

Studies on Migration Pathway from the Japan Sea  
to the Sea of Okhotsk of Radioactive Cesium  
Derived from the Fukushima Daiichi Nuclear  
Power Plant

( 福島第一原子力発電所に由来する放射性セシ  
ウムが日本海からオホーツク海への移行経路  
に関する研究 )

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## Chapter 1      General Introduction

## 1.1 Purpose of this study

Fukushima Daiichi Nuclear Power Plant (FDNPP) accident occurred on March 11, 2011, and a large quantity of radioactive material was released in environment, and it became a serious disaster. Refuge dwellers more than 400,000 appeared <sup>1-1)</sup>.

Because radioactive material was released in environment, Fukushima prefectural government office carried out Fukushima Health Management survey. The Fukushima Health Management survey is aimed for evaluation of exposure dose, grasp of health condition, prevention of illness and early treatment of illness. The final purpose is maintenance of health and its increase until the future. The interim report in the Fukushima Health Management survey was announced in March, 2016. Approximately 300,000 people received thyroidal inspection, and 113 people received a judgment of thyroid cancer. The influence of radiation was suspected. But the influence caused by radioactivity is not completely denied at this stage, and accumulation of long-term information is indispensable for the impact statement <sup>1-2)</sup>.



When a radiation accident occurs, radioactive materials are released in environment, and healthy damage may occur. It is necessary to minimize radiological influence on human body. Therefore, environmental radioactivity surveys are carried out, and air dose rate and migration pathway are surveyed. However, there are few studies of migration pathway at the Japan Sea and the Sea of Okhotsk, and the migration pathway is not clarified. A purpose of this study is to clarify migration pathway of radioactive cesium derived from FDNPP at the Japan Sea and the Sea of Okhotsk.

I used a radiochemical method to clarify the migration pathway. Many kind of radioactive materials were released from FDNPP in environment, but the nuclides that half-life is relatively long are  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ .  $^{134}\text{Cs}$  is a nuclide peculiar to nuclear power plant accident, and  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  are also released by the nuclear test in the atmosphere.  $^{137}\text{Cs}$  is generated in a nuclear reactor by nuclear fission, but  $^{134}\text{Cs}$  is not generated by the nuclear fission.  $^{133}\text{Cs}$  formed by nuclear fission in the nuclear reactor becomes  $^{134}\text{Cs}$  by the irradiation of the neutron. Therefore, the production of  $^{134}\text{Cs}$  is controlled by the driving time for nuclear reactor and the type

of reactor. The  $^{134}\text{Cs}/^{137}\text{Cs}$  radioactivity ratio was approximately 0.5 in the Chernobyl accident, while it was approximately 1 in the case of the FDNPP accident. When the  $^{134}\text{Cs}/^{137}\text{Cs}$  radioactivity ratio is used for analysis, radioactive material derived from the nuclear test in the atmosphere and the radioactive material derived from FDNPP accident can be estimated. Six years passed since FDNPP accident, and the radioactive materials derived from FDNPP are decreasing. However, there are many kinds of radioactive materials in environment, and each nuclide emits the radiation. If energy distribution and strength of the  $\gamma$ -ray are measured in the case of  $\gamma$ -ray emitter, its nuclide and concentration can be estimated. In this way, the radiochemistry is essential to study the migration pathway of radioactive material derived from FDNPP.

It is also thought that the result of this study contributes to the evaluation of the impact to the human body, the prevention of the radiation exposure, and decontamination of radioactive materials.

This thesis is mainly referred to the next three next papers.

(1) Nabaе, Y., Miyashita, S., Nakashima, S.: Observation of radiocesium in

seabed soil at the Notsuke Strait of the southern Sea of Okhotsk derived from the Fukushima Daiichi Nuclear Power Plant, Radiation Safety Management, 15, 9 - 15 (2016).

(2) Nabae, Y., Miyashita, S., Nakashima, S.: Observation of radioactive cesium in seabed soil at the Soya Strait derived from the Fukushima Daiichi Nuclear Power Plant, Radiation Safety Management, 16, 8 - 12 (2016).

(3) Nabae, Y., Tsujimoto, M., Miyashita, S., Nakashima, S.: Distribution of radioactive cesium from the Fukushima Daiichi Nuclear Power Plant in seabed soil from the Niigata Prefecture and Yamagata Prefecture offings, in preparation.

## 1.2 The 2011 off the Pacific coast of Tohoku Earthquake

The 9.0-magnitude earthquake occurred at the Sanriku offing on 14:46, March 11, 2011. The maximum seismic intensity of each place was Miyagi -7, Fukushima, Ibaraki, and Tochigi – 6 upper. Seismic intensity of 6 lower to 1 was observed in

the wide areas from Hokkaido to the Kyushu district. In addition, a tsunami occurred by this earthquake. The height of tsunami was over 9.3 m at Soma and over 8.6 m at Ayukawa. It was very high in Pacific side of North Kanto region, and this tsunami was observed from Hokkaido to Okinawa <sup>1-3)</sup>. This tsunami damaged the town at the Sanriku Coast. The area suffered the damage from the tsunami spread to the Sendai plain where is a grain belt approximately 5 km from the shoreline. In addition, the tsunami went up in the Natori River or the Abukuma River, and water level changed from the river mouth to approximately 49 km from the shoreline in the Kitakami River. Geographical Information Authority of Japan investigated the situation of the damage by aerial photos and local surveys in the stricken area. The inundation area of 62 cities, towns and villages in Aomori, Iwate, Miyagi, Fukushima, Ibaraki and Chiba was about 535 km<sup>2</sup>, and 40 % or more of the area were flooded higher than 2 m <sup>1-4)</sup>.

## 1.3 FDNPP accident

### 1.3.1 FDNPP

FDNPP locates in Okuma-machi and Futaba-machi, Futaba-gun, Fukushima, the east faces the Pacific. This power station has an area of approximately 3,500,000 m<sup>2</sup>, and is the Tokyo Electric Power Company's first nuclear power plant. The first nuclear reactor started commercial operation on March, 1971, and there are six nuclear reactors now, and total generation facilities capacity is 4,696,000 kW<sup>1-5</sup>).

### 1.3.2 Driving situation of FDNPP on March 11, 2011

FDNPP the first reactor, the second reactor and the third reactor were running on March 11, 2011. The fourth reactor was stopped for periodical inspection, and all nuclear fuels were transferred from a nuclear reactor to a used nuclear fuel pool. Periodical inspection was carried out for the fifth reactor, however the nuclear fuel was loaded in the nuclear reactor. The sixth reactor was also in periodical inspection,

but nuclear fuel was loaded in the nuclear reactor, and it was the state of cold shutdown. Because the external power supplies for the third reactor and the fourth reactor were under construction on the time of earthquake, the usable external power supplies were 6 lines <sup>1-5</sup>).

### 1.3.3 Disaster caused by the earthquake and the tsunami

The earthquake occurred at 14:46 on March 11, 2011. Because acceleration exceeded a standard at FDNPP, all reactors stopped automatically. Steel towers for power lines collapsed outside a site, and outside power lines were cut off. The emergency diesel generators started automatically, the cooling function of nuclear reactors and used nuclear fuel pools were maintained. Afterwards, the tsunami caused by the earthquake hit FDNPP, and emergency diesel generators and distributors became under water. All emergency diesel generators except for the sixth nuclear reactor stopped, and the 1-5 nuclear reactor lost all AC power supplies. In a turbine building of the first reactor, it was confirmed that radiation dose rose at

about 23:00 on March 11. The pressure of the storage container exceeded the limit pressure at 0:49 on March 12, and the explosion such as the hydrogen explosion occurred in the upper first nuclear reactor at 15:36 on March 12. Nuclear reactor water level decreased in the second reactor at 13:25 on March 14. The large impulsive sound such as the hydrogen explosion was confirmed in the neighborhood of pressure control room at about 6:00 on March 15, and the pressure of the pressure control room decreased rapidly. A nuclear reactor cooling system stopped in the third reactor at 11:36 on March 12, a wet vent was operated to reduce the pressure of the reactor pressure vessel. Because the pressure of the reactor pressure vessel rose, vents of the reactor pressure vessel were operated several times. The explosion such as the hydrogen explosion occurred in the upper third nuclear reactor at 11:01 on March 14. In addition, because a radioactivity accident occurred, Nuclear and Industrial Safety Agency temporarily estimated it to be level 3 (serious abnormality phenomenon) of International Nuclear Event Scale, and reported it to IAEA on March 12<sup>1-5)</sup>.

### 1.3.4 The radioactivity released by the accident

Nuclear and Industrial Safety Agency announced a gross value of radioactive material released in the atmosphere by FDNPP accident on April 12, 2011, and announced again an estimation value based on a new test calculation on June 6, 2011. On February 1, 2012, Nuclear and Industrial Safety Agency revised the estimation value of total radioactive material released in the seventh Tokyo Electric Power Company FDNPP accident technical knowledge hearing society. Because the situation of the accident of 2-3 nuclear reactor was changed, Nuclear and Industrial Safety Agency announced  $^{131}\text{I}$ -150,000 TBq and  $^{137}\text{Cs}$  -8,200 TBq as the estimation value of radioactive materials released in the atmosphere. When these are converted into iodine, it becomes 480,000 TBq.

$$\text{Iodine reduced value} = {}^{137}\text{Cs}(\text{Bq}) \times 40.$$

Nuclear Safety Commission estimated radiation dose around FDNPP by monitoring results and SPEEDI in cooperation with JAEA. Nuclear Safety Commission also estimated the radioactive materials gross value released in the



atmosphere in the process. According to this, the value released in the atmosphere by FDNPP accident was  $^{131}\text{I}$  - 150,000 TBq and  $^{137}\text{Cs}$  - 12,000 TBq. The value converted into iodine becomes 630,000 TBq. Nuclear Safety Commission announced the both values on April 12. After that, Nuclear Safety Commission acquired environmental monitoring data before March 15, 2011, and analyzed it again. As a result, radioactive materials gross value which was released in the atmosphere were estimated to be  $^{131}\text{I}$ -130,000 TBq,  $^{137}\text{Cs}$ -11,000 TBq. When these are converted into iodine, it becomes 570,000 TBq. The Nuclear Safety Commission announced the both values on August 24.

Tokyo Electric Power Company estimated radioactive materials gross value released in the atmosphere by Dose Information Analysis for Nuclear Accident (DIANA). DIANA is a program to calculate air dose rate when radioactive material was released in the atmosphere. Tokyo Electric Power Company calculated the radioactive materials gross value based on monitoring data and weather data reversely. The radiological gross value released in the atmosphere became  $^{131}\text{I}$  - 500,000 TBq,  $^{137}\text{Cs}$ - 10,000 TBq. The value converted into iodine becomes 900,000

TBq. In addition, Tokyo Electric Power Company estimated radioactive materials gross value released in the ocean in cooperation with Central Research Institute of Electric Power Industry. A program to calculate concentration of radioactive material when radioactive material was released in the ocean was used, and this program is based on monitoring data of the seawater. According to this, the radioactive materials gross value released in the ocean by FDNPP became  $^{131}\text{I}$  - 11,000 TBq,  $^{137}\text{Cs}$  - 3600 TBq. Tokyo Electric Power Company announced these results of estimation on May 24, 2012 <sup>1-6)</sup>.

#### 1.4 Migration of radioactive cesium in the atmosphere

The radioactive cesium was released from FDNPP to the atmosphere, migrated by wind to leeward, and was spread to horizontally and vertically. The radioactive cesium was migrated in this way in the atmosphere, decayed by half-life, and gradually decreases. The radioactive cesium in atmosphere is deposited in the earth surface with the dry deposition by the current of air and wet deposition by the

rainfall. When wind blows to one direction, radioactive cesium is migrated such as smoke in the atmosphere. This is radioactive plume. However, the wind rarely blows constantly for a long time, the wind is affected by the topography, earth surface state, land breeze, sea breeze, mountain breeze, valley breeze, high atmospheric pressure, low atmospheric pressure, and the wind is changed complicatedly. Around FDNPP, the western mountains and local wind formed a complicated current of air. Therefore, it is thought that radioactive cesium migrated complicatedly. The radioactive cesium migrated in the atmosphere, and it is eliminated by dry deposition and wet deposition in the atmosphere. When precipitation occurs at radioactive plume area of radiological particle, high pollution area appears. This is a hotspot. Dry deposition of  $^{131}\text{I}$  and wet deposition of  $^{137}\text{Cs}$  are easy to happen<sup>1-7)</sup>.

$^{134}\text{Cs}$  from FDNPP was detected in surface seawater at the Shimane offing (35 ° 41 ' N 133 ° 04 ' E) and the Ishikawa offing (37 ° 08 ' N 136 ° 26 ' E) of the Japan Sea<sup>1-19)</sup>. However, because  $^{134}\text{Cs}$  was not detected in the neighboring land,

it is thought that  $^{134}\text{Cs}$  migrated from the atmosphere to the Japan Sea at the Shimane offing and the Ishikawa offing.

### 1.5 Influence of radioactive materials on soil of Japan as a whole

Ministry of Education, Culture, Sports, Science and Technology and United States Department of Energy carried out plane monitoring jointly. The purpose is a survey of the air dose rate of the height of 1 m from earth surface, and the survey of radioactive materials in the wide area at the evacuation zone in a range of 80 km from FDNPP. The plane equips a large and high sensitivity radiation detector. This is a method that gamma ray can be measured from the radioactive material which deposited on the ground, a large area is measured quickly.

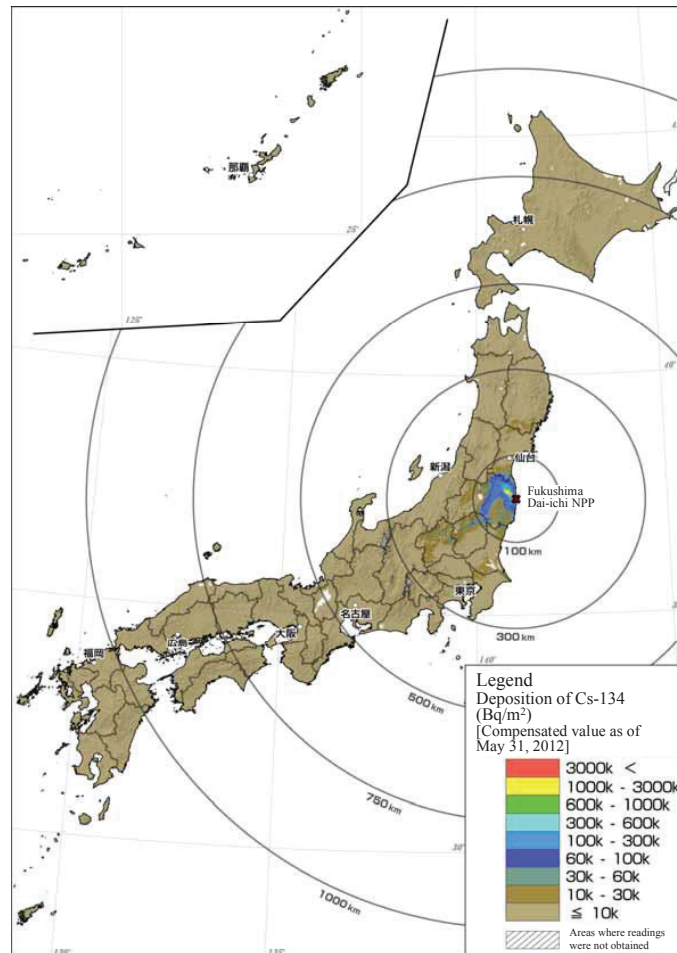
Atomic Energy Security Technology Center which belonged to Ministry of Education, Culture, Sports, Science and Technology and United States Department of Energy carried out the first plane monitoring during 6-29 April, 2011. The Atomic Energy Security Technology Center carried out investigation at area of 60-

80 km from FDNPP using private helicopter (BELL412). United States Department of Energy carried out investigation in area of 60 km from FDNPP using small plane (C-12) and helicopter (UH-1). The flight altitude is 150-700 m from the surface of the earth. Air dose rate is used average as value of air dose rate in footprint of 300-1500 m in diameter. Because flight of the low altitude is difficult, the mountains were not measured. However, the Atomic Energy Security Technology Center measured the mountains by monitoring car, and it was confirmed that air dose rate was less than 1  $\mu\text{Sv/h}$ . The plane monitoring was not performed in the sky of FDNPP, because this area was measured for the direct air dose rate from the power plant. Air dose rate and radioactive materials deposition were decay-corrected on April 29, 2011 that was the last day when monitoring was carried out. After the first plane monitoring, the next plane monitoring was carried out in the whole area of Japan. In addition, the plane monitoring is continued in outskirts area of Fukushima. Figure 1-1 and Figure 1-2 show result of the plane monitoring<sup>1-8)</sup>.

The high deposition was distributed in the neighboring area of FDNPP. The high deposition was shown at Nakadouri and Iidate, and the depositions were also

shown at Yamagata, Gunma and Tochigi.  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio in all soil at the neighboring area of FDNPP was approximately 1, and it is thought that the behavior of  $^{137}\text{Cs}$  was the same as  $^{134}\text{Cs}$ . When radioactive material was released, it rained. The aerosol and the water-soluble gas were washed by rain. According to AMEDAS, it rained in Fukushima from 17:00 March 15 to 4:00 March 16, and rained in Ibaraki, Chiba, Tochigi, Saitama, and Tokyo from 8:00 March 21 to 6:00 on March 23. Most of radioactive materials on the surface of the earth deposited at this time <sup>1-9)</sup>.

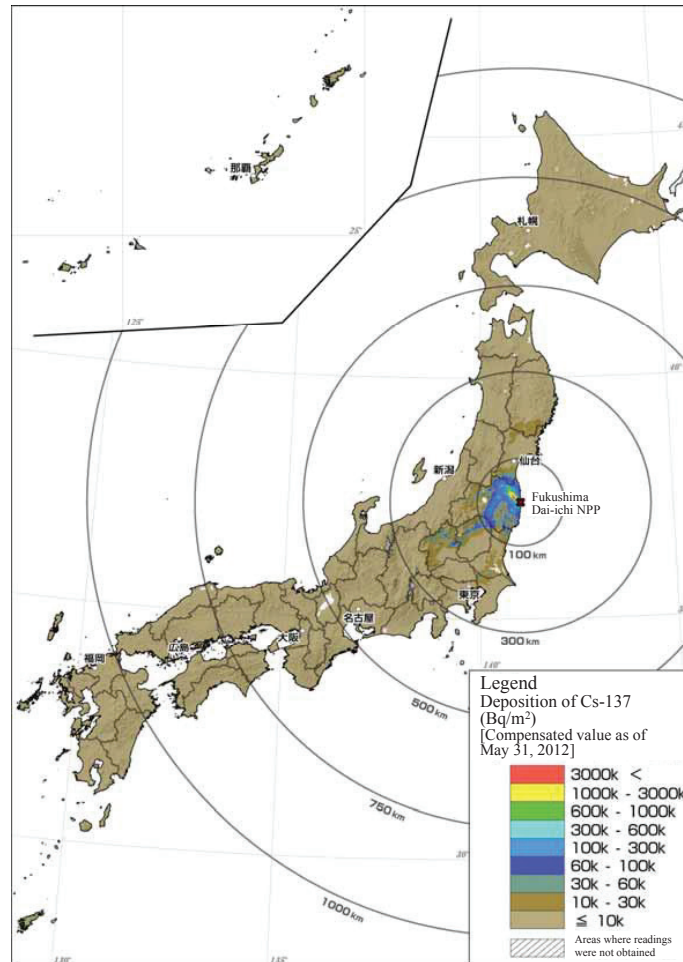
Measurement Results of the Airborne Monitoring Surveys Conducted by MEXT Nationwide  
(Deposition of Cs-134 on the ground surface nationwide)



\* With regard to the areas targeted in the Second Airborne Monitoring (within an 80 to 100km-range (or 120km-range on the southern part) from Fukushima Dai-ichi NPP), and part of the areas targeted in the Fourth Airborne Monitoring (within a 40 to 80km-range from Fukushima Dai-ichi NPP), as well as Aichi, Aomori, Akita, Ishikawa, Iwate, Kanagawa, Gifu, Saitama, Shizuoka, Chiba, Tokyo, Toyama, Nagano, Niigata, Fukui, and Yamanashi prefectures, and the western part of Fukushima prefecture, out of areas in the eastern part of Japan, the past measurement results of the monitoring were revised using the method to assess the influence of natural radionuclides in detail, which was developed for the airborne monitoring survey in the western part of Japan.

Figure 1-1 Deposition of  $^{134}\text{Cs}$ .

Measurement Results of the Airborne Monitoring Surveys Conducted by MEXT Nationwide  
(Deposition of Cs-137 on the ground surface nationwide)



\* With regard to the areas targeted in the Second Airborne Monitoring (within an 80 to 100km-range (or 120km-range on the southern part) from Fukushima Dai-ichi NPP), and part of the areas targeted in the Fourth Airborne Monitoring (within a 40 to 80km-range from Fukushima Dai-ichi NPP), as well as Aichi, Aomori, Akita, Ishikawa, Iwate, Kanagawa, Gifu, Saitama, Shizuoka, Chiba, Tokyo, Toyama, Nagano, Niigata, Fukui, and Yamanashi prefectures, and the western part of Fukushima prefecture, out of areas in the eastern part of Japan, the past measurement results of the monitoring were revised using the method to assess the influence of natural radionuclides in detail, which was developed for the airborne monitoring survey in the western part of Japan.

Figure 1-2 Deposition of <sup>137</sup>Cs.



## 1.6 Influence of FDNPP accident on soil of Hokkaido

The Hokkaido Prefectural Government Agricultural Administration Department carried out monitoring of the soil in farmland of Hokkaido. In September, 2011, the soil was collected in central agricultural experimental station (Naganuma-cho), Kamikawa agricultural experimental station (Pippu-cho), Southern Hokkaido agricultural experimental station (Hokuto-cho), Kamikawa agricultural experimental station – Tenhoku branch (Hamatonbetsu-cho), Tokachi agricultural experimental station (Memuro-cho), Kitami agricultural experimental station (Kunneppu-cho), Kosen agricultural experimental station (Nakashibetsu-cho). Hokkaido Inst. of Public Health analyzed the soil.  $^{131}\text{I}$  and  $^{134}\text{Cs}$  were not detected in all soils. The concentration of  $^{137}\text{Cs}$  was 4.9-13.3 Bq/kg (drying), and the value was lower than environmental radioactivity standard survey for the past 3 years of 2008 - 2010 <sup>1-10</sup>).

## 1.7 Migration of radioactive cesium at the land

Most deposited radioactive cesium by rain is trapped to canopy in the forest. Afterwards it is estimated that radioactive cesium migrated from canopy to the forest floor by the rain in the forest mainly. In an initial stage, the ratio that canopy traps radioactive cesium is different between evergreen and deciduous trees, and the ratio changed approximately 60-90 %. The radioactive cesium which deposited in the tree drops by rain, and the quantity of fall is strongly affected by rain strength and frequency<sup>1-11)</sup>.

Radioactive cesium flows from the forest to the river, and is transported in dissolved state or suspension state from the upper stream to the downstream<sup>1-12)</sup>. In addition, it is thought that a part of radioactive cesium is deposited on riverbed and river bank in transportation process. The movement situation of radioactive cesium was observed in the Natsui River and the Same River of Fukushima. The suspension state radioactive cesium was 21-56 % in normal river water, but occupied approximately 100 % in the river water of the typhoon. One heavy rain moves 30-50 % of the annual suspension state radioactive cesium. The suspension

state radioactive cesium influences considerably migration of radioactive cesium in the river. On the other hand, from June, 2011 to July, 2013, dissolved state radioactive cesium concentration in mountain stream of Kuchibuto River water and groundwater were investigated. Dissolved state radioactive cesium concentration in mountain stream water suddenly decreased until November, 2011, and gradually decreased afterwards.

In the river mouth neighborhood, the salinities of the river water increase, and the concentration of dissolved state radioactive cesium increases to approximately 2 times. Because suspension state / dissolved state decreased, it was suggested that a part of the suspension state radioactive cesium was detached and became to dissolved state radioactive cesium.

## 1.8 Migration of radioactive cesium in the sea

In seawater, radioactive cesium is comprised of 99 % of dissolved state and 1 % of suspension states. The sedimentation rate of the suspension state radioactive cesium

is around 10-100 m per a day. In the study of the Chernobyl accident, there are reports that radioactive cesium was adsorbed to organic matter. In the study of the FDNPP accident, there were reports that radioactive cesium was adsorbed to mineral such as aluminum silicate. In the coast and a continental shelf, seabed soil which adsorbed radioactive cesium rises again, and it may be transported horizontally towards continental shelf slope<sup>1-13</sup>).

Japan Coast Guard carries out radioactive surveys in the ocean for marine pollution prevention and marine environmental conservation. Radioactivity surveys at Japanese peripheral sea area were carried out in 2011, and radioactive cesium derived from FDNPP was detected in a part of samples. Concentration of <sup>137</sup>Cs in surface seawater at west sea area of Hokkaido increased by 2.42 mBq/kg at the maximum in 2011 compared with 2010, and also concentration of <sup>137</sup>Cs in surface seawater at the Sea of Okhotsk increased by 2.18 mBq/kg<sup>1-14, 1-15</sup>). <sup>134</sup>Cs of 0.7 Bq/kg was detected in seabed soil at Ishikari Bay in 2012, and influence by the FDNPP was clarified<sup>1-16</sup>).

## 1.9 The detection of radioactive cesium from the Agano River mouth, the Mogami River and the Shinano River offing

Niigata prefectural government investigated the Agano River and the offing from August 2011 to May 2012 to know the radioactive contamination by FDNPP accident<sup>1-17)</sup>. In the Agano River,  $6.9 - 1.2 \times 10^2$  Bq/kg of radioactive cesium was detected from sediment, and the muddy sediment had high radioactive cesium concentration.  $4.2 \times 10^2$  Bq/kg of radioactive cesium at the maximum was detected from seabed soil of the Agano River offing by Niigata-Fukushima heavy rain in July, 2011. The highly-concentrated radioactive cesium of seabed soil migrated from the shallow seabed to the deep seabed in the sea. At the Agano River and Agano River mouth offing, much radioactive cesium was included in the particles less than 250  $\mu\text{m}$ .

The survey was carried out in the Sakata offing in this study. The Sakata offing is the sea area adjacent to the Mogami River mouth. Yamagata prefectural government office measured the sediment of Mogami River and announced the result<sup>1-18)</sup>.  $^{134}\text{Cs}$  of 490 Bq/kg and  $^{137}\text{Cs}$  of 760 Bq/kg were detected in sediment

from the Osawa River of the Mogami River branch on July 18 - 21, 2012, and these concentrations were the highest values of the river in Yamagata prefecture.

Radioactivity survey was also conducted at May 22, 2011 at the Shinano river mouth offing (37 ° 150 'N, 138 ° 135' E) <sup>1-19)</sup>. 0.55 mBq/L of <sup>134</sup>Cs from surface seawater, 0.10 mBq/L of <sup>134</sup>Cs from near-bottom water and 7.7 mBq/g of <sup>134</sup>Cs from the seabed soil were detected. The source of <sup>134</sup>Cs detected from near-bottom water is thought to be due to change from sedimentary particles to buoyant particles, leaching from sediment or organic matter.

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## Chapter 2      Sampling and Experiments

## 2.1 Sampling

Seabed soil was collected from the Notsuke Strait, the Soya Strait, the Sakata offing, the Kamo offing and the Naoetsu offing. Figure 2-1 shows the sea survey area.

The radioactivity survey was carried out by small ships and the bottom sampler, and the surface layer of seabed soil was collected. Table 2-1 shows the sampling position, depth and sampling day of seabed soil. The stones and the visible creature with naked eye were removed, and it was transported from the sea survey area to the laboratory. Seabed soil was dried at 95 °C for more than 24 h and passed through a 2 mm sieve in the laboratory.

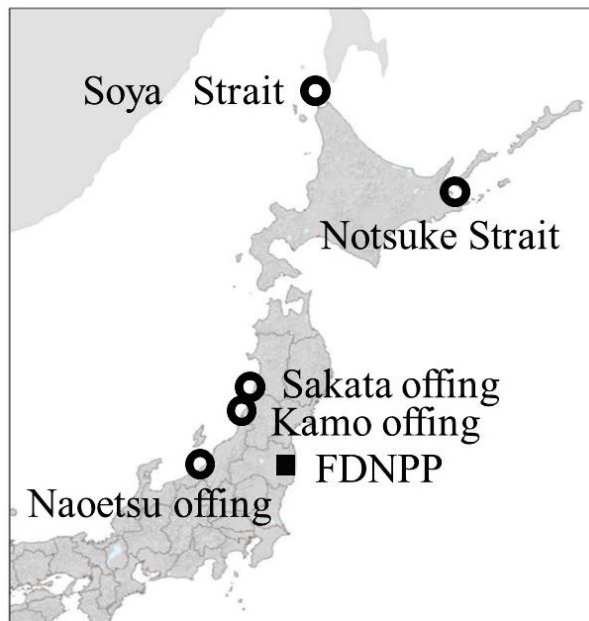


Figure 2-1 Sampling sea area.

Table 2-1 Sampling day, position and depth

Survey arias	Sampling day	No.	Latitude	Longitude	Depth [m]
Notsuke Strait	23-Feb-15	St.1	43° 40' 12" N	145° 12' 15" E	8.0
	23-Feb-15	St.2	43° 38' 21" N	145° 13' 23" E	8.6
	23-Feb-15	St.3	43° 38' 46" N	145° 14' 31" E	5.4
	23-Feb-15	St.4	43° 38' 43" N	145° 17' 14" E	5.0
	23-Feb-15	St.5	43° 38' 14" N	145° 19' 26" E	8.4
	23-Feb-15	St.6	43° 40' 36" N	145° 17' 7" E	10.4
	23-Feb-15	St.7	43° 39' 30" N	145° 17' 31" E	12.3
	23-Feb-15	St.8	43° 40' 44" N	145° 14' 21" E	15.2
	23-Feb-15	St.9	43° 41' 12" N	145° 11' 26" E	13.2
	6-Feb-16	St.27	43° 40' 20" N	145° 7' 56" E	4.0
	6-Feb-16	St.28	43° 41' 30" N	145° 7' 7" E	5.0
	6-Feb-16	St.29	43° 43' 42" N	145° 5' 37" E	9.0
	6-Feb-16	St.30	43° 45' 32" N	145° 4' 2" E	5.0
6-Feb-16	St.31	43° 47' 21" N	145° 3' 55" E	5.0	
6-Feb-16	St.32	43° 38' 21" N	145° 13' 23" E	10.0	
6-Feb-16	St.33	43° 39' 56" N	145° 8' 8" E	5.5	
Soya Strait	9-Sep-15	St.19	45° 29' 09" N	141° 52' 39" E	2.8
	17-Sep-15	St.20	45° 28' 58" N	141° 50' 59" E	20.4
	17-Sep-15	St.21	45° 29' 12" N	141° 50' 46" E	22.6
	17-Sep-15	St.22	45° 28' 48" N	141° 50' 26" E	23.5
	17-Sep-15	St.23	45° 29' 43" N	141° 50' 16" E	28.2
	9-Sep-15	St.24	45° 29' 06" N	141° 52' 43" E	3.2
	10-Sep-15	St.25	45° 25' 28" N	141° 40' 26" E	0.5
10-Sep-15	St.26	45° 26' 59" N	141° 38' 44" E	0.5	
Sakata Offing	13-Jul-15	St.10	38° 59' 06" N	139° 50' 12" E	8.2
	13-Jul-15	St.11	38° 59' 18" N	139° 48' 30" E	26.8
	13-Jul-15	St.12	38° 59' 30" N	139° 47' 00" E	44.7
	13-Jul-15	St.13	39° 00' 06" N	139° 45' 12" E	53.6
	13-Jul-15	St.14	39° 00' 18" N	139° 43' 28" E	55.9
	13-Jul-15	St.15	38° 59' 59" N	139° 43' 31" E	55.5
	13-Jul-15	St.16	38° 58' 23" N	139° 44' 24" E	55.9
	13-Jul-15	St.17	38° 58' 12" N	139° 46' 18" E	45.7
13-Jul-15	St.18	38° 58' 00" N	139° 48' 14" E	22.4	
Kamo Offing	6-Aug-16	St.40	38° 45' 46" N	139° 43' 58" E	13.2
	6-Aug-16	St.41	38° 46' 22" N	139° 43' 27" E	17.0
	6-Aug-16	St.42	38° 46' 55" N	139° 41' 27" E	39.6
	6-Aug-16	St.43	38° 48' 02" N	139° 37' 48" E	59.6
	6-Aug-16	St.44	38° 48' 32" N	139° 36' 02" E	79.7
Naoetsu Offing	5-Aug-16	St.34	37° 14' 30" N	138° 20' 01" E	6.8
	5-Aug-16	St.35	37° 15' 38" N	138° 19' 16" E	20.9
	5-Aug-16	St.36	37° 15' 55" N	138° 18' 18" E	39.5
	5-Aug-16	St.37	37° 17' 01" N	138° 16' 53" E	62.3
	5-Aug-16	St.38	37° 17' 37" N	138° 15' 54" E	79.4
	5-Aug-16	St.39	37° 18' 23" N	138° 14' 42" E	99.7

## 2.2 Method for measuring radioactive cesium

Seabed soil was put in a U-8 or a 2 L Marinelli vessel and the radioactivity was measured. A germanium semiconductor (GEM40-76-XLB-C, Seiko EG&G Co., Ltd.) was used to measure the radioactivity of the sample. The  $\gamma$  radioactivity of all samples were measured for 80,000 s. When  $^{134}\text{Cs}$  was suggested but the measurement value is less than the three times of standard deviation (SD), the sample was measured again for 250,000 s. The seabed soil of St33 was measured for 250,000 seconds, it was, however, measured more for 600,000 seconds because the value was not more than three times of SD for 250,000 s. The concentrations of radioactivity were calculated by the official method <sup>2-1)</sup>, and the results of the measurements were decay-corrected to the quantity of radioactivity of the sampling day. The peak search, the detection lower limit and the Sum effect were calculated by the second smoothing differentiation, cooper method, revision calculating formula.  $\gamma$ -ray nuclide analysis program DS-P200/W32 (Seiko EG&G Co., Ltd.) was used for the calculation of the radioactivity. The analyzed nuclides are  $^{134}\text{Cs}$ ,

and  $^{137}\text{Cs}$ . Figure 2-2 shows the disintegration schemes of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ .  $\gamma$ -ray of 604.70 keV is emitted in  $^{134}\text{Cs}$ , and  $\gamma$ -ray of 661.65 keV is emitted in  $^{137}\text{Cs}$ . Because the emission probability is high and the separation from peaks of other radionuclides is easy, 604.70 and 661.65 keV were chosen for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , respectively.



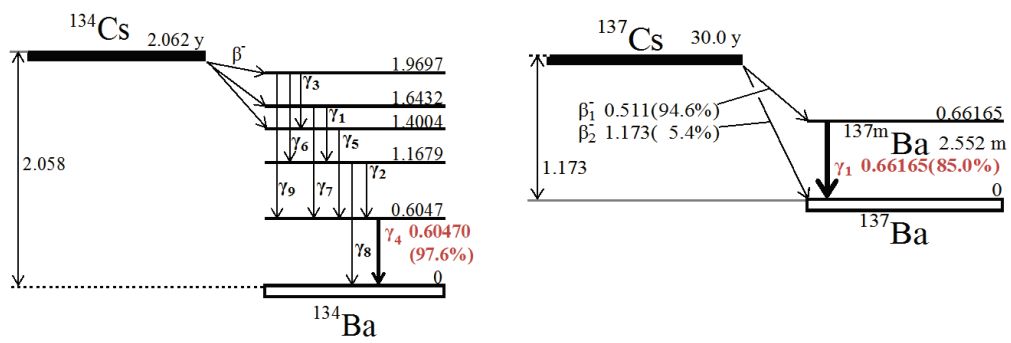


Figure 2-2 Disintegration schemes of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  (2-3).

### 2.3 Method for measuring grain size distribution

After the radioactivity measurements, the grain size distribution of the seabed soil was determined with sieves. According to the precedent study, when the sieving was carried out for two hours, the soil was fractionated more than 95 %<sup>2-2)</sup>. Therefore, this study carried out the sieving for three hours to get enough result. Portions of seabed soil (30 g) were separated by using 850, 250, and 75  $\mu\text{m}$  sieves, and the seabed soils were classified as coarse sand, medium sand, fine sand, and clay-silt. The sample isolation, sieving, and weighing were carried out twice for each sample. The average was calculated and the grain size distribution was determined.

### 2.4 References

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Chapter 3            Observation of Radioactive Cesium in  
Seabed Soil at the Notsuke Strait of the Southern Sea of  
Okhotsk Derived from the Fukushima Daiichi Nuclear Power  
Plant

### 3.1 Introduction

Tokyo Electric Power Company FDNPP accident occurred on March 2011. Large quantity of radioactive material was spread in environment, and the area around Fukushima was contaminated with the radioactive material <sup>3-1)</sup>. 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 <sup>3-2)</sup>. 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 <sup>3-3)</sup>. 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 <sup>134</sup>Cs was detected at the Agano River mouth mud in the investigation of August 2011. In addition, 72 Bq/kg of <sup>134</sup>Cs was observed in the seabed soil of 20 m in depth at the sea area of the Agano River mouth offing <sup>3-4, 3-5)</sup>.

On June 2011, 1.9 mBq/L of  $^{134}\text{Cs}$  was detected at surface seawater of Ishikari Bay in the northern Japan Sea, and 3.14 mBq/L of  $^{137}\text{Cs}$  which was 3.3 times of the preceding year was detected in the NO-10 point in the southern Sea of Okhotsk <sup>3-6, 3-7</sup>). In addition, some investigations were carried out in the Japan Sea and the Sea of Okhotsk, and  $^{134}\text{Cs}$  was detected at the northeastern part of the Japan Sea, Hokkaido coast and Tohoku coast <sup>3-8, 3-9</sup>).

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

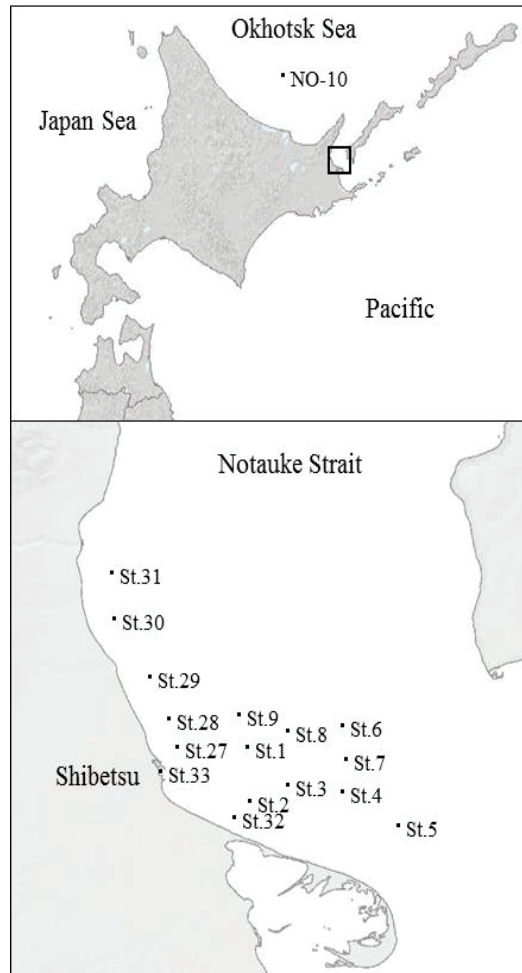


Figure 3-1 Sampling map at Notsuke Strait.

Seabed soil was sampled at the Notsuke Strait in the Sea of Okhotsk. Seabed soil at St.1-9 were collected in February 23, 2015. It of St.27-33 were collected in February 6, 2016. Japan Coast Guard investigates at NO-10 every year.

## 3.2 Sampling and experimental method

### 3.2.1 Sampling

On February 23, 2015, we collected seabed soil at the Notsuke Strait of the Sibetsu-cyo, Hokkaido. Figure 3-1 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.

### 3.2.2 Method for measurement of radioactive cesium

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 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  $^{134}\text{Cs}$  in all samples, therefore it was re-measured as St.2\* for 250,000 seconds. The concentrations of



$^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were calculated by official method <sup>3-10)</sup>, and the results of the measurements were decay-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  $^{134}\text{Cs}$ . And 662 keV of a peak was chosen for  $^{137}\text{Cs}$ . 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<sup>3</sup>) were measured for 250,000 seconds. And the sample of St.33 was measured again as St.33\* for 600,000 seconds, because the  $^{134}\text{Cs}$  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.

### 3.3 Results and discussion

Table 3-1 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 3-2 showed the sampling day, the volume of measurement, the time of measurement,  $^{134}\text{Cs}$  concentration and  $^{137}\text{Cs}$  concentration for the sample of the seabed soil.  $0.033 \pm 0.0084$  Bq/kg of  $^{134}\text{Cs}$  was detected for a sample of St.33.

Table 3-2 Position and Depth of sampling points at Notsuke Strait

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

Table 3-3 Results of the measurement of the sample at Notsuke Strait

No.	Sampling day	Volume [cm <sup>3</sup> ]	Times [s]	<sup>134</sup> Cs [Bq/kg]	<sup>137</sup> Cs [Bq/kg]
St.1	23-Feb-15	100	86,400	ND (0.19)	ND (0.33)
St.2	23-Feb-15	100	86,400	ND (0.33)	ND (0.20)
St.2*	23-Feb-15	100	250,000	ND (0.14)	0.41 ± 0.14
St.3	23-Feb-15	100	86,400	ND (-)	ND (0.25)
St.4	23-Feb-15	100	86,400	ND (0.22)	ND (0.39)
St.5	23-Feb-15	100	86,400	ND (-)	ND (0.16)
St.6	23-Feb-15	-	-	-	-
St.7	23-Feb-15	100	86,400	ND (0.19)	ND (0.19)
St.8	23-Feb-15	100	86,400	ND (-)	ND (0.02)
St.9	23-Feb-15	100	86,400	ND (0.09)	ND (-)
St.27	6-Feb-16	2,000	250,000	ND (0.01)	0.35 ± 0.012
St.28	6-Feb-16	2,000	250,000	ND (0.02)	0.95 ± 0.018
St.29	6-Feb-16	2,000	250,000	ND (0.01)	0.86 ± 0.015
St.30	6-Feb-16	-	-	-	-
St.31	6-Feb-16	-	-	-	-
St.32	6-Feb-16	2,000	250,000	ND (0.02)	0.28 ± 0.013
St.33	6-Feb-16	2,000	250,000	LTD (0.03)	1.45 ± 0.023
St.33*	6-Feb-16	2,000	600,000	0.034 ± 0.0086	1.43 ± 0.015

γ- Ray of the seabed soil of the Notsuke Strait was measured. \* shows the results measured for longer time again. Although the concentrations of <sup>134</sup>Cs except for St.33\* were judged to be ND, the obtained values were included in ( ) 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.

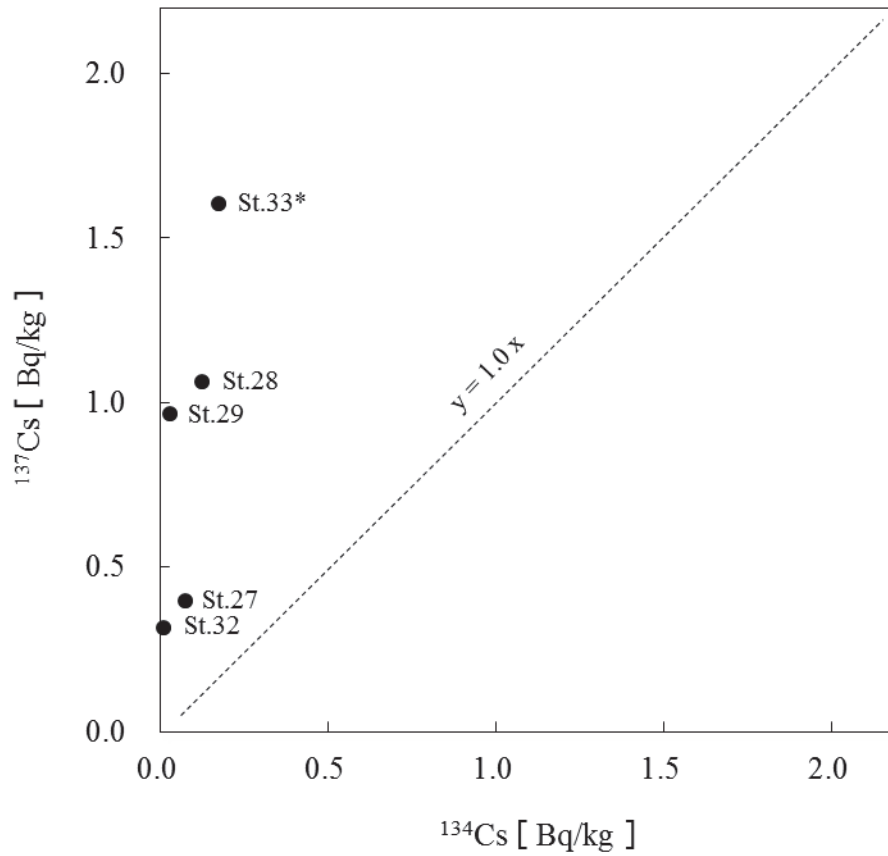
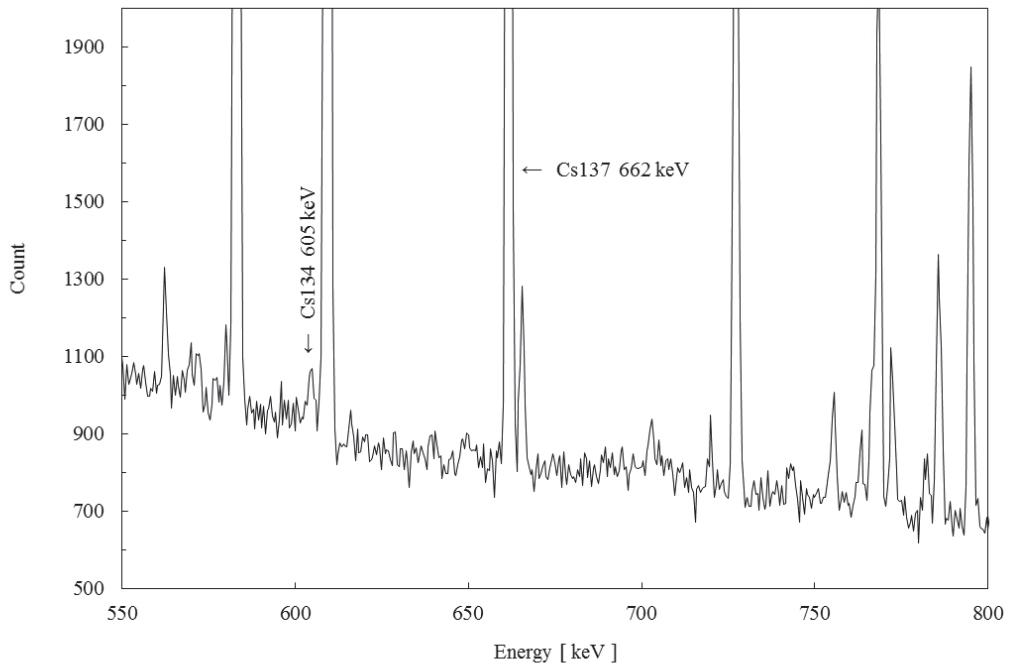


Figure 3-2 Relation of concentrations between  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  at seabed soil of the Notsuke Strait.

The data were decay-corrected on the values of March 11, 2011. Although the concentrations of  $^{134}\text{Cs}$  except for St. 33 were judged to be ND, the obtained values were included in Figure 3-2 to know the relation.



Energy and Counts of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  are shown in the Figure 3-3.  
 Figure 3-3 Gamma-ray spectrum of the seabed soil from St.33\* at the Notsuke Strait.

Measurement date: February 26, 2016. Measurement time: 600,000 s.

Germanium semiconductor: GEM40-76-XLB-C made by SEIKO EG & G Co., Ltd

Figure 3-2 shows relation between the concentrations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  for each sample for the additional investigation. Although the concentrations of  $^{134}\text{Cs}$  except for St. 33 were judged to be ND, the obtained values were included in Figure 3-2 to know the relation. The values were decay-corrected on the values of March 11, 2011 to estimate the concentration of the global fallout.

The  $^{134}\text{Cs}/^{137}\text{Cs}$  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)<sup>3-11)</sup>. The  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio means that we can estimate concentration of  $^{137}\text{Cs}$  from the concentration of  $^{134}\text{Cs}$  in the Fukushima accident. At St.33 on February 6, 2016, the concentration of  $^{134}\text{Cs}$  was  $0.034 \pm 0.0086$  Bq/kg, and the concentration of  $^{137}\text{Cs}$  was  $1.43 \pm 0.015$  Bq/kg. When these concentrations were decay-corrected to the

values of March 11, 2011, the concentration of  $^{134}\text{Cs}$  is expected to be  $0.17 \pm 0.045$  Bq/kg, and the concentration of  $^{137}\text{Cs}$  is  $1.60 \pm 0.017$  Bq/kg. When radioactive cesium was released by the Fukushima accident, the  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio is expected to be approximately 1.0 at the time of accident. From this fact, the concentration of  $^{137}\text{Cs}$  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 3-2, the vertical lengths to St.27, 28, 29, 32 points from line of  $y = 1.0 x$  are supposed to be the concentration of  $^{137}\text{Cs}$  from global fallout.

The Notsuke Strait is located at the offing of Shibetsu-cho, Hokkaido in southern part of the Sea of Okhotsk. 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 <sup>3-12)</sup>, and this shows sedimentation of radioactive cesium on the earth surface soil. According to the report of this investigation,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were less than 10 kBq/m<sup>2</sup> 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  $^{134}\text{Cs}$  was all ND in the earth surface soil radioactivity investigation including farmland of the whole Hokkaido. And the radioactivity concentration of  $^{137}\text{Cs}$  was less than the value before the FDNPP accident <sup>3-13</sup>). From the result above, it is thought that there were very little  $^{134}\text{Cs}$  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,  $44^{\circ} 50' \text{ N}$ ,  $144^{\circ} 00' \text{ E}$ ) in the southern Sea of Okhotsk, and investigated radioactivity. Concentration of  $^{137}\text{Cs}$  was  $0.96 \pm 0.04 \text{ mBq/L}$ , but in June 2011 of the next year,  $3.14 \pm 0.07 \text{ mBq/L}$  that was 3.3 times higher than the preceding year was detected in the investigation of the same sea area <sup>3-6, 3-7</sup>). The method of Japan Coast Guard was that the Cs of the surface seawater was adsorbed by ammonium phosphomolybdate 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 3-4<sup>3-14</sup>). 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<sup>3-15</sup>). 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 <sup>137</sup>Cs of the seabed soil was 2.5 Bq/kg on the Japanese peripheral sea area. However, the concentration of <sup>137</sup>Cs of the seabed soil at the Notsuke Strait was extremely low than the average concentration of <sup>137</sup>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  $^{137}\text{Cs}$  at NO-10 point (Figure 3-1) increased in 2011<sup>3-16)</sup>. It is thought that some of these  $^{137}\text{Cs}$  were adsorbed in the seabed soil at the Notsuke Strait. It can be judged that the  $^{134}\text{Cs}$  obtained in the present study is reached from the FDNPP accident. We can suppose that radioactive material released by Tokyo Electric Power Company 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.

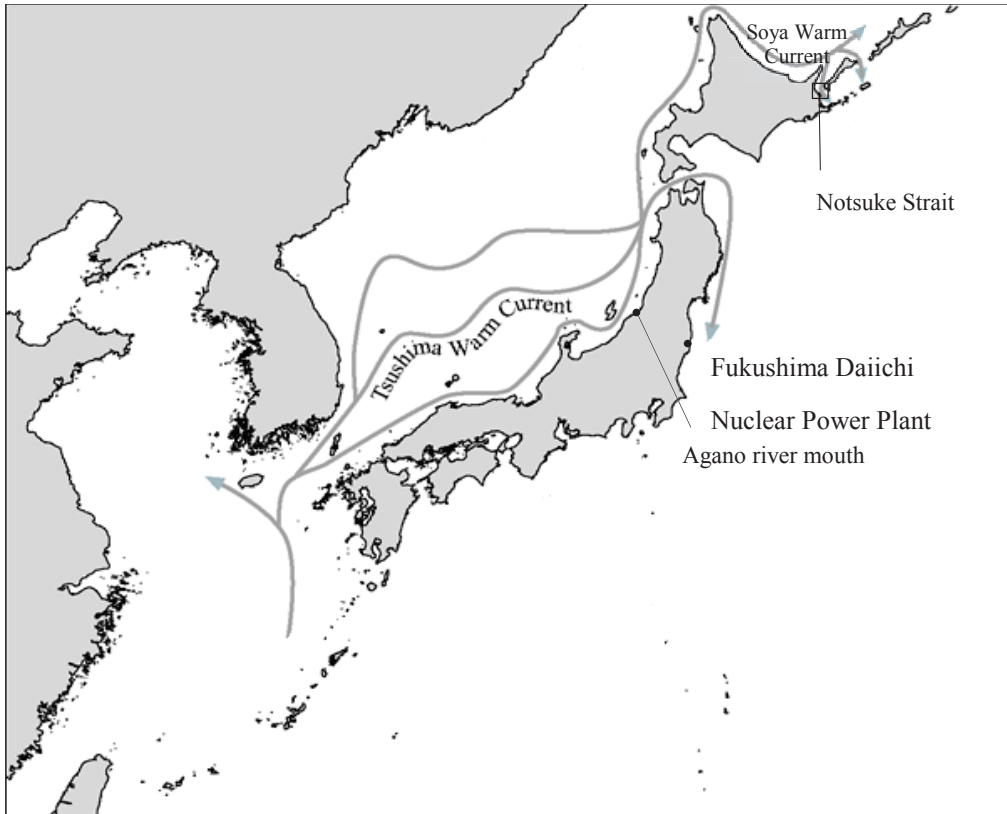


Figure 3-4 Tsushima warm current and Soya warm current<sup>3-14</sup>).

The Tsushima warm current and the Soya warm current contribute to the migration of Cs from the Agano river mouth to the Notsuke Strait.

### 3.4 Conclusions

For the investigation of the migration pathway, we sampled the seabed soil at the Notsuke Strait and measured the  $\gamma$ -Ray spectrum with the Ge semiconductor detector. The  $\gamma$ -Ray spectrum had peak of  $^{134}\text{Cs}$  at 605 keV and that of  $^{137}\text{Cs}$  at 662 keV. We measured concentration of  $^{134}\text{Cs}$  with  $0.033 \pm 0.0084$  Bq/kg, and the concentration of  $^{137}\text{Cs}$  with  $1.4 \pm 0.015$  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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Chapter 4      Observation of radioactive cesium in seabed  
soil at the Soya Strait derived from the Fukushima Daiichi  
Nuclear Power Plant

## 4.1 Introduction

FDNPP accident occurred in March, 2011, and large quantity of radioactive material was spread in environment. Ministry of Education, Culture, Sports, Science and Technology carried out radioactivity surveys with air planes, and announced that Fukushima and neighboring areas were polluted with radioactive materials <sup>4-1)</sup>. Niigata prefectural government also carried out radioactivity survey to know the pollution with radioactive materials at Niigata. <sup>134</sup>Cs was detected in seabed soil of the Agano River Mouth offing in surveys of August, 2011 <sup>4-2)</sup>. In June, 2011, Japan Coast Guard detected <sup>137</sup>Cs, which is higher by 2.18 Bq/kg at most compared with the previous year in surface seawater at the southwestern Sea of Okhotsk <sup>4-3)</sup>. These surveys suggested that radioactive materials were transported to the Japan Sea, the Sea of Okhotsk and then the Pacific by ocean currents.

In order to identify the migration pathway of radioactive materials from the Japan Sea to the Pacific via the Sea of Okhotsk, we started the radioactivity survey of cesium in the seabed soil. First survey was carried out at the Notsuke Strait and

$^{134}\text{Cs}$  derived from FDNPP was detected in the seabed soil<sup>4-4)</sup>. This result suggested that the radioactive material derived from FDNPP migrated to the Sea of Okhotsk from the Japan Sea. The present paper reports the second survey of radioactive materials at the Soya Strait and discusses the migration of radioactive cesium from the Japan Sea to the Sea of Okhotsk.

## 4.2 Sampling and Measurement

### 4.2.1 Sampling

In September 9, 10 and 17, 2015, seabed soil at the Soya Strait of the Wakkanai-city, Hokkaido offing was collected. Figure 4-1 shows the position of Soya Strait and the sampling points. Seabed soils were collected by using a dredge sampler.

The stones and the visible creature in the seabed solids were removed. Seabed soil was dried at 95 °C for more than 24 hours and passed through a sieve of 2 mm  $\phi$ .

The seabed soil was put in 2 liters of Marinelli type vessel and was used for the measurement of the radioactivity.

#### 4.2.2 Method for measurement of the radioactive cesium

A germanium semiconductor detector (GEM40-76-XLB-C, SEIKO EG & G Co., Ltd) was used for the measurement of the radioactivity of the sample. The  $\gamma$  radioactivity of the sample was measured for 250,000 seconds. The concentrations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were calculated by official method <sup>4-5)</sup>, and the results of the measurements were corrected to the quantity of radioactivity of the sampling day. Because the emission probability is high and the separation with peaks of other radionuclides is easy, 605 and 662 keV were chosen for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , respectively.

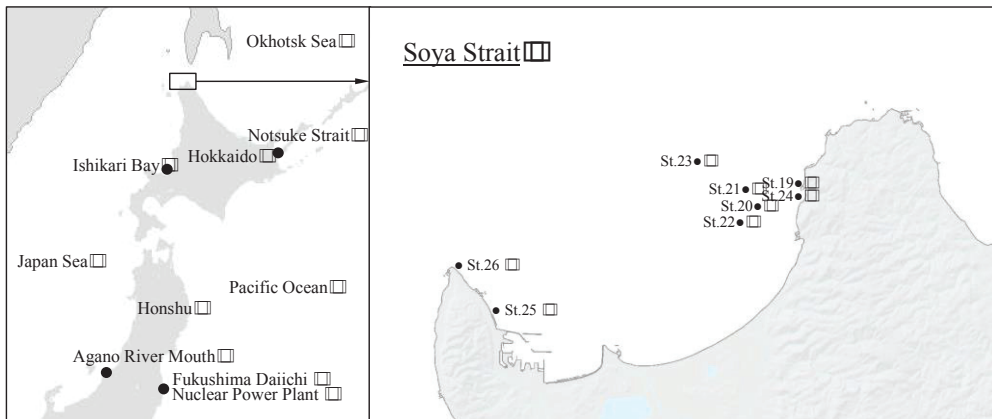


Figure 4-1 Survey sea area and sampling points.

## 4.3 Results and Discussion

### 4.3.1 Radioactivity and geological feature of seabed soil

Table 4-1 shows latitude, longitude, depth and the concentrations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  of seabed soils at the sampling points. Depth was 0.5 - 28.2 m, the deepest point was St.23. The condition of sampled seabed soil at St.19 was muddy, and those at other sampling points were sandy. The concentration of  $^{137}\text{Cs}$  was 0.11 - 2.3 Bq/kg, and St.19 showed the highest value in this sampling area.  $^{134}\text{Cs}$  was detected only at St.19. Figure 4-2 shows the  $\gamma$ -ray spectrum of the sample at St.19. There are clear peaks at the position of 605 and 662keV in this spectrum.

The geological feature is that the seabed is mainly rock in the survey sea area. There is a submarine canyon in the west direction from St.24, and sand is distributed over there. There is a little seabed soil of mud and the mud is slightly distributed around St.19 and St.24<sup>4-6, 4-7</sup>). As a result of sampling, St.19 had seabed soil of the only mud, and  $^{134}\text{Cs}$  was detected at only St.19. In the previous survey at the Notsuke Strait,  $^{134}\text{Cs}$  was detected with the seabed soil of the mud<sup>4-4</sup>). The present

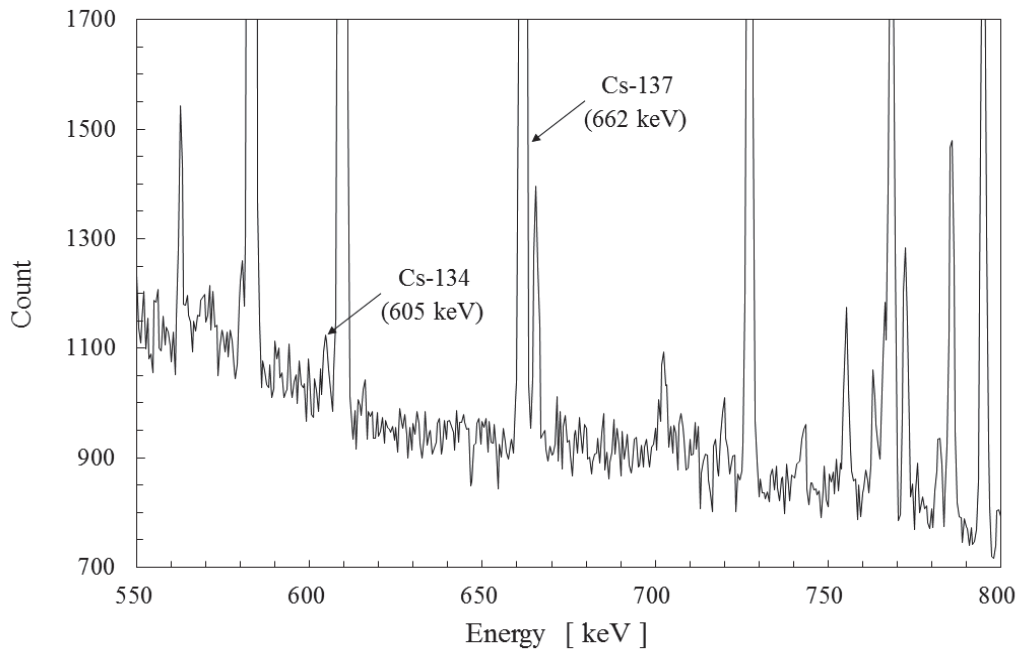
situation is similar to that at the Notsuke Strait. This result suggested that particle size of seabed soil is an important factor of adsorption behavior of the radioactive cesium from seawater to seabed soil, as the same as the adsorption behavior of the radioactive cesium on sediment soil in ponds and rivers. In the investigation into ponds at the peripheral area of FDNPP, the sediment of the small particle size adsorbed much  $^{137}\text{Cs}$  (4-8). In the investigations into sediment of the river, the sediment was separated to clay, silt, sand and stone. As a result, the radioactive cesium was adsorbed to clay and silt, and there was little adsorption to sand (4-9).



Table 4-1 Position, depth, sampling day and the concentration of radioactive cesium at each sampling point

No	Latitude	Longitude	Depth [m]	Sampling day	<sup>134</sup> Cs [Bq/kg]	<sup>137</sup> Cs [Bq/kg]
St.19	45° 29' 09"	141° 52' 39"	2.8	9-Sep-15	0.059 ± 0.019	2.3 ± 0.1
St.20	45° 28' 58"	141° 50' 59"	20.4	17-Sep-15	N.D	0.47 ± 0.02
St.21	45° 29' 12"	141° 50' 46"	22.6	17-Sep-15	N.D	0.46 ± 0.02
St.22	45° 28' 48"	141° 50' 26"	23.5	17-Sep-15	N.D	0.53 ± 0.02
St.23	45° 29' 43"	141° 50' 16"	28.2	17-Sep-15	N.D	0.65 ± 0.02
St.24	45° 29' 06"	141° 52' 43"	3.2	9-Sep-15	N.D	1.2 ± 0.1
St.25	45° 25' 28"	141° 40' 26"	0.5	10-Sep-15	N.D	0.42 ± 0.03
St.26	45° 26' 59"	141° 38' 44"	0.5	10-Sep-15	N.D	0.11 ± 0.02

- a) The concentration of radioactive cesium was corrected to the value at sampling day.
- b) N.D shows “Not detected”. Measurement time was 250,000 seconds.



Energy and Counts of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  are shown in the figure.

Sampling Figure 4-2  $\gamma$ -Ray spectrum of seabed soil at St.19. position:  
St.19 of the Soya Strait (45 ° 29'09" N, 141 ° 52'39" E)

Sampling date: September 9<sup>th</sup>, 2015.

Amount for the measurement: 2707.38 g.

Measurement date: November 10, 2015.

Measurement time: 250,000 s.

Germanium semiconductor: GEM40-76-XLB-C, SEIKO EG & G Co., Ltd.

### 4.3.2 Contribution of global fallout on seabed soil

In East Japan Great Earthquake Disaster, the radioactive materials were released to the environment from No.1, No.2 and No.3 nuclear reactors at FDNPP. The  $^{134}\text{Cs}/^{137}\text{Cs}$  radioactivity ratio when the FDNPP accident occurred was almost 1, which is similar among three reactors<sup>4-10</sup>. When the concentrations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were corrected to the values as of March 11, 2011, the ratio of  $^{134}\text{Cs}/^{137}\text{Cs}$  at St.19 was 0.11. This value is lower than typical value (about 1) of FDNPP.  $^{137}\text{Cs}$  derived from global fallout should be considered in the surface of seabed soil at Japanese peripheral sea area. Calculated  $^{137}\text{Cs}$  concentration at St.19 on the day of FDNPP accident becomes 0.27 Bq/kg by assuming that the ratio of  $^{134}\text{Cs} / ^{137}\text{Cs}$  is 1. The difference between measured concentration and calculated concentration for  $^{137}\text{Cs}$  becomes 2.28 Bq/kg. This value corresponds to typical value of  $^{137}\text{Cs}$  concentration of global fallout. The average concentration of  $^{137}\text{Cs}$  of global fallout is about 2 Bq/kg and maximum value of concentration of  $^{137}\text{Cs}$  was 5.6 Bq/kg at the Shinano River mouth in 2010<sup>4-11</sup>. Average value of  $^{137}\text{Cs}$  concentration at the

Soya Strait is 0.77 Bq/kg. The concentration of  $^{137}\text{Cs}$  at St.19 is higher than average value of the Soya Strait.

#### 4.3.3 Migration pathway of radioactive cesium from the Japan Sea to the Sea of Okhotsk

During from April to May in 2012, the radioactivity survey of the earth surface of all Japan was carried out by Ministry of Education, Culture, Sports, Science and Technology. Hokkaido was the lowest level in all Japan <sup>4-12</sup>). Radioactivity of the soils on 7 agricultural experimental stations in Hokkaido was investigated by Hokkaido Prefectural Government.  $^{134}\text{Cs}$  was not detected in the soil of agricultural experimental stations collected on September 5, 2011. In addition, the concentration of  $^{137}\text{Cs}$  was less than those of 2008-2010 <sup>4-13</sup>). These results suggest that little radioactive cesium was derived from FDNPP to the soil at Hokkaido. There is little radioactive cesium which deposited from the atmosphere to the earth surface of Hokkaido, and there may be very little radioactive cesium which flowed

out from the land of the neighborhood of St.19 into the sea area. It is suggested that  $^{134}\text{Cs}$  detected in seabed soil at St.19 migrated from the other sea area.

Figure 4-3 shows the ocean currents of the Soya Strait peripheral sea area and the sampling points of surveys in the Sea of Okhotsk and the Hokkaido western sea area. The Tsushima warm current flows along the west coast of the Honshu, and flows along Hokkaido west coast. A part of the Tsushima warm current flows into the Soya Strait and becomes the Soya warm current. The Soya warm current flows through the southeastern direction along Hokkaido northeast coast and reaches the Shiretokomisaki offing. The Soya warm current separates to the Sea of Okhotsk central part, the Nemuro Strait part and the Kunashiri Strait part <sup>4-14)</sup>.  $^{137}\text{Cs}$  derived from FDNPP was detected by Japan Coast Guard in surface seawater around the Soya Strait.

The concentration of  $^{137}\text{Cs}$  for surface seawater in 2010 was 0.96 - 1.5 Bq/kg, while that in 2011 was 2.41 – 3.82 Bq/kg <sup>4-3, 4-9)</sup>. It increased by 2.18 Bq/kg at most. The method of Japan Coast Guard was that the radioactive cesium of the surface seawater was adsorbed by ammonium phosphomolybdate and was

separated with the cation exchange resin. It can be judged that ionic cesium exists in surface seawater at that period. In addition,  $^{134}\text{Cs}$  was detected in July, 2012 at seabed soil of the Ishikari Bay, and the concentration was 0.7 Bq/kg<sup>4-15)</sup>.

A part of the surface seawater of the Hokkaido western sea area flows into the Sea of Okhotsk via the Soya Strait by ocean currents.

Figure 4-4 shows the relationship between the concentration of  $^{134}\text{Cs}$  and distance from Ishikari Bay. The concentration of  $^{134}\text{Cs}$  decreased with increasing of distance from Ishikari Bay. Those results suggest that the radioactive cesium was transported by ocean current and adsorbed in seabed soil in the pathway of current. The radioactive cesium migrated along ocean currents from the Ishikari Bay via the Soya Strait to the Notsuke Strait.

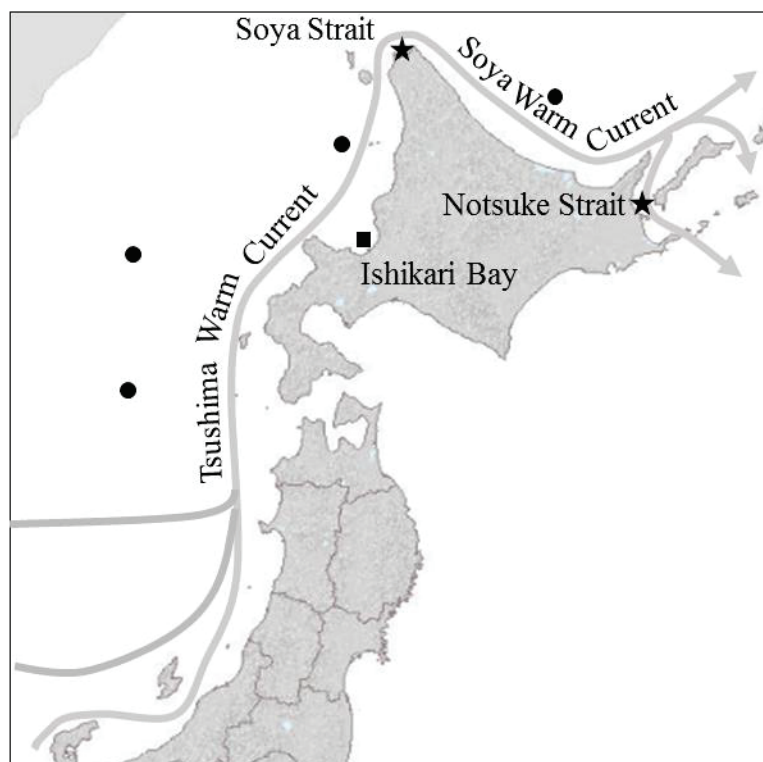


Figure 4-3 Current and sampling points of seawater and seabed soil.

Closed circles and closed square show the radioactivity survey of seawater and seabed soil by Japan Coast Guard <sup>4-3</sup>). Closed stars show our sampling sea areas of seabed soil. The curve shows a part of the Tsushima warm current and the Soya warm current <sup>4-16</sup>).

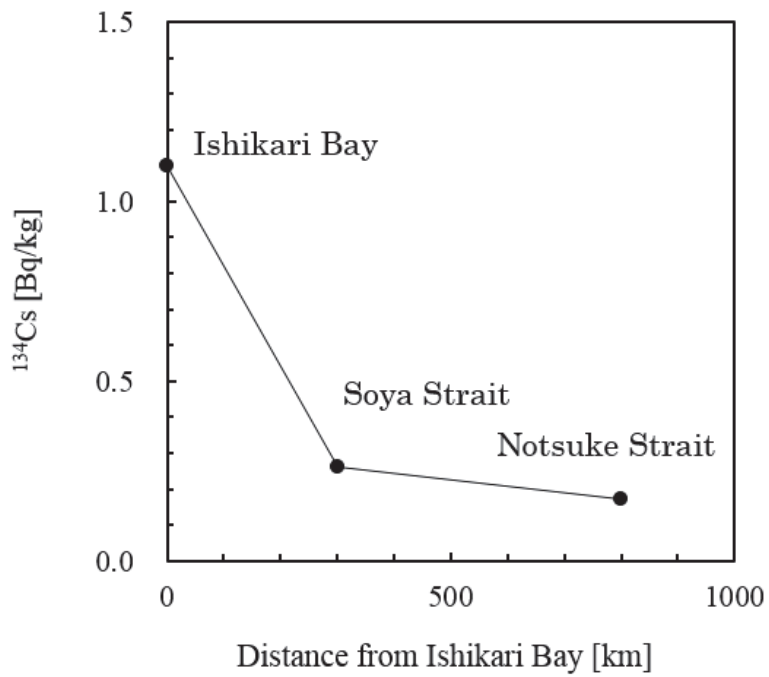


Figure 4-4 Relationship between the distance from Ishikari Bay and the concentration of  $^{134}\text{Cs}$  in seabed soil.

The concentration of radioactive cesium was corrected to the value at March 11, 2011.



#### 4.4 Conclusion

We investigated radioactivity of the seabed soil at the Soya Strait for identification of migration pathway of the radioactive cesium derived from FDNPP.  $^{134}\text{Cs}$  was detected in muddy seabed soil, which is similar to the result of ponds and rivers.

$^{137}\text{Cs}$  from the global fallout was detected at all sampling points in this survey.

The contribution of FDNPP was separated from that of global fallout. The radioactive cesium migrated to the Soya Strait by the ocean currents and was adsorbed to seabed soil. It is estimated that radioactive cesium of the Hokkaido western sea area passed through the Soya Strait and migrated to the southwestern part of the Sea of Okhotsk. It is suggested that there is a relation between the migration distance by the ocean currents and the concentration of  $^{134}\text{Cs}$  deposited at the seabed soil in the Hokkaido peripheral sea area.

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Chapter 5      Distribution of radioactive cesium from the  
Fukushima Daiichi Nuclear Power Plant in seabed soil from  
the Niigata Prefecture and Yamagata Prefecture offings

## 5.1 Introduction

After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011, the Ministry of Education, Culture, Sports, Science and Technology carried out aerial radioactivity surveys, which showed that Fukushima and neighboring areas were polluted with radioactive materials<sup>5-1)</sup>. Niigata Prefecture's government also carried out a radioactivity survey to investigate pollution by radioactive materials in the Agano River basin. <sup>134</sup>Cs was detected in seabed soil from the Agano River mouth offing by surveys in August 2011<sup>5-2)</sup>. In June 2011, the Japan Coast Guard detected 3.14 Bq/kg of <sup>137</sup>Cs in surface seawater in the southwestern Sea of Okhotsk, which is 2.18 Bq/kg higher than the previous year<sup>5-3), 5-4)</sup>. These surveys suggest that radioactive cesium from FDNPP was transported to the Japan Sea and the Sea of Okhotsk.

There have been few radioactivity surveys in the Japan Sea and the Sea of Okhotsk compared with the Fukushima peripheral sea area and the Pacific. In addition, it has not been confirmed whether the radioactive materials migrated via

ocean currents in the Japan Sea and the Sea of Okhotsk. It has been reported that radioactive cesium migrated from the atmosphere to the Japan Sea <sup>5-5)</sup>. Our research suggests that radioactive cesium migrated from the Japan Sea to the Soya Strait and the Notsuke Strait <sup>5-6, 5-7)</sup>, although the origin of the radioactive cesium is unknown. In the present study, we conducted radioactivity surveys at the Sakata, Kamo, and Naoetsu offings to clarify the origin and migration pathway of radioactive cesium. The results show that the origin is limited to the region from Naoetsu to Sakata, including the Agano and Mogami Rivers.

## 5.2 Sampling and Measurement

### 5.2.1 Sampling

Seabed soil was collected from the Sakata Port offing in July 2015, and from the Naoetsu Port and Kamo Port offings in August 2016. Figure 5-1 shows the sea survey area and sampling points. Seabed soils were collected (~3 L) at each sampling point by using a Smith-Mcintyre grab sampler. Stones and visible

creatures in the seabed solids were removed. Seabed soil was dried at 95 °C for more than 24 h and passed through a 2 mm sieve. Seabed soil was put in a 2 L Marinelli vessel and the radioactivity was measured.



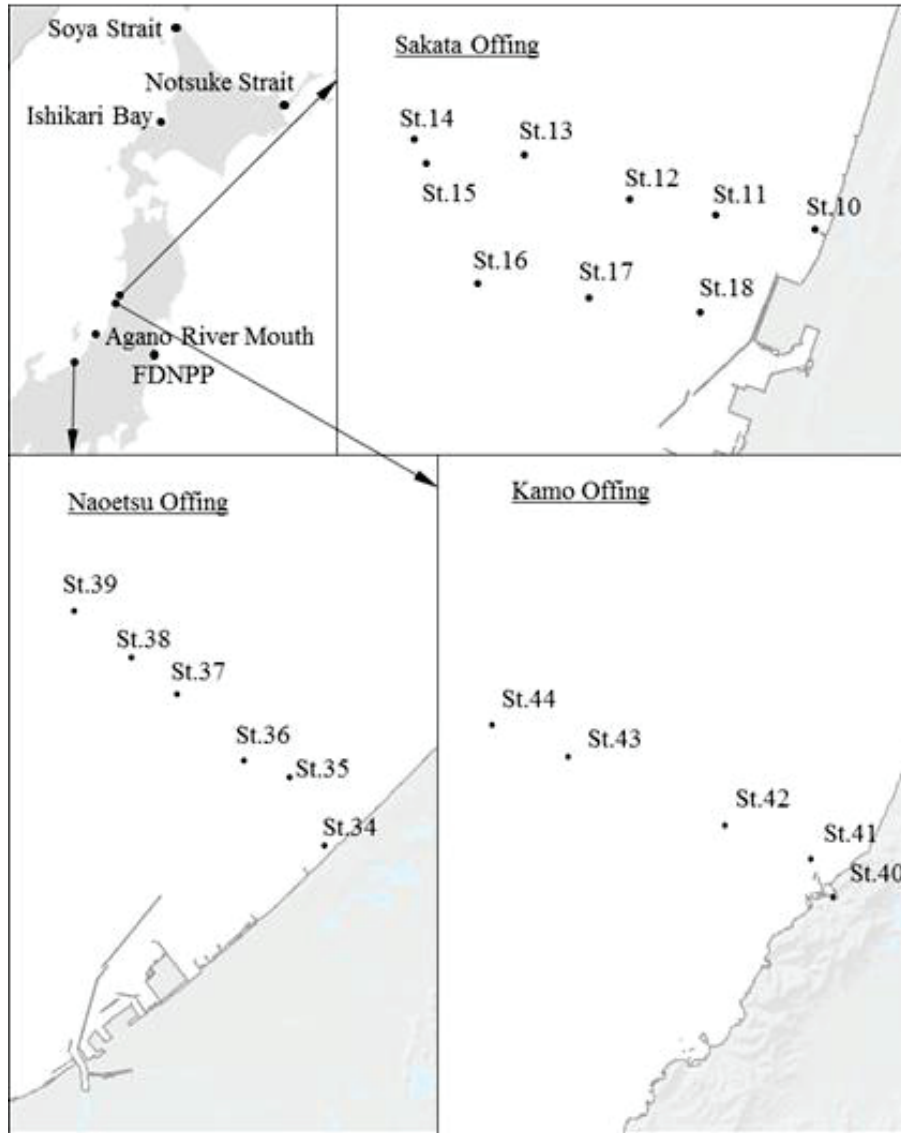


Figure 5-1 Maps showing survey sea areas and sampling points.

### 5.2.2 Method for measuring radioactive cesium

A germanium semiconductor (GEM40-76-XLB-C, Seiko EG&G Co., Ltd.) was used to measure the radioactivity of the sample. The  $\gamma$  radioactivity of seabed soils was measured for 80,000 s. The concentrations of radioactive cesium were calculated by the official method<sup>5-8)</sup>, and the results of the measurements were decay-corrected to the quantity of radioactivity of the sampling day. Because the emission probability is high and the separation from peaks of other radionuclides is easy, 605 and 662 keV were chosen for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , respectively.

### 5.2.3 Method for measuring grain size distribution

After the radioactivity measurements, the grain size distribution of the seabed soil was determined with sieves<sup>5-9)</sup>. Portions of seabed soil (30 g) were separated by using 850, 250, and 75  $\mu\text{m}$  sieves, and the seabed soils were classified as coarse sand, medium sand, fine sand, and clay-silt. The sample isolation, sieving, and

weighing were carried out twice for each sample. The average was calculated and the grain size distribution was determined.

### 5.3 Results and discussion

#### 5.3. 1 Radioactivity and geological feature of seabed soil

Table 5-1 shows the latitude, longitude, and depth along with the concentrations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  for seabed soils at the sampling points.

In the Sakata offing sampling area,  $^{134}\text{Cs}$  was detected in the seabed soil at four sampling points, whereas  $^{137}\text{Cs}$  was detected in seabed soil at all sampling points.

The depth was 8.2–55.9 m and the deepest points were stations (Sts.) 14 and 16.

The seafloor topography of the Sakata offing is slanted from the shoreline to the northwest. The seabed soil is mainly composed of sand at the Sakata offing,

although the soil also contains shell and stone<sup>5-10</sup>). There is a natural fishing bank

called Akashi-guri west of St. 14. Because sea life is protected in the sea area around

Akashi-guri, the position of St. 15 was irregular.

In the Kamo offing sampling area,  $^{134}\text{Cs}$  was detected in seabed soil at two sampling points, whereas  $^{137}\text{Cs}$  was detected in seabed soil at all sampling points. The depth of the Kamo offing is 13.2–79.7 m and the deepest point is St. 44. The seabed soil is mainly composed of sand, and the seafloor topography of the Kamo offing is slanted from the shoreline to the northwest<sup>5-11</sup>).

At the Naoetsu offing sampling point,  $^{134}\text{Cs}$  was not detected in all seabed soil even if the soil was composed of mud, whereas  $^{137}\text{Cs}$  was detected in seabed soil at all sampling points. The depth is 6.8–99.7 m and the deepest point is St. 39. The seafloor topography of the Sakata offing is slanted from the shoreline to the northwest. The seabed soil is composed of mud, sand, shell, and stone at the Naoetsu offing<sup>5-11</sup>).

Table 5-1 Position, depth, sampling day and the concentration of radioactive cesium at each sampling point

Survey areas	No	Latitude	Longitude	Depth [m]	Sampling day	<sup>134</sup> Cs [Bq/kg]	<sup>137</sup> Cs [Bq/kg]
Sakata Offing	St.10	38° 59' 06"	139° 50' 12"	8.2	13-Jul-15	N.D	0.28 ± 0.04
	St.11	38° 59' 18"	139° 48' 30"	26.8	13-Jul-15	0.32 ± 0.04	1.5 ± 0.1
	St.12	38° 59' 30"	139° 47' 00"	44.7	13-Jul-15	0.68 ± 0.02	4.0 ± 0.1
	St.13	39° 00' 06"	139° 45' 12"	53.6	13-Jul-15	0.16 ± 0.03	0.75 ± 0.04
	St.14	39° 00' 18"	139° 43' 28"	55.9	13-Jul-15	N.D	0.43 ± 0.04
	St.15	38° 59' 59"	139° 43' 31"	55.5	13-Jul-15	N.D	0.52 ± 0.16
	St.16	38° 58' 23"	139° 44' 24"	55.9	13-Jul-15	N.D	0.86 ± 0.04
	St.17	38° 58' 12"	139° 46' 18"	45.7	13-Jul-15	0.64 ± 0.05	3.8 ± 0.1
	St.18	38° 58' 00"	139° 48' 14"	22.4	13-Jul-15	N.D	2.5 ± 0.3
Kamo Offing	St.40	38° 45' 46"	139° 43' 58"	13.2	6-Aug-16	0.61 ± 0.06	4.7 ± 0.1
	St.41	38° 46' 22"	139° 43' 27"	17	6-Aug-16	N.D	0.49 ± 0.04
	St.42	38° 46' 55"	139° 41' 27"	39.6	6-Aug-16	N.D	0.29 ± 0.03
	St.43	38° 48' 02"	139° 37' 48"	59.6	6-Aug-16	N.D	0.32 ± 0.04
	St.44	38° 48' 32"	139° 36' 02"	79.7	6-Aug-16	0.33 ± 0.04	3.2 ± 0.1
Naoetsu Offing	St.34	37° 14' 30"	138° 20' 01"	6.8	5-Aug-16	N.D	0.13 ± 0.04
	St.35	37° 15' 38"	138° 19' 16"	20.9	5-Aug-16	N.D	0.19 ± 0.06
	St.36	37° 15' 55"	138° 18' 18"	39.5	5-Aug-16	N.D	0.52 ± 0.04
	St.37	37° 17' 01"	138° 16' 53"	62.3	5-Aug-16	N.D	0.97 ± 0.04
	St.38	37° 17' 37"	138° 15' 54"	79.4	5-Aug-16	N.D	1.1 ± 0.1
	St.39	37° 18' 23"	138° 14' 42"	99.7	5-Aug-16	N.D	0.96 ± 0.04

a) The concentration of radioactive cesium was decay-corrected to the value on the sampling day.

b) N.D shows “Not detected”. Measurement time was 80,000 second.

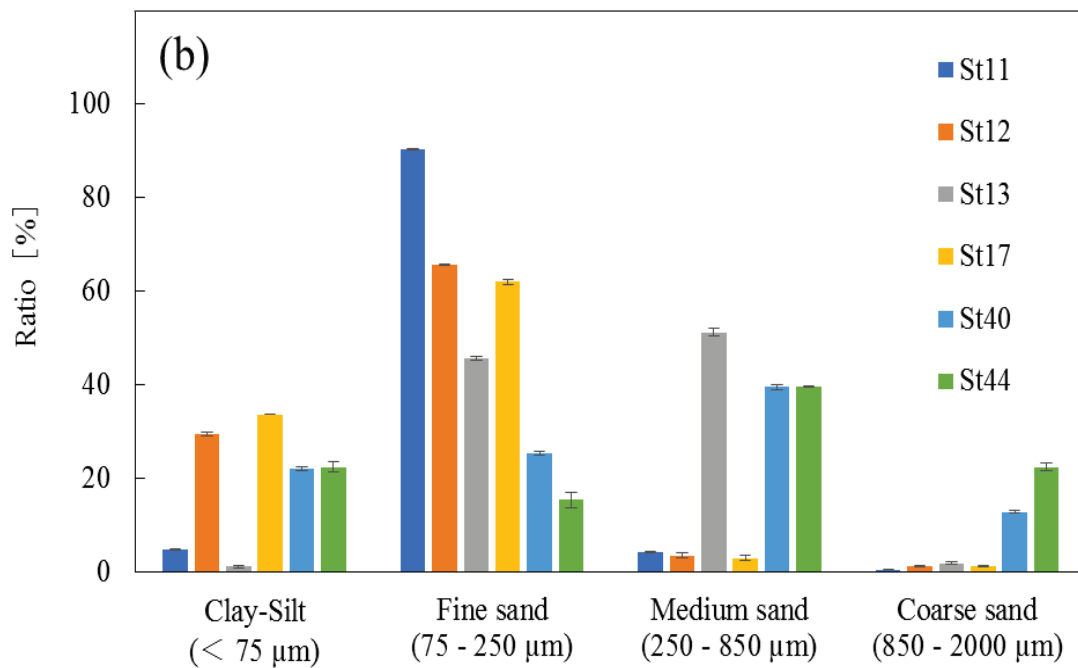
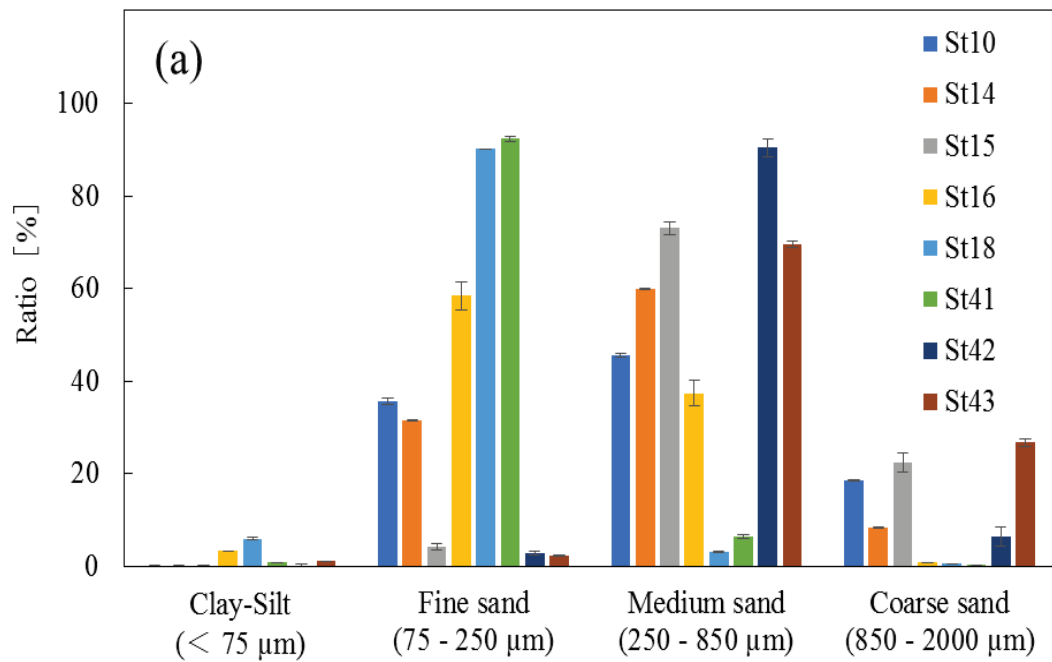
### 5.3.2 Grain size distribution of seabed soil

The grain size distribution of seabed soil was investigated to clarify the relationship between the distribution and the adsorption behavior of the radioactive cesium.

Figure 5-2 (a) shows the grain size distribution of the seabed soil from the Kamo and Sakata offings where  $^{134}\text{Cs}$  was not detected (see Section 3.3). Figure 5-2 (b) shows the grain size distribution of seabed soil from the Kamo and the Sakata offings where  $^{134}\text{Cs}$  was detected (see Section 3.3). Figure 5-2 (c) shows the grain size distribution of seabed soil from the Naoetsu offing. Figure 5-2 (d) shows the relationship between the radioactive cesium concentration and the clay-silt ratio.

$^{134}\text{Cs}$  was not detected at the Naoetsu offing (see Section 3.3). There was a significant difference in the clay-silt and medium sand ratios between Figures 5-2 (a) and (b). The clay-silt was only 0.1%–6.0% in Figure 5-2 (a), where  $^{134}\text{Cs}$  was not detected. However, the clay-silt ratio was 1.2%–33.7% in Figure 5-2 (b), where  $^{134}\text{Cs}$  was detected. In addition, the ratio of medium sand was lower in Figure 5-2 (b) than in Figure 5-2 (a). The ratio of clay-silt increased proportionally with the

$^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations in Figure 5-2 (d).  $^{134}\text{Cs}$  was not detected in seabed soil that did not contain clay-silt, whereas  $^{134}\text{Cs}$  was detected in seabed soil that contained clay-silt. This difference suggests that the particle size of seabed soil is an important factor in the adsorption of radioactive cesium from seawater to seabed soil. In a previous survey, ponds around the periphery of FDNPP, sediment with small particle sizes adsorbed a large amount of  $^{137}\text{Cs}$  <sup>5-12</sup>). Furthermore, other work found that radioactive cesium was adsorbed to clay and silt river sediment, and there was little adsorption to sand <sup>5-13</sup>). Therefore, radioactive cesium is adsorbed to clay-silt selectively in fresh water and seawater. However,  $^{134}\text{Cs}$  was not detected in seabed soil that contained more than 20% clay-silt at St. 38–39 (Figure 5-2 (c)), suggesting that the water mass containing the radioactive cesium did not reach the Naoetsu offing.





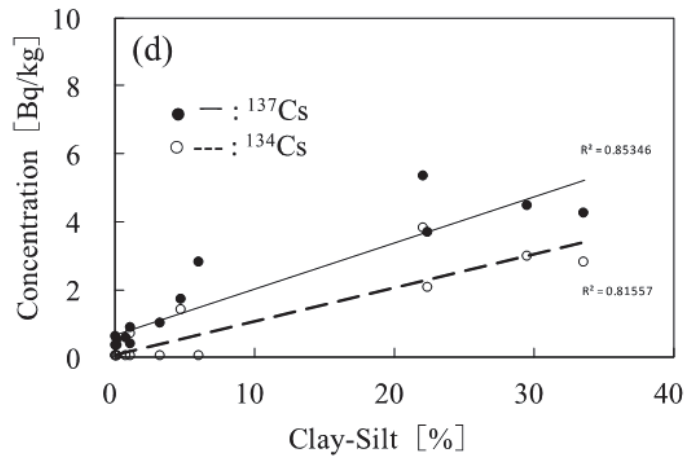
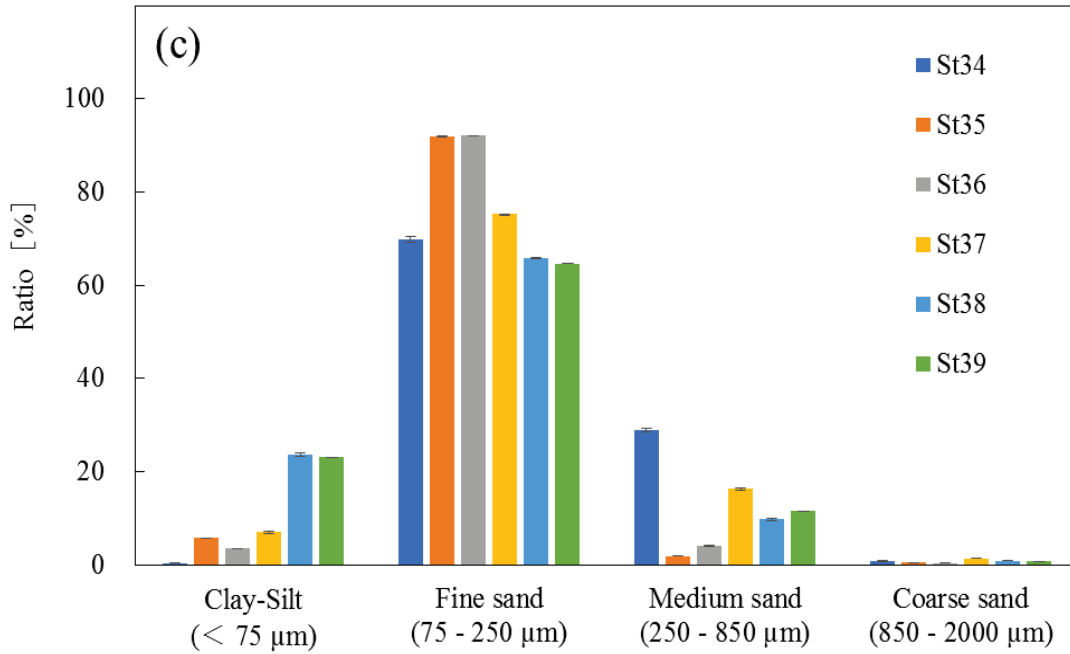


Figure 5-2 Grain size distributions in dry seabed soil.

Error bars are shown at the top of the bar graphs.

(a): Seabed soil where <sup>134</sup>Cs was not detected at Sakata and Kamo offings.

(b): Seabed soil where <sup>134</sup>Cs was detected at Sakata and Kamo offings.

(c): Seabed soil at Naoetsu offing (<sup>134</sup>Cs was not detected).

(d): Relations between <sup>134</sup>Cs, <sup>137</sup>Cs concentration and Clay-Silt ratio

### 5.3.3 $^{134}\text{Cs}$ concentration in seabed soil

Table 5-1 shows the  $^{134}\text{Cs}$  concentration in seabed soil, which is decay-corrected to the sampling time. Figure 5-3 shows the relationship between the distance from the Agano River mouth to the sampling point and the radioactive cesium concentration in the seabed soil for each sea area. The  $^{134}\text{Cs}$  concentration was decay-corrected to the value on March 11, 2011.  $^{134}\text{Cs}$  concentrations of 3.75 and 2.92 Bq/kg were detected at the Kamo offing (K) and Sakata offing (S), respectively. However,  $^{134}\text{Cs}$  was not detected at the Naoetsu offing (N). Thus, the radioactive cesium may not have reached the Naoetsu offing. The Niigata Prefecture government performed a radioactivity survey of seabed soil at the Agano River mouth offing in August 2011. The  $^{134}\text{Cs}$  concentration was 194 Bq/kg (decay-corrected to the value on March 11, 2011), which was higher than those of the Kamo and Sakata offings<sup>5-2</sup>). The  $^{134}\text{Cs}$  concentration in seabed soil decreased in the order of the Kamo offing, the Sakata offing, and Ishikari Bay, which indicated a decrease with migration distance.

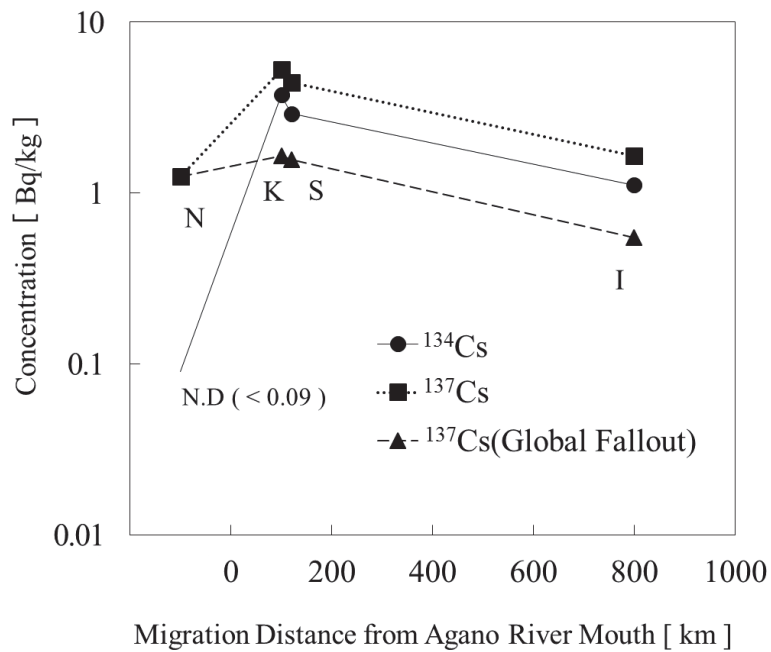


Figure 5-3 Relationship between the distance from Agano River mouth and the concentration of radioactive cesium in seabed soil.

The concentration of radioactive cesium was decay-corrected to the value on the time of March 11, 2011. The meanings of letters in Figure 5-3 are as follows. N: Naoetsu offing, K: Kamo offing, S: Sakata offing, I: Ishikari Bay. The concentration of sea areas is the maximum value in each sampling point. The concentration of the Ishikari Bay was excerpted from the survey of Japan Coast Guard <sup>5-19</sup>).

#### 5.3.4 Contribution of global fallout

Table 5-2 shows the results for seabed soil where  $^{134}\text{Cs}$  was detected in this radioactivity survey. The sampling point of the Naoetsu offing is not listed because  $^{134}\text{Cs}$  was not detected. The radioactive cesium concentration was decay-corrected to March 11, 2011.

The radioactive cesium was released into the environment from nuclear reactors 1–3 at FDNPP during the Great East Japan Earthquake. The  $^{134}\text{Cs}/^{137}\text{Cs}$  radioactivity ratio when the FDNPP accident occurred was almost 1, which was similar for all three reactors<sup>5-14</sup>). However, the  $^{134}\text{Cs}/^{137}\text{Cs}$  ratios are from 0.56 to 0.83 in Table 5-2, which are lower than the typical value (about 1) of FDNPP. This is due to the  $^{137}\text{Cs}$  from global fallout deposited on the seabed soil of the survey sea area.

$^{137}\text{Cs} - ^{134}\text{Cs}$  in Table 5-2 is the calculated global fallout value. Seabed soil at St. 11 and 13 was low in clay-silt, and the  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were low in concentration. In contrast, the seabed soil at St. 12, 17, 40, 44 was mud and the  $^{134}\text{Cs}$

concentrations were higher than those at St. 11 and 13. The seabed soil with small grain sizes adsorbed a larger amount of the global fallout.

Figure 5-4 shows the relationship between  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations in seabed soil at the Sakata, Kamo, and Naoetsu offings. The radioactive cesium concentration was decay-corrected to March 11, 2011. A broken line representing  $^{137}\text{Cs}/^{134}\text{Cs} = 1$  is shown in Figure 5-4. Because the  $^{137}\text{Cs}/^{134}\text{Cs}$  radioactivity ratio when the FDNPP accident occurred was almost 1, all data should be plotted on the broken line. However, all  $^{137}\text{Cs}$  values were plotted above the line. The differences between the  $^{137}\text{Cs}$  concentration and the broken line at each data point are the estimated  $^{137}\text{Cs}$  concentration from global fallout, which is 0.14–1.60 Bq/kg. In the radioactivity survey in 2010, which is the year before the Great East Japan Earthquake, the  $^{137}\text{Cs}$  concentration in seabed soil from global fallout was 0.6–5.6 Bq/kg in the adjacent Japan Sea<sup>5-4)</sup>, which is similar to the calculated value in the present study.

Table 5-2 Concentration ratios of radioactive cesium and global fallout

Survey areas	No	$^{134}\text{Cs}$ [Bq/kg]	$^{137}\text{Cs}$ [Bq/kg]	$^{134}\text{Cs} / ^{137}\text{Cs}$	$^{137}\text{Cs} - ^{134}\text{Cs}$ [Bq/kg]
Sakata Offing	St.11	1.37	1.66	0.83	0.28
	St.12	2.92	4.42	0.66	1.50
	St.13	0.69	0.83	0.83	0.14
	St.17	2.75	4.20	0.65	1.45
Kamo Offing	St.40	3.75	5.29	0.71	1.54
	St.44	2.03	3.63	0.56	1.60

The concentration of radioactive cesium was decay-corrected on the time of March 11, 2011.

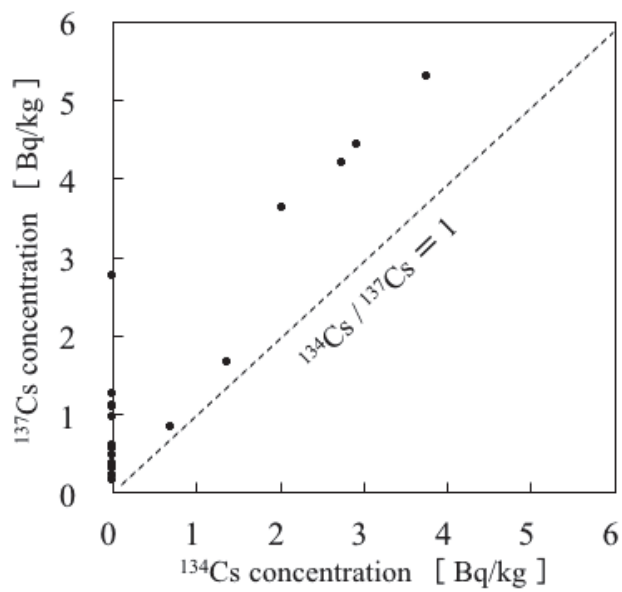


Figure 5-4 Relation between  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations in seabed soil. The  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  values were detected in the seabed soil from the Sakata, Kamo, and Naoetsu offings. The data were decay-corrected to March 11, 2011. A concentration of 0 means no  $^{134}\text{Cs}$  or  $^{137}\text{Cs}$  was detected.

### 5.3.5 Concentration of $^{137}\text{Cs}$ in seabed soil

Table 5-1 also shows the  $^{137}\text{Cs}$  concentration in seabed soil, which was decay-corrected to the sampling time.  $^{137}\text{Cs}$  was detected in all seabed soil at the Kamo, Sakata, and Naoetsu offings, and the concentration was 0.13–4.7 Bq/kg. The maximum concentration was in the seabed soil from St. 40 in the Kamo offing. Because  $^{134}\text{Cs}$  was detected in seabed soil from the Kamo and Sakata offings,  $^{137}\text{Cs}$  from FDNPP may also have migrated in these sea areas. However, because  $^{134}\text{Cs}$  was not detected in seabed soil at the Naoetsu offing, the  $^{137}\text{Cs}$  from FDNPP probably did not affect the Naoetsu offing.

Figure 5-3 also shows the relation between the  $^{137}\text{Cs}$  concentration in seabed soil and the migration distance from the Agano River mouth. The maximum  $^{137}\text{Cs}$  concentrations in each sea area decay-corrected to March 11, 2011 were used. The  $^{137}\text{Cs}$  concentration in seabed soil decreased in the order of the Kamo offing, the Sakata offing, and Ishikari Bay.



### 5.3.6 Migration of radioactive cesium from FDNPP to the Japan Sea and the Sakata offing

Naoetsu Port is approximately 200 km from Kamo Port. In this region there are several rivers, including the Agano and Mogami Rivers. In August 2011, radioactive cesium from FDNPP was detected in seabed soil from the Agano River mouth offing <sup>5-2)</sup>. From August to September 2011, the Ministry of Education, Culture, Sports, Science and Technology performed aerial radioactivity surveys in Niigata Prefecture, and they found that the surface soil from Niigata Prefecture contained radioactive cesium from FDNPP <sup>5-1)</sup>. This radioactive cesium migrated from FDNPP to forest areas via the atmosphere and was washed into the river by rainfall. Radioactive cesium is dissolved and suspended in the river, and some of the suspended cesium becomes dissolved in the river mouth and the neighboring sea area <sup>5-15, 5-16)</sup>. The radioactive cesium dissolved in seawater is moved by ocean currents, adsorbed to mineral particles, and settles to the seabed <sup>5-17)</sup>.

Figure 5-5 shows the Tsushima warm current and the Soya warm current. The Tsushima warm current flows north along the northwest coast to the sea area of the

Sakata offing from the Naoetsu offing. Radioactive cesium from FDNPP was detected in seabed soil at the Kamo offing and the Sakata offing, whereas it was not detected in seabed soil at the Naoetsu offing. The present results show that the origin is limited to the region from Naoetsu to Sakata, including the Agano and Mogami Rivers. However, because  $^{134}\text{Cs}$  was detected in surface seawater from the Fukui prefecture offing higher in the current during early days<sup>5-5)</sup>, we should also consider the migration pathway of  $^{134}\text{Cs}$  from the atmosphere.

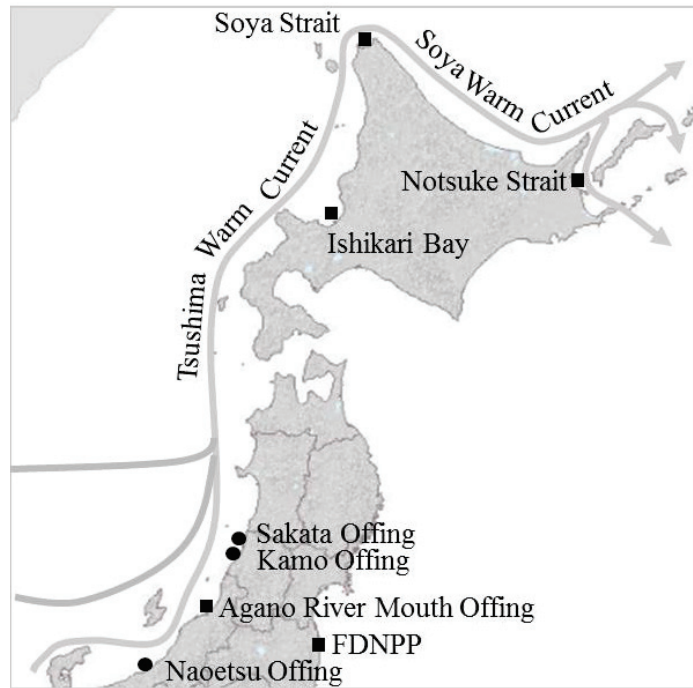


Figure 5-5 Ocean currents and sampling areas.

The curve shows part of the Tsushima warm current and the Soya warm current<sup>5-18)</sup>. The black circles indicate the sampling areas.

## 5.4 Conclusion

We performed radioactivity surveys in seabed soil from the Sakata, Kamo, and Naoetsu offings to identify the origin and the migration pathway of radioactive cesium from FDNPP in these offings.  $^{134}\text{Cs}$  was detected in seabed soil containing mud and sand at the Sakata and the Kamo offing, which contained 1.2% - 33.7% of clay-silt. There was a proportional relationship between the clay-silt ratio and radioactive cesium concentration. This result suggested that the particle size of the seabed soil is an important factor in the adsorption of radioactive cesium from seawater to seabed soil.  $^{134}\text{Cs}$  was not detected in seabed soil from the Naoetsu offing, even though seabed soils contained more than 20% clay-silt, showing that the radioactive cesium from FDNPP did not migrate to the Naoetsu offing. The  $^{134}\text{Cs}$  concentration in seabed soil decreased in the order of the Kamo offing, the Sakata offing, and Ishikari Bay.  $^{137}\text{Cs}$  from global fallout was detected at all sampling points in this survey, in which the concentration was 0.28–1.6 Bq/kg. Radioactive cesium from FDNPP was detected in seabed soil from the Kamo and

Sakata offings, whereas it was not detected in seabed soil from the Naoetsu offing.

The present results show that the origin is limited to the region from Naoetsu to Sakata, including the Agano and Mogami Rivers.

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## Chapter 6      Simulation of Cesium Concentration

## 6.1 Change of the $^{134}\text{Cs}$ concentration depending on migration distance

Seabed soils were collected, and the radioactivity was measured at sea areas of the Notsuke Strait, the Soya Strait, the Sakata offing, the Kamo offing and the Naoetsu offing. Table 3-2, Table 4-1, Table 5-1 show the result of the measurements.  $^{134}\text{Cs}$  is detected at 8 points of seabed soil, the maximum concentration was 0.68 Bq/kg (concentration was corrected to the collection day). Collection day of seabed soils passed 3-5 years from Great East Japan Earthquake. Because half-life of  $^{134}\text{Cs}$  is approximately 2 years,  $^{134}\text{Cs}$  decayed very much in seabed soil.

Figure 6-1 shows relations between migration distance from the Agano River mouth and the maximum concentration of  $^{134}\text{Cs}$  concentration at each collection sea area. Concentration was decay-corrected on March 11, 2011. The square shows measurement values in Figure 6-1. The value of the Agano River mouth and the Ishikari Bay were listed in addition to the measurements by this study to know concentration at whole sea area.

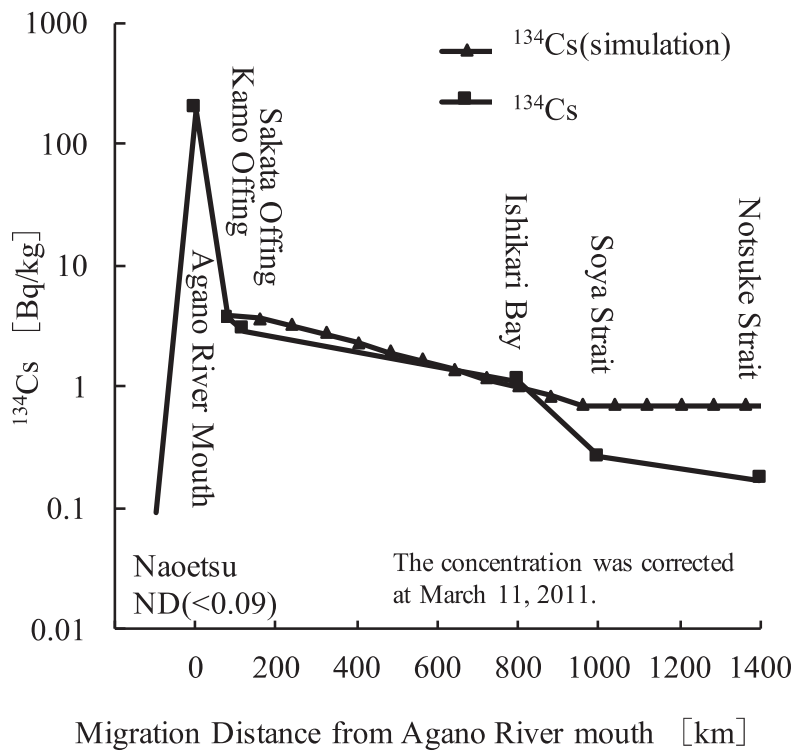


Figure 6-1 Relations between migration distance from the Agano River mouth and the maximum concentration of  $^{134}\text{Cs}$  at each sampling sea area.

According to the survey by Niigata prefectural government office,  $^{134}\text{Cs}$  was detected at the Agano River mouth offing in seabed soil. Similarly,  $^{134}\text{Cs}$  was detected in seabed soils at the Kamo offing, the Sakata offing, the Ishikari Bay, the Soya strait, the Notsuke Strait. As distance from the Agano River mouth increases, concentration of  $^{134}\text{Cs}$  in seabed soil becomes smaller. This result shows that the origin of  $^{134}\text{Cs}$  is limited to the region from Naoetsu to Sakata, which includes Shinano River, Agano River and Mogami River. The result suggests that water mass including  $^{134}\text{Cs}$  migrated by ocean currents.

The triangle in Figure 6-1 shows a value of the simulation. This simulation estimated  $^{134}\text{Cs}$  concentration in the seabed soil by an advection-diffusion equation. The simulation was referenced to Akira Wada "Diffusion predictive analysis of the radionuclide in the sea (2012 Maruzen publication)"<sup>6-1)</sup>.

### 6.1.1 Advection-Diffusion calculation model

This simulation is performed to confirm that the source of  $^{134}\text{Cs}$  is a river and the  $^{134}\text{Cs}$  concentration in the seabed soil decreases according to the advection-diffusion equation. The diffusion calculation of  $^{134}\text{Cs}$  concentration in seawater is expressed by next expression.

$$\frac{\partial C}{\partial t} + U_x \frac{\partial C}{\partial x} + U_y \frac{\partial C}{\partial y} + U_z \frac{\partial C}{\partial z}$$

①            ②            ③

$$= \frac{\partial}{\partial x} \left( K_x \frac{\partial C_D}{\partial x} \right) + \frac{\partial}{\partial y} \left( K_y \frac{\partial C_D}{\partial y} \right) + \frac{\partial}{\partial z} \left( K_z \frac{\partial C_D}{\partial z} \right) + V_s \frac{\partial C_s}{\partial z} - B \cdot C + C_o$$

④                    ⑤                    ⑥                    ⑦            ⑧            ⑨

-----No.1

$t$ : Time (s),

$x, y, z$ : Coordinate axis (m),

$U_x, U_y, U_z$ : Velocity component (m/s) to  $x, y, z$  direction,

$C$ : Material concentration ( $C=C_D$ (dissolved state) +  $C_s$ (suspension state)) ( $\text{g/m}^3$ ),

$K_x, K_y, K_z$ : Diffusion coefficient to the  $x, y, z$  direction ( $\text{m}^2/\text{s}$ ),

$V_s$ : Sedimentation velocity of the suspension state (m/s),

$B$ : Decay constant (1/s),

$C_0$ : value of fallout ( $\text{g}/\text{m}^3/\text{s}$ )

Each term has a meaning shown below.

① ② ③: advection

④ ⑤ ⑥: diffusion

⑦: sedimentation

⑧: decay

⑨: fallout.

Condition of this simulation is shown below.

- This simulation estimated  $^{134}\text{Cs}$  concentration in surface seabed soil.
- This simulation estimated the value under ocean currents axis. Because the ocean current is  $X$  direction and this is dominant, we ignore the contributions of ② and ⑤ which are  $Y$  direction.
- Concentration was decay-corrected on March 11, 2011. Because ⑧ is decay-corrected value, it is not necessary to calculate.

- ⑨ is term of Fallout. Plume did not reach the Japan Sea, and it is thought that the fallout to the Japan Sea is little. Therefore, the term ⑨ was omitted in the present simulation.
- Sea area of calculations is the Tsushima Warm Current and the Soya Warm Current from the Agano River mouth to the Notsuke Strait.

### 6.1.2 Discretization

The advection-diffusion equation (No.1) was described in partial differential equation. Discretization was calculated by finite differential method to enable numerical computation.

$$\begin{aligned} & \frac{C(t + \Delta t, x, z) - C(t, x, z)}{\Delta t} + U_x \frac{C(t, x, z) - C(t, x - \Delta x, z)}{\Delta x} \\ & = K_x \frac{C_D(t, x + \Delta x, z) - 2C_D(t, x, z) + C_D(t, x - \Delta x, z)}{\Delta x^2} \\ & + K_z \frac{C_D(t, x, z + \Delta z) - 2C_D(t, x, z) + C_D(t, x, z - \Delta z)}{\Delta z^2} \\ & + V_S \frac{C_s(t, x, z + \Delta z) - C_s(t, x, z)}{\Delta z} \end{aligned}$$

----- No.2



$$C = C_D + C_S \text{ (g/m}^3\text{)}$$

$$C_D = A_D \times C$$

$$C_S = A_S \times C$$

Material concentration is described by dissolved state ratio and suspension state ratio.

No. 2 is converted as follows.

$$C(t + \Delta t, x, z)$$

$$= C(t, x, z)$$

$$\begin{aligned}
 & + \Delta t \times \left\{ -U_x \times \frac{C(t, x, z) - C(t, x - \Delta x, z)}{\Delta x} \right. \\
 & \quad + A_D \times K_x \frac{C(t, x + \Delta x, z) - 2C(t, x, z) + C(t, x - \Delta x, z)}{\Delta X^2} \\
 & \quad + A_D \times K_z \frac{C(t, x, z + \Delta z) - 2C(t, x, z) + C(t, x, z - \Delta z)}{\Delta Z^2} \\
 & \quad \left. + A_S \times V_S \frac{C(t, x, z + \Delta z) - C(t, x, z)}{\Delta Z} \right\}
 \end{aligned}$$

----- No.3

The numerical value used for this calculation is as follows.

- Velocity of the ocean current:

$$0-1000 \text{ km} \quad U_x = 0.3 \text{ kt (13.3 km/day),}$$

1000-2000 km       $U_x = 2.0$  kt (88.8 km/day)

- Diffusion coefficient:

$1.3 \times 10^{-9}$  m<sup>2</sup>/s ( $1.1 \times 10^{-10}$  km<sup>2</sup>/day).

- Dissolved state / Suspension state:

0.9/0.1. (The previous study shows 0.99 / 0.01. If this ratio is used in the present simulation, the result diverges. Therefore, the ratio 0.9/0.1 was used at the present stage.)

- Division width of x direction:

80 km.

- Division of time:

0.5 days.

- Division of depth:

3 layers    0, 50, 100 m.

- Sedimentation velocity of suspension state of <sup>134</sup>Cs:

40 m/day.

- Initial condition:

130 Bq/kg of  $^{134}\text{Cs}$  is included in the surface seawater in the neighborhood of Agano River mouth.

- Suspension state of the bottom layer:

All suspension states are adsorbed by seabed soil.

130 Bq/kg was given in the initial condition to surface seawater at the Agano River mouth. The reason is to conform the estimated concentration to measured concentration in the Kamo offing.  $^{134}\text{Cs}$  concentration after 180 days was compared with that after 120 days, but there were not many differences. Therefore, steady state was regarded as 180 days (360steps). In these conditions, concentration of  $^{134}\text{Cs}$  in seabed soil at Ishikari Bay was calculated as 1.16 Bq/kg. The measured value was 1.1 Bq/kg, and the difference is 0.06 Bq/kg.

The concentrations measured in the Soya Strait and the Notsuke Strait are lower than simulation, and these differences are 0.45 Bq/kg and 0.53 Bq/kg, respectively. The Okhotsk Surface Low Salinity Water and the Intermediate Cold Water exist in the northern part of the Soya warm current. The Okhotsk Surface Low Salinity

Water, the Intermediate Cold Water and the Soya warm current were mixed, and the concentration of  $^{134}\text{Cs}$  might be diluted. It is supposed that the difference of concentration appeared because the inflow seawater from the outside is not calculated on this simulation. A tendency to decrease with currents was seen in both the simulation and measurements. It is suggested that radioactive cesium flows out from the areas from the Naoetsu to the Sakata into the Japan sea which was migrated along the Tsushima warm current and the Soya Warm current without swerving from currents.

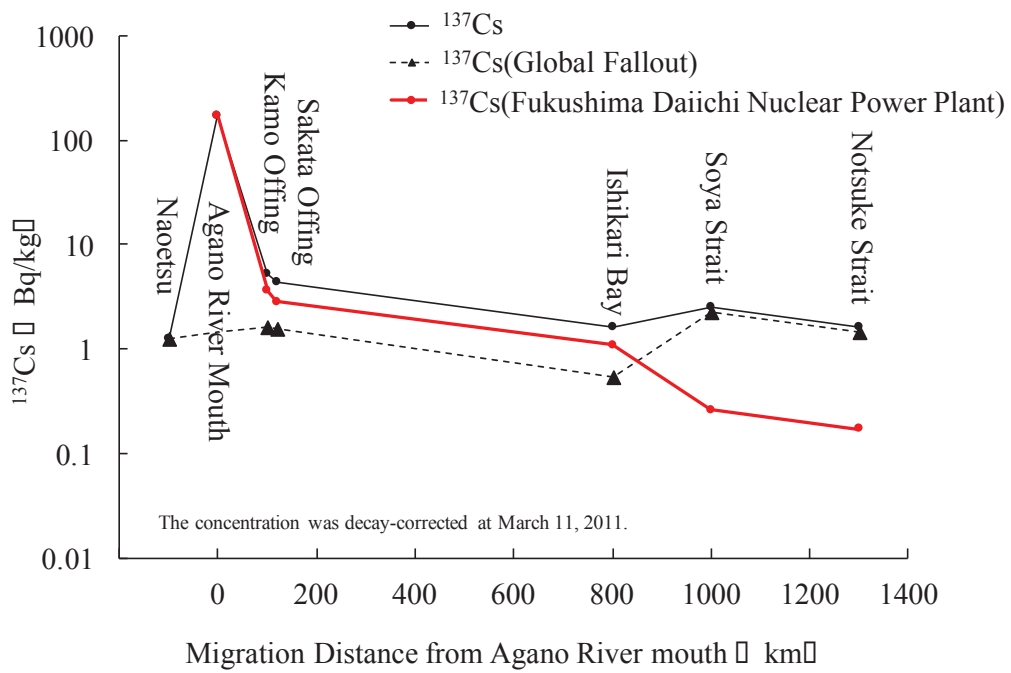


Figure 6-2 Relations between migration distance from the Agano River mouth and the maximum concentration of  $^{137}\text{Cs}$  at each sampling sea area.

## 6.2 Change of $^{137}\text{Cs}$ concentration depending on migration distance

Figure 6-2 shows the concentration of  $^{137}\text{Cs}$  in seabed soil collected on this study.

The values of the Agano River mouth and the Ishikari Bay were written jointly in

Figure 6-1. Because  $^{134}\text{Cs}$  was not detected in seabed soil at the Naoetsu offing, it

is thought that there is little influence of  $^{137}\text{Cs}$  derived from FDNPP. Because the

Tsushima warm current flows from the west to the east at the Naoetsu offing, the

radioactive cesium does not flow out into the Japan Sea at western region of

Naoetsu. Niigata prefectural government office carried out radioactivity survey at

the Agano River mouth of northeast approximately 100 km of the Naoetsu offing<sup>6-</sup>

2).  $1.7 \times 10^2$  Bq/kg of  $^{137}\text{Cs}$  was detected in surface seabed soil on August, 2011.

This concentration was the maximum in survey at sea areas of this study. In the

Kamo offing, the Sakata offing and the Ishikari Bay, as migration distance increases

depending on currents,  $^{137}\text{Cs}$  concentration in seabed soil gradually decreased.

Tendency to decrease in concentration becomes different in the Soya Strait. The

Okhotsk Surface Low Salinity Water and the Intermediate Cold Water exist in the

northern part of the Soya warm current <sup>6-3)</sup>. The Okhotsk Surface Low Salinity Water, the Intermediate Cold Water and the Soya warm current were mixed, and the concentration of <sup>137</sup>Cs might be diluted. This phenomenon was similar to <sup>134</sup>Cs.

### 6.3 References

6-1) Wada, A.: Marine diffusion predictive analysis of the radionuclide, Maruzen Press (2012).

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## Chapter 7      General Conclusions



## General Conclusions

A radioactivity survey was carried out at the Notsuke Strait, the Soya Strait, the Sakata offing and the Kamo offing, and  $^{134}\text{Cs}$  was detected in seabed soil. It was found that radioactive cesium derived from FDNPP was migrated to these 4 sea areas. The radioactive survey at the Naoetsu offing was also carried out, but  $^{134}\text{Cs}$  was not detected in seabed soil on the survey at Naoetsu offing. The source of the outflow of the radioactive cesium came from FDNPP was limited to the area east than Naoetsu.

The result of advection diffusion simulation by ocean currents almost corresponded with measurements, and it was confirmed that an ocean current was the important factor of migration of the radioactive cesium. The Tsushima warm current and the Soya warm current, where the Kuroshio Current of the East China Sea is a source, exist at the sea areas from the Naoetsu offing to the Notsuke Channel.

In conclusion, a part of the radioactive cesium derived from FDNPP flowed out into the Japan Sea from the region east than Naoetsu, and migrated to Notsuke Strait by the Tsushima warm current and the Soya warm current.

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