

# Chapter 2

## Speciation of $^{137}\text{Cs}$ and $^{129}\text{I}$ in Soil After the Fukushima NPP Accident

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### 2.1 Introduction

A number of radionuclides (including  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{136}\text{Cs}$ ,  $^{131}\text{I}$ ,  $^{132}\text{Te}$ ) were released into the atmosphere from the Fukushima Daiichi NPP accident in March, 2011. On March 15–17 and 21–23, deposition increased in the areas surrounding Fukushima prefecture because north–easterly, easterly, and south–easterly winds under a low-pressure system transported the radionuclides from the Fukushima NPP, and subsequent precipitation associated with the same system washed radioactive materials out of the radioactivity plume, thereby effectively depositing them on land [1–5]. In addition to the radioactive plume that covered the Fukushima Prefecture, two other large plumes suffered severe radioactive contamination over north Japan. Precipitation from these plumes caused high-radioactive spots across wide areas including the Tokyo metropolitan [1].

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Although the eastern parts in the Tokyo metropolitan area are located far from the Fukushima NPP, high  $^{137}\text{Cs}$  and  $^{131}\text{I}$  deposition was observed in the areas. Therefore, the Tokyo metropolitan area is one of the hot spots of radioactive fallout from the Fukushima NPP accident [4]. The Tokyo metropolitan hot-spot area has a high-density population, and many residents have been worried about the radiation exposure from the Fukushima NPP. The released  $^{131}\text{I}$  from the Fukushima NPP contaminated tap water with rainfall that precipitated in the Tokyo metropolitan hot-spot areas on March 23, 2011, whereas  $^{131}\text{I}$  was also detected in the tap water of the Kanto and Tohoku regions through mid-April [1].

At present,  $^{131}\text{I}$  released from the Fukushima NPP accident in March, 2011 is not detected in the environment, because its short half-life is only 8 days. On the contrary,  $^{129}\text{I}$  with a half-life of 15.7 Ma is easily found. The  $^{129}\text{I}/^{127}\text{I}$  ratio measured in surface soil could provide information on the local deposition of  $^{131}\text{I}$  released from the Fukushima NPP, if the ratio of  $^{131}\text{I}$  and  $^{129}\text{I}$  before release from the broken reactors can be estimated [6, 7]. The  $^{131}\text{I}$  behavior in the unsaturated zone and shallow groundwater immediately after the accident may be inferred from the  $^{129}\text{I}$  content.

This study has two objectives: (1) to examine the behavior of  $^{137}\text{Cs}$  and  $^{131}\text{I}$  in the surface soil of the Kanto loam in the Tokyo metropolitan hot-spot areas and Fukushima immediately after the accident and (2) to determine the  $^{137}\text{Cs}$  and  $^{129}\text{I}$  speciation in the soil to evaluate  $^{137}\text{Cs}$  and  $^{131}\text{I}$  contamination in the shallow groundwater which residence time is short.

## 2.2 Material and Methods

### 2.2.1 Soil Samples

One surface-soil sample within a depth of 0.5 cm and four surface-soil samples within a depth of 1 cm were collected on March 29, 2011 at the western Tokyo metropolitan area (WTMA, W1) and on March, 30 and 31, 2011 at the eastern Tokyo metropolitan hot-spot area (ETMA, E1-4), respectively. The soil samples collected at western Tokyo were placed in a polypropylene vessel (vessel A), 5 cm in diameter and 10 cm in height, and each of the four soil samples collected at eastern Tokyo was packed into a polypropylene vessel (vessel B), 2.2 cm in diameter and 1.2 cm in height.

Surface-soil core samples ( $5\text{ cm}^2 \times 10\text{ cm}$ ) were collected at the WTMA on August 25, 2011 and ETMA, on October 14–18, 2011, respectively. The undisturbed soil cores were sampled using a cylindrical stainless steel core sampler, 0–10 cm in depth. The soil core samples were cut into the lengths of 0–2.5, 2.5–5 cm, and 5–10 or 0–2.5 and 2.5–5.0 cm. Each sample was stored in plastic vessels (vessels A). We also collected soil in the same manner in Nagadoro, Fukushima (N1) in May, 2012. Then we measured the vertical profiles of  $^{137}\text{Cs}$  in soil.

### ***2.2.2 Column-Infiltration Experiments Using the Rainwater from the Tokyo Metropolitan Hot-Spot Area***

It rained in the southern Ibaraki Prefecture from 0:00 to 2:00 LT on March 31, 2011, resulting in a total precipitation of 5.5 mm in Tsukuba. A rainwater sample was collected during precipitation at the ETMA (Japan Meteorological Agency). The rainwater sample (101 mL) was placed in a polypropylene vessel (5 cm in diameter and 10 cm in height, vessel A).

We investigated the infiltration of  $^{137}\text{Cs}$  from the rainwater in the soil environment via column experiments on April 1, 2011. Sand (Toyoura Standard Sand, Toyoura Keiseki Kogyo Co., Ltd., Yamaguchi, Japan) and soil were used in the columns. The soil, classified by the *World Reference Base* as a haplic stagnosol, was collected from a depth of approximately 30 cm from an outcrop at the campus of Kyoto University Research Reactor Institute (KURRI), Osaka, Japan. The soil contained a small amount of organic matter (organic carbon 2.7–2.8 %, pH 5.7–5.8) and was devoid of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ .

The sand was rinsed several times to remove clay minerals using deep groundwater that did not contain  $^{137}\text{Cs}$ . The sand and soil samples were packed into two columns (20 mm in diameter and approximately 1 cm in depth). After filling, columns A (sand) and B (soil) were soaked in pure water for 3 h. Next, the rainwater sample (50 mL) was passed through each column at a rate of  $0.7 \text{ mL} \cdot \text{min}^{-1}$ , and then, each column was rinsed with ultrapure water (84 mL). Although the flow rate used was faster than the actual rate of rainwater infiltration in the Kanto district (approximately  $1.5 \text{ m} \cdot \text{y}^{-1}$ ), this provides a conservative estimation of radionuclide migration. After infiltration, the sand and soil samples were stored separately in polypropylene containers (vessel B). Then, these vessels were placed into a different container, and the radioactivity of the samples in each container was measured using a gamma-ray spectrometer.

### ***2.2.3 Leaching of Radionuclides from Soils Using the Batch Method***

Radioactivities of the 100-g soil samples collected on March 29, 2011 at WTMA were immediately measured by the following method. The soil samples were mixed with the ultrapure water and shaken by hand for 30 min. The mixture was stored for 15 min, and the solution was separated by ultra-centrifuge (Kokusan Co., Ltd, Japan), and then, radioactivity in the supernatant was measured by gamma-ray spectrometry.

Approximately 10 g of the surface-soil samples collected from the WETA were added to ultrapure and groundwater samples, and then, they were agitated at a speed of 100 rpm for 90 and 270 days. The groundwater sample was discharged from the Kanto-loam layer in the Tokyo metropolitan hot-spot area, and was sampled in

**Table 2.1** Concentrations of major ions in groundwater

Element	Concentration (mg·L <sup>-1</sup> )	Element	Concentration (mg·L <sup>-1</sup> )
Na	6.92	Cl <sup>-</sup>	5.83
K	7.00	SO <sub>4</sub> <sup>2-</sup>	13.4
Ca	22.0	HCO <sub>3</sub> <sup>-</sup>	110
Mg	7.87	pH	8.3

March, 2010 before the Fukushima NPP accident. The concentrations of Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, and Ca<sup>2+</sup> in the groundwater were measured by cation chromatography and those of Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> were measured by anion chromatography, whereas HCO<sub>3</sub><sup>-</sup> was measured by titration using HCl. Table 2.1 lists the major cation and anion of the groundwater samples.

After leaching, solid-liquid separation was conducted by centrifugation at 2000 rpm for 5 min. Then, the solution was filtered through a membrane filter with a pore size of 0.45 μm (Advantech, Co., Ltd., Japan). After storing each fraction in a U-8 vessel, radioactivities of <sup>131</sup>I, <sup>134</sup>Cs, <sup>136</sup>Cs, <sup>137</sup>Cs, and <sup>132</sup>Te were measured by gamma-ray spectrometry.

### 2.2.4 Separation of <sup>137</sup>Cs and <sup>129</sup>I in Soil Samples

We extracted <sup>137</sup>Cs and <sup>129</sup>I from three surface soil samples from the Kanto-loam layer in the ETMA and the Nagadoro in Fukushima. The soil of depths of 0–2.5 cm (E5, E6, N1) was well mixed and approximately 10 g soil sample was taken by cone and quartering method. Approximately 10 g of the samples were used in the sequential-extraction experiment [8, 9]. A ratio of solution to sample of 5 (v/w) was used for extraction in each step.

Fraction 1: After ultrapure water was added to the soil sample, the suspension was shaken for 24 h at room temperature, and then the suspension was stored overnight. After extraction, solution was separated from the soil residue by centrifugation at 2000 rpm for 5 min. The solution was filtered through a membrane filter with a pore size of 0.45 μm (Advantech, Co., Ltd., Japan). The fraction of the filtrate represents water-soluble species. The remaining solid on the membrane filter was combined with the residue for the next leaching step.

Fraction 2: 1 M of NaAc was added to the residue from Fraction 1. The suspension was shaken for 12 h at room temperature and stored overnight. The fraction of the filtrate represents exchangeable species.

Fraction 3: 1-M NaAc – HAc (pH 5) was added to the residue from Fraction 2, and the suspension was shaken for 12 h at room temperature. The fraction of the filtrate represents carbonate-bound species.

Fraction 4: 0.04-M NH<sub>2</sub>OH·HCl in 25 % (v/v) HAc (pH 2) was added to the residue from Fraction 3 and stirred in a hot-water bath at 80 °C for 4 h. The

fraction of the filtrate represents species associated with solids via chemical-sorption mechanisms that can be released into the extraction solution with a weak reducing agent, and they mainly include species bound to Fe/Mn oxides.

Fraction 5: 30 %  $\text{H}_2\text{O}_2$  was added to the residue, in which  $\text{HNO}_3$  had already been added to adjust the final pH to 2. The suspension was agitated for 2 h at 85 °C. After the suspension was cooled to room temperature, 1.8-M  $\text{NH}_4\text{Ac}$  in 11 %  $\text{HNO}_3$  (v/v) was added, and the extraction continued for 30 min at room temperature. The fraction of the filtrate is associated with organic matter.

After each fraction was stored into a U-8 vessel,  $^{137}\text{Cs}$  was measured by gamma-ray spectrometry.

### ***2.2.5 Purification of Iodine Isotopes for Accelerator Mass Spectrometry (AMS) Measurement***

One-milliliter solution of nitric acid (Kanto Chemical Co., Ltd.) and 0.5 mL of  $\text{H}_2\text{O}_2$  (Kanto Chemical Co., Ltd.) were added to sample solutions separated from each fraction. The dissolved iodine was oxidized to  $\text{I}_2$  and was then separated from the sample solution into 10 mL of chloroform (Wako Co., Ltd.). The chloroform was separated from the sample solution, and then 10 mL of 0.1 M of  $\text{NaHSO}_3$  (Wako Co., Ltd.) was added to the chloroform to extract  $\text{I}^-$  into the  $\text{NaHSO}_3$  solution. The  $\text{NaHSO}_3$  solution was separated from the chloroform, and 1 mL of 6 M  $\text{NaCl}$  (Aldrich Co., Ltd.) was added to the solution. A 0.1 mL portion of 1 M  $\text{AgNO}_3$  (Aldrich Co., Ltd.) was then added to the solution, and the solution was agitated, causing  $\text{AgI}$  to precipitate with  $\text{AgCl}$ . This precipitation was allowed to continue for 30 min before the mixture was centrifuged for 5 min at 3000 rpm. The mixture of  $\text{AgCl}$  and  $\text{AgI}$  precipitate was separated from the solution.  $\text{AgCl}$  was separated from  $\text{AgI}$  by adding 4 mL of concentrated  $\text{NH}_3$  to dissolve  $\text{AgCl}$  only. The  $\text{AgI}$  precipitate was rinsed with 5 mL of ultrapure water to yield a pure  $\text{AgI}$  sample, which was then dried in an electric oven at 70 °C for 40 min. The  $\text{AgI}$  sample was added to Nb powder at an Nb and  $\text{AgI}$  ratio of 4 (w/w). The detail chemical separation was described by a previous paper [10].  $^{129}\text{I}$  and  $^{127}\text{I}$  were measured by AMS (Malt, Tokyo Univ., Japan), and detailed procedure of measurement of  $^{129}\text{I}/^{127}\text{I}$  atomic ratios were described by Matsuzaki et al. [11].

### ***2.2.6 Measurement of Radioactivity in Environmental Samples by Gamma-Ray Spectrometry***

The radioactivity of the rainwater and soil sample was measured using a p-type high-purity germanium detector (IGC-309, Princeton Gamma-Tech) with 40 % relative efficiency and a multichannel analyzer (7600-000, Seiko EG&G) with a

high-voltage circuit (7600-310) and pulse height analyzer (7600-510) in KURRI. The gamma-ray counting efficiency of the detector was estimated by constructing a relative gamma-ray-counting-efficiency curve using a certified mixed-radionuclide gamma-ray reference source containing  $^{57}\text{Co}$  (122.1 keV),  $^{137}\text{Cs}$  (661.7 keV), and  $^{60}\text{Co}$  (1173 and 1332 keV), which were normalized to the 1460-keV gamma-ray peak of  $^{40}\text{K}$  in KCl. A quadratic function was fitted to the logarithmic relationship between the relative counting efficiency and gamma-ray energy using the least-squares method and was normalized at 1460 keV. The radiation energies of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ ,  $^{136}\text{Cs}$ ,  $^{137}\text{Cs}$ , and  $^{132}\text{Te}$  were 364, 796, 818, 662, and 228 keV, respectively. The sum effect of gamma rays from  $^{134}\text{Cs}$  was corrected by measuring the  $^{134}\text{Cs}$  solution.  $^{134}\text{Cs}$  was produced by the neutron activation of CsCl at KURRI. CsCl was dissolved in the ultrapure water and the prepared  $^{134}\text{Cs}$  solution. The detection limits of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  with a measuring time of 10,000 s were 0.11, 0.099, and 0.12 Bq for vessel B and 0.20, 0.19, and 0.23 Bq for vessel A, respectively.

The radioactivity of undisturbed core samples from the ETMA and radio-Cs samples leaching into the ultrapure water and groundwater samples was measured in the Isotopes Centre, Hokkaido University using a p-type, high-purity germanium detector (model IGC-309, Princeton Gamma-Tech) with 40 % relative efficiency. The detection limits of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  with a measuring time of 864,000 s were 0.02 and 0.02 Bq for the U-8 vessel, respectively.

## 2.3 Results and Discussion

Table 2.2 shows the concentrations of  $^{134}\text{Cs}$ ,  $^{136}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{131}\text{I}$ , and  $^{132}\text{Te}$  in the surface soils collected in March, 2011 from the Tokyo metropolitan hot-spot area (but only W6 was collected in August, 2011). Short half-lives for  $^{136}\text{Cs}$ ,  $^{131}\text{I}$ , and  $^{132}\text{Te}$  were detected in the surface-soil samples except W6. The radioactivities of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  in the surface-soil samples except W6 collected at ETMA were measured, and W6 measured only the radioactivities of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ .  $^{131}\text{I}$  was the source of the maximum radioactivity in the soil in March, 2011 in the Tokyo metropolitan hot-spot area. The concentration of  $^{131}\text{I}$  ranged from 9.4 to 13 k Bq·kg<sup>-1</sup> before rain (March 30, 2011) and from 7.2 to 11 k Bq·kg<sup>-1</sup> after rain, whereas that of  $^{137}\text{Cs}$  ranged from 0.72 to 3.3 k Bq·kg<sup>-1</sup> before rain and 1.7–2.9 k Bq·kg<sup>-1</sup> after rain (March 31). As the fallout radionuclides on the surface soils were mainly washed out from the atmospheric aerosol plume by precipitation,  $^{137}\text{Cs}$  concentrations being similar to  $^{131}\text{I}$  in the soil before rain on March 30 and after rain on March 31 suggest that the rainfall on March 21 and 22 (and the very small precipitation on March 15 and 16) was major source to most radionuclides in the soil.

The concentrations of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ , and  $^{137}\text{Cs}$  in the rainwater collected at the ETMA toward the end of March, 2011 were  $66 \pm 3$ ,  $28 \pm 2$ , and  $31 \pm 2$  Bq·L<sup>-1</sup>, respectively. The concentration of each radionuclide was corrected at 2:00 LT, 31 March 2011.

**Table 2.2** Concentrations of  $^{134}\text{Cs}$ ,  $^{136}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{131}\text{I}$ , and  $^{132}\text{Te}$  in surface soils in Tokyo metropolitan area

Sampling site number	Sampling sites	Depth	Sampling date	$^{134}\text{Cs}$	$^{137}\text{Cs}$
		cm		$\text{kBq}\cdot\text{kg}^{-1}$	$\text{kBq}\cdot\text{kg}^{-1}$
W1	WTMA	0–0.5	March 29, 2011	$0.517 \pm 0.01$	$0.530 \pm 0.07$
E1	ETMA	0–1	March 30, 2011	$2.86 \pm 0.07$	$3.25 \pm 0.07$
E2	ETMA	0–1	March 30, 2011	$2.70 \pm 0.14$	$2.90 \pm 0.12$
E3	ETMA	0–1	March 31, 2011	$1.01 \pm 0.06$	$0.720 \pm 0.06$
E4	ETMA	0–1	March 31, 2011	$1.80 \pm 0.07$	$1.70 \pm 0.18$
W6	WTMA	0–5	Aug. 25, 2011	$0.078 \pm 0.002$	$0.093 \pm 0.002$
		$^{131}\text{I}$	$^{136}\text{Cs}$	$^{132}\text{Te}$	$^{134}\text{Cs}/^{137}\text{Cs}$
		$\text{kBq}\cdot\text{kg}^{-1}$	$\text{kBq}\cdot\text{kg}^{-1}$	$\text{kBq}\cdot\text{kg}^{-1}$	
W1	WTMA	$3.43 \pm 0.01^a$	$0.051 \pm 0.002^a$	$0.373 \pm 0.05^a$	$1.03 \pm 0.13$
E1	ETMA	$9.36 \pm 0.10^b$	$0.301 \pm 0.360^b$	$0.338 \pm 0.050^b$	$1.14 \pm 0.03$
E2	ETMA	$11.1 \pm 0.21^b$	$0.220 \pm 0.056^b$	$0.168 \pm 0.074^b$	$1.07 \pm 0.07$
E3	ETMA	$13.4 \pm 0.2^b$	$0.128 \pm 0.050^b$	$0.271 \pm 0.056^b$	$0.71 \pm 0.10$
E4	ETMA	$7.32 \pm 0.12^b$	$0.632 \pm 0.076^b$	$0.328 \pm 0.049^b$	$0.94 \pm 0.11$
W6	WTMA		–	–	$1.19 \pm 0.03$

WTMA Western Tokyo metropolitan area, ETMA Eastern Tokyo metropolitan area

<sup>a</sup>Correction at 5:00 March 29, 2011 at local time

<sup>b</sup>Correction at 2:00 March 31, 2011 at local time

**Table 2.3** Radioactivities collected in sand and soil (column A and B), and collection efficiencies

Sample ID	Sand column	Soil column
Material	Sand	Soil
Weight (g)	3.2	4.7
$^{131}\text{I}$ (Bq)	ND	$0.82 \pm 0.06$
$^{137}\text{Cs}$ (Bq)	$1.44 \pm 0.09$	$1.26 \pm 0.08$
$^{131}\text{I}$ collection efficiency (%)	–	$31 \pm 3$
$^{137}\text{Cs}$ collection efficiency (%)	$93 \pm 6$	$81 \pm 5$

ND: below detection limit

Table 2.3 lists radioactivities trapped in both columns of sand and soil, and the  $^{131}\text{I}$ - and  $^{137}\text{Cs}$ -trapping efficiencies in both columns. In column A (sand), almost all of the  $^{137}\text{Cs}$  was trapped on sand with the trapping efficiency of  $93 \pm 6$  %. In column B (soil), the  $^{137}\text{Cs}$ -trapping efficiency was  $81 \pm 5$  %. Therefore, we considered almost all  $^{137}\text{Cs}$  in rainwater to be adsorbed on sand and the Haplic Gray Upland soil within a depth of 1 cm from the ground, while the trapping efficiency of  $^{131}\text{I}$  in both sand and soil columns was below the detection limit and 30 %, respectively. These

**Table 2.4** Concentrations of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  in surface soil collected in ETMA and Nagadoro

Sample No.	Depth cm	$^{134}\text{Cs}$ $\text{kBq}\cdot\text{m}^{-2\text{a}}$	$^{137}\text{Cs}$ $\text{kBq}\cdot\text{m}^{-2}$	$^{134}\text{Cs}/^{137}\text{Cs}$ Activity ratio
E5	0.0–2.5	$173 \pm 4$	$175 \pm 3$	$0.99 \pm 0.03$
	2.5–5.0	$0.911 \pm 0.059$	$0.722 \pm 0.041$	$1.26 \pm 0.11$
	5.0–10	$3.94 \pm 0.15$	$3.66 \pm 0.12$	$1.08 \pm 0.05$
Total	0.0–10	$178 \pm 1$	$179 \pm 1$	$0.99 \pm 0.01$
E6	0.0–2.5	$147 \pm 4$	$133 \pm 4$	$1.11 \pm 0.04$
	2.5–5.0	$22.7 \pm 0.8$	$20.3 \pm 0.6$	$1.12 \pm 0.05$
	5.0–10	$12.0 \pm 1.0$	$16 \pm 1$	$0.75 \pm 0.08$
Total	0.0–10	$182 \pm 1$	$169 \pm 1$	$1.07 \pm 0.01$
N1	cm	$\text{MBq}\cdot\text{m}^{-2}$	$\text{MBq}\cdot\text{m}^{-2}$	Activity ratio
	0.0–1.0	$26.6 \pm 0.5$	$26.4 \pm 0.3$	$1.01 \pm 0.02$
	1.0–2.0	$17.3 \pm 0.4$	$17.1 \pm 0.2$	$1.01 \pm 0.03$
	2.0–3.0	$18.6 \pm 0.4$	$18.9 \pm 0.2$	$0.98 \pm 0.02$
	3.0–4.0	$5.50 \pm 0.16$	$5.73 \pm 0.10$	$0.98 \pm 0.03$
	4.0–5.0	$2.76 \pm 0.06$	$2.82 \pm 0.04$	$0.98 \pm 0.03$
	5.0–10	$1.44 \pm 0.05$	$1.48 \pm 0.03$	$0.97 \pm 0.04$
	10–15	$0.08 \pm 0.01$	$0.09 \pm 0.01$	$0.90 \pm 0.13$
	15–20	ND	ND	–
	20–25	ND	ND	–
	25–29	ND	ND	–
Total	0.0–29	$72.3 \pm 0.7$	$72.5 \pm 0.4$	$1.00 \pm 0.01$

<sup>a</sup>Correction on March 11, 2011

data indicate that retardation of  $^{131}\text{I}$  in both sand and soil downward was smaller than that of  $^{137}\text{Cs}$ .

Table 2.4 shows the concentrations of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  in the surface soil collected at the ETMA in October, 2011, 7 months after the accident. Almost all  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in the Kanto-loam soil were within a depth of 2.5 cm from the ground. The column test and the distribution of  $^{137}\text{Cs}$  obtained from the core sample in the field indicated that  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were strongly adsorbed on the surface soil within a depth of several centimetres from the surface. Vertical profiles of radionuclides in soil in Koriyama, Fukushima, showed that more than 90 % of  $^{131}\text{I}$  was found to be within about 5 cm depth from the surface in soil layer after the accident [12].

Following the leaching tests from the surface soil collected at WTMA into the pure water immediately after sampling in March, 2011, the leachate was separated from soil.  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{131}\text{I}$ ,  $^{136}\text{Cs}$ , and  $^{132}\text{Te}$  in soil did not leach into the water (Table 2.5). Table 2.5 lists the amount of radioactive Cs in the leachate when the surface soil of the ETMA or WTMA was mixed with the ultrapure water and groundwater during 90 and 270 days. Both  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  in each soil of the Kanto-loam layer and the Nagadoro were not released by leaching with both the ultrapure water and groundwater after mixing during 90 and 270 days.



**Table 2.5** Extraction of cesium isotopes in soil by pure water and groundwater (in Bq)

Nuclides/media	Pure water	Pure water	Groundwater	Groundwater
Sampling site	W1	W6	W6	E5
$^{134}\text{Cs}$	ND (0.17)	ND (0.02)	ND (0.02)	ND (0.02)
$^{137}\text{Cs}$	ND (0.16)	ND (0.02)	ND (0.02)	ND (0.02)
$^{131}\text{I}$	ND (0.19)	–	–	–
$^{136}\text{Cs}$	ND (0.15)	–	–	–
$^{132}\text{Te}$	ND (0.14)	–	–	–
Period of extraction	30 min	90 day	90 day	90 day
Sampling date of soil	March 29, 2011	Aug. 25, 2011	Aug. 25, 2011	Aug. 25, 2011
Measuring time	10,000 s	864,000 s	864,000 s	864,000 s
Nuclides/media	Pure water	Pure water	Pure water	Groundwater
Sampling site	E5	E6	N1	N1
$^{134}\text{Cs}$	ND (0.02)	ND (0.02)	ND (0.02)	ND (0.02)
$^{137}\text{Cs}$	ND (0.02)	ND (0.02)	ND (0.02)	ND (0.02)
Period of extraction	270 day	270 day	90 day	270 day
Sampling date of soil	August 25, 2011	August 25, 2011	May 24, 2012	May 24, 2012
Measuring time	864,000 s	864,000 s	864,000 s	864,000 s

ND: below the detection limit, () : detection limit value

**Table 2.6** Distribution of  $^{137}\text{Cs}$  and  $^{129}\text{I}$  in the soil dissolved in each fraction solution

Fraction	E5 (%)	E6 (%)	N1 (%)
$^{137}\text{Cs}$			
F1: Water	0.0	0.0	0.0
F2: Exchangeable	0.4	0.9	1.67
F3: Bound to carbonates	1.3	0.9	1.43
F4: Bound to Fe-Mn oxides	6.0	2.1	1.12
F5: Bound to organic matter	37.8	33.1	5.27
Residual	54.4	63.1	90.5
Total	100	100	100
$^{129}\text{I}$			
F1: Water	9.5	0.8	0.74
F2: Exchangeable	3.8	1.0	2.40
F3: Bound to carbonates	5.4	1.3	3.92
F4: Bound to Fe-Mn oxides	72	20.9	82.3
F5: Bound to organic matter	9.2	66.4	0.91
Residual	–	9.6	9.74
Total	100	100	100

Table 2.6 shows the leaching fractions of  $^{137}\text{Cs}$  from each soil in the Kanto-loam layer of the ETMA and in the Nagadoro, Fukushima according to the sequential-extraction procedure described previously. The amount of  $^{137}\text{Cs}$  leached into water (F1) from the Kanto-loam soil and Fukushima soil was below the detection limit. This has been demonstrated by the leaching results of radioactive Cs from the

**Table 2.7** Concentrations of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  in groundwater and radioactivities of suspended matter (SS) of the groundwater sampled at the Tokyo metropolitan area after the accident

Nuclides	$^{131}\text{I}$ ( $\text{Bq}\cdot\text{kg}^{-1}$ )	$^{137}\text{Cs}$ ( $\text{Bq}\cdot\text{kg}^{-1}$ )	$^{134}\text{Cs}$ ( $\text{Bq}\cdot\text{kg}^{-1}$ )
Groundwater	ND (0.007)	ND (0.008)	ND (0.008)
SS ( $\sim 80\ \mu\text{m}$ )	ND	ND	ND
SS ( $80\ \mu\text{m} \sim 3\ \mu\text{m}$ )	ND	ND	ND
SS ( $3\ \mu\text{m} \sim 0.45\ \mu\text{m}$ )	ND	ND	ND

ND: below detection limit, ( ): detection limit value

soil by the ultrapure water and groundwater, as shown in Table 2.5. The ratio of exchangeable  $^{137}\text{Cs}$  from the two soil samples (E5 and E6) was less than 1 %, with 50–60 % of  $^{137}\text{Cs}$  remaining in the residue. Approximately 98 % of  $^{137}\text{Cs}$  was in the fraction of Fe–Mn oxide, organic matter, and the residue. The  $^{137}\text{Cs}$  in the water fraction (F1) is undetectable and this is the same results of the extraction test obtained in Table 2.5. The small fraction of exchangeable  $^{137}\text{Cs}$  suggests that immediately after the  $^{137}\text{Cs}$  in rainwater dropping on the surface soil,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were strongly adsorbed onto the soil of the Kanto loam, and they were not readily leached into the soil water. The amount of  $^{129}\text{I}$  in fraction of F1 and F2 were 0.7–9.5 % and 1–3.8 %, respectively; therefore,  $^{131}\text{I}$  in unsaturated soil moved downward faster than  $^{137}\text{Cs}$ . More than 90 % of  $^{137}\text{Cs}$  was in the fraction of organic matter and the residue, while  $^{129}\text{I}$  was mainly fixed by Fe–Mn oxidation and organically bound.

The estimated  $^{137}\text{Cs}$  migration rate for the Nishiyama loam soil, which was obtained in situ at Nishiyama (Nagasaki), is  $1.0\ \text{mm}\cdot\text{y}^{-1}$  [13]. This is considerably less than the rainwater infiltration rate of  $2.5\ \text{m}\cdot\text{y}^{-1}$ . Furthermore,  $^{137}\text{Cs}$  was not detected in the groundwater of the Nishiyama area, suggesting that  $^{137}\text{Cs}$  has not yet migrated to the groundwater table [14]. We collected 4 L of shallow groundwater in the Kashiwa city in the ETMA on August 17, 2012. The groundwater was filtered through membrane filter with a pore size of 80, 3, and  $0.45\ \mu\text{m}$ . The groundwater after the filtration was gradually reduced to 5 mL by a mantle heater. Both  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were not detected in the shallow groundwater and the suspended matters collected from the filter (Table 2.7).

Although the  $^{131}\text{I}$  moving velocity deduced from that of  $^{129}\text{I}$  was greater than that of  $^{137}\text{Cs}$ , the  $^{129}\text{I}$  migration rate is lower than the water infiltration rate, due to the  $^{129}\text{I}$  absorption on soil in the unsaturated zone.

In Kanto loam, soil water reaches depths of 20–30 cm, considering a  $1\text{--}1.5\ \text{m}\cdot\text{y}^{-1}$  infiltration rate, after 80 days corresponding to the time length of 10 times of the  $^{131}\text{I}$  half-lives. Mainly  $^{137}\text{Cs}$  was detected in litters in forest [5] but  $^{137}\text{Cs}$  was detected to depth of 10 cm in soil without litter [15]. Because rainy force is buffered in litters when the surface of soils has litters, rain is hard to directly enter to the deep part of the soil. Water-soluble  $^{131}\text{I}$  would merely move downward to depths of 30–40 cm, even if  $^{131}\text{I}$  penetrated a depth of 10 cm in the soil without litters and/or grass immediately after the accident. Therefore,  $^{131}\text{I}$  could never reach the depths of 50 cm in the groundwater table.

## 2.4 Conclusion

The sequentially chemical fractionations of  $^{129}\text{I}$  and  $^{137}\text{Cs}$  in soil indicate that most part of  $^{137}\text{Cs}$  and  $^{129}\text{I}$  were insoluble. Traces of  $^{131}\text{I}$  in the soil water did not reach the 50-cm depth by late June, 2011, corresponding to the time length of 10 times of  $^{131}\text{I}$  half-lives after the Fukushima NPP accident. Therefore, shallow groundwater could be safely useful water resource after the accident.

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