Concentration of radiocesium in stream water from a mountainous catchment area during rainfall events

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Terrestrial and aquatic systems were contaminated with radioactive materials following the nuclear accident at Fukushima Daiichi Nuclear Power Station on 11 March, 2011. It is important that levels of radiocesium (Cs) in stream water from affected areas be monitored as this water is used for paddy irrigation and domestic water. Additionally, soil particles and organic matter from the streams are deposited in rivers, estuaries and into the ocean. Predictions suggest that Cs levels will increase during intense rainfall-runoff events. To check this prediction, we monitored temporal changes in runoff events and Cs levels in stream water from a mountainous catchment area northwest of the Fukushima plant. In March and April, 2012, the concentrations of Cs and suspended solids (SS) in stream water taken from low-level water flow were found to be 0.2–0.3 Bq/L and 2–7 mg/L, respectively. A heavy rainfall event in July 2012 resulted in an increase and subsequent decrease of both the runoff volume and SS concentration. At the beginning of the rainfall event the concentration of Cs absorbed in the SS was measured to be 23 Bq/L, this decreased gradually to 0.3 Bq/L over the course of the event. The concentration of Cs dissolved in the water was 0.1 Bq/L, this decreased only slightly during the runoff event. During a low rainfall event in September 2012 the concentration of Cs absorbed in the SS at the beginning of the rainfall event was found to be 15 Bq/L, this decreased gradually to 0.5 Bq/L as the amount of SS in the water decreased. The concentration of Cs dissolved in the water was 0.2 Bq/L, again this decreased only slightly over the course of the runoff event. The Cs levels in stream water, during rainfall-runoff events, were primary influenced by the concentration of SS. The amount of Cs dissolved in the water, on the other hand, was roughly constant at 0.1–0.2 Bq/L. The results of this study indicate that, although the concentration of Cs in stream water is below the critical value of 10 Bq/L, monitoring Cs levels is important to ensure that irrigation water and domestic water remain safe.

Key Words: *Radiocesium, stream water, suspended solids, dissolved cesium, Fukushima*

1. Introduction

Contamination of terrestrial and aquatic systems by radioactive materials occurred following the nuclear accident at Fukushima Daiichi Nuclear Power Station in 2011. The accident was caused by a tsunami, off the

Pacific coast of Japan, induced by the Tohoku Earthquake on 11 March 2011. In 2010, the percentage of forest and agricultural land in the Fukushima prefecture was 70 % and 11 %, respectively[1]. Rice-paddies accounted for 64 % of the agricultural land with an acreage of 77,728 ha[2]. The paddies main irrigation source is stream water from local mountainous areas. River water is also used for domestic purposes. Radiocesium (Cs) in rivers and streams could influence the entire aquatic ecological system in the areas through which they flow. It is therefore important that the concentration of Cs be monitored in water catchment areas local to the power plant. The amount of Cs found in runoff events is predicted to increase during intense rainfall. Here we studied the Cs levels during rainfall events in a small catchment area located in a mountainous region that was affected by the nuclear accident. Burrough *et al.*[3] showed that during flood events the drainage of the Cs isotope ^{137}Cs intensifies; increased transport of dissolved and adsorbed ^{137}Cs was observed in the Pripyat catchment near Chernobyl Nuclear Power Plant in Belarus and Ukraine. Hilton *et al.*[4] studied the release of dissolved ^{137}Cs from a catchment area into a small lake (Devoke Water) in the Cumbria, UK due to a small amount of illite, which tightly bind Cs, in most catchments and a presence of organic molecules which prevent the development of irreversible sorption. In this study, we measured the levels of ^{134}Cs and ^{137}Cs adsorbed in suspended solids (SS) and the levels of ^{134}Cs and ^{137}Cs dissolved in the stream water.

2. Materials and Methods

The temporal changes in runoff water levels and the concentration of Cs were monitored in stream water from a catchment area northwest of Fukushima Daiichi Nuclear Power Station. Measurements were started in March 2012. The catchment area is 0.06 km^2 in size and contains forested land (90%) and abandoned agricultural fields (10%). The main tree species in the forest is Japanese cedar. The air radiation dose rate in the town which contains the catchment area was 0.4–4 μSv/h at a height 1 m above the soil according to a report published by the municipal government on September 8, 2012.

The amount of water flowing in the stream was estimated by measuring the water level at the outlet of a

Table 1. Concentration of Cs in stream water during low-level water flow events in 2012.

	March 10	March 28	April 17
Discharge (mm/10 min)	0.015	0.018	0.012
^{134}Cs (Bq/L)	0.13	0.06	0.089
^{137}Cs (Bq/L)	0.19	0.09	0.16
Total Cs (Bq/L)	0.32	0.15	0.25
SS (mg/L)	5.4	7.3	2.0

Table 2. Hydrological characteristics and concentrations of Cs in stream water during three rainfall events in 2012.

		Event 1	Event 2	Event 3
		July 23–25	September 4	September 12
Rainfall (mm)		71.0	37.4	17.6
Duration of rainfall event (h)		2.3	4.3	1.2
Rainfall intensity (mm/h)		28.4	8.6	15.1
Discharge during an event (mm)		16.3	6.6	1.6
Peak discharge (mm/10 min)		0.57	0.43	0.078
SS (mg/L)	Immediately after the start of rainfall	1100	3400	960
	↓	Max. 6890	Max. 36,000	↓
	After rainfall event	20	340	30
Concentration of Cs absorbed by SS (Bq/L)	Immediately after the start of rainfall	23	52	15
	After rainfall event	0.3	1.1	0.5
Concentration of Cs dissolved in water (Bq/L)	Immediately after the start of rainfall	0.13	0.24	0.20
	After rainfall event	0.10	0.21	0.16

small rectangular concrete settling pond. The settling pond was set up at the end of the catchment area and was connected to a concrete water channel. The water level was recorded at 10-min intervals using a water-level gauge (WT-HR, Sencom Inc., Saitama, Japan). Stream water at the end of the catchment area was manually sampled on a monthly basis. When the water level exceeded a threshold level of 2cm automatically sampling was carried out at 2-h intervals using an automatic water sampler (6712, ISCO, Inc., NE). This occurred during rainfall events. The concentration of suspended solids (SS) was measured by filtering sampled water through 0.45 μm membrane filters. The concentration of the Cs isotopes ^{134}Cs and ^{137}Cs in the suspended solid residue was measured using gamma-ray spectrometry with a Ge semiconductor detector (SEG-EMS GEM20-70, Seiko EG&G Co. Ltd., Tokyo, Japan). The amount of Cs adsorbed by the SS in sampled water was calculated from the concentration of the residue. The concentration of the filtrate (dissolved Cs) was measured after evaporation of the sampled water by heating. The precipitation level was measured using a tipping bucket type rain gauge in the immediate proximity of the water sampling site.

3. Results

(1) Low-level water flow

The concentration of Cs in stream water during low-level water flow from March to April 2012 is shown in Table 1. The total Cs concentration is 0.2–0.3 Bq/L and the SS concentration is 2–7 mg/L. The water quality standard for tap water is a total Cs concentration of 10 Bq/L. The concentration of Cs in stream water during the low-level water flow was much lower than this standard.

(2) Rainfall events

Table 2 shows the hydrological characteristics of monitored rainfall events and the corresponding changes in the Cs concentration in the stream water. Figure 1 shows the temporal changes in SS, the concentration of Cs absorbed by the SS, and the concentration of Cs dissolved in the stream water during the three rainfall events. Also included are the rainfall and runoff discharge levels.

During heavy rainfall events, the peak concentration of SS occurs at roughly the same time as water flow is

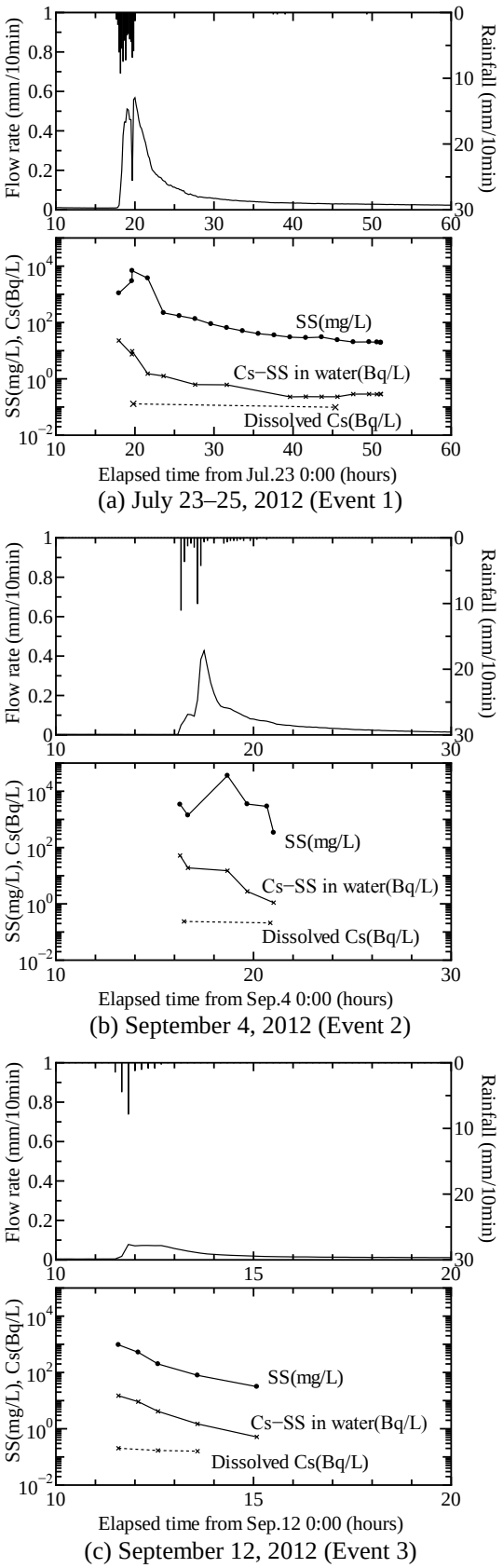

Figure 1. Concentration of SS, radioactive Cs absorbed by SS (Cs-SS in water) and dissolved radioactive Cs (Dissolved Cs) in stream water during rainfall events.

seen to peak. The maximum SS concentration measured on July 23–25, 2012 after 71.0 mm of rainfall (rainfall duration = 2.3 h) is 6,890 mg/L and on September 4, 2012 after 37.4 mm of rainfall (rainfall duration = 4.3 h) is 36,000 mg/L. The concentration of Cs absorbed by the SS at the beginning of event 1 is 23 Bq/L. This decreases gradually to 0.3 Bq/L over the duration of the event. For event 2, the initial concentration is 52 Bq/L, decreasing to 1.1 Bq/L. The concentration of dissolved Cs is 0.10–0.13 Bq/L and 0.21–0.24 Bq/L for events 1 and 2, respectively. A small decrease in the concentration of dissolved Cs is seen over the course of each event.

During event 3 (a low rainfall event) the concentration of Cs absorbed by the SS decreases from 15 Bq/L at the beginning of the event to 0.5 Bq/L at the end. The concentration of Cs dissolved in the water is 0.16–0.20 Bq/L. This is a similar concentration to that observed in the heavy rainfall events (events 1 and 2).

4. Discussion

The concentration of Cs in stream water is thought to be dependent on the amount of soil particles suspended in the stream water flow. The concentration of SS is related to the runoff discharge for high rainfall events but not for low rainfall events. Some clay minerals strongly absorb Cs; the reported Cs distribution coefficients of vermiculite, biotite, and illite are high.[5] Specifically, Cs can adsorb onto the frayed edges of vermiculite and illite.[6] The results suggest that small, Cs absorbing, clay minerals are suspended in stream water at the beginning and end of each rainfall event. The concentration of SS increases with increasing discharge. Since larger particles can be suspended by the higher flow rates the composition of SS is dominated by soil particles of large size (sand and silt). The concentration of Cs within the SS is thus seen to decrease during a rainfall event.

The results show that Cs is dissolved in stream water, however, only at low levels. Burrough *et al*.[3] showed that ^{137}Cs is highly mobile in both river water and poorly drained organic soils. Leaf litter and organic soil are both found in the flow path of streams. It is however unclear if these materials are the source of the dissolved Cs found in stream water. Investigations to identify the source of the dissolved radioactive materials are currently being conducted.

5. Conclusions

The majority of Cs found in stream water, during rainfall events, was seen to arise from the presence of suspended solids. The concentration of suspended solids was dependent on the composition of suspended solids, which changes during the rainfall events. Dissolved Cs, which can be easily absorbed by crops, was present in the stream water at concentrations of 0.1–0.2 Bq/L. This result makes it necessary to clarify the origins of dissolved Cs.

Although the concentration of Cs in stream water is below the critical concentration, it is important that the levels of Cs in stream water be monitored to ensure that irrigation water and domestic water remain safe.

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