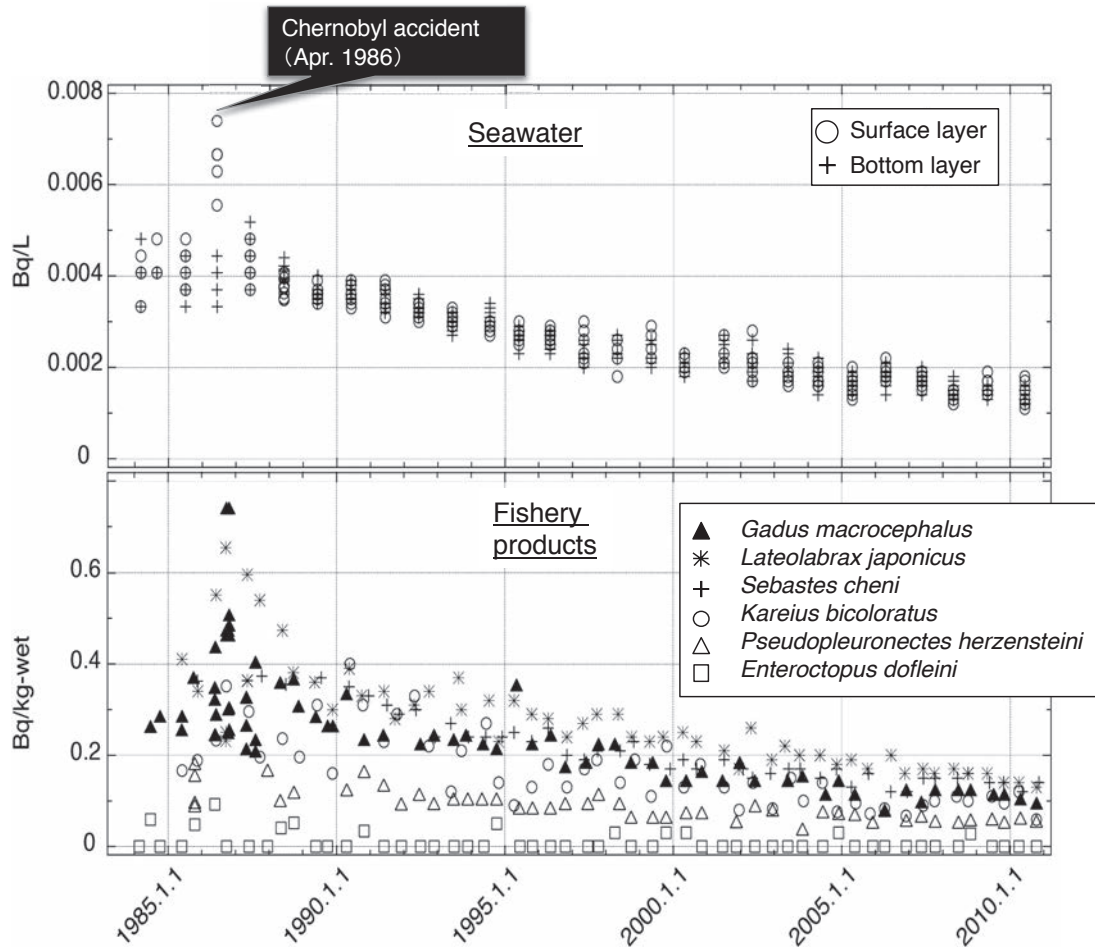


Fig.7 History of  $^{137}\text{Cs}$  sea surface concentrations in the northwest portion of the North Pacific. The information is to be added to Povinec *et al.* (2013).



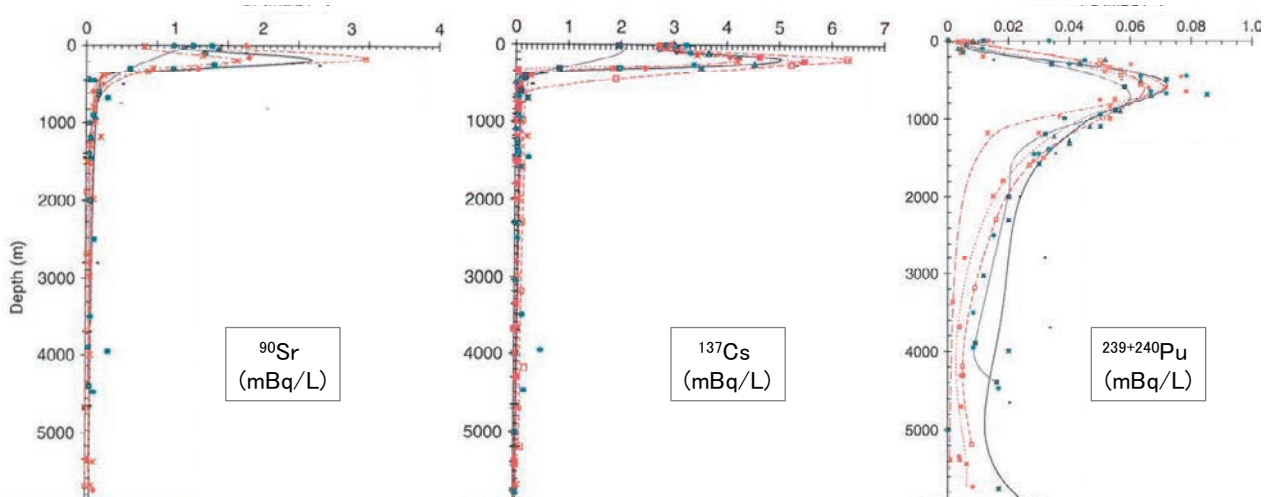
**Fig.8** Temporal changes in the  $^{137}\text{Cs}$  concentrations in sea water and marine animals in the sea off the coast of Fukushima Prefecture (1984-2010). Adapted based on the results of studies by the Marine Ecology Research Institute.

areas that have relatively less pollution. Figure 7 shows the change over time of  $^{137}\text{Cs}$  concentration in the northern Pacific Ocean. In the early 1960s, it reached several tens of millibecquerels per liter; since then it has been decreasing exponentially. Since 1984, the Marine Ecology Research Institute of Japan has conducted detailed studies of radioactivity in the waters offshore of Japan's nuclear power plants; for example, off the coast of Fukushima Prefecture (Fig. 8). It is evident that the effects of the 1986 Chernobyl accident (to be discussed later) reached as far as the Pacific, and that there were 1 to 2 mBq/L of  $^{137}\text{Cs}$  in the seawater even before the Fukushima Daiichi reactor accident.

Radionuclides from the nuclear tests existed in the surface layers of the oceans immediately after their emission. As time passed, they were mixed with deeper water and transported downward. The vertical distribution of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in the northern Pacific (Fig. 9) shows that these isotopes reached a depth of

500 m over approximately 30 years after 1963. However, the  $^{239+240}\text{Pu}$  that was emitted at the same time was carried even deeper. The apparent half-lives of these nuclides in the surface layers of the oceans have been observed for the entire Pacific Ocean (Table 5). The apparent half-lives of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in the surface oceans were mostly in a range from 13 to 14 years. In contrast, those of  $^{239+240}\text{Pu}$  were only about half that. These marked differences in vertical distribution patterns and apparent half-lives are ascribed to the respective chemical characteristics. The downward movement of elements in seawater is not only because of the diffusion of the dissolved elements themselves but is also closely tied to the precipitation of particles. Qualitatively, if a dissolved element (or an element in the form of suspended fine particles) in seawater tends to be immediately incorporated into settling particles, it will have a short half-life.

Reactions between dissolved elements and



**Fig.9** Vertical distributions of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ , and  $^{239+240}\text{Pu}$  concentrations in sea water in the North Pacific (1973-1997). Adapted from Aoyama (2010) with the addition of some text.

**Table 5** Apparent half-lives of artificial radionuclides in the surface water of the Pacific Ocean

	half-life (yrs)		
	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{239+240}\text{Pu}$
North Pacific	$12 \pm 1$	$13 \pm 1$	$7 \pm 1$
Equatorial Pacific	$21 \pm 2$	$23 \pm 1$	$10 \pm 1$
South Pacific	$18 \pm 1$	$22 \pm 1$	$12 \pm 4$
entire Pacific	$13 \pm 1$	$14 \pm 1$	$7 \pm 1$

Adoped from Povinec *et al.* (2005)

**Table 6**  $K_d$  values for open ocean and ocean margin

Element	Recommended $K_d$ values	
	Open ocean	Ocean margin
Sr	$2 \times 10^2$	$8 \times 10^0$
I	$2 \times 10^2$	$7 \times 10^1$
Cs	$2 \times 10^3$	$4 \times 10^3$
Pu	$1 \times 10^5$	$1 \times 10^5$

Adoped from IAEA (2004)

particles in seawater are dynamic, including reactions between particles of various sizes (for example, Bacon, 2004), so that it is not easy to accurately describe these interactions kinetically. Here, for a first approximation, we use the distribution coefficient ( $K_d$ )—that is, the ratio of the concentration of a nuclide in particles to the concentration of the nuclide in seawater—as an index of the ease of incorporation of an element into particles.  $K_d$  is calculated using the following equation:

$$K_d = \frac{\text{Concentration of nuclide in particles}}{\text{Concentration of nuclide in seawater}}$$

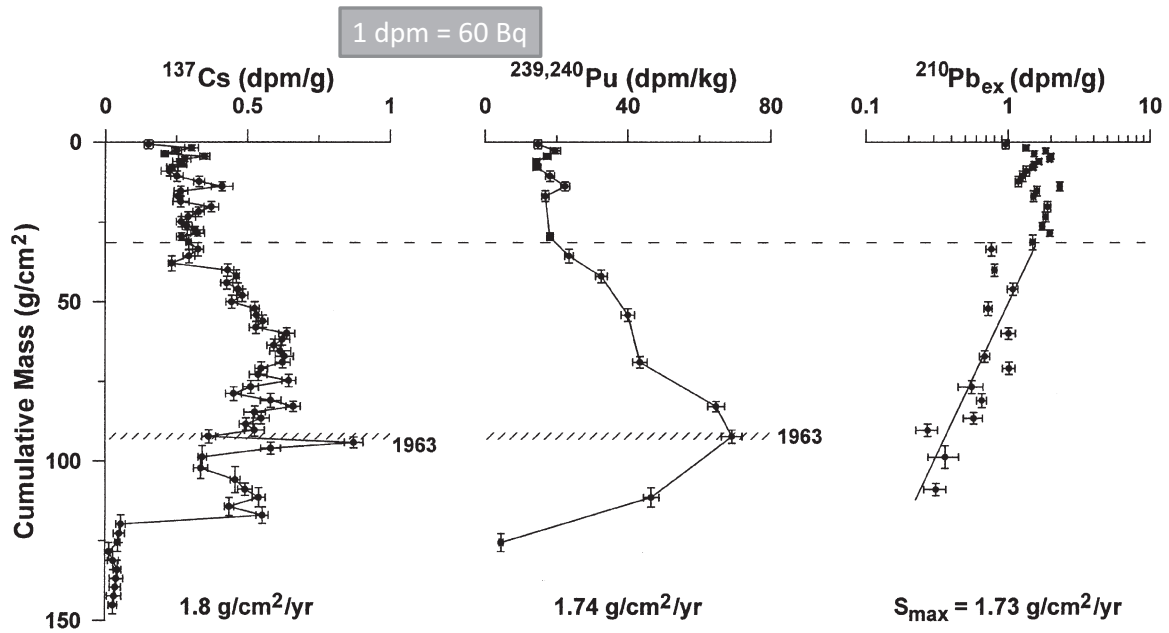
Table 6 shows the values of  $K_d$  recommended by the IAEA.  $K_d$  increases in the order  $\text{Sr} < \text{Cs} < \text{Pu}$ . These values represent that Pu is the easiest to remove from the oceans, and Sr is the hardest of these three nuclides. In fact, experiments using equipment that collects settling particles (sediment traps) have confirmed that settling particles play an important role in the downward migration of Pu in the oceans (Livingston and Anderson, 1983). Thus, by understanding the biological and chemical properties of nuclides, it is possible to predict the future distribution of artificial radionuclides in the oceans.

The impact of the nuclear tests has also been recorded in marine sediments (especially in areas of shallow seas). In the marine sediments of the East China Sea, sediment layers from 1963, as determined from natural radionuclides, show a concentration peak for  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$  due to the nuclear tests (Fig. 10). This type of vertical distribution is not evident in deep-sea sediments because the sedimentation rate in the deep sea is not high enough for sufficient time resolution.

### 3.2 Artificial radionuclides from accidents at nuclear power-related facilities

Large amounts of artificial radionuclides have been released to the environment because of accidents





**Fig.10** Vertical distributions of  $^{137}\text{Cs}$ ,  $^{239+240}\text{Pu}$ , and  $^{210}\text{Pb}$  concentrations in marine sediments of the East China Sea. The vertical axis does not show depth but rather the accumulated mass of marine sediments, considering factors such as water content. Adapted from Su and Huh (2002).

at nuclear power plants and nuclear fuel reprocessing facilities. The types of nuclides released depend on the accident, so a general comparison is difficult. With respect to the total amount of nuclides released and the range of impacts to the environment, however, the most serious accident was the one at the former Soviet Union's Chernobyl power plant in 1986 (Table 7). The amount of  $^{137}\text{Cs}$  released (Table 3) from this accident was approximately 85 PBq, whereas that from the FDNPP accident was 19 to 24 PBq. There is an outstanding difference of the Chernobyl accident from others in terms of extent of the impact. After the Chernobyl accident,  $^{137}\text{Cs}$  traveled through the atmosphere and appeared in the surface waters of the ocean off the coast of Fukushima on the opposite side of the globe (Fig. 8). The nuclides from the accident did not show up in the bottom water near Japan, and those in surface water disappeared in the following year, so the radiological impact to the marine environment near Japan could be regarded as minimal. It is worth noting that the  $^{137}\text{Cs}$  concentration in some marine animals increased following the accident and persisted into the following year (Fig. 8, bottom). The actual value of the concentration increase was not of a level to endanger human health, but it is interesting in terms of the

behavior of Cs in the marine environment.

Kusakabe *et al.* (1988) also found  $^{137}\text{Cs}$  from the Chernobyl accident in settling particles in the intermediate depths of the North Pacific. The accident occurred on 26 April 1986, and large amounts of radionuclides were released into the atmosphere. Around 7 May they reached the upper atmosphere of the northwestern North Pacific, and some settled to the sea surface. Data from sediment traps showed that the nuclides reached a depth of approximately 780 m in at most one or two months, with a settling velocity of 60–190 m/day. Because Cs is usually an element that dissolves relatively easily in seawater, this relatively fast settling velocity implies that the  $^{137}\text{Cs}$  derived from the accident had a chemical form that was relatively hard to dissolve, or that it was incorporated into particulate matter, perhaps via plankton, and transported as settling particles.

The most important accident in Japan in terms of artificial radionuclides was evidently the one that occurred at FDNPP. Its impact on the ocean environment and fishery products is detailed by Takata *et al.* and Yokota *et al.* (this special issue). Here, briefly, the scale of the FDNPP accident can be appreciated by comparing it with that of the nuclear tests. Basically, the impact of

the FDNPP accident on the ocean environment was limited to the North Pacific. Before the accident (in 2010),  $^{137}\text{Cs}$  in the North Pacific from nuclear tests amounted to 69 PBq. The FDNPP accident added 3.5 PBq by direct discharge, and 12–15 PBq via the atmosphere. As a result, the amount of  $^{137}\text{Cs}$  in the waters of the North Pacific increased by 22–27% from the accident (Aoyama *et al.*, 2016). It is expected that the concentration of  $^{137}\text{Cs}$  in the future will decrease with a half-life of more than 10 years, as shown in Table 5, as the result of seawater movement into other areas, precipitation of  $^{137}\text{Cs}$  onto marine sediments, and radioactive decay.

### 3.3 Artificial radionuclides discharged from nuclear power-related facilities

There have been some radionuclides discharged from nuclear power-related facilities; in particular, facilities related to nuclear fuel processing. Generally, care is taken to try to ensure that there is no significant effect on local ecology but there have probably been cases in the past where the amounts emitted exceeded the currently accepted levels. Among these, the largest sources of radionuclides were two reprocessing facilities in Europe: Sellafield (formerly Windscale, as in Table 7) in England and La Hague in France. Table 8 shows the change over time (1970–1998) in the  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  discharged from these two reprocessing

facilities. The cumulative amount of  $^{137}\text{Cs}$  discharged from Sellafield into the ocean for over 28 years has been approximately 40 PBq. Considering radioactive decay, the corrected amount for the year 2000 is 23 PBq. Through the middle of the 1970s, the annual discharge rate reached a maximum of 5.2 PBq/year, but it had been reduced to 0.008 PBq/year by 1998. Since 2000, the rate has been further reduced, to less than 0.005 PBq/year in 2013 (Fig. 11). The total amount of  $^{137}\text{Cs}$  discharged from La Hague is less than 3% that of Sellafield.

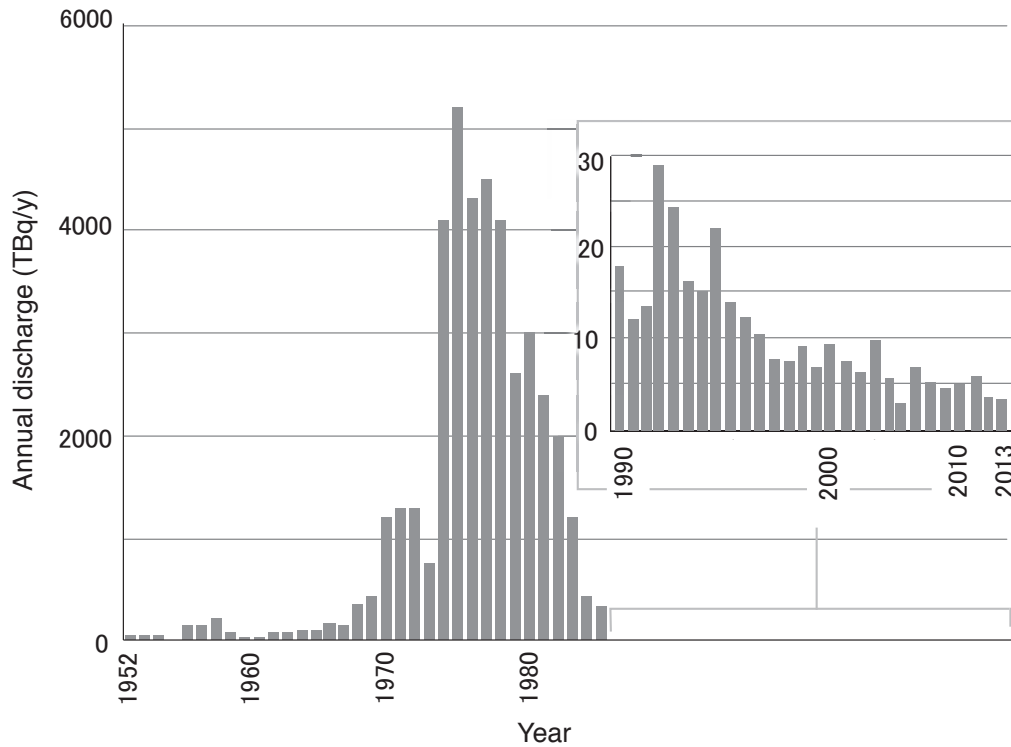
The Irish Sea, which faces Sellafield, has less active water exchange with other seawater than does the water off the coast of Fukushima Prefecture. Furthermore, because of persistent discharge of waste with high radionuclide level until the middle of the 1980s, the concentration of  $^{137}\text{Cs}$  in the water of the Irish Sea remained high (Fig. 12). The seawater of the English Channel that is affected by La Hague has a concentration that is three orders of magnitude lower than that of the Irish Sea. It is believed that the difference in concentration is because of the differences in the amounts discharged and in the seawater mixing. The  $^{137}\text{Cs}$  from Sellafield flows northward on the ocean currents to enter the North Sea (Fig. 13). The increase in concentration seen in the Baltic Sea after 1986 was mainly because of  $^{137}\text{Cs}$  that was transported through the atmosphere from the Chernobyl accident.

**Table 7** Amount of radionuclides released during accident at nuclear power-related facilities

nuclides	place	Chelyabinsk	Windscale	Three Mile Islans	Chernobyl	Fukushima Pref.
	country	Soviet Union	England	United States	Soviet Union	Japan
	year	1957	1957	1979	1986	2011
$^{90}\text{Sr}$		4	$7 \times 10^{-5}$		~10	0.09~0.9 (ocean)
$^{131}\text{I}$			0.74	$5.55 \times 10^{-4}$	~1,760	200 (atmosphere) 11.1 (ocean)
$^{137}\text{Cs}$		0.03	0.022		~85	5~20 (atmosphere) 3.6 (ocean)
$^{239}\text{Pu}$			$1.6 \times 10^{-6}$		0.03	~0
$^{240}\text{Pu}$					0.042	~0
$^{241}\text{Pu}$					~6	~0
	The facilities were located inland. Few impact to the ocean.		Impact mainly in inland	Major nuclides released: $^{85}\text{Kr}$ and $^{133}\text{Xe}$	$^{137}\text{Cs}$ to the ocean: 15~20 PBq	$^{137}\text{Cs}$ to the ocean through the atmosphere : 12~15

Adapted from UNSCEAR (2000), Casacuberta *et al.* (2013), Kobayashi *et al.* (2013)

unit : PBq (=  $10^{15}\text{Bq}$ )



**Fig.11** History of emitted amounts per year of  $^{137}\text{Cs}$  at Sellafield. Adapted from the Environmental Protection Agency (2015).

The effects of the discharge from Sellafield have been recorded in nearby marine sediments. The concentration of  $^{137}\text{Cs}$  in the surface sediment was approximately 400 Bq/kg-dry in 1992, whereas it exceeded 1000 Bq/kg-dry in deeper layers (Fig. 14; MacKenzie *et al.*, 1998). This pattern of vertical distribution is not seen in the studies outside the 30-km

radius from the FDNPP conducted by the Marine Ecology Research Institute of Japan under contracts from the Nuclear Regulatory Agency of the Japanese Government (see Takata *et al.* [this issue] for details). The difference in vertical distribution patterns can be attributed to the differences in oceanic material cycling; that is, whereas the Fukushima coast directly faces the

**Table 8** Annual discharge rates of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  at Sellafield (England) and La Hague (France)

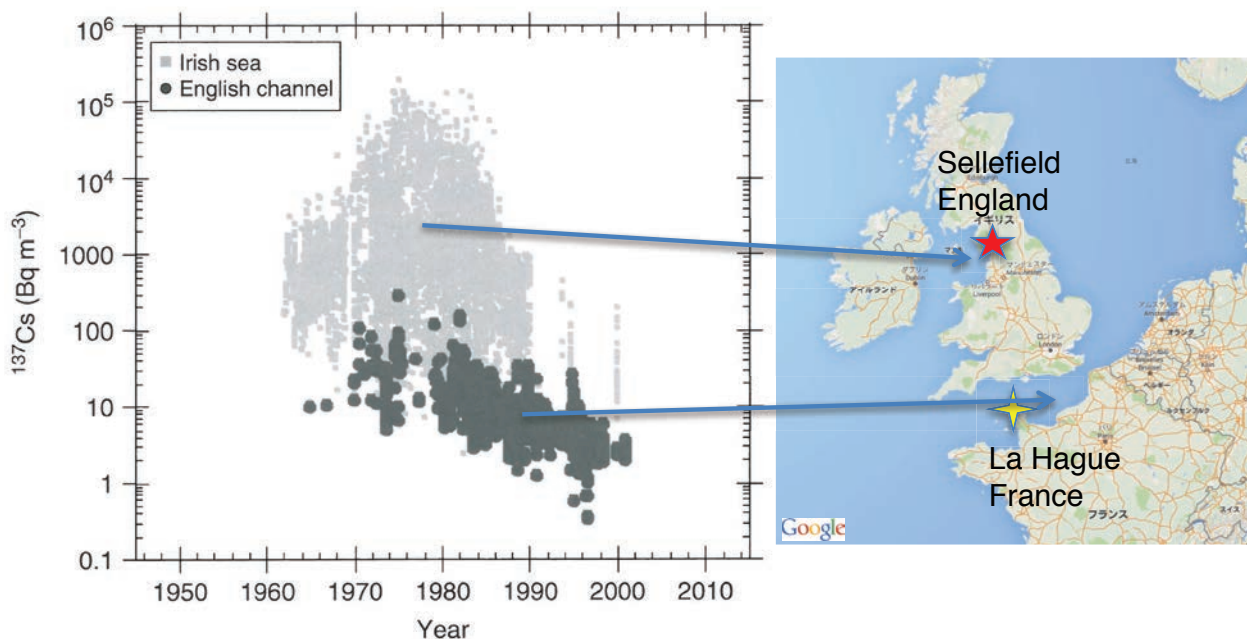
year	Sellafield				La Hague			
	$^{137}\text{Cs}$	$^{90}\text{Sr}$	$^{137}\text{Cs ac}^*$	$^{90}\text{Sr ac}^*$	$^{137}\text{Cs}$	$^{90}\text{Sr}$	$^{137}\text{Cs ac}^*$	$^{90}\text{Sr ac}^*$
1970	1,200	230	600	112	89	2	45	1
1975	5,230	466	2,936	255	34	38	19	21
1980	2,970	352	1,871	217	27	29	17	18
1985	325	52	230	36	29	47	21	33
1990	24	4	19	3	13	16	10	13
1995	12	28	11	25	5	30	4	27
1998	8	18	7	17	3	3	3	3
sum	38,837	5,528	22,912	3,193	961	1,106	566	746

Adopted from Aarkrog (2003)

Sum was calculated based on the data of every year from 1970 to 1998

unit :  $10^{12}\text{Bq}$

\* Radioactivity was corrected for decay to the year 2000



**Fig.12** History of  $^{137}\text{Cs}$  concentrations in the surface water of the Irish Sea and the English Channel. The map shows the locations of the two fuel reprocessing facilities (see the main text) as well as the Irish Sea and the English Channel. The data are from Inomata (2010). Concentrations are shown as  $\text{Bq}/\text{m}^3$ , but this is equivalent to  $\text{mBq}/\text{L}$ .

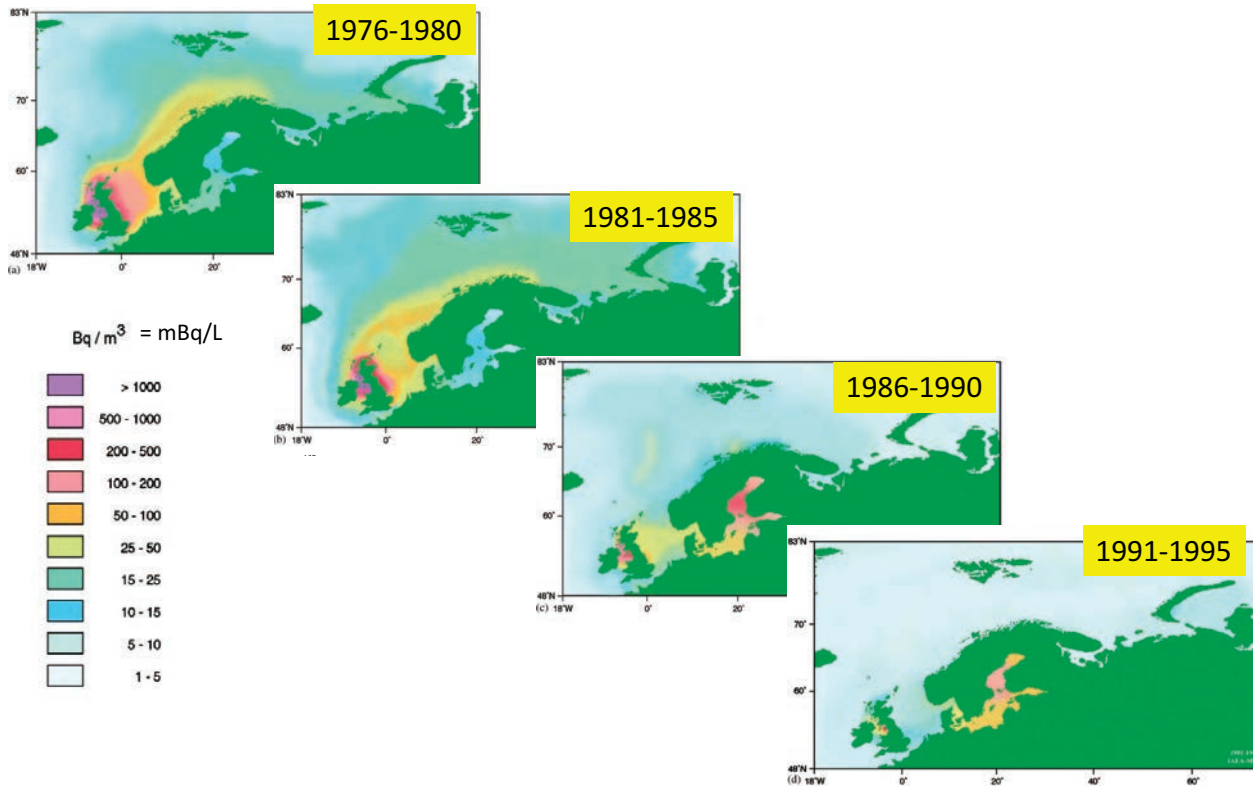
open ocean, the sea off the coast of Sellafield is semi-enclosed. Future comparative studies of these two areas should allow more precise predictions regarding the recovery of the ocean near Fukushima.

#### 4. Summary

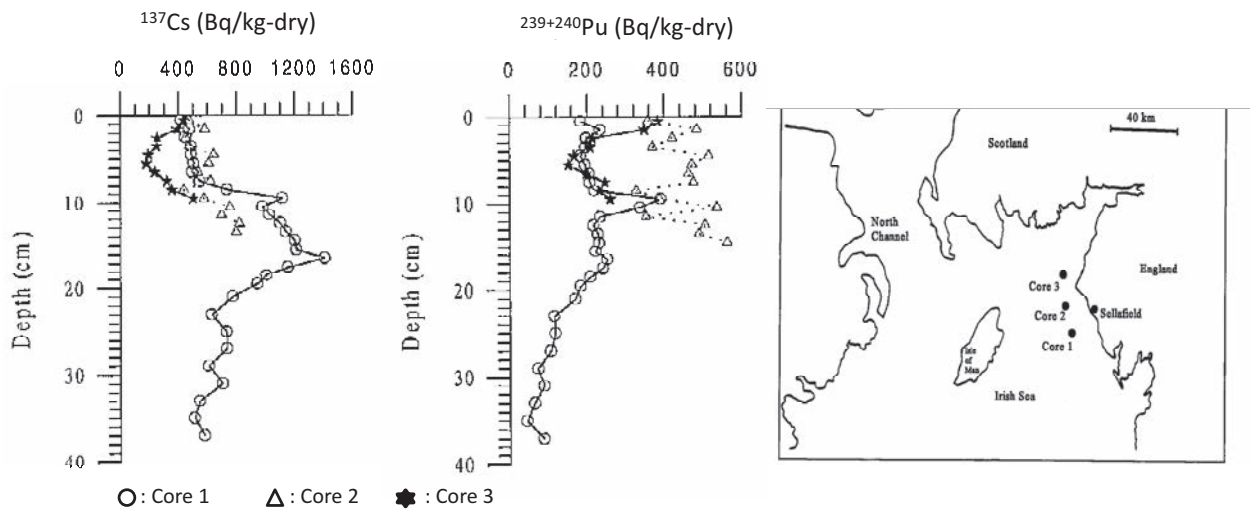
- Humans have always been living surrounded by various types of natural radionuclides. At the same time, certain radionuclides have accumulated within their bodies.
- The largest source of radioactive pollution in the ocean environment is atmospheric nuclear tests.
- The largest accident at a nuclear power-related facility occurred at the Chernobyl nuclear power plant.
- Planned discharges during daily operations at nuclear fuel processing facilities are currently extremely low compared to the two preceding sources.

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**Fig.13** History of  $^{137}\text{Cs}$  concentration in the surface water of the northeastern Atlantic. Adapted from Povinec *et al.* (2003).



**Fig.14** Distribution of  $^{137}\text{Cs}$  in marine sediments because of Sellafield. Samples are collected in January 1992. Adapted from MacKenzie *et al.* (1998).

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