Export of ^{134}Cs and ^{137}Cs in the Fukushima river systems at heavy rains by Typhoon Roke in September 2011

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Abstract

Effects of a heavy rain event on radiocesium export were studied at stations on the Natsui River and the Same River in Fukushima Prefecture, Japan after Typhoon Roke during September 21–22, 2011, six months after the Fukushima Daiichi Nuclear Power Plant accident. Radioactivity of ^{134}Cs and ^{137}Cs in river waters was 0.011–0.098 Bq/l at normal flow conditions during July–September in 2011, but it increased to 0.85 Bq/l in high flow conditions by heavy rains occurring with the typhoon. The particulate fractions of ^{134}Cs and ^{137}Cs were 21–56% in the normal flow condition, but were close to 100% after the typhoon. These results indicate that the pulse input of radiocesium associated with suspended particles from land to coastal ocean occurred by the heavy rain event. Export flux of ^{134}Cs and ^{137}Cs by the heavy rain accounts for 30–50% of annual radiocesium flux in 2011. Results show that rain events are one factor controlling the transport and dispersion of radiocesium in river watersheds and coastal marine environments.

1 Introduction

A nuclear accident at the Fukushima Daiichi Nuclear Power Plant (NPP) occurred after the 2011 Tohoku earthquake and tsunami. About 15 PBq of both ^{134}Cs and ^{137}Cs was released from the NPP as a result of venting operations and hydrogen explosions (Japanese Government, 2011; Chino et al., 2011). Surface deposition of ^{134}Cs and ^{137}Cs reveals considerable external radioactivity in a zone extending northwest from the NPP, about 20 km wide and 50–70 km long inside the 80 km zone of the NPP (MEXT, 2011; Yoshida and Takahashi, 2012). Moderate radioactivity (100–600 kBq/m^2) was also found in the Naka-dori region. The deposition pattern is explained by emission rates of ^{134}Cs and ^{137}Cs coupled with wind direction and precipitation (Morino et al., 2011).

A major part of radiocesium deposited on the ground surface is present at the surface to 5 cm depth (MEXT, 2012a; Koarashi et al., 2012). Chemical extraction of ^{134}Cs and ^{137}Cs from selected soil samples has indicated that both radionuclides in the soil are only slightly water soluble. Even the fraction extracted with 1 M ammonium acetate was only approximately 10% (Matsunaga et al., 2013). However, ^{134}Cs and ^{137}Cs have been transported from contaminated watersheds to rivers in Fukushima Prefecture after the Fukushima Daiichi NPP accident (MEXT, 2012a; Sakaguchi et al., 2012). Similar outcomes were observed at the Pripyat and Dnieper Rivers in Ukraine after the Chernobyl accident in 1986 (IAEA, 2006a). The migration of ^{137}Cs has decreased markedly over time in river waters from Ukraine (UHMI, 2004; IAEA, 2006b) and Finland (Saxén and Ilus, 2001). The radioactivity of ^{137}Cs shows little change from upstream to downstream of the exclusion zone in the Pripyat River of the Chernobyl area (IAEA, 2006a). An increase in radioactivity

of ^{134}Cs and ^{137}Cs in river waters was also found in the Chernobyl area during a spring flood event (IAEA, 2006a, b) and in northwest Italy from a delayed release in summer during ice and snow melting in mountain areas (Spezzano et al., 1994). The transport of materials generally depends on watershed conditions such as vegetation, slope, soil types, and spring snow-melting. It is important to clarify migration behavior of radiocesium and its controlling factors for future prediction of its dispersion in Fukushima Prefecture, Japan.

To elucidate the short-term to long-term impacts of the Fukushima Daiichi NPP accident on ecosystems of river and coastal marine environments, the Japanese Government has been monitoring the radioactivity of ^{134}Cs and ^{137}Cs in river systems in Fukushima Prefecture (MEXT, 2012b). Japanese rivers have a short length, high riverbed slope, and high river regime coefficient, which is the ratio of maximum /minimum discharge (Suetsugu, 2005). Annual mean precipitation was high, 1718 mm during 1971–2000 because of *tsuyu* (the rainy season in Japan), typhoon and snow-melting events in spring (MLTI, 2012). Matsunaga et al. (1991) reported that the radioactivity of ^{137}Cs derived from fallout increased at high flow conditions in the Kuji River because of rain events. Nagano et al. (2003) pointed out that variations of suspended and dissolved form concentrations of elements in the Kuji River waters were formulated as a function of the water discharge rates. Therefore, it is important to evaluate effects of rain events on export of radiocesium from land to ocean in the Fukushima area.

This study was conducted to investigate the transport of ^{134}Cs and ^{137}Cs in river systems in Fukushima Prefecture after rain events. This report describes the monitoring results of

radioactivity of ^{134}Cs and ^{137}Cs in river waters at two rivers after the heavy rain event by a typhoon in September 2011. Field experiments were conducted at the Natsui River and the Same River in the southern part of Fukushima Prefecture, Japan. We discussed transport behavior of radiocesium and estimated its export flux from inland to coastal areas.

2 Materials and methods

Typhoon Roke (T1115) struck Japan on September 21 and subsequently weakened to an extra-strong tropical cyclone on September 22, 2011 (JMA, 2011). The typhoon precipitated more than 400 mm of rain daily in parts of eastern and western Japan (JMA, 2011). Fukushima Prefecture had rainfall of 100–200 mm during September 15–22. The daily rainfall on September 21 was 137 mm by the impact of Typhoon Roke at Iwaki city, located in the southern coastal region of Fukushima and in a watershed of the Natsui River. This value is about one-tenth of the annual mean rainfall (1409 mm for 1981–2010: JMA, 2012).

The sampling location is presented in Fig. 1. This study investigated Natsui River and Same River flowing through a less-contaminated area (<100 kBq/m^2: MEXT, 2011) to the Pacific coast. The Natsui River watershed area is 749 km^2. That of the Same River is 600 km^2. The Natsui River length is 67 km. That of the Same River is 58 km. The annual mean water discharge data in 2011 were 17.6 m^3/s for the Natsui River and 21.4 m^3/s for the Same River (Fukushima Prefectural Government, 2012). The water discharge data are presented in Fig. 2. River water samples (10–20 L) were collected at normal flow conditions on July 12 and 27, September 14, November 22, December 6, and September 22 at high-flow

conditions after Typhoon Roke last year. The sampling was conducted at the Iwaki-bashi Bridge of the Natsui River and the Eguri-Ohashi Bridge of the Same River.

In normal flow conditions on July 12 and September 14, dissolved and particulate forms of radiocesium were separated using cartridge filters with pore sizes of 10 μm, 1 μm, and 0.45 μm. In river waters after the heavy rain event with the typhoon, particles were separated using centrifugation and filtration with No. 5A (approximate pore size of 7 μm) filters and a pore size of 0.45 μm membrane filters. Filtration was conducted using No. 5A filters and then filtered with membrane filters. In this study, suspended solids using centrifugation are designated as "deposits". The suspended solids on the filters are designated as "No. 5A" for those filtered with No. 5A filters and "membrane" for those collected with 0.45 μm filters.

The radioactivity of ^{134}Cs and ^{137}Cs in the filtered river waters was measured as dissolved forms of radiocesium with gamma-ray spectrometry using ammonium molybdophosphate (AMP)/Cs compound method (Tanaka et al., 2006). The ^{134}Cs and ^{137}Cs were measured using gamma-ray spectrometry with a low BKG Ge detector at the Low Level Radioactivity Laboratory and the Ogoya Underground Laboratory of Kanazawa University for 1–3 days (Hamajima and Komura, 2010). The gamma-lines were used for the activity calculation 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. The decay correction of radioactive concentration for ^{134}Cs and ^{137}Cs was done at each sampling date. The deposited solids and suspended solids on the filters were also measured using gamma-ray spectrometry after drying them at room temperature.

The mineral composition of riverine suspended solids on the filters and of deposited solids was analyzed using X–ray diffraction (XRD). XRD analysis of powdered samples mounted in glass slides was conducted with an Ultima IV (Rigaku Co. Ltd.) diffractometer using Cu*K*α radiation operated at 40 kV and 30 mA from 2° to 65°. Suspended solids on GF/F and membrane filters were also mounted on glass slides and measured for clay mineral composition.

3 Results and discussion

3.1 Radioactivity of ^{134}Cs and ^{137}Cs

Radioactivity of ^{134}Cs and ^{137}Cs in the river waters is shown in Table 1 and Fig. 3. The respective radioactivity of ^{134}Cs and ^{137}Cs was 0.009 Bq/l – 0.089 Bq/l and 0.011 Bq/l – 0.098 Bq/l in the normal flow condition. The radioactivity decreased concomitantly with increasing time after the Fukushima Daiichi NPP accident. However, the radioactivity of ^{134}Cs and ^{137}Cs in the high flow condition after the typhoon was about one order higher than that at normal flow conditions, which indicates that high export of ^{134}Cs and ^{137}Cs occurred after the heavy rain event. The ^{134}Cs/^{137}Cs activity ratio for all samples is about 1.0, corrected to March 11, 2011, so that radiocesium derived from the Fukushima Daiichi NPP was transported from deposited surface to rivers. The highest radioactivity is 2–3 orders higher than that of the Japanese rivers collected in 1985–1988 (Hirose et al., 1980; Matsunaga et al., 1991).

3.2 Existing forms of ^{134}Cs and ^{137}Cs

Fig. 4 shows the total radioactivity of ^{137}Cs versus radioactivity of dissolved forms of ^{137}Cs. In a normal water flow condition, dissolved forms of ^{137}Cs were 0.011-0.064 Bq/l, but in the high flow condition, that is after the heavy rain event, dissolved ^{137}Cs was about 0.005 Bq/l. These results indicate that the heavy rain affects export of radiocesium deposited on the ground surface derived from the Fukushima Daiichi NPP accident. Similar results have been reported for Ukraine river systems after the Chernobyl accident (Matsunaga et al., 1998).

Table 2 presents percentages of particulate phase ^{137}Cs at normal and high flow conditions in the Natsui River and the Same River after the Fukushima Daiichi NPP accident, together with results for the Kuji River before the Fukushima Daiichi NPP accident. At normal flow conditions, the percentage of particulate ^{137}Cs is 21–56% on average of 41 ± 17%. The other rivers in central to northern Japan were 11–47% (Hirose et al., 1990; Matsunaga et al., 1991). The Pripyat River had approximately 40–60% of radiocesium in the particulate phase during the decade after the Chernobyl accident (Voitsekhovich et al., 1997). Heavy rains show that a major part of ^{137}Cs is present as particulate phase in the Natsui River and the Same River. The particulate form of ^{137}Cs is predominant in river waters after the rain event in the Kuji River (Matsunaga et al., 1991).

As Table 3 shows, radioactivity contents of suspended solids are about 2000 Bq/kg-dried suspended solids (ss). This value corresponds to that of river-bottom sediment (about 2000 Bq/kg: Ministry of the Environment, 2012) and soil in watershed (230–2400 Bq/kg: MEXT,

2012b). ^{134}Cs and ^{137}Cs of fine particle fraction trapped on the filters in the river waters also show higher radioactivity content of 1550–2640 Bq/ss-kg for the Natsui River and 7200–8660 Bq/kg-ss for the Same River. Therefore, radiocesium associated with suspended solids is an important pathway from land to coastal areas.

Fig. 5 shows X-ray diffraction analysis for the suspended solids filtered with Advantec No. 5A filters, membrane filters with a pore size of 0.45 μm, and deposited suspended solids. Blank GF/F and membrane filters have their own characteristics such as a broad peak between 13 and 35 ˚2θ and three peaks of 13–25 degree 2θ, respectively. All samples include clay minerals such as chlorite, mica, and/or kaolin. Clay mineral type in fixation of radiocesium has been widely claimed (e.g., Facchinelli et al., 2001; Korobova et al., 2007). For example, an increase in ^{137}Cs specific activity was observed in floodplain soil with increased smectite content in clay fractions (Korobova et al., 2007). Selective sorption of ^{137}Cs has been reported for illite and mica at laboratory experimental systems (Brouwer et al., 1983; Staunton and Roubaud, 1997). Therefore, clay minerals in suspended solids from the Natsui River and the Same River appear to have fixation and/or association ability for ^{134}Cs and ^{137}Cs.

3.3 Migration behavior of ^{134}Cs and ^{137}Cs in river systems

Distribution coefficient (Kd) between suspended solids and river water is defined as

$$K\mathrm{d} = C_{\mathrm{solid}}/C_{\mathrm{dissolved}}\ , \qquad (1)$$

where C_{solid} and $C_{dissolved}$ respectively denote the ^{137}Cs concentrations in the suspended solids (Bq/g) and dissolved phase (Bq/ml). The fate and bioavailability depend strongly on the K_d and strength of the particle-contaminant association. Estimation of K_d values was conducted using measurement data presented in Tables 1 and 3. The K_d is 0.43–0.55 × 10^6 ml/g for the Natsui River and 4.1–5.0 × 10^6 ml/g for the Same River. These values are 1–2 orders higher than those of other Japanese rivers such as the Tone River and the Ishikari River (Hirose et al., 1990) and the Kuji River (Matsunaga et al., 1991) before the Fukushima Daiichi NPP accident. The K_d values of the Fukushima rivers are two orders higher than that of Ukraine after the Chernobyl accident (Matsunaga et al., 1998), which is regarded as supplying suspended solids from the watershed and resuspension of river bottom sediments by rain events.

The cumulative ^{134}Cs and ^{137}Cs inventory from the surface down to depth in undisturbed soils in Fukushima Prefecture confirms that >90% of the total ^{134}Cs and ^{137}Cs in the soil profile was found within the upper 5 cm layer at the cropland and grassland sites (Koarashi et al., 2012). The surface erosion processes in watershed have been studied using ^{137}Cs derived from fallout as a tracer of suspended solids. Surface runoff generally does not occur in forested areas, but unmanaged Japanese cypress plantations often have little surface cover, and surface runoff is generated during large rainstorms (Miyata et al., 2007; Gomi et al., 2008). Fukuyama et al. (2010) have shown that, for different stand species, surface coverage and forest management practices affect the runoff of the surface-derived suspended solids at the catchment scale. To understand the effects of input of suspended solids on K_d value, we compared the K_d values after heavy rain events due to typhoon in

September 2011 and July 2012. The suspended solids concentration was 0.20–0.41 g L^{-1} for the September samples and 0.26–0.34 g L^{-1} (Nagao unpublished data) for the July samples. The dissolved radiocesium concentration was almost constant, 0.0025–0.0046 Bq L^{-1} in both sampling date. The K_d value of ^{134}Cs and ^{137}Cs for the July samples is 0.20–0.43 x 10^6 ml g^{-1} and still high, though the values are 1/2–1/10 lower than that of 2011 September samples. These results suggest that the sources of particulate forms of radiocesium are important factors controlling K_d values at the second dispersion after the nuclear accident. Direct input of suspended solids eroded from the ground surface may be reflected in the higher values found for the Natsui River and the Same River after the heavy rain at the initial stage of the Fukushima Daiichi NPP accident.

The radioactivity of ^{134}Cs and ^{137}Cs in the surface soil of riverside at Samegawa-Ohashi in the downstream of Same River ranged from 22 to 970 Bq kg^{-1} during September 2011–November 2012 (Ministry of Environment in Japan, 2012). The radioactivity at Idosawa-Ohashi in the upper downstream was from 70 to 900 Bq kg^{-1} during September 2011–July 2012, but 1500-7100 Bq kg^{-1} on September 2 and November 9 in 2012 at high water level condition. The results suggest that ^{134}Cs and ^{137}Cs were deposited on the ground surface in the riverside due to the effects of rain events.

The radioactive content of ^{134}Cs and ^{137}Cs deposited in river bottom sediments was approx. 770 Bq/kg-sediment for the Natsui River and approx. 2000 Bq/kg for the Same River during May-July 2011, but 10-250 Bq kg^{-1} during September 2011-November 2012 (Ministry of the Environment, 2012). The radioactivity varied with sampling, although the samplings

were conducted at fixed stations in each river. The river bottom sediments are sandy, so the apparent residence time of fine particles might be short in the Natsui River and the Same River. Therefore, the contaminated area around the river basin and river bottom sediments plays an important role as sources of particulate phase of ^{134}Cs and ^{137}Cs in river waters.

Fig. 6 presents a schematic illustration showing the export of radiocesium from the watershed to a river. Erosion of riverbank and the ground surfaces, and re-suspension of river bottom sediments occurs at rain events so that the amount of suspended solids increases and the radioactivity of ^{134}Cs and ^{137}Cs associated with riverine suspended solids also increases. Similar results have been reported for the Kuji River, running through Ibaraki and Fukushima prefectures (Matsunaga et al. 1998). Increased erosion and radioactivity of ^{137}Cs derived from global fallout have also been observed at the Kuzuryuu River in 2009 after rain events (Nagao, unpublished data).

3.4 Estimation of radiocesium flux from land to ocean

To understand the impacts of heavy rains on the transport of radiocesium and its dispersion to coastal ocean, flux of radiocesium from land through river to ocean was estimated simply based on our measurements, as shown in Table 1 and from water discharge data (Fukushima Prefectural Government, 2011). In the Natsui River, the averaged radioactivity was 0.0493 ± 0.0276 Bq/l for ^{134}Cs and 0.0566 ± 0.0315 Bq/l for ^{137}Cs, except for the sample taken after the heavy rain. Mean monthly water discharge was 17.6 ± 30.8 m^3/s during March–December 2011. The fluxes (F) of ^{134}Cs and ^{137}Cs are defined as

$$F = C \times WD \times T, \quad (2)$$

where C stands for the mean radioactivity of ^{137}Cs except for the heavy rain sample, WD signifies the mean monthly water discharge, and T denotes the period of March 11 – December 31 in 2011. The fluxes of radiocesium from land to ocean are estimated as 2.3 ± 4.2 Bq/305 days for ^{134}Cs and 2.6 ± 4.8 × 10^{10} Bq/305 days for ^{137}Cs. The radioactivity of ^{137}Cs during January 1 – March 10 might be similar to that before the Fukushima Daiichi NPP accident and 2–3 order lower values. Therefore, the estimated value corresponds to the annual export flux.

Regarding the heavy rain event, a mean daily water discharge was 350 ± 264 m^3/s calculated from hourly water discharge on September 22, 2011. The radioactivity levels of ^{134}Cs and ^{137}Cs were, respectively, 0.673 ± 0.004 Bq/l and 0.853 ± 0.004 Bq/l. Therefore the fluxes from river to ocean were 2.0 ± 1.5 × 10^{10} Bq/day for ^{134}Cs and 2.6 ± 1.9 × 10^{10} Bq/day for ^{137}Cs on September 22, 2011. This value, which corresponds to that in March–December 2011, represents a rough estimation because of a lack of consideration for the six rain events with precipitation greater than 50 mm monitored at the Onahama site (JMA, 2012). However, we can understand the importance of rain events on transport of radiocesium from inland to the ocean in a year.

Table 4 shows flux data of ^{137}Cs for the Natsui River and the Same River together with other river systems. The flux at the Same River was estimated using a similar simple

method in this study. As Table 4 shows, the export flux in Fukushima rivers in 2011 is one order higher than that of the Tone River in 1985–1986 and the Kuji River in 1988–1989. The heavy rain event by Typhoon Roke on September 22 occupied 30–50% of the annual export flux. These results indicate that the heavy rain event is one factor determining the export of ^{134}Cs and ^{137}Cs in the Natsui River and the Same River.

4 Summary

To realize transport of ^{134}Cs and ^{137}Cs from land to ocean after the Fukushima Daiichi NPP accident, river research was performed at the Natsui River and the Same River running through a contaminated watershed in Fukushima Prefecture during July–December, 2011. The radiocesium concentration was measured for river water samples at a normal flow condition and a high flow condition by the heavy rain event, daily precipitation of 137 mm, with Typhoon Roke in September 2011. The ^{134}Cs and ^{137}Cs radioactivity in a normal flow condition was 0.009–0.098 Bq/l, but at a high flow condition, it was 0.36–0.85 Bq/l. Particulate phase of ^{134}Cs and ^{137}Cs was 21–56% in a normal flow condition and ca. 100% in a high flow condition. Therefore, radiocesium associated with suspended solids is exported in large quantities from river to coastal ocean by heavy rain. Export of ^{134}Cs and ^{137}Cs after the heavy rain event is estimated as roughly 2.0–2.6 × 10^{10} Bq/day in the Natsui River and 0.74–0.87 × 10^{10} Bq/day in the Same River. These values account for 30–50% of the export of radiocesium for the 10 months of March 11– December 31 in 2011 in both rivers. Therefore, results show that pulse input by heavy rain events is one important pathway of radiocesium from inland to coastal ocean areas in the southern part of the

Fukushima Prefecture, Japan.

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Table 1 Water quality and radioactivity of ^{134}Cs and ^{137}Cs in water samples from the Natsui River and the Same River

Sampling date	pH	WT (°C)	^{134}C (×10^{-3}Bq/l)	^{137}Cs (×10^{-3} Bq/l)	^{134}Cs/^{137}Cs*

Natsui River					
2011/7/12	7.6	29.5	49.0 ± 1.3	52.0 ± 2.5	1.06 ± 0.06
2011/7/27	7.4	26.0	89.0 ± 1.6	98.0 ± 2.5	1.02 ± 0.03
2011/9/13	7.3	25.5	25.2 ± 1.2	26.0 ± 1.2	1.14 ± 0.07
2011/9/22	7.8	18.6	673.0 ± 4.0	853.0 ± 4.0	1.00 ± 0.01
2011/11/24	7.6	10.9	61.5 ± 1.8	78.7 ± 1.2	0.97 ± 0.03
2011/12/6	7.4	6.5	22.0 ± 1.4	27.7 ± 1.6	1.04 ± 0.06
Same River					
2011/7/12	7.5	25.4	74.5 ± 2.4	81.0 ± 2.2	1.03 ± 0.04
2011/7/27	7.5	23.4	47.5 ± 5.9	52.0 ± 5.7	0.99 ± 0.16
2011/9/13	7.8	22.9	15.9 ± 1.3	18.9 ± 1.2	1.00 ± 0.10
2011/9/22	7.9	18.6	360.0 ± 3.0	424.0 ± 3.0	1.01 ± 0.01
2011/11/24	7.4	11.8	13.2 ± 0.7	15.3 ± 0.6	1.00 ± 0.05
2011/12/6	7.5	8.5	8.9 ± 1.0	11.4 ± 1.5	1.10 ± 0.13

*Decay correction of radioactive concentration for ^{134}Cs and ^{137}Cs was done on March 11 in 2011.

Table 2 Percentage of ^{134}Cs and ^{137}Cs associated with suspended solids in the Natsui and Same River waters at low and high flow conditions

River	Flow Condition	^{137}C ($\times 10^{-3}$Bq/l)	Particulate ^{137}Cs (%)	No. sample	Reference

Natsui[1]	High	49.0 ± 1.3	~100	1	This study
	Normal	89.5 ± 1.6	40 ± 17	2	This study
Same[1]	High	25.2 ± 1.2	99	1	This study
	Normal	673.0 ± 4.0	26 ± 5	2	This study
Kuji[2]	High	1.0*	77	1	Matsunaga et al. (1991)
	Normal	0.12 ± 0.06	41 ± 17	12	Matsunaga et al. (1991)
Kitakami		0.14	39	1	Hirose et al. (1990)
Tone		0.77 ± 0.59	21 ±9	6	Hirose et al. (1990)
Ishikari		0.37 ± 0.12	47 ± 31	2	Hirose et al. (1990)
Kuziryu		1.17	11	1	Hirose et al. (1990)

[1] Water discharge was 238–350 m^3/s in a high flow condition and 11.0–23.7 m^3/s in a normal flow condition.

[2] Water discharge was 75.4 m^3/s in a high flow condition and 8.1–59.4 m^3/s in a normal flow condition.

* Preceding precipitation was above 30 mm/day.

Table 3 Radioactivity of ^{134}Cs and ^{137}Cs of suspended solids in water samples from the Natsui River and the Same River collected on September 22, 2011 after Typhoon Roke

Sample	Suspended solid (g)	^{134}C (Bq/kg-ss)	^{137}Cs (Bq/kg-ss)	$^{134}Cs/^{137}Cs$*

Natsui River				
Deposit	6.88	1980 ± 11	2336 ± 11	1.00 ± 0.01
No.5A	0.096	1548 ± 58	1817 ± 64	1.03 ± 0.04
Membrane	0.362	1934 ± 70	2303 ± 76	0.97 ± 0.04
Same River				
Deposit	3.42	1569 ± 14	1865 ± 14	1.01 ± 0.01
No.5A	0.062	7176 ± 113	8268 ± 127	1.03 ± 0.02
Membrane	0.182	7691 ± 81	8649 ± 93	1.03 ± 0.01

ss = suspended solids in river water.

* Decay correction of radioactive concentration for ^{134}Cs and ^{137}Cs was done on March 11, 2011.

Table 4 Export fluxes of ^{137}Cs from land to ocean in the Natsui, Same, Kuji and Tone Rivers

River	Catchment area	Export flux of ^{137}C		Contribution of heavy rain	Year	Major origin
		Annual	Heavy rain			

	(km^2)	($\times 10^{10}$ Bq/yr)	($\times 10^{10}$ Bq/day)	(%)		of ^{137}Cs
Natsui	749	5.2 ± 1.3	2.6 ± 1.9	50	2011	NA
Same	600	2.9 ± 1.2	0.87 ± 0.92	30	2011	NA
Kuji*	1490	0.26	—	—	1988	GF
Tone**	16840	0.50 ± 0.38	—	—	1985	GF

NA = Nuclear accident, GF = Global fallout.

* Flux estimates from ^{137}Cs annual load (Matsunaga et al., 1991) and watershed area of the Kuji River.

** Flux estimates from the average radioactivity of ^{137}Cs (Hirose et al., 1990) and mean annual water discharge in 1985 (MLIT, 2012).

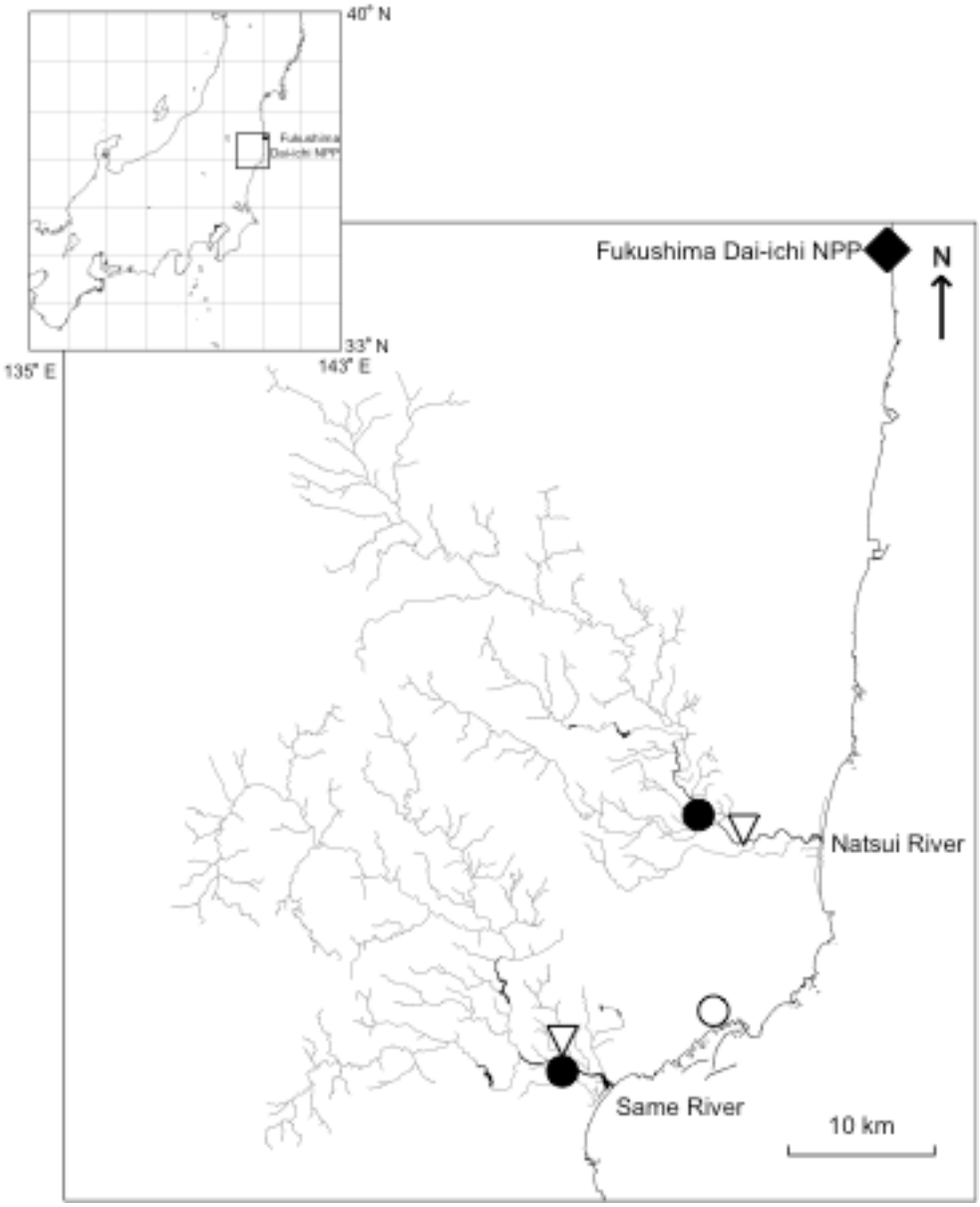

Figure 1 Sampling location of the Natsui River and the Same River. Closed circles represent sampling stations. Open circles show a monitoring station at Onahama for precipitation. Open inverted triangles represent water level observatory sites.

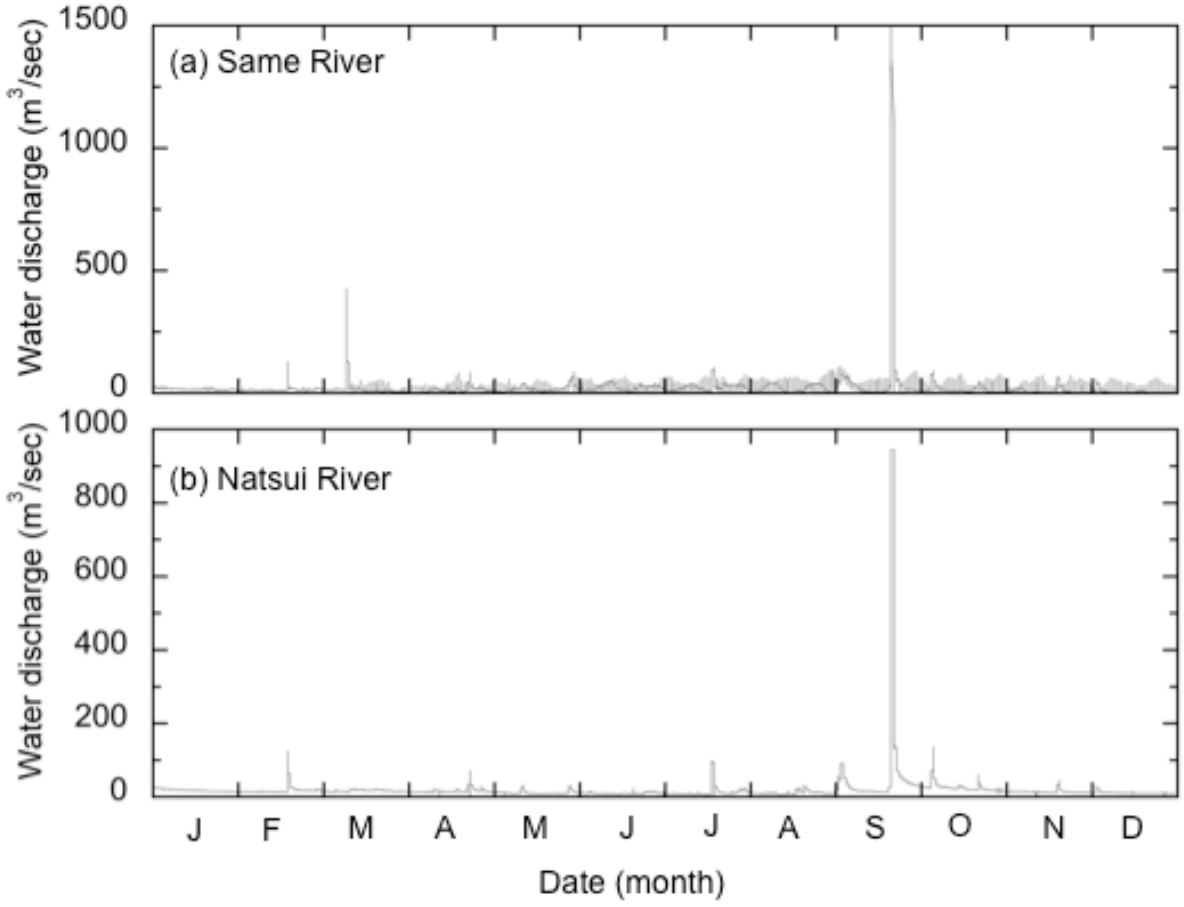

Figure 2 Water discharge of the Natsui River and the Same River in 2011. Data were referred from the Fukushima Prefectural Government.

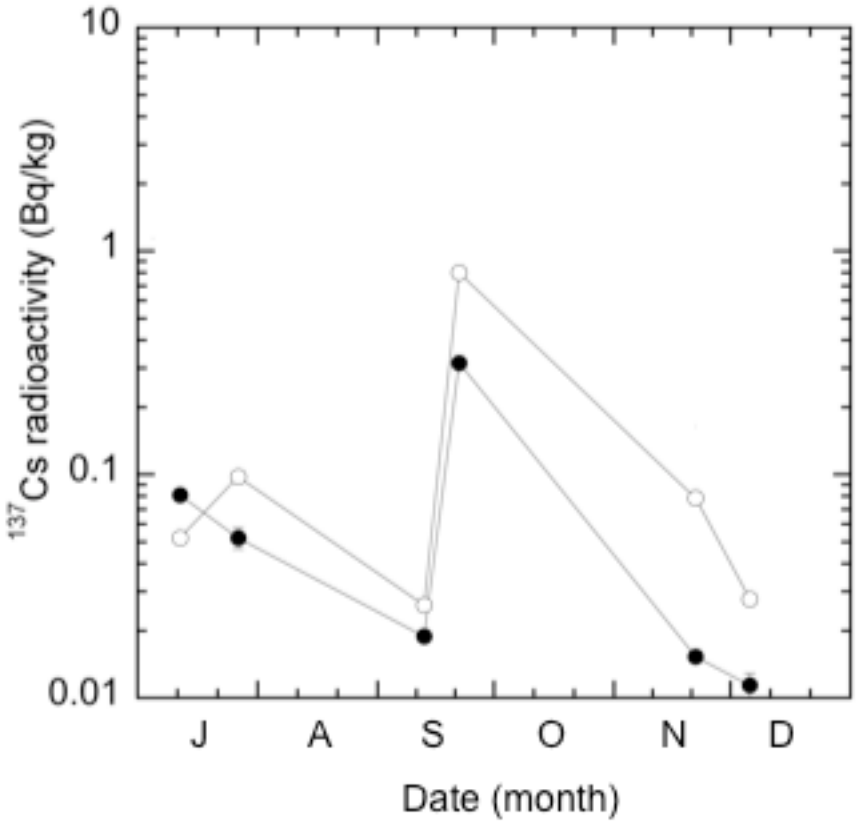

Figure 3 Radioactivity of ^{137}Cs in water samples from the Natsui River (○) and the Same River (●) during July–December 2011.

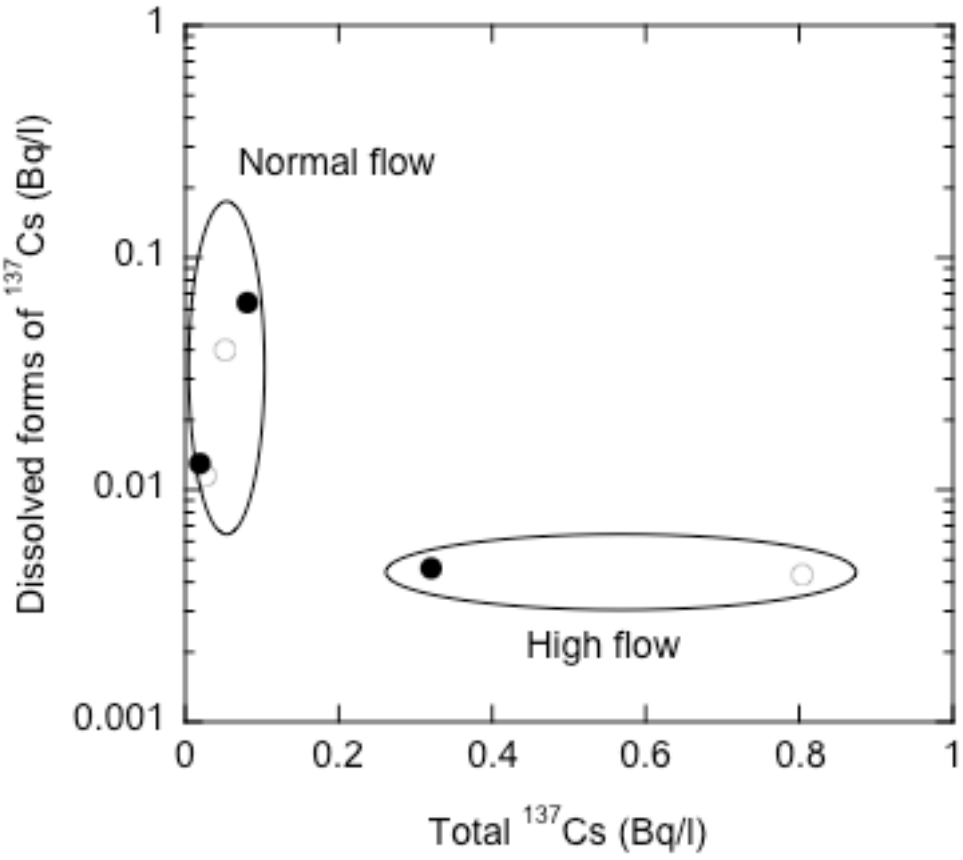

Figure 4 Total and dissolved radioactivity of ^{137}Cs in river waters from the Natsui River (○) and the Same River (●).

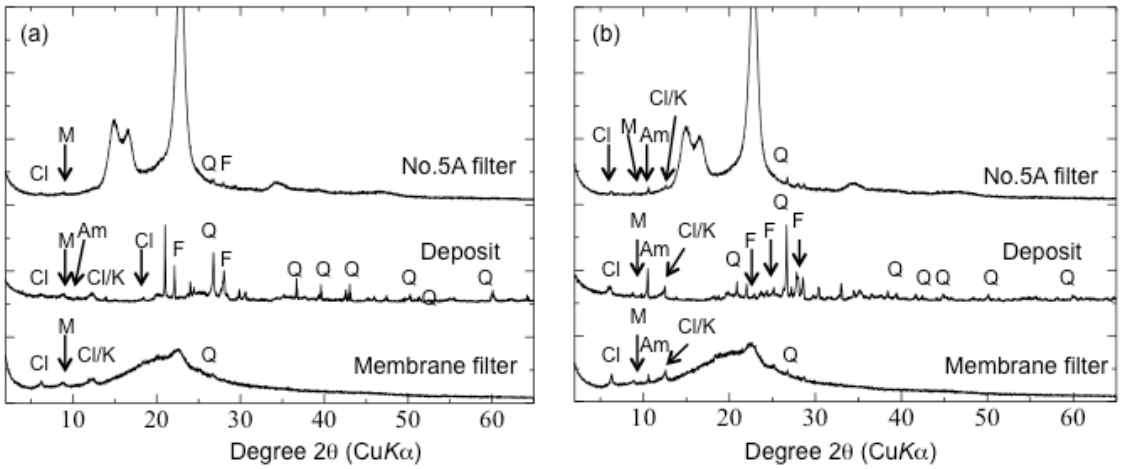

Figure 5 X-ray diffraction patterns of suspended solids in the Natsui River (a) and Same River (b) after the heavy rain event by Typhoon Roke in 2011.

Legend for abbreviations of minerals is as follows: Cl, Chlorite; M, Mica; Am, amphibole; F, feldspar; Q, quartz; Cl/K, chlorite and/or kaolin.

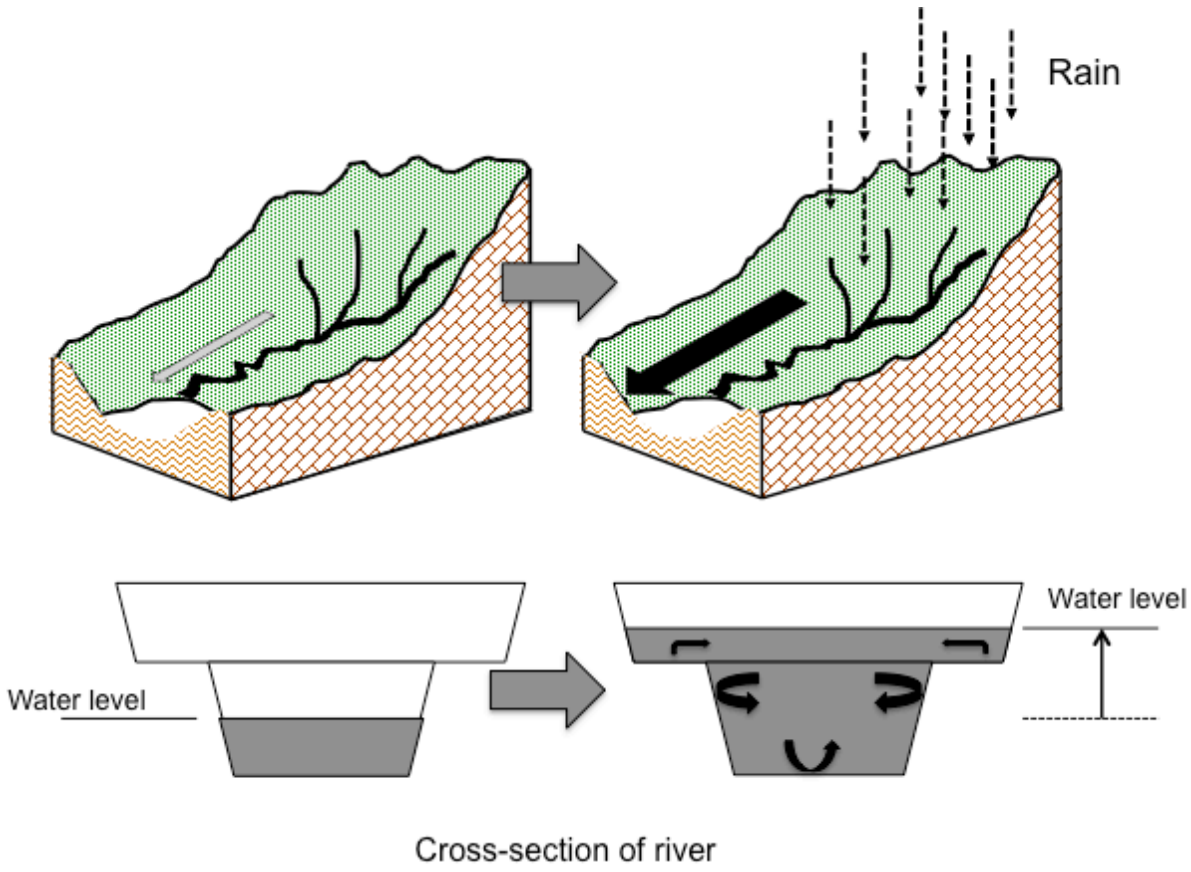

Figure 6 Schematic illustrations of export of ^{134}Cs and ^{137}Cs from watersheds to rivers.