Observation of radiocesium in seabed soil at the Notsuke Strait of the southern Okhotsk Sea derived from the Fukushima Daiichi Nuclear Power Plant

Yasushi NABAE[1), 2)], Sunao MIYASHITA[1)], and Satoru NAKASHIMA[1), 3)]

[1)] Department of Chemistry, Graduate School of Science, Hiroshima University,
1-3-1 Kagamiyama, Higashi-Hiroshima, Hiroshima 739-8526, Japan
[2)] Hydrographic and Oceanographic Department, Japan Coast Guard,
3-1-1 Kasumigaseki, Chiyoda-ku, Tokyo 100-8932, Japan
[3)] Natural Science Center for Basic Research and Development, Hiroshima University,
1-4-2 Kagamiyama, Higashi-Hiroshima, Hiroshima 739-8526, Japan

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Seabed soils of the southern Okhotsk Sea at the Notsuke Strait, Hokkaido were collected, and the radioactivity was measured with a Ge semiconductor detector. Trace amount of radiocesium derived from the Fukushima Daiichi Nuclear Power Plant was detected. It was suggested by combining other results that this radiocesium was originally deposited in western Fukushima from the Fukushima Daiichi Nuclear Power Plant, and migrated to the Japan Sea with the Agano River, and then carried to the Notsuke Strait by Tsushima Warm Current and Soya Warm Current.

Key words: fukushima daiichi nuclear power plant, okhotsk sea, radiocesium, agano river, notsuke strait

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1. Introduction

Tokyo Electric Power Company (TEPCO) Fukushima Daiichi Nuclear Power Plant (FDNPP) accident occurred in March 2011. Large quantity of radioactive material was spread in environment, and the area around Fukushima was contaminated with the radioactive material[1)]. The direction of the wind at the time of the release, a passage course of plume and its precipitation were related to the quantity of sediment of radioactive material in each place[2)]. The radioactive material deposited on the earth surface soil by rain or snow migrates with the wind or flow of water. Ministry of Education, Culture, Sports, Science and Technology detected radioactive material deposited in the upper reaches of the Agano River by plane monitoring[3)]. The Agano River flows from Fukushima and Gunma to Niigata.

Niigata Prefecture continued the environmental radioactivity investigation to know the influence by FDNPP accident from the early time. 38 Bq/kg (wet) of ^{134}Cs was detected at the Agano River mouth mud in the investigation of August 2011. In addition, 72 Bq/kg of ^{134}Cs was observed in the seabed soil of 20 m in depth at the sea area of the Agano River mouth offing[4–5)].

In June 2011, 1.9 mBq/L of ^{134}Cs was detected at surface seawater of Ishikari Bay in the northern Japan Sea, and 3.14 mBq/L of ^{137}Cs which was 3.3 times of the preceding year was detected in the NO-10 point in the southern Okhotsk Sea[6–7)]. In addition, some investigations were carried out in the Japan Sea and the Okhotsk Sea, and ^{134}Cs was detected at the northeastern part of the Japan Sea, Hokkaido coast and Tohoku coast[8–9)].

In the present study, seabed soils at the Notsuke Strait in the southern Okhotsk Sea were collected, and an extremely small amount of the radioceasium migrated from FDNPP was confirmed.

Natural Science Center for Basic Research and Development,
Hiroshima University,
1-4-2 Kagamiyama, Higashi-Hiroshima 739-8526, Japan
TEL: 082-424-6291 FAX: 082-424-0700
E-mail: snaka@hiroshima-u.ac.jp

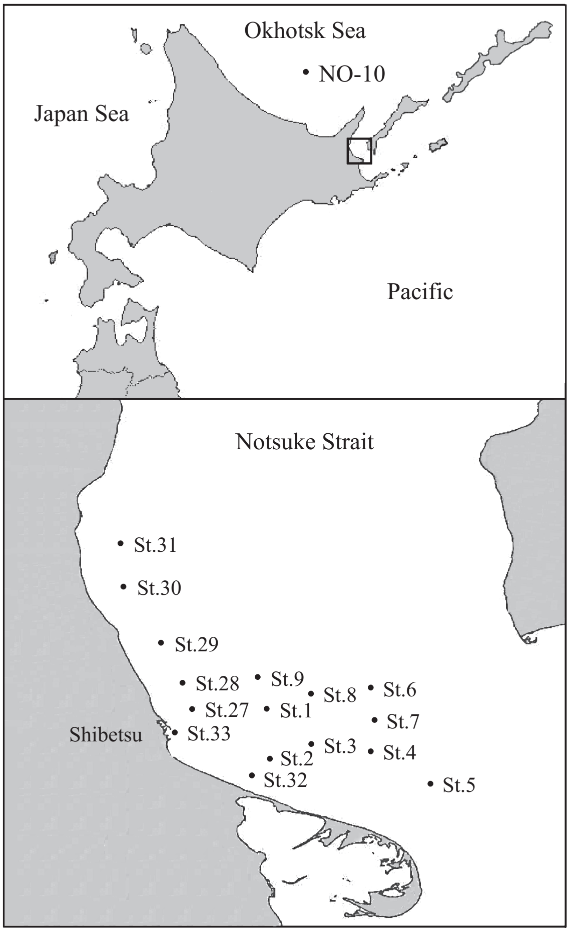

Fig. 1. Sampling points.
Seabed soils were collected at the Notsuke Strait in Okhotsk Sea. Seabed soils of St.1–9 were collected on February 23, 2015. Those of St.27–33 were collected on February 6, 2016. The Japan Coast Guard investigates NO-10 every year.

2. Sampling and experimental method

2-1. Sampling

On February 23, 2015, we collected seabed soil at the Notsuke Strait of the Sibetsu-cho, Hokkaido. The Notsuke Strait is a sea area where a part of the Tsushima Warm Current of the Japan Sea flows out into the Pacific. Figure 1 shows detailed sampling points of seabed soil. The seabed soil samples were collected using Ekman-Berge bottom sampler, and the stones and the visible creature with naked eye were removed. Samples were stirred and were put in U8 container in a wet state. As an additional investigation, we collected seabed soil of surface layer (2 cm) by a diver on February 6, 2016.

2-2. Method for measurement of radiocesium

A germanium semiconductor (GEM40-76-XLB-C made by SEIKO EG & G Co., Ltd) was used for the measurement of the radioactivity of the sample.

The γ radioactivity of samples (St.1–St.9) on February 23, 2015 was measured for 86,400 seconds. Sample of St.2 showed the highest count of ^{134}Cs in all samples, therefore it was re-measured as St.2* for 250,000 seconds. The concentrations of ^{134}Cs and ^{137}Cs were calculated by official method[10], and the results of the measurements were corrected to the quantity of radioactivity of the sampling day. Because the emission ratio is high and the separation with other nuclides is easy, 605 keV of a peak was chosen for ^{134}Cs. And 662 keV of a peak was chosen for ^{137}Cs. The samples were dried after the measurement and the weights were measured to obtain water ratio of the sample, and then the radioactivity concentration in dry sample was calculated. This measurement was carried out in Hiroshima approximately 800 km away from Fukushima. In addition, this detector was not affected by the Fukushima accident.

The samples (St.27–St.33) of the additional investigation on February 6, 2016 of dry seabed soil (2,000 cm^3) were measured for 250,000 seconds. And the sample of St.33 was measured again as St.33* for 600,000 seconds, because the ^{134}Cs concentration was the highest among them. This measurement was carried out in Tokyo. The background measurement was carried out, and the influence from environment was removed.

3. Results and discussion

Table 1 shows the latitude, longitude, and depth of the seabed soil sampling points. The depth in this sea area is particularly shallow in the Notsuke Strait, and the sea area shallower than 20 m is widely spread. The sampling points were chosen in the southwest side of the Notsuke Strait, because these sampling points were supposed that the sediment is thick. The depth of the shallowest sampling point was 4.0 m, and that of the deepest sampling point was 15.2 m. Some seabeds of the sampling points included sand, some included few clay and silt, and some included small stones. Because the seabed soils of St.6, 30, 31 were small in quantity and did not include the clay, the seabed soils were not collected. A lot of clay and silt were included in St.32 and St.33.

Table 2 showed the sampling day, the volume of measurement, the time of measurement, ^{134}Cs concentration and ^{137}Cs concentration for the sample of the seabed soil. 0.033 ± 0.0084 Bq/kg of ^{134}Cs was detected for a sample of St.33.

Table 1 Position and Depth at each sampling points

No.	Latitude	Longitude	Depth [m]
St. 1	43°40′12″N	145°12′15″E	8.0
St. 2	43°38′21″N	145°13′23″E	8.6
St. 3	43°38′46″N	145°14′31″E	5.4
St. 4	43°38′43″N	145°17′14″E	5.0
St. 5	43°38′14″N	145°19′26″E	8.4
St. 6	43°40′36″N	145°17′ 7″E	10.4
St. 7	43°39′30″N	145°17′31″E	12.3
St. 8	43°40′44″N	145°14′21″E	15.2
St. 9	43°41′12″N	145°11′26″E	13.2
St. 27	43°40′20″N	145° 7′56″E	4.0
St. 28	43°41′30″N	145° 7′ 7″E	5.0
St. 29	43°43′42″N	145° 5′37″E	9.0
St. 30	43°45′32″N	145° 4′ 2″E	5.0
St. 31	43°47′21″N	145° 3′55″E	5.0
St. 32	43°38′21″N	145°13′23″E	10.0
St. 33	43°39′56″N	145° 8′ 8″E	5.5

Figure 2 shows relation between the concentrations of ^{134}Cs and ^{137}Cs for each sample for the additional investigation. Although the concentrations of ^{134}Cs except for St. 33 were judged to be ND, the obtained values were included in Figure 2 to know the relation. The values were corrected on the values of March 11, 2011 to estimate the concentration of the global fall-out.

The ^{134}Cs/^{137}Cs radioactivity ratio when the radioactive materials were spread in environment by FDNPP accident was approximately 1. And the slight difference in the ratio among FDNPP 1–3 nuclear reactors was reported. The ratio of the first nuclear reactor was around 0.89–0.93 (average: 0.91). The ratio of the second nuclear reactor was around 0.96–1.05 (average: 1.0). The ratio of the third nuclear reactor was around 0.97–1.04 (average: 1.01)[11]. The ^{134}Cs/^{137}Cs ratio means that we can estimate concentration of ^{137}Cs from the concentration of ^{134}Cs in the Fukushima accident. At St.33 on February 6, 2016, the concentration of ^{134}Cs was 0.034 ± 0.0086 Bq/kg, and the concentration of ^{137}Cs was 1.43 ± 0.015 Bq/kg. When these concentrations were corrected to the values of March 11, 2011,

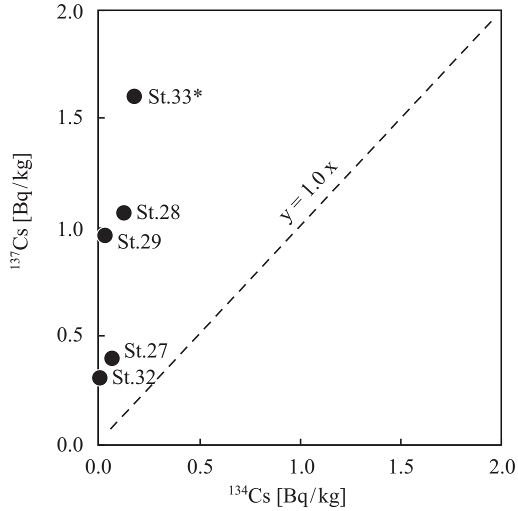

Fig 2. Relation of concentrations between ^{134}Cs and ^{137}Cs at seabed soil of Notsuke Strait.
The data were corrected on the values of March 11, 2011. Although the concentrations of ^{134}Cs except for St.33* were judged to be ND, the obtained values were included in Fig. 2 to know the relation.

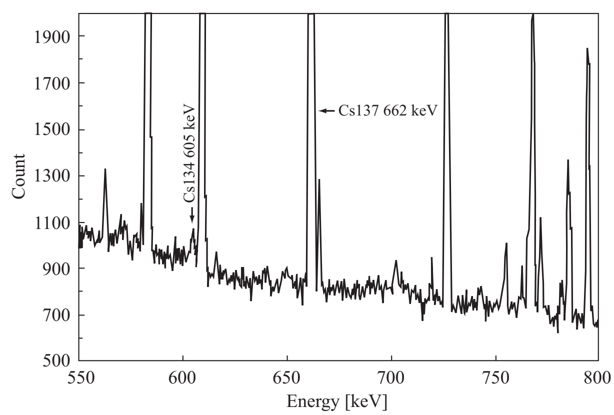

Fig 3. Gamma-ray spectrum of the seabed soil from St.33* of the Notsuke Strait.
Energy and Counts of ^{134}Cs and ^{137}Cs are shown in the figure.
Sampling position: St.33* of the Notsuke Strait (43 ° 39′ 56′′ N, 145 ° 8′ 8′′ E)
Sampling date: February 6, 2016. Amount for the measurement: 2187.56 g.
Measurement date: February 26, 2016. Measurement time: 600,000 s.
Germanium semiconductor: GEM40-76-XLB-C made by SEIKO EG & G Co., Ltd

the concentration of ^{134}Cs is expected to be 0.17 ± 0.045 Bq/kg, and the concentration of ^{137}Cs is 1.60 ± 0.017 Bq/kg. When radiocesium was released by the Fukushima accident, the ^{134}Cs/^{137}Cs ratio is expected to be approximately 1.0 at the time

Table 2 Results of the measurement of the sample

No.	Sampling day	Volume [cm^3]	Time [s]	^{134}Cs [Bq/kg]	^{137}Cs [Bq/kg]
St. 1	Feb 23, 2015	100	86,400	ND (0.19)	ND (0.33)
St. 2	Feb 23, 2015	100	86,400	ND (0.33)	ND (0.20)
St. 2*	Feb 23, 2015	100	250,000	ND (0.14)	0.41 ± 0.14
St. 3	Feb 23, 2015	100	86,400	ND (—)	ND (0.25)
St. 4	Feb 23, 2015	100	86,400	ND (0.22)	ND (0.39)
St. 5	Feb 23, 2015	100	86,400	ND (—)	ND (0.16)
St. 6	Feb 23, 2015	—	—	—	—
St. 7	Feb 23, 2015	100	86,400	ND (0.19)	ND (0.19)
St. 8	Feb 23, 2015	100	86,400	ND (—)	ND (0.02)
St. 9	Feb 23, 2015	100	86,400	ND (0.09)	ND (—)
St. 27	Feb 6, 2016	2,000	250,000	ND (0.01)	0.35 ± 0.012
St. 28	Feb 6, 2016	2,000	250,000	ND (0.02)	0.95 ± 0.018
St. 29	Feb 6, 2016	2,000	250,000	ND (0.01)	0.86 ± 0.015
St. 30	Feb 6, 2016	—	—	—	—
St. 31	Feb 6, 2016	—	—	—	—
St. 32	Feb 6, 2016	2,000	250,000	ND (0.02)	0.28 ± 0.013
St. 33	Feb 6, 2016	2,000	250,000	LTD (0.03)	1.45 ± 0.023
St. 33*	Feb 6, 2016	2,000	600,000	0.034 ± 0.0086	1.43 ± 0.015

Gamma ray of the seabed soil of the Notsuke Strait was measured. Asterisk(*) shows the results measured for longer time again. Although the concentrations of ^{134}Cs except for St. 33* were judged to be ND, the obtained values were included in the parenthesis to know the relation. We were not able to measure the samples of St. 6, St. 30, and St. 31 because the amount is very few.

of accident. From this fact, the concentration of ^{137}Cs from the global fallout at St.33 is supposed to be 1.60–0.17 = 1.43 Bq/kg on March 11, 2011. In addition, on Figure 2, the vertical lengths to St.27, 28, 29, 32 points from line of y = 1.0 x are supposed to be the concentration of ^{137}Cs from global fallout.

The Notsuke Strait is located at the offing of Shibetsu-cho, Hokkaido in southern part of the Okhotsk Sea. Ministry of Education, Culture, Sports, Science and Technology investigated the radioactivity of the whole Japan by plane and carried it out in Hokkaido from April to May, 2012. The air dose rate of the altitude of 1 m from the earth surface of Hokkaido is shown in the investigation report[12], and this shows sedimentation of radiocesium on the earth surface soil. According to the report of this investigation, ^{134}Cs and ^{137}Cs were less than 10 kBq/m^2 in whole Hokkaido. It was the lowest level in the whole Japan. The remarkable deposition was not reported in it. According to the report of the Hokkaido Government Agricultural Administration Department, the radioactivity of ^{134}Cs was all ND in the earth surface soil radioactivity investigation including farmland of the whole Hokkaido. And the radioactivity concentration of ^{137}Cs was less than the value before the FDNPP accident[13]. From the result above, it is thought that there were very little ^{134}Cs which flowed out from the river of the neighborhood of Shibetsu-cho into the sea area.

In the sea area, in April 2010 of the preceding year of the FDNPP accident, the Japan Coast Guard collected surface seawater in offing of Monbetsu, Hokkaido (northeast offing approximately 74 km far from Monbetsu, 44 ° 50′ N, 144 ° 00′ E) in the southern Okhotsk Sea, and investigated radioactivity. Concentration of ^{137}Cs was 0.96 ± 0.04 mBq/L, but in June 2011

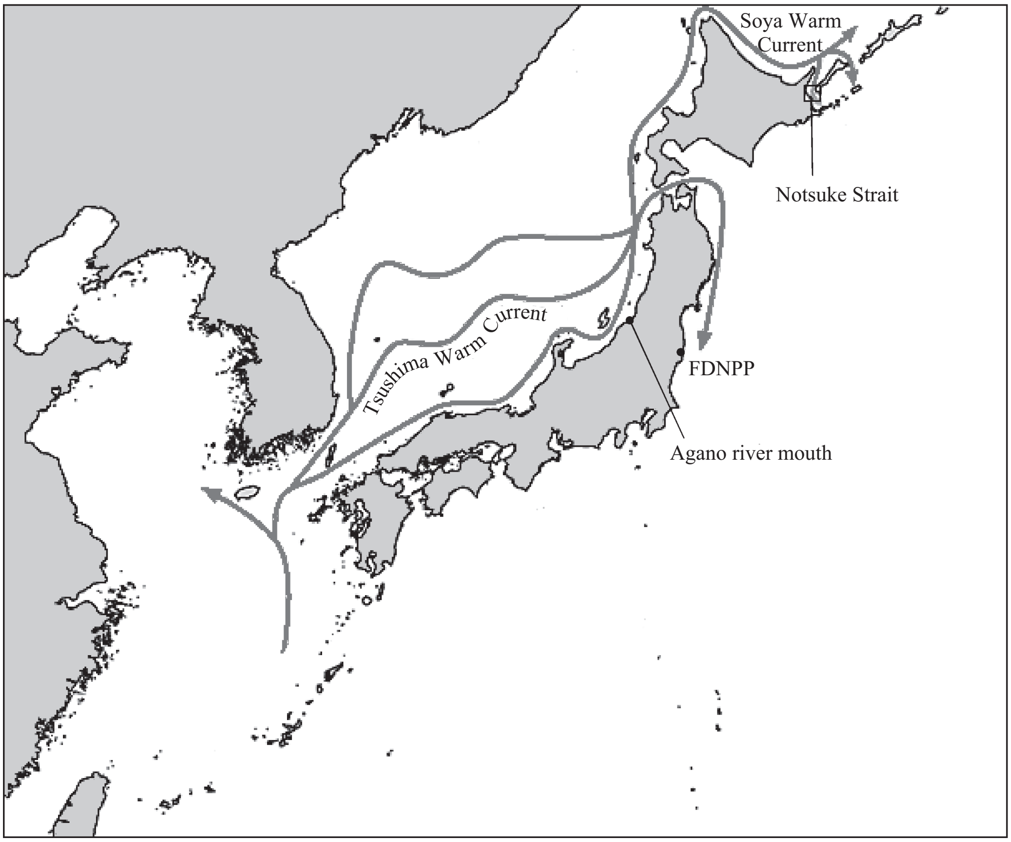

Fig 4. Tsushima Warm Current and Soya Warm Current [14].
The Tsushima Warm Current and the Soya Warm Current contribute to the migration of Cs from the Agano River mouth to the Notsuke Strait.

of the next year, 3.14 ± 0.07 mBq/L that was 3.3 times higher than the preceding year was detected in the investigation of the same sea area[6–7]. The method of Japan Coast Guard was that the Cs of the surface seawater was adsorbed by ammonium phoshomolybdate and was separated with the cation exchange resin. From this, it can be judged that ionic cesium exists in surface seawater, and Cs of NO-10 point migrated to the Notsuke Strait by the ocean currents and was adsorbed to the seabed soil. In addition, there is the ocean current in this nearby sea area, which is shown in Figure 4[14]. The Tsushima Warm Current flows near the Agano River mouth and a part of the Tsushima Warm Current flows through the Soya Strait. The Soya Warm Current flows to the Pacific. The physical property of these ocean currents gradually changes, but these ocean currents are a consecutive ocean current for the material transportation[15]. It is known that the radioactive materials by the global fallout deposited in the seabed soil of the Japanese peripheral sea area. According to a Report of Radioactivity Surveys (Results of Surveys in 2010) by the Japan Coast Guard, the average concentration of ^{137}Cs of the seabed soil was 2.5 Bq/kg on the Japanese peripheral sea area. However, the concentration of ^{137}Cs of the seabed soil at the Notsuke Strait was extremely low than the average concentration of ^{137}Cs of the seabed soil on the Japanese peripheral sea area. It is known that Cs is easily adsorbed by clay. A lot of coarse sand and shells were included in the seabed soil collected in the Notsuke Strait, but there were few clay and silt. Therefore, it is supposed that there was little quantity of adsorption of the Cs by the global fallout. Similarly, the Cs derived from FDNPP is supposed that there was a little quantity of adsorption. Japan Coast Guard investigates the radioactivity of the surface water every year. The concentration of ^{137}Cs at NO-10 point (Figure 1) increased in 2011[16]. It is thought that some of these ^{137}Cs were adsorbed in the seabed soil at the Notsuke Strait. It can be judged that the ^{134}Cs obtained in the present study is reached from the FDNPP accident. We can suppose that radioactive material released by TEPCO FDNPP accident reached this sea area by the Tsushima Warm Current and the Soya Warm Current from the Agano River mouth. We continue the sampling and the radioactivity measurement of the seabed soils at Sakata offing and Soya offing.

4. Conclusions

For the investigation of the migration pathway, we sampled the seabed soil at the Notsuke Strait and measured the γ-ray spectrum with the Ge semiconductor detector. The γ-ray spectrum had peak of ^{134}Cs at 605 keV and that of ^{137}Cs at 662 keV. We measured concentration of ^{134}Cs with 0.033 ± 0.0084 Bq/kg, and the concentration of ^{137}Cs with 1.4 ± 0.015 Bq/kg. The radioactivity investigation with the plane and the farmland earth surface soil radioactivity investigation did not show the remarkable deposit of artificial radioactive materials in Hokkaido. From these results and consideration of ocean current, it was suggested that the radioactive material released from FDNPP accident reached the Notsuke Strait from the Agano River mouth by the Tsushima Warm Current and the Soya Warm Current.

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