Chapter 6
Migration Behavior of Particulate ^{129}I in the Niida River System

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Abstract This study investigates the source and flux of particulate ^{129}I in the downstream reaches of the Niida River system in Fukushima. The upper watershed is a relatively highly contaminated zone located 30–40 km northwest of the Fukushima Daiichi Nuclear Power Plant. Samples of total suspended substance (SS) were collected continuously at Haramachi (5.5 km upstream from the river mouth) from December 2012 to January 2014 using a time-integrative SS sampler. Activity of ^{129}I and the ^{129}I/^{127}I ratio in SS were 0.9–4.1 mBq kg^{-1} and (2.5–4.4) × 10^{-8}, respectively, and were strongly correlated with the total dry weight of SS samples with R^2 of 0.79–0.88. High SS ^{129}I activity and ^{129}I/^{127}I ratios were found in March, April, September, and October 2013. SS ^{129}I activity and ^{129}I/^{127}I ratios are considered to reflect the SS source, i.e., the more contaminated upper watershed or the less contaminated downstream area. The flux of particulate ^{129}I at the Haramachi site was estimated to be 7.6–9.0 kBq month^{-1} during September–October 2013. A relatively high amount of particulate ^{129}I may have been transported from the upstream to the downstream reaches of the Niida River by high rainfall over this period.

Keywords River system • Suspended substance • ^{129}I activity • ^{129}I/^{127}I ratio • Rain event • Migration behavior • Accelerator mass spectrometry

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6.1 Introduction

The nuclear accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP), Japan, resulted in the massive release of high-volatility fission products to the environment, including ^{129}I ($T_{1/2} = 15.7$ million years) and ^{131}I ($T_{1/2} = 8.02$ days). The total amount of radionuclides discharged into the atmosphere was estimated to be 8.1 GBq for ^{129}I [1] and 120–150 PBq for ^{131}I [2, 3]. Approximately 13 % of the total amount of released ^{131}I was deposited over Japan via radioactive plumes [4]. Any short-lived ^{131}I deposited in the soil decays after a few months, however, long-lived ^{129}I derived from the FDNPP accident must be traced from land to the marine environment via river systems owing to its relatively high fission yield, high chemical reactivity, biological concentration in the marine ecosystem, and affinity for the thyroid gland although it is less radiologically harmful than ^{131}I.

This study aims to elucidate the source and flux of particulate ^{129}I in the downstream reaches of the Niida River, a small river in Fukushima Prefecture with an upper watershed located in a relatively high-contamination zone 30–40 km northwest of the FDNPP. We investigated temporal changes in ^{129}I activity and ^{129}I/^{127}I ratios in total suspended substance (SS) collected during December 2012–January 2014 at Haramachi on the Niida River. Particular attention is given to quantifying the monthly flux of particulate ^{129}I in the downstream reaches of the Niida River system.

6.2 Materials and Methods

The Niida River system in northeastern Fukushima Prefecture has a 78-km-long watershed with an area of 585 km^2 [5]. The upper watershed is located 30–40 km northwest of the FDNPP and is affected by relatively high levels of radioactive contamination, with ^{137}Cs inventory of >300 kBq m^{-2} (Fig. 6.1) [6]. In contrast,

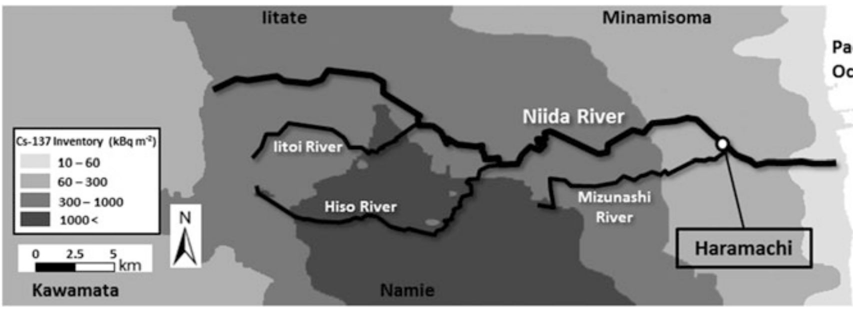

Fig. 6.1 Map showing the sampling site (Haramachi) located 5.5 km upstream from the Niida river mouth along with the spatial ^{137}Cs inventory from MEXT [6]. Total SS was collected continuously from December 2012 to January 2014 using a time-integrative SS sampler

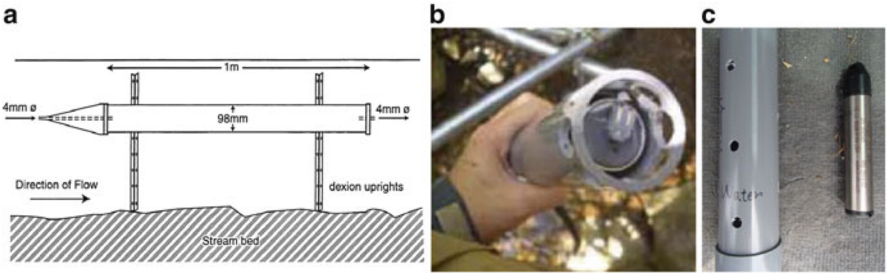

Fig. 6.2 (**a**) Schematic diagram of the time-integrative SS sampler. (**b**) Photographs of the tuner turbidity sensor, and (**c**) water level sensor for measuring discharge

the downstream reaches are characterized by low levels of contamination, with ^{137}Cs inventories of 10–300 kBq m^{-2} [6]. Mean annual precipitation is ~1900 mm at Haramachi station in the downstream reaches and 1700–1800 mm at Iitate and Tsushima stations located near the upper watershed [7].

SS was collected continuously at Haramachi (5.5 km upstream from the river mouth) from December 2012 to January 2014 using a time-integrative SS sampler with an intake diameter of 4 mm (Fig. 6.2). This sampler was used successfully in a previous study of particulate ^{137}Cs flux in the Fukushima river system [8, 9]. Monthly turbidity (kg m^{-3}) and water discharge (m^3 month^{-1}) were calculated at sampling sites using data from a tuner turbidity sensor and water level sensor (Fig. 6.2). The flux of particulate ^{129}I at Haramachi can be estimated using the following function:

$$F\left(^{129}\mathrm{I}\right) = A\left(^{129}\mathrm{I}\right) \cdot T \cdot D \tag{6.1}$$

where F (^{129}I) is the flux of ^{129}I (Bq month^{-1}), A (^{129}I) is the SS ^{129}I activity (Bq kg^{-1}), T is turbidity (kg m^{-3}), and D is water discharge (m^3 month^{-1}).

Samples for measurements of ^{129}I activity and ^{129}I/^{127}I ratios were prepared following Muramatsu et al. [10] and analyzed using an accelerator mass spectrometer (AMS) configured by Matsuzaki et al. [11]. Dried SS samples (~0.5 g) were combusted with V_2O_5 at 1000 °C in a quartz tube for 30 min under a constant flow of pure O_2 and water vapor. Volatilized iodine was trapped in an organic alkaline solution. Stable iodine (^{127}I) in trap solutions was measured using an inductively coupled plasma-mass spectrometer (ICP–MS, Agilent 8800). After adding 2 mg of iodine carrier to the trap solution, the iodine was isolated and precipitated as AgI. The ^{129}I/^{127}I ratio of AgI targets was measured using an AMS system at the Micro Analysis Laboratory Tandem Accelerator (MALT), University of Tokyo. A terminal voltage of 3.47 MV and a charge state of 5+ were chosen for acceleration and detection. Measurement ratios were normalized against the Purdue-2 reference material, which has an ^{129}I/^{127}I ratio of 6.54×10^{-11} [12] and was obtained from the Purdue Rare Isotope Measurement Laboratory (PRIME Lab) at Purdue University. The overall precision of the system was better than 5 %, and the blank levels, which included the iodine carrier, were $2.2–4.9 \times 10^{-13}$ during all experimental procedures. The original $^{129}I/^{127}I$ ratio and ^{129}I activity in SS samples were calculated using the ^{127}I concentration obtained by ICP–MS and the $^{129}I/^{127}I$ ratio obtained by AMS.

6.3 Results and Discussion

6.3.1 *Source of Particulate ^{129}I in the Niida River System*

Table 6.1 lists the total dry weight, ^{129}I activity, and $^{129}I/^{127}I$ ratio for SS samples collected at Haramachi. As shown in Fig. 6.3, SS weights increased abruptly in March and April 2013, then increased continuously from May to October 2013. Based on meteorological data provided by the Japan Meteorological Agency, monthly precipitation was relatively high at Haramachi, Iitate, and Tsushima stations in April (140–160 mm), September (150–220 mm), and October (240–350 mm) [7]. Thus, the increased SS dry weights are thought to be related to higher-than-average precipitation in 2013.

SS ^{129}I activity and $^{129}I/^{127}I$ ratios in Niida River samples were 0.9–4.1 mBq kg^{-1} and $(2.5–4.4) \times 10^{-8}$, respectively. These values are 2–10 and 2–3 times larger, respectively, than the pre-accident level for ^{129}I activity of 0.42 mBq kg^{-1} and $^{129}I/^{127}I$ ratio of 1.6×10^{-8} at Fukushima before the FDNPP accident [13]. Higher SS ^{129}I activity and $^{129}I/^{127}I$ ratios were found in March, April, September,

Table 6.1 Suspend substance (SS) weight, ^{129}I activity, and $^{129}I/^{127}I$ ratios measured in samples collected at Haramachi in the downstream reaches of the Niida River system

Sampling month		Sampling period		Suspended substance	^{129}I activity	$^{129}I/^{127}I$ ratio
		Start date	End date	(g dry weight)	(mBq kg^{-1})	($\times 10^{-8}$)
2012	December	2012/12/6	2012/12/18	0.003	n.a.	n.a.
2013	January	2012/12/18	2013/1/22	0.35	n.a.	n.a.
	February	2013/1/22	2013/2/26	0.06	n.a.	n.a.
	March and April	2013/2/26	2013/4/18	6.70	2.06 ± 0.05	4.02 ± 0.11
	May	2013/4/18	2013/5/21	0.95	n.a.	n.a.
	June	2013/5/21	2013/6/18	2.16	0.92 ± 0.03	2.52 ± 0.09
	July	2013/6/18	2013/7/25	3.24	1.71 ± 0.06	3.24 ± 0.11
	August	2013/7/25	2013/8/22	5.51	2.15 ± 0.07	4.14 ± 0.14
	September	2013/8/22	2013/9/26	9.25	4.07 ± 0.20	4.39 ± 0.22
	October	2013/9/26	2013/10/30	7.47	2.86 ± 0.11	4.83 ± 0.20
	November	2013/10/30	2013/11/20	0.83	n.a.	n.a.
	December	2013/11/20	2013/12/23	0.26	n.a.	n.a.
2014	January	2013/12/23	2014/1/17	0.05	n.a.	n.a.

n.a. not analyzed

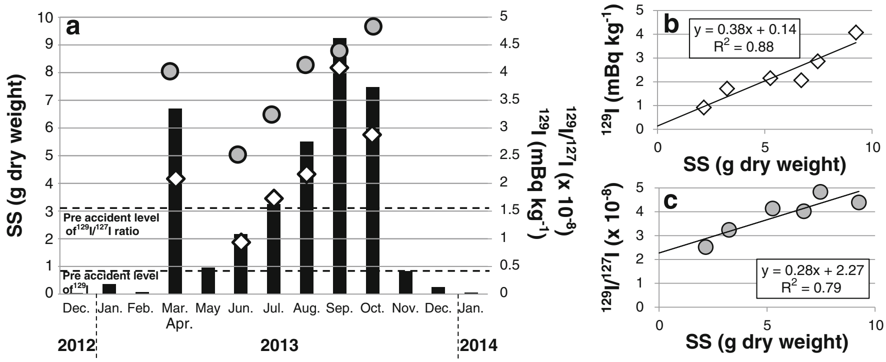

Fig. 6.3 (**a**) Temporal changes in monthly SS weight (*black bars*), ^{129}I activity (*open diamonds*), and $^{129}I/^{127}I$ ratio (*gray circles*) at Haramachi. (**b**) Correlations between ^{129}I activity and SS, and (C) the $^{129}I/^{127}I$ ratio and SS

and October 2013, when the SS weights were relatively high. The ^{129}I activity and $^{129}I/^{127}I$ ratios are strongly correlated with SS weight ($R^2 = 0.79$–0.88). As described in Sect. 6.2, the Niida River flows through highly contaminated areas in the upper watershed and medium–low contamination areas in the middle to lower reaches [6]. Therefore, it is possible that SS ^{129}I activity and $^{129}I/^{127}I$ ratios reflect the source of SS, i.e., either the more contaminated upper watershed or less contaminated downstream area. Further study is needed to clarify the differences in ^{129}I activity, $^{129}I/^{127}I$ ratios, and the ^{129}I inventory in soil between the upper watershed and downstream areas.

6.3.2 *Flux of Particulate ^{129}I in the Niida River System*

Table 6.2 lists the monthly flux of SS and associated ^{129}I at Haramachi. The SS flux and particulate ^{129}I are estimated to be 30–3200 ton month^{-1} and 0.1–9.0 kBq month^{-1} from March to October 2013, respectively. A higher ^{129}I flux of 7.6–9.0 kBq month^{-1} was recorded in September and October 2013, when high monthly precipitation (150–350 mm) was observed in the Niida River watershed [7]. As discussed in Sect. 6.3.1, particulate ^{129}I in September–October 2013 is considered to have originated mainly from the more highly contaminated upper watershed area. Therefore, a relatively high amount of particulate ^{129}I was transported from the upstream to downstream reaches of the Niida River by a rain event during September and October 2013. Further investigation is needed to better understand the ^{129}I flux in the river system.

Table 6.2 Monthly suspended substance flux and associated ^{129}I at Haramachi in the downstream reaches of the Niida River system

Analysis period		Analysis period		Suspended substance flux	^{129}I activity	^{129}I flux
		Start date	End date	(ton month^{-1})	(mBq kg^{-1})	(kBq month^{-1})
2013	March	2013/3/1	2013/4/1	45.1	2.06	0.09
	April	2013/4/1	2013/5/1	318	2.06	0.66
	May	2013/5/1	2013/6/1	30.3	n.a.	–
	June	2013/6/1	2013/7/1	110	0.92	0.10
	July	2013/7/1	2013/8/1	–	1.71	–
	August	2013/8/1	2013/9/1	–	2.15	–
	September	2013/9/1	2013/10/1	1860	4.07	7.58
	October	2013/10/1	2013/10/30	3160	2.86	9.03

n.a. not analyzed

6.4 Conclusions

Monthly SS ^{129}I activity and ^{129}I/^{127}I ratios measured from March to October 2013 in the downstream reaches of the Niida River system were 0.9–4.1 mBq kg^{-1} and (2.5–4.4) $\times 10^{-8}$, respectively. These values are strongly correlated with the total SS dry weight ($R^2 = 0.79$–0.88). The SS ^{129}I activity and ^{129}I/^{127}I ratio are considered to reflect the source of SS, i.e., the more contaminated upper watershed or the less contaminated downstream area. The particulate ^{129}I flux at Haramachi was estimated to be 7.6–9.0 kBq month^{-1} from September to October 2013. Relatively large amounts of particulate ^{129}I were transported by heavy rain from the upstream to downstream reaches of the Niida River over this period.

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