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ARTICLE

Dispersion of Cs-134 and Cs-137 in river waters from Fukushima and Gunma prefectures at nine months after the Fukushima Daiichi NPP accident

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To estimate short-term and long-term impacts of radiation dose, dynamics of ^{134}Cs and ^{137}Cs in river watershed environments were studied in Fukushima and Gunma prefectures at nine months after the Fukushima Daiichi Nuclear Power Plant (NPP) accident. Radioactivity of ^{134}Cs and ^{137}Cs in the river waters ranged respectively from 0.007 Bq/l to 0.149 Bq/l and 0.008 Bq/l to 0.189 Bq/l. Their horizontal distributions were consistent with those of material that had been deposited on the ground surface. The percentages of particulate forms of ^{134}Cs and ^{137}Cs were 56–89% for the Fukushima river samples. Results show that the transport of ^{134}Cs and ^{137}Cs from the watershed to the river waters occurred mainly as particulate forms and that their radioactivity depends on the levels of radioactivity at the watersheds of the respective river systems.

Keywords: ^{134}Cs; ^{137}Cs; radiocesium; existence forms; lake waters; transport behavior

1. Introduction

A nuclear accident at the Fukushima Daiichi Nuclear Power Plant (NPP) occurred after the 2011 Tohoku Earthquake and Tsunami. About 15 PBq from both ^{134}Cs and ^{137}Cs were released from the Fukushima Daiichi NPP because of venting operations and hydrogen explosions [1-2]. The Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT) reported total surface deposition of ^{134}Cs and ^{137}Cs in the 80 km zone from the Fukushima Daiichi NPP [3]. Surface deposition results reveals significant external radioactivity in a zone extending northwest from the NPP: about 20 km wide and 50–70 km long. However, a mountainous area in Gunma Prefecture, located about 220 km distance from Fukushima Daiichi NPP south of Fukushima Prefecture, shows similar accumulation of ^{134}Cs and ^{137}Cs [3].

To estimate short–term and long–term impacts of the radiation dose in Japan, it is important to understand the dynamics of radionuclides, especially those of ^{134}Cs and ^{137}Cs, on river watershed environments. The study was undertaken to investigate ^{134}Cs and ^{137}Cs radioactivity in river systems in Fukushima and Gunma prefectures, Japan. The secondary radioactive dispersion of ^{134}Cs and ^{137}Cs from the contaminated watershed to the river waters is reported for both research areas with widely various radiocesium deposition on ground surfaces [3] at nine months after the Fukushima Daiichi NPP accident.

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2. Materials and methods

2.1. Sampling area

Five river systems–the Abukuma River, Uta River, Niida River, Natsui River and Same River– were set up as monitoring sites in Fukushima Prefecture, Japan. The Abukuma River has river length of 239 km and a watershed area of 5400 km^2. The other rivers have short river lengths of 43–78 km and small watersheds of 106–749 km^2. The annual mean water discharge of the Abukuma River is 192 m^3/s, as observed in 2008–2009 at Iwanuma Observatory in the lower river area [4]. The other rivers have annual mean discharge of 1.8–14.0 m^3/s for 2000–2005 [5].

In Gunma Prefecture, river research was conducted at the Tone River system, with watershed area of 16840 km^2 and river length of 322 km. Its branch rivers, Watarase River and Karasu River, have respective watershed areas of 2621 km^2 and 470 km^2. The annual mean water discharge of the Tone River was 146 m^3/s during 1998-2010 at Maebashi Observatory in Gunma Prefecture [4]. The Karasu River was 14.2 m^3/s at Takasaki for 2002–2010. The Watarase River flow was 20.9 m^3/s at Takatsudo for 1998–2010 [4].

Lake Akagi-Ohnuma, a holomictic lake [6], is located around the top of Mt. Akagi at about 1350 m altitude. The lake has a surface area of 0.83 km^2 and a watershed area of 4.82 km^2. Lake Akagi-Konuma, located near the top of Mt. Choshichiro at altitude of 1470 m, has a lake surface of 0.1 km^2. The lake waters overflowing from both lakes run through tributaries to the Tone River.

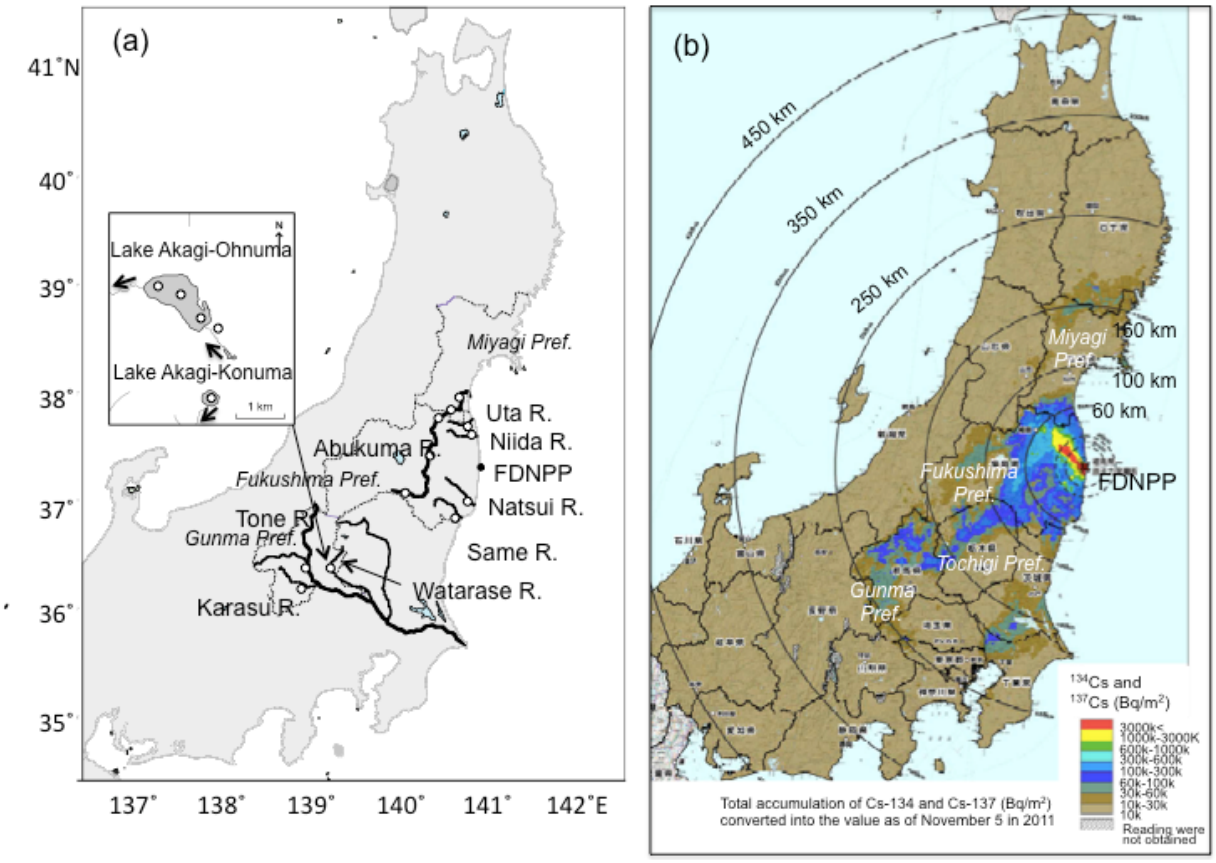

Figure 1. Sampling locations in this study (a) and distribution of ^{134}Cs + ^{137}Cs measured by the fourth airborne monitoring by MEXT (b). Open circles indicate the sampling sites for the measurements of ^{134}Cs and ^{137}Cs radioactivity in the river and lake waters. Solid lines drawn on the map in Figure 1(a) indicate the distribution of each river. Dashed lines indicate prefectural border. FDNPP indicates Fukushima Daiichi Nuclear Power Plant. Figure 1(b) was modified from [3].

2.2. Sampling

Figure 1 shows monitoring sites in the river systems in Fukushima Prefecture and Gunma Prefecture and the accumulation of ^{134}Cs + ^{137}Cs on the ground surface. The surface river water in the streamline was collected from a bridge at a monitoring site in the Uta River, Niida River, Natsui River, and Same River on 6–7 December 2011. The Abukuma River has five such monitoring sites from upstream (Shirakawa) through midstream (Motomiya and Date) to downstream (Marumori and Iwanuma). River research was also conducted at monitoring sites in the Tone River, the Karasu River, and the Watarase River in Gunma Prefecture on 15–16 December 2011. Surface water samples from Lake Akagi-Ohnuma and Lake Akagi-Konuma were collected using a bucket on 11 December 2011. Water samples in middle and bottom layers at the center of Lake Akagi-Ohnuma were collected using a Van Dorn water sampler. The water discharge was a normal flow condition in each river during the samplings [4,7].

2.3. Measurements

^{134}Cs and ^{137}Cs were concentrated onto ammonium molybdophosphate (AMP) [8] for 10–20 l of raw river waters and filtered river waters using cartridge filters with pore sizes of 10µm, 1µm and 0.45 µm sequentially. The AMP/Cs compound was dried at room temperature and weighed to determine the weight yield of AMP/Cs compound. The averaged yield was 91 ± 5% for all water samples. The dried AMP/Cs compound was packed into a polyethylene bag (3.5 cm × 7.0 cm).

The radioactivity of ^{134}Cs and ^{137}Cs was measured for the AMP/Cs compound using gamma-ray spectrometry with low background Ge detectors (GEM-251855 of ORTEC® and other similar models) equipped with a multichannel analyzer (Model 7700 of SEIKO EG&G Co. Ltd. and other similar models) at the Low Level Radioactivity Laboratory and the Ogoya Underground Laboratory of Kanazawa University during 1–3 days. Gamma emission peaks were used for calculating activity at 605 keV and 795 keV for ^{134}Cs and 661 keV for ^{137}Cs. The cascade summing effect was corrected for ^{134}Cs using a Fukushima contaminated soil sample. Decay correction of radioactivity for ^{134}Cs and ^{137}Cs was done at each sampling date. The ^{134}Cs and ^{137}Cs radioactivity is designated as the total (dissolved + particulate phase) radioactivity for the raw river and lake

Table 1. Physicochemical parameters and radioactivity of ^{134}Cs and ^{137}Cs in river and lake waters in Fukushima Prefecture and Gunma Prefecture, Japan.

Location	Site	Sampling date (Year/Month/Day)	pH	WT (C°)	Turbidity (NTU)	Conductivity (mS/m)	DO (mg/l)	^{134}Cs (Bq/l)	^{137}Cs (Bq/l)
River									
Abukuma	Shirakawa	2011/12/6	7.13	7.4	5.3	9.5	10.0	0.048 ± 0.002	0.063 ± 0.002
	Motomiya	2011/12/6	7.34	7.0	8.7	9.8	10.9	0.031 ± 0.002	0.041 ± 0.002
	Date	2011/12/7	7.16	6.9	9.6	16.5	9.7	0.091 ± 0.004	0.120 ± 0.004
	Marumori	2011/12/7	7.40	7.4	13.3	16.4	9.7	0.081 ± 0.002	0.105 ± 0.002
	Iwanuma	2011/12/7	7.19	7.8	8.6	16.0	10.1	0.067 ± 0.002	0.085 ± 0.003
Uta	Souma	2011/12/7	7.40	8.2	1.0	8.9	10.0	0.030 ± 0.002	0.037 ± 0.002
Niida	Minamisouma	2011/12/7	6.88	7.6	6.6	6.2	10.2	0.149 ± 0.004	0.189 ± 0.004
Natsui	Iwaki	2011/12/6	7.38	6.5	9.2	10.8	11.6	0.022 ± 0.001	0.028 ± 0.002
Same	Iwaki	2011/12/6	7.47	8.5	6.2	13.5	10.7	0.009 ± 0.001	0.011 ± 0.002
Tone	Maebashi	2011/12/15	7.09	7.0	10.1	17.5	9.8	0.007 ± 0.001	0.008 ± 0.001
Karasu	Takasaki	2011/12/15	7.51	9.7	16.2	29.4	8.7	0.009 ± 0.001	0.011 ± 0.001
Watarase	Midori	2011/12/16	7.26	6.5	17.5	10.1	9.5	0.011 ± 0.001	0.013 ± 0.001
Akagi-Ohnuma	inflow river	2011/12/16	6.76	3.2	5.0	4.2	9.1	0.014 ± 0.001	0.014 ± 0.001
Lake									
Akagi-Ohnuma	Inflow	2011/12/11	7.1	4.9	n.a.	n.a.	10.3	0.112 ± 0.001	0.146 ± 0.001
	Outflow	2011/12/11	7.3	5.2	n.a.	n.a.	10.7	0.101 ± 0.001	0.129 ± 0.001
	Center-0.5m	2011/12/11	7.2	5.0	n.a.	n.a.	10.6	0.120 ± 0.001	0.153 ± 0.001
	Center-8.0m	2011/12/11	n.a.	4.6	n.a.	n.a.	n.a.	0.123 ± 0.005	0.162 ± 0.005
	Center-16.2m	2011/12/11	n.a.	4.5	n.a.	n.a.	n.a.	0.104 ± 0.001	0.129 ± 0.001
Akagi-Konuma	Center-0.5m	2011/12/11	n.a.	n.a.	n.a.	n.a.	n.a.	0.057 ± 0.002	0.075 ± 0.002

WT = water temperature. DO = dissolved oxygen. n.a. = not analysis. Errors of radioactivity indicate the counting errors (1σ).

waters, and is as dissolved radioactivity for filtered river waters. The ^{134}Cs and ^{137}Cs particulate phase was estimated by subtracting the radioactivity of dissolved phase from the total value for raw river waters.

3. Results and discussion

3.1. Water chemistry

The physicochemical parameters of river and lake waters are presented in **Table 1**. The river water pH was 6.76–7.51. The conductivity was 4.2–29.4 mS/m. The turbidity was 1.0–17.5 NTU. The dissolved oxygen (DO) concentration was 8.7–11.6 mg/l.

3.2. Radioactivity of ^{134}Cs and ^{137}Cs

Table 1 shows ^{134}Cs and ^{137}Cs radioactivity in surface river water samples from Fukushima Prefecture on December 6–7, 2011. The ^{137}Cs radioactivity ranges from 0.011 Bq/l to 0.189 Bq/l and decreases downward from the Shirakawa site to the Motomiya site and then shows maximum value at the Date site in the Abukuma River.

The ^{137}Cs radioactivity ranges from 0.008 Bq/l to 0.014 Bq/l for river water samples from the Tone River systems and a small river flowing into Lake Akagi-Ohnuma. These values resemble those of the Same River and the Natsui River in the south part of Fukushima. However, surface water samples in Lake Akagi-Ohnuma and Lake Akagi-Konuma show ^{137}Cs radioactivity ranging from 0.075 Bq/l to 0.153 Bq/l. Their values are 1–2 orders of magnitude higher than those of river waters in Gunma Prefecture. The radioactivity of ^{137}Cs at the center of Lake Akagi-Onuma ranges from 0.129 Bq/l to 0.162 Bq/l, and is almost constant with water depth. The $^{134}Cs/^{137}Cs$ activity ratio of all samples was nearly 1.0 within measurement error after the decay correction to March 11 in 2011. Therefore, a major part of radiocesium is derived from the Fukushima Daiichi NPP accident.

The higher radioactivity for the waters of Lake Akagi-Ohnuma and Lake Akagi-Konuma was probably attributed to the stability of cesium in lake water as dissolved forms [9-10] and the longer residence time (2.4 years) of lake waters [6]. The still high radioactivity of ^{137}Cs in waters was observed in closed lakes, with no marked surface inflow and outflow of water, in the Chernobyl exclusion zone [11].

Table 2 presents the range of ^{137}Cs radioactivity for the river waters in these areas (Fukuhsima, Miyagi, Ibaraki and Gunma prefectures, see Figure 1) before and after the Fukushima Daiichi NPP accident. Before the accident, the radioactivity ranges from 0.05–1.89 x 10^{-3} Bq/l [12-13] for the Tone River and the Kuji River, which originate from the southern end of mountainous region of Abukuma. The radioactivity increased to 0.01–0.19 Bq/l after the accident. These values are 2–3 orders higher than those before the accident. The differences in radioactivity of ^{137}Cs before and after the accident was probably attributed to the transport of radiocesium from the contaminated watershed.

Table 2. Radioactivity of ^{137}Cs in river waters before and after the Fukushima Daiichi NPP accident in Fukushima, Ibaraki and Gunma prefectures, Japan.

River	Date	^{137}Cs (Bq/l)	Reference
After the Fukushima Diichi NPP accident			
Abukuma	2011	0.041–0.120	This study
Niida	2011	0.189	This study
Uta/Same/Natsui	2011	0.011–0.037	This study
Tone/Karasu/Watarase	2011	0.008–0.013	This study
Before the Fukushima Daichi NPP accident			
Tone	1985–1987	(0.16–1.89) x 10^{-3}	Hirose et al. [12]
Kuji	1987–1989	(0.05–1.00) x 10^{-3}	Matsunaga et al. [13]

3.3. Existing forms of radiocesium

To ascertain the present circumstances and transport behavior of radiocesium, we analyzed existing forms of radiocesium in river waters using the filtration method described in *2.3 Measurements*. The percentage of particulate form of ^{137}Cs was estimated for the Fukushima river water samples. The particulate form of radiocesium was 89% for the Shirakawa sample upstream, 61–62% for Motomiya and Date midstream, 56% for Marumori, and 76% for Iwanuma downstream of the Abukuma River. The fraction of particulates varied 20 percentage points from upstream to downstream in the Abukuma River. The Uta and Niida rivers were, respectively, 58% and 80%.

Table 3 shows averaged percentages of particulate ^{137}Cs in river waters before and after the Fukushima Daiichi NPP accident. The percentage of particulate ^{137}Cs for the Fukushima rivers after the Fukushima Daiichi NPP accident became 20–30 percentage points higher than those of rivers before the accident [12-14]. Differences reported at the Pripyat River with 40–60% [15] and other river systems with 4–80% [16] after the Chernobyl accident, depending on water chemistry and composition, and on the concentration of suspended solids in river waters.

Table 3. Averaged percentages of ^{137}Cs radioactivity in particles to the total radioactivity for river waters.

River	Particulate ^{137}Cs (%)		No. of	Reference
	Rnage	Averaged valu	samples	
After the Fukushima Daiichi NPP accident				
Abukuma	56–89	71 ± 12	6	This study
Niida	80	80	1	This study
Uta	58	58	1	This study
Before the Fukushima Daiichi NPP accident				
Tone	11–36	21 ± 9	5	Hirose et al. [12]
Kuji	22–80	43 ± 19	8	Matsunaga et al. [13]
Pripyat/Sakhan	2–50	16 ± 20	5	Matsunaga et al. [14]

3.4. Transport of ^{134}Cs and ^{137}Cs in river systems

Figure 2 shows the distribution of ^{137}Cs in river waters from Fukushima and Gunma prefectures in December 2011. A highly contaminated area (>60 kBq/m^2) is the eastern area of Fukushima Prefecture, extending into the northern parts of Tochigi and Gunma prefectures, as shown in Figure 1(b). The distribution pattern of river water ^{137}Cs is consistent with the distribution of ^{134}Cs and ^{137}Cs accumulated on surface soil in Fukushima and Gunma prefectures [3]. The Niida River shows the highest ^{137}Cs radioactivity among the rivers, but the Niida River is a small watershed area. The lower Niida River and Uta River show less accumulation (<100 kBq/m^2) for radiocesium, but the upper Niida River shows accumulation of 1000–3000 kBq/m^2 (Figure 1(b)). Ueda et al. [17] reported that the ^{137}Cs radioactivity in river waters from the tributaries of upper Niida River was 0.25–0.49 Bq/l on November 21–22, 2011. These values are 1–3 times higher than the downstream Niida River in this study. Particulate forms of ^{137}Cs were 48 ± 10% for the upper Niida [17] and 80% for the lower Niida. These results suggest that radiocesium deposited on the surface is mainly exported as existing forms adsorbed with suspended particles, clay minerals [18], and organic matter [13] in the rivers.

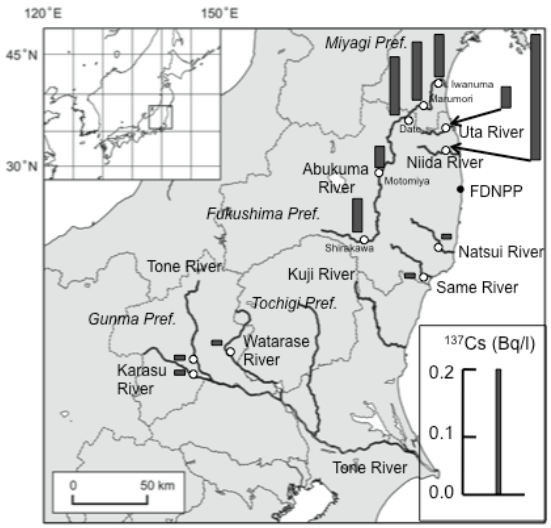

Figure 2. Distribution of ^{137}Cs radioactivity in the river waters from Fukushima and Gunma Prefecture, Japan on December 6–16, 2011. FDNPP = Fukushima Daiichi Nuclear Power Plant. The map data was used from Global Map Japan version 2 of the Geospatial Information Authority of Japan [19].

4. Conclusion

To assess secondary radioactive migration of ^{134}Cs and ^{137}Cs in river watersheds, we measured the radioactivity for river waters collected from the six river systems in Fukushima and Gunma Prefecture, Japan at nine months after the Fukushima Daiichi NPP accident. The radioactivity level of ^{137}Cs in the river waters at Fukushima Prefecture was 0.011 Bq/l to 0.189 Bq/l, and 0.008 Bq/l to 0.014 Bq/l for Gunma rivers. The radioactivity depends on the level of ^{134}Cs and ^{137}Cs contamination in the watershed. Particulate forms of ^{134}Cs and ^{137}Cs at normal flow conditions were 58–89% of total radioactivity on average of 71 ± 12%. Therefore, radiocesium deposited on the ground surface in these river watersheds was exported dominantly from watershed to river as particulate forms.

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