Article

Effect of Fukushima Daiichi Nuclear Power Plant Accident on Lakes in Fukushima Prefecture and Niigata City

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The Fukushima Daiichi Nuclear Power Plant Accident happened in 12 – 15 th March 2011. This accident caused to the radioactive contamination in the near area. In this work, some lake waters and the sediments were collected in Fukushima Prefecture and Niigata City to clarify the effect of the accident on the area contaminated. As to the collected lake water, an enrichment of “the specific activity of tritium (T specific activity)” in each water collected was carried out by the solid polymer electrolyte (SPE) apparatus, and the T specific activity was measured by a liquid scintillation counter. Based on the T specific activity thus measured, the influence of the accident to the environment was investigated. At the same time, specific activities of radioactive caesiums in the lake sediments were also measured. Consequently, the influence of the Fukushima Daiichi Nuclear Power Plant Accident to the adjacent lakes (e.g., in Fukushima Prefecture and Niigata City) was quantitatively clarified. From the above-mentioned matters, it is found that the effect of the accident on Niigata City is so small, and the accident gradually decreases in Fukushima Prefecture.

Key Words : Fukushima Daiichi Nuclear Power Plant Accident, tritium, caesium-137, caesium-134, solid polymer electrolyte apparatus

1. Introduction

The accident of the Fukushima Daiichi Nuclear Power Plant occurred in March 2011. This accident caused a level 7 event of the International Nuclear Event Scale that means the worst classification. It is the second scale of the nuclear power plant accident next to Chernobyl disaster. This accident caused a lot of radioactive fallouts, and the fallouts were accumulated not only in near the area but also in the adjacent prefectures. According to our previous research¹⁾, Tritium (T or ³H) specific activity (obtained in Niigata City) was temporary in-

creased by the accident. The highest T content was also found in the first rainfall after the accident²⁾.

As to radioactive fallouts, T is one of them. Its half-life is 12.32 years, emits β^- ray, and its maximum energy is 18.6 keV (ave. 5.7 keV). Therefore external exposure from T can be neglected. However, internal exposure cannot be neglected because T invades into the body and emits β^- ray to certain cell directly³⁾. In environment, T behaves with water because most of the chemical form of T is HTO. Total amount of T in the world is estimated to be approximately 1 300 PBq (3.5 kg), and this value

Fig. 1 Locations of Niigata Prefecture (A) and Fukushima Prefecture (B), and of sampling points in (A) and (B).

(A) Niigata Prefecture

(B) Fukushima Prefecture

① Lake Inawashiro

② Lake Akimoto

③ Goshikinuma tarn

④ Lake Hayama

⑤ Sakata lagoon

⑥ Lake Toyanogata

is similar to the natural equilibrium level^(4),5). T is naturally produced in the upper atmosphere. When cosmic rays interact with nitrogen nuclei in air^(6),7), following (1) occurs, and finally T decays as (2).

In general, radioactive caesium (^{134}Cs and ^{137}Cs) are not discharged from a nuclear power plant. However, when serious nuclear power plant accident like a Chernobyl Accident is

happened, these caesium may be discharged in an environment. The half-life of ^{137}Cs is 30.1 years, and the caesium compounds are generally water-soluble. Thus caesium taken into a body are distributed in whole of the body, and many of them are accumulated into the muscle because it is an alkali metal, and because its physical and chemical properties are similar to those of rubidium and potassium. If a human takes radioactive caesium, about 10% of them is excreted from the body as sweat and urine. The remaining 90% ones decrease with a bio-

Fig.2 The sampling points at Lake Inawashiro, Lake Akimoto, Goshikinuma tarn and Lake Hayama in Fukushima Prefecture.

Fig.3 The sampling points at Lake Toyanogata and Sakata lagoon in Niigata Prefecture.

logical half-life between 50 and 150 days⁸⁾.

Considering the above-mentioned, the following two were aimed in this study: (1) to research the influence of the accident to several lakes in Fukushima Prefecture and to Niigata City, (2) to clarify the effect of the accident on Niigata City.

2. Experimental

2.1 Sampling

Fukushima Daiichi Nuclear Power Plant locates in Okuma town in Fukushima Prefecture, Japan (Coordinates : $37^{\circ}25'22.7''\text{N}$ $141^{\circ}01'58.5''\text{E}$). We chose some sampling points in order to research the influence of the accident to some lakes in Fukushima and Niigata. Each location is shown in Fig.1. There are Lake Inawashiro, Lake Akimoto, Goshikinuma tarn, and Lake Hayama in Fukushima Prefecture shown in Figs.2. At Lake Inawashiro, we picked up the samples in the three spots (Recreation park, Sakka beach and Tenjin beach). Lake Toyanogata and Sakata lagoon in Niigata City is also shown in Fig.3. Lake Hayama is the nearest to the Fukushima Daiichi Nuclear Power

Table 1-1 Several sampling conditions

Sampling point	2013.May			2013.Sep		
	distance	lake water	depth / cm	distance	lake water	depth / cm
	from shore / m	temperature / °C		from shore / m	temperature / °C	
Lake Inawashiro						
Recreation park	5	16	30	5	21.2	10
Sakka beach	5	14	30	5	22.3	50
Tenjin beach	5	15.6	50	5	21.7	30
Lake Akimoto	3	17	10	3	23.8	20
Goshikinuma tarn	1	16	10	1	20.2	20
Lake Hayama	10	16	30	5	23.8	10
Lake Toyanogata	-	-	-	1	26	30
Sakata lagoon	1	23.1	20	1	23.6	30
Sampling point	2014.May			2014.Oct		
	distance	lake water	depth / cm	distance	lake water	depth / cm
	from shore / m	temperature / °C		from shore / m	temperature / °C	
Lake Inawashiro						
Recreation park	5	20.6	40	5	25.2	20
Sakka beach	5	18.6	30	5	20.9	20
Tenjin beach	5	19.4	40	5	21.6	30
Lake Akimoto	3	26.2	5	3	21.7	10
Goshikinuma tarn	1	20.6	10	1	17.2	5
Lake Hayama	10	26.1	10	10	20.6	20
Lake Toyanogata	1	29.4	15	1	18.6	20
Sakata lagoon	1	22.5	50	1	20.8	30

Plant in the sampling points chosen in this work. Sample water and sediment were collected at each lake. The sample water was picked up about 5 meter from beach and depth is roughly 30 centimeter under the surface. The sediment was also picked up using increment scoop (approximately 0 – 10 centimeter from the bottom of a lake). We collected these samples in spring and in autumn (in 2013 and 2014). Tables 1-1 and 1-2 show several sampling conditions and geographic coordinates. Dissolved Oxygen (DO), pH and ORP in the

water were immediately measured at each sampling point. As to a reference data, we used the collected precipitation in Niigata City collected every one month. The collecting apparatus was set on a roof of Faculty of Engineering Niigata University⁹⁾. The collected sample is called “Simulated Infiltrating Water”, which is similar to grand filtration water¹⁰⁾.

2.2 Sample pretreatment

As to each collected lake water, an enrichment of T specific activity was carried out by

Table 1-2 Geographic coordinates sampling points

Sampling point	north latitude			east longitude		
Lake Inawashiro						
Recreation park	37°	30′	55″	140°	1′	17″
Sakka beach	37°	27′	45″	140°	2′	48″
Tenjin beach	37°	31′	18″	140°	6′	57″
Lake Akimoto	37°	39′	37″	140°	7′	28″
Goshikinuma tarn	37°	39′	10″	140°	5′	22″
Lake Hayama	37°	43′	42″	140°	48′	27″
Lake Toyanogata	37°	53′	49″	139°	4′	1″
Sakata lagoon	37°	49′	10″	138°	52′	41″

using the solid polymer electrolyte (SPE) apparatus. Because of the difficulty of measuring the weak β^- radiation (of T) and of the low levels of the T specific activity, it is usually necessary to carry out an isotopic enrichment before measurement^{(1), (11), (12)}. In this work, T in precipitation was measured by liquid scintillation counter (LSC) after both distillation and electrolytic enrichment⁽¹³⁾.

2•3 Measurements of activity of T and of various ions

The samples were distilled to prevent the contaminations of other materials and to minimize quenching⁽¹⁴⁾ before electrolytic enrichment. This process was performed by adding both 0.3 g of KMnO_4 and 0.3 g of Na_2O_2 to 1 380 mL of each water sample.

In general, when the T specific activity in an aqueous sample is low ($<1 \text{ Bq} \cdot \text{L}^{-1}$), the direct counting of the distilled water using LSC is very difficult, accordingly, the sample needs to be enriched. In the enrichment of T with electrolysis, a 1 080 mL of sample water is electrolytically concentrated to 36 mL. Consequently, 30 fold of volume enrichment were performed.

In this work, analysis procedure of T enrichment and the cell design were closely similar to the Masaaki SAITO's description⁽¹³⁾.

Moreover, major ions (Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- , NO_3^- , and SO_4^{2-}) in the sample were measured by using both ion chromatography and atomic adsorption spectrometry. In addition, the radioactivities of ^{137}Cs and ^{134}Cs in sediment samples were determined by gamma-ray spectroscopy.

3. Results and discussions

3•1 T specific activity

T specific activity in the precipitation in Niigata City was about $0.6 - 1.0 \text{ Bq}^9$. Fig. 4 shows the activity in the water in each lake. This figure indicates that deposition of T is easily affected by the topography and may accumulate at the place where convection does not occur. Especially, Lake Akimoto is surrounded by mountains, and it seems that the lake has an almost closed system. Therefore, T specific activity at Lake Akimoto showed higher value than other samples in Fukushima. In addition, Sakata lagoon in Niigata Prefecture has also closed system, and not connected to Agano

Fig. 4 Specific activity of T in each lake in Fukushima or Niigata.

river (this river connected to Fukushima). From our previous research¹⁵⁾, it revealed that T specific activity in Niigata City was increased by the accident. However, this research showed that T specific activity in Sakata lagoon was not so large, and Sakata lagoon is closed itself. Accordingly, based on T specific activity, effect of the accident on Sakata lagoon could not be observed.

3.2 Specific activities of ^{134}Cs and ^{137}Cs

Specific activities of ^{134}Cs and ^{137}Cs in the lake sediments were shown in Figs. 5 and 6. From the figures, it is found that the ratio of ^{134}Cs to ^{137}Cs was almost 1 : 2. The Nuclear Power Plant has been worked for a long time. And ^{134}Cs and ^{137}Cs would not be generated in the nuclear reactors after the accident. So, the ratio of ^{134}Cs to ^{137}Cs might be 1 : 1 at the first of the accident. Caesium-134 has a half-life of about 2 years. On the other hand, that of ^{137}Cs is 30 years. When no radioactive material continued to be fallout

on these lakes after the accident, the ratio of ^{134}Cs to ^{137}Cs may be estimated almost 1 : 2 after about 2 years. Moreover, we had to consider about influence of aboveground nuclear testing in 1950 – 1960s and Chernobyl disaster. However, these effects can be neglected because the effects were only a few becquerel. Accordingly, it can be deduced that both ^{134}Cs and ^{137}Cs were emitted from the accident, and ^{134}Cs and ^{137}Cs have not discharged since the accident occurred. As to the value of specific activity of each caesium in Lake Inawashiro (see Fig. 2), the activity at outflow place (Recreation Park at Lake Inawashiro) was higher than that at inflow places (Sakka beach at Lake Inawashiro, and Tenjin beach at Lake Inawashiro). The other hand, the influence of the accident to Sakata lagoon in Niigata City could not be found because radioactive caesium could not be observed.

Fig. 7 shows the map of radioactive caesiums on soil in East Japan¹⁶⁾. From the figure, it

Fig. 5 Specific activity of ^{134}Cs in sediment of each lake in Fukushima or Niigata.

Fig. 6 Specific activity of ^{137}Cs in sediment of each lake in Fukushima or Niigata.

found that east Japan was influenced by the accident directly, especially Fukushima, and adjacent prefectures. In this work, it also seems that the sampling points in Fukushima are influenced by the accident.

On the other hand, in Niigata, the caesium could not be detected in Sakata lagoon. But, the caesium was a little detected in the sediment at Lake Toyonogata. The reason may be that Lake Toyonogata is connected to Agano River,

Fig. 7 Map of radioactive caesium on soil in East Japan¹⁶⁾.

and the river also connected to Lake Inawashiro through some rivers, consequently, the detected caesium came from Fukushima, especially, Lake Inawashiro.

3.3 DO, pH, ORP and various ions

Dissolved Oxygen (DO), pH, ORP and vari-

ous ions in the sample were measured. DO, pH and ORP in the sample were directly measured at each sampling point (Table 2). There are many hot springs near Goshikinuma tarn. Therefore, the values of these ion concentrations in the sample collected at Goshikinuma tarn were largely different from others, and

Table 2-1 Several data measured on May 11 – 12th in 2013 (mg/L)

Sampling point	DO / mg · L ⁻¹	pH	orp / mV	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
Lake Inawashiro										
Recreation park	9.2	7.6	209	8.89	1.82	2.40	10.8	9.83	0.74	30.9
Sakka beach	10.3	8.0	220	8.86	1.90	2.43	10.2	9.10	0.89	25.0
Tenjin beach	9.8	7.6	206	10.9	2.27	3.27	11.4	12.1	0.81	40.8
Lake Akimoto	9.2	8.3	243	5.75	1.27	1.28	38.4	4.49	0.66	5.71
Goshikinuma tarn	9.2	7.3	209	65.0	10.1	20.2	71.0	58.8	0.00	247
Lake Hayama	9.6	7.9	230	6.74	1.09	2.46	10.8	4.23	0.92	3.43
Lake Toyanogata	–	–	–	–	–	–	–	–	–	–
Sakata lagoon	13.5	9.8	21.0	21.0	4.58	14.9	25.5	25.8	0.23	41.2

Table 2-2 Several data measured on Sep. 28 – 29th in 2013 (mg/L)

Sampling point	DO / mg · L ⁻¹	pH	orp / mV	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
Lake Inawashiro										
Recreation park	9.8	7.3	146	9.79	1.04	2.14	7.5	9.85	0.78	31.6
Sakka beach	8.3	8.6	156	8.67	1.02	2.12	7.8	9.72	0.91	31.5
Tenjin beach	9.3	9.0	148	9.9	1.10	2.34	8.2	10.6	0.77	31.7
Lake Akimoto	9.7	7.3	134	55.7	5.69	0.58	57.5	58.2	0.00	254
Goshikinuma tarn	7.9	7.0	156	5.7	0.7	0.8	3.2	6.0	0.00	6.0
Lake Hayama	7.9	7.6	155	5.75	0.52	1.77	8.2	3.91	0.73	3.42
Lake Toyanogata	6.5	6.4	72	74.6	3.0	10.7	15.3	13.6	1.8	31.6
Sakata lagoon	11.8	6.5	121	–	2.97	9.8	27.8	49.5	0.00	22.1

the concentrations of the ions may be influenced by hot spring. In addition, the value of pH in Sakata lagoon was larger than others because there were a lot of plants in the lagoon at the sampling day and the water sampled was so muddy.

3・4 Comparing the caesium in the samples with each other

The samples were also collected in autumn

(Sep., 2013 and Oct., 2014). Base on Figs. 5 and 6, comparing the specific activity of ¹³⁷Cs at spring sample with autumn one, it is found that the specific activity of ¹³⁷Cs was increased at closed-system place (Lake Akimoto) and at outflow ones (Sakka beach and Tenjin beach at Lake Inawashiro (see Fig. 2)). However, the activities at Lake Hayama were low (Sep., 2013). The reason is that water level at Lake Hayama was much higher than the level at the day sam-

Table 2-3 Several data measured on May 30 – 31th in 2014 (mg/L)

Sampling point	DO / mg · L ⁻¹	pH	orp / mV	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
Lake Inawashiro										
Recreation park	9.1	7.8	557	8.39	2.03	2.38	7.6	11.1	0.73	32.3
Sakka beach	10.5	7.5	492	8.04	1.92	2.31	8.2	10.8	1.10	32.2
Tenjin beach	9.5	6.9	420	8.4	2.21	2.51	8.8	11.4	1.12	36.6
Lake Akimoto	8.0	7.3	398	2.89	0.87	0.81	3.6	3.22	1.47	4.82
Goshikinuma tarn	8.0	6.6	212	39.7	7.3	13.9	57.3	46.5	0.13	216
Lake Hayama	10.3	8.4	268	4.64	0.71	1.85	4.5	4.37	1.09	3.16
Lake Toyanogata	7.0	8.1	92	74.3	4.04	9.42	10.2	161	3.68	21.8
Sakata lagoon	11.0	8.2	591	20.2	4.78	13.2	26.4	25.8	6.18	33.2

Table 2-4 Several data measured on Oct. 9 – 10th in 2014 (mg/L)

Sampling point	DO / mg · L ⁻¹	pH	orp / mV	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
Lake Inawashiro										
Recreation park	9.7	6.8	130	7.76	1.89	2.25	7.9	10.3	0.51	31.2
Sakka beach	9.0	7.5	202	7.99	1.97	2.34	77.0	10.6	0.60	32.1
Tenjin beach	12.0	7.2	163	11.8	3.66	3.31	11.4	15.3	0.16	34.7
Lake Akimoto	10.0	7.4	216	5.43	1.89	1.02	4.3	5.73	0.00	6.34
Goshikinuma tarn	8.1	6.6	199	48.2	8.4	15.6	58.3	59.5	0.12	227
Lake Hayama	9.6	7.7	244	5.65	0.95	2.04	5.3	4.34	0.26	3.75
Lake Toyanogata	4.5	6.7	166	46.3	6.27	7.09	38.4	68.2	6.68	14.2
Sakata lagoon	11.3	7.9	221	20.4	6.25	1.0	19.4	32.3	0.00	41.4

pled (comparing with the sampling days (May, 2013, May, 2014, and Oct., 2014)), therefore, we could not sample at the same place.

4. Conclusions

In order to reveal the effect of the Fukushima Daiichi Nuclear Power Plant Accident on some lakes in Fukushima and Niigata, the lake waters and sediments were collected and measured. From the result, the following

conclusions are clarified.

- (1) The effect of the accident on several lakes was clarified because the ratio of ¹³⁴Cs to ¹³⁷Cs was almost 1 : 2 after about 2 years. As to the lakes, specific activities of the caesium at outflow place apt to be higher than in inflow places.
- (2) The deposition of each caesium was greatly affected by the topography and it seems that each caesium was deposited in

closed-system place.

- (3) The measurement results show the T specific activity was similar to that of the precipitation recently collected at Niigata City. Therefore, based on T specific activity, the effect of the nuclear accident may be no longer observed in Niigata City after 5 months.
- (4) It seems that if the lake does not circulate, T cannot move, and be accumulated there.

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要 旨

福島県と新潟市の湖沼に及ぼす福島第一原子力発電所事故の影響

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2011 年 3 月 12 ～ 15 日に、福島第一原子力発電所事故が起こった。この事故は、近県に放射性物質汚染を引き起こした。本研究では、汚染された地域に及ぼすこの事故の影響を明らかにするため、新潟市と福島県における幾つかの湖沼の湖沼水と湖沼泥を採取した。湖沼水においては、固体高分子膜電解 (SPE) 装置によりトリチウム (T) 比放射能の濃縮を行い、T-比放射能を液シンで測定した。このように測定した T-比放射能に基づき、環境に及ぼすこの事故の影響を調査した。それと同時に、湖沼泥中の放射性セシウムの比放射能も測定した。その結果、福島第一原子力発電所事故が近くの湖沼 (例えば、福島県や新潟市) に及ぼす影響が定量的に明らかになった。以上のことから、新潟市に及ぼすこの事故の影響はかなり小さく、事故は徐々に収束していることがわかった。