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Radioactivity concentrations of ^{131}I, ^{134}Cs and ^{137}Cs in river water in the Greater Tokyo Metropolitan area after the Fukushima Daiichi Nuclear Power Plant Accident

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Radioactivity concentrations of ^{131}I, ^{134}Cs and ^{137}Cs were determined for the river water sampled from the Edogawa River, the Arakawa River and the Tamagawa River, three major rivers spreading in or surrounding Tokyo Metropolis. Data from March 28 to April 7 are presented. Among the three radioactive nuclides, ^{131}I recorded the highest radioactivity concentrations of 31 Bq/L and 11 Bq/L in water samples collected on March 28 from the Edogawa and the Arakawa Rivers, respectively. Two trends of the ^{131}I concentration change occur from March 28 to April 7 for the Edogawa River water; the ^{131}I concentration decreased abruptly from March 28 to March 30 and then decreased gradually until April 7. These trends suggest that ^{131}I in the Edogawa River in late March primarily originated from fallout in the Tokyo-Chiba area, which dropped to the ground together with rain from March 21 to 24. The ^{137}Cs concentration in river water from the Edogawa River changed differently from the ^{131}I concentration. The highest ^{137}Cs concentration was measured for the sample collected on March 31. Apparently, fractionation between ^{131}I and ^{137}Cs occurred during the transportation from FDNPP to the metropolitan area and also during the transportation from the ground where radionuclides deposited to the sampling site of river water.

Keywords: Fukushima, ^{131}I, ^{137}Cs, river, Tokyo

INTRODUCTION

On March 11, 2011, a gigantic earthquake and resulting tsunami hit Japan. A total of 20,000 people were deceased and rendered missing by these two related disasters. In addition, more than 80,000 people remain displaced from their homes. As reported by various worldwide media outlets, Fukushima Daiichi Nuclear Power Plant (FDNPP) of Tokyo Electric Power Company was severely damaged; eventually, a large amount of radioactive material was released from the reactor facilities into the environment. The contamination affected not only the areas around the reactor site itself, but also a relatively large area of eastern mainland Japan, with some specific areas becoming heavily contaminated (e.g., Yoshida and Takahashi, 2012). In addition, a small amount of the radioactive material became airborne and was dispersed into the atmosphere (Manolopoulou *et al*., 2011; Pittauerová *et al*., 2011; Leon *et al*., 2011; Bolsunovsky and Dementyev, 2011) and in the oceans (Honda *et al*., 2012), spreading all over the world.

The Greater Tokyo Metropolitan area is approximately 200 km southwest from FDNPP. Until March 14, no significant radioactivity was detected in the suspended particulate matter (SPM) sampled in the Metropolitan area; the air dose rate was in the normal range even at Tokai in the Ibaraki prefecture (Ibaraki Prefecture, 2011), which is closer to FDNPP compared with Tokyo. On March 15, the first increase of air dose rate was observed at 1 a.m. at Tokai, followed by the second and larger increase at 4 to 5 a.m. (Ibaraki Prefecture, 2011). On the same day, the SPM sample collected at Komazawa in the midtown area of Tokyo showed high radioactivity of ^{132}Te, ^{131}I, ^{132}I, ^{134}Cs and ^{137}Cs, with their peak values being recorded for the sample collected from 10 a.m. to 11 a.m. (Nagakawa *et al*., 2012). After a gradual decrease in their radioactivity in the following days, a large increase was again observed on March 21 to 23, with a coincidence of rain on these days. After March 25, the radioactivity in SPM has mostly remained near or below the detection limit (TMITRI, 2011).

On March 23, 2011, Tokyo Metropolitan Government officially announced that high radioactivity of ^{131}I (210 Bq/kg) was detected in purified water that was sampled at 9 a.m. on March 22 at the Kanamachi water purification plant (BW, 2011a). The radioactivity measured was

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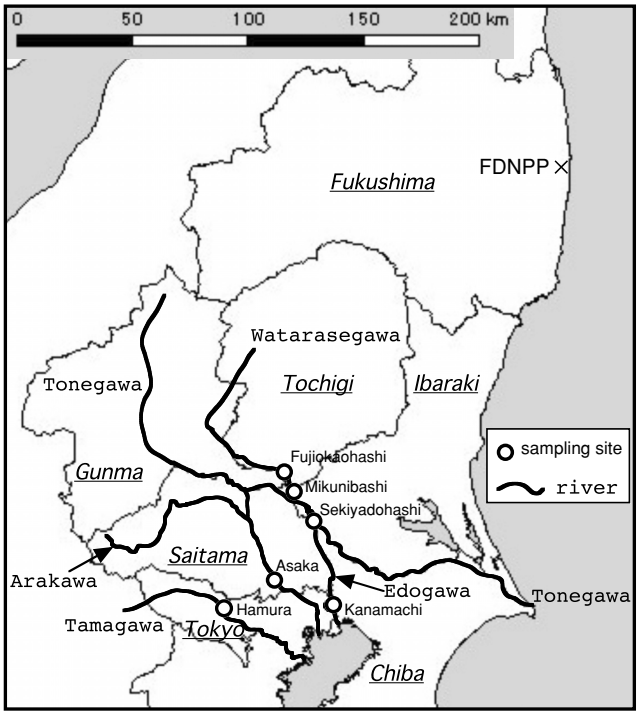

Fig. 1. Sampling sites of river water. The Edogawa River and the Watarasegawa River are tributaries of the Tonegawa River. The Edogawa River, the Arakawa River and the Tamagawa River flow into Tokyo Bay, while the Tonegawa River goes to the Pacific Ocean.

lower than the provisional regulation value (300 Bq/kg) guided by the Ministry of Health, Labor, and Welfare but was above the provisional regulation value for infants (100 Bq/kg). Thus, uneasiness was aroused in many people living not only in the Tokyo Metropolitan area but also in the neighboring prefectures such as Chiba. Fortunately, the activity level of ^{131}I in the purified water decreased in a couple of days (Nagakawa *et al*., 2012). However, the radioactive materials in food and especially drinking water remain of great concern.

We initiated measuring the radioactivity in river water in collaboration with the Bureau of Waterworks, Tokyo Metropolitan Government from March 28. The water sample was obtained at three sampling sites, Kanamachi, Asaka and Hamura, where raw water was taken from the Edogawa, the Arakawa, and the Tamagawa Rivers, respectively, for supplying purified water to citizens of Metropolitan Tokyo. In this study, activity concentrations of ^{131}I, ^{134}Cs and ^{137}Cs are reported for such water samples taken from March 28 to April 7. Some supplemental data are also presented for river water sampled from the Watarasegawa River, a tributary of the Tonegawa River, from which water flows into the Edogawa River and the Arakawa River to some extent. Based on the data

Table 1. Radioactivity (Bq/L) in the Edogawa River water sampled at Kanamachi*

Sampling Date and Time	^{131}I	^{134}Cs	^{137}Cs	^{131}I/^{137}Cs
2011/3/28 9:00	31 ± 2	0.8 ± 0.2	0.6 ± 0.2	52 ± 18
2011/3/29 9:00	23 ± 2	<0.4	0.7 ± 0.2	33 ± 10
2011/3/30 9:00	7 ± 1	1.0 ± 0.2	0.8 ± 0.2	9 ± 3
2011/3/31 9:00	7 ± 1	2.3 ± 0.2	2.5 ± 0.2	3 ± 1
2011/4/1 6:00	5 ± 1	<2	2.0 ± 0.7	3 ± 1
2011/4/5 7:30	4 ± 1	<2	<2	
2011/4/5 16:00	2 ± 1	<2	<3	
2011/4/6 16:00	3 ± 1	<0.5	<0.5	
2011/4/7 16:00	<2	<2	<3	

**Corresponding to the radioactivity at the time of sampling.*

obtained, the characteristics of radioactivity concentration variations for the three radioactive nuclides are determined, and their source and behavior are considered. Numerous data have become available for radioactive materials released by the FNDPP accident in environmental and food samples. However, radioactivity data for river samples are limited, especially for samples taken in March 2011. Therefore, the data obtained in this study must be highly beneficial in considering the release and the distribution of radioactive materials in the environment from the FDNPP accident, especially in Tokyo and its surrounding area.

EXPERIMENTAL

Sampling of river water

River water was sampled from three major rivers in or surrounding Tokyo Metropolis; the Edogawa, the Arakawa, and the Tamagawa Rivers, from which most water consumed in Tokyo Metropolis is obtained. Water from the Edogawa, the Arakawa, and the Tamagawa Rivers was sampled at Kanamachi, Asaka, and Hamura, respectively. River water was sampled in a polypropylene bottle (500 mL) without filtration. After bottling, a few mL of 1 M NaOH solution was added to prevent the dissolved iodine from being lost by evaporation. In addition to these three major sampling sites, river water was sampled at the Fujiokaohashi and Mikunibashi Bridges on the Watarasegawa River, and Sekiyadobashi Bridge on the Edogawa River. Sampling sites of river water are shown in Fig. 1, where major rivers concerned in this study are also shown.

Gamma-ray spectrometry

The water sample was transferred to Tokyo Metropolitan University (TMU) on the day of or one day after the sampling. Samples were assayed to gamma-ray counting by using a coaxial-type Ge semiconductor detector at Radioisotope Research Laboratory of TMU. Relative

Table 2. Radioactivity (Bq/L) in the Arakawa River water sampled at Asaka*

Sampling Date and Time	^{131}I	^{134}Cs	^{137}Cs	^{131}I/^{137}Cs
2011/3/28 8:30	11 ± 1	0.8 ± 0.2	0.7 ± 0.5	16 ± 11
2011/3/29 9:00	8.2 ± 0.9	<0.5	<0.4	
2011/3/30 9:00	6 ± 1	0.6 ± 0.2	0.4 ± 0.2	15 ± 8
2011/3/31 9:00	3 ± 1	2.0 ± 0.2	1.4 ± 0.2	2 ± 1
2011/3/31 22:00	5 ± 1	<2	<2	
2011/4/4 17:00	5 ± 1	<0.5	<0.5	
2011/4/5 17:00	<2	<0.5	0.6 ± 0.3	
2011/4/6 17:10	2 ± 1	<0.5	<0.5	
2011/4/7 17:05	3 ± 1	<2	<2	

**Corresponding to the radioactivity at the time of sampling.*

Table 3. Radioactivity (Bq/L) in the Tamagawa River water sampled at Hamura*

Sampling Date and Time	^{131}I	^{134}Cs	^{137}Cs
2011/3/28 9:00	<0.6	<0.4	<0.4
2011/3/29 9:00	<2	<0.5	<0.5
2011/3/30 9:00	<2	0.6 ± 0.2	0.4 ± 0.2
2011/3/31 9:00	<2	1.5 ± 0.2	0.9 ± 0.2
2011/4/1 8:00	<2	<2	<2
2011/4/4 21:30	<2	<3	<2
2011/4/5 21:30	2 ± 1	<2	<2
2011/4/6 21:30	<2	<2	<2
2011/4/7 21:30	<2	<2	<3

**Corresponding to the radioactivity at the time of sampling.*

counting efficiency and energy resolution of the Ge detector used are 25 % and 2 keV, respectively, for 1332 keV ^{60}Co gamma-ray. The water sample was all measured for 3600 s. Some samples were remeasured for 12 h to 24 h to obtain improved counting statistics, lowering the detection limit. To convert count rates (cps) of gamma-ray emitted by a nuclide of interest to activity concentrations in Bq/L, a function of counting efficiency vs. gamma-ray energy was determined using a homemade reference solution (500 mL in dil. HCl) containing known amounts of 9 radioactive nuclides such as ^{51}Cr, ^{54}Mn, ^{57}Co, ^{60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{137}Cs, and ^{139}Ce (7900, Eckert & Ziegler). The function (with logarithmic values for both parameters) consisted of a quadric function and a liner function in low energy (<200 keV) and high energy (>200 keV) regions, respectively.

Results and Discussion

Sampling of river water started on March 28, 2011 and has been continued since then. By using gamma-ray spectrometry, three radioactive nuclides, ^{131}I, ^{134}Cs and ^{137}Cs were confirmed to be present in river water samples collected at the three sampling sites (Kanamachi, Asaka and Hamura) in March. Activity concentrations (in Bq/L) of these three nuclides in water samples collected from March 28 to April 7, 2011 at these sampling sites are summarized in Tables 1, 2 and 3, respectively. In these tables, hour and date of the water sampling are shown. The gamma-ray counting normally started in three hours after sampling. The activity values given in Tables 1 to 3 correspond to those at the time of sampling. An uncertainty coupled with each value of activity concentration is due to counting statistics (1σ). An upper limit is defined to be a value corresponding to 3σ of background counts at the peak area of a corresponding gamma-ray (365 keV for ^{131}I, 605 keV for ^{134}Cs and 662 keV for ^{137}Cs). Under the present condition of gamma-ray spectrometry, typical values of detection limits are 2 Bq/L and 0.5 Bq/L for 3600 s and 12 h countings, respectively. The gamma-ray counting data for raw water sampled after April 12 at three water purification plants of the Bureau of Waterworks, Tokyo Metropolitan Government, which is locate near the three sampling sites of river water measured in this study, are open to the public on the web site (BW, 2011b). After April 12, concentrations of radioactive nuclides mentioned above were lower than the detection limit (2 Bq/kg to 3 Bq/kg) except for a few samples (BW, 2011b).

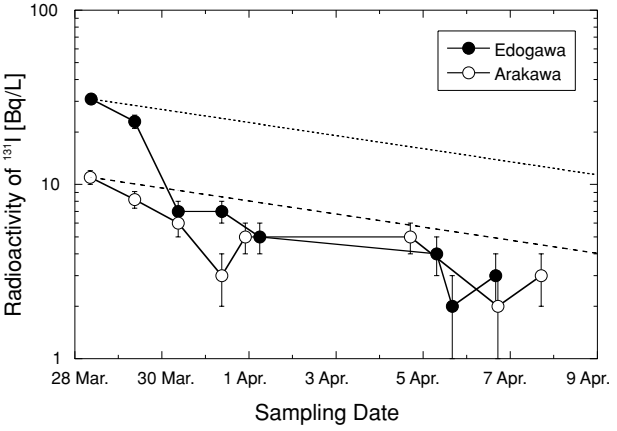

Fig. 2. Time-series changes of the ^{131}I radioactivity concentrations (in Bq/L) in river water sampled at Kanamachi on the Edogawa River and Asaka on the Arakawa River. Dotted and broken lines indicate the decay curve of ^{131}I with a half-life of 8.02 d.

Concentration variation of ^{131}I in water samples collected in late March to early April

Among the three radioactive nuclides detected in the water samples measured in this study, ^{131}I recorded the highest radioactivity concentration; 31 Bq/L and 11 Bq/L of ^{131}I were detected in water samples collected on March 28 from the Edogawa River and the Arakawa River, re-

Table 4. Radioactivity (Bq/L) in the water sampled upstream of the Edogawa River*

Site	Sampling Date and Time	^{131}I	^{134}Cs	^{137}Cs	^{131}I/^{137}Cs
Fujiokaohashi Bridge	2011/3/31 10:15	24 ± 2	1.8 ± 0.2	1.9 ± 0.3	13 ± 2
(Watarasegawa River)	2011/4/9 9:20	1.4 ± 0.4	<0.4	<0.5	
	2011/4/10 9:55	<2	<2	<2	
	2011/4/12 10:15	<2	<2	<2	
Mikunibashi Bridge	2011/3/31 10:40	13 ± 1	1.6 ± 0.3	1.5 ± 0.3	9 ± 2
(Watarasegawa River)	2011/4/9 10:25	<2	<1	<2	
	2011/4/10 11:00	2.3 ± 0.8	1.7 ± 0.4	2.2 ± 0.6	1.0 ± 0.5
Sekijyadobashi Bridge	2011/3/31 11:30	2.2 ± 0.6	<0.5	<0.5	
(Edogawa River)	2011/4/9 11:15	<2	<2	<2	
	2011/4/10 11:45	<2	<3	<2	
	2011/4/12 12:15	<2	<2	<2	

**Corresponding to the radioactivity at the time of sampling.*

spectively. In contrast, ^{131}I activity in river water sampled at Hamura along the Tamagawa River was lower than the detection limit (<0.6 Bq/L). Concentration changes of ^{131}I radioactivity in water samples collected from the Edogawa River and the Arakawa River from March 28 to April 7 are shown in Fig. 2. For comparison, decay curves of ^{131}I ($T_{1/2}$ = 8.02 days) are also shown. Concentration of ^{131}I decreased drastically from March 28 to March 30 in the Edogawa River. A similar but less drastic decrease was observed for the Arakawa River water. After March 31, the ^{131}I concentration decreased gradually for both rivers and their decrease changes appear to be consistent with the decay rate of ^{131}I.

The Edogawa River is one of branches of the Tonegawa River, the second longest river in Japan that originates in northern Gunma. Leaving the main stream of the Tonegawa River, the Edogawa River flows through between Tokyo and Chiba and reaches to Tokyo Bay. The sampling site of the Edogawa River water, Kanamachi, is located on the border of Tokyo, Chiba and Saitama. Based on the concentration map of ^{134}Cs and ^{137}Cs in farmland soil samples, Gunma Prefecture, especially the northern mountain area, was heavily contaminated with radioactive materials after the FDNPP accident (Gunma Prefecture, 2012). The radioactivity survey carried out by aircraft confirmed an areal heterogeneous distribution of gamma-ray-emitting radioactive materials (MEXT, 2011). The Tonegawa River originates in and passes through such contaminated area in Gunma. The aircraft survey further confirmed that west and south part of the Kanto district (Kanagawa, Tokyo, Chiba and Saitama) was scarcely contaminated with radioactive materials with an exception of several patchy areas in northwest Chiba and eastern Tokyo (neighboring to Chiba), which were relatively heavily contaminated with radioactive materials compared with major area of Saitama and Tokyo. As the Edogawa River passes through these contaminated areas, ^{131}I in the Edogawa River must have been highly affected by the radioactive materials distributed in such areas in addition to the basin of the main stream, the Tonegawa River.

The Arakawa River originates in Chichibu Mountains, western Saitama and crosses Saitama Prefecture and then eastern Tokyo, finally flowing into Tokyo Bay. The sampling site of the Arakawa River water, Asaka, is located in central Saitama. The Arakawa River is artificially linked to the Tonegawa River through the Musashi-suiro Channel, which is located in the upstream from Asaka and water flows from the Tonegawa River down to the Arakawa River. Thus, some or most ^{131}I observed in the water sampled at Asaka likely originated from the Tonegawa River.

^{131}I concentrations in river water sampled at three additional sampling sites (Fujiokaohashi Bridge, Mikunibashi Bridge and Sekiyadobashi Bridge) are summarized in Table 4. Data are available only for selected dates after March 31 for these three sampling sites; no data are available for March 28. The first two sampling sites are on the Watarasegawa River, which is a branch stream of the Tonegawa River and whose basin was fairly contaminated with radioactive materials. Although Sekiyadobashi Bridge is over the Edogawa River, it is located at the fork of the Tonegawa River and the Edogawa River. Obviously, ^{131}I concentration values for water samples taken from the Watarasegawa River are higher than that from the Edogawa River on March 31. Furthermore, the river water sampled at the upper Watarasegawa River exhibited higher ^{131}I concentration than that sampled at the lower, confirming that the ^{131}I concentration in the river water was highly affected by the radioactive materials spread over the basin of the corresponding river.

Source and behavior of ^{131}I detected in water samples from several rivers in the Kanto Plain

Two trends of the ^{131}I concentration change occur from March 28 to April 7 (Fig. 2). The ^{131}I concentration decreased abruptly from March 28 to 30 and then decreased gradually until April 7. As confirmed in Fig. 2, the degree of decrease for the first trend apparently differs between the Edogawa River water and the Arakawa River water, while no difference in the ^{131}I concentration change can be observed between the two rivers for the second trend. The first trend can be retroacted to an extremely high concentration of ^{131}I (210.5 Bq/kg water; Nagakawa *et al*., 2012) in purified water at the Kanamachi water filtration plant measured on March 22. Notably, the ^{131}I concentration could be decreased down to approximately 50% at the Kanamachi water purification plant on the Edogawa River (personal communication). According to the monitoring data of radioactive nuclides in fallout including rain at Shinjuku (TMIPH, 2011a), daily ^{131}I inventory was approximately 50 Bq/m^2/day from March 18 to March 20 and suddenly increased from 2900 Bq/m^2/day up to 36000 Bq/m^2/day in the next few days. The gamma-ray dose rate (TMIPH, 2011b) also increased abruptly from March 21. This coincident increase of ^{131}I concentration in fallout and gamma-ray dose rate is likely to have been caused by rainfall on March 21 to 23. The high concentration of ^{131}I in water purified at Kanamachi measured on March 22 was apparently caused by the Edogawa River water in which ^{131}I-enriched rain precipitating from March 21 to the following day in the area of east Tokyo and northwest Chiba flowed. ^{131}I was also distributed also in the watershed of the Tonegawa River in Gunma and Tochigi as described above. Thus, such ^{131}I must have streamed into the Edogawa River but its contribution could not be significant because ^{131}I concentration in purified water did not increase from March 22 to 28 (Nagakawa *et al*., 2012). This fact further suggests that relatively high concentration of ^{131}I in the Edogawa River water observed from March 28 to 30 primarily originated from the fallout from March 21 to 23 in the Tokyo-Chiba area.

After the sudden decrease of ^{131}I concentration in the Edogawa River water (and to a lesser extent, in the Arakawa River water) until March 29, ^{131}I concentrations in both rivers decreased gradually in late March 30 to early April, characterizing the second trend in Fig. 2. The decrease rate of ^{131}I activity appears to follow the radioactive decay of ^{131}I with a half-life of 8.02 d, which is drawn in Fig. 2 for reference. From these two trends confirmed in the ^{131}I concentration change in water samples, we speculate the following concerning the behavior of radioactive ^{131}I:

(i) ^{131}I was released from FDNPP to the surrounding environment, possibly in the forms of gas (I_2 and CH_3I; e.g., Santschi *et al*., 1988) and tiny solid particles. Such radioactive substances including ^{131}I were transferred from FDNPP to the Kanto district from March 15. The second largest increase of ^{131}I activity in aerosol was recorded in the downtown area of Tokyo on March 21 (Nagakawa *et al*., 2012).

(ii) Coincidentally, rain occurred in the vicinity of Tokyo in the morning on March 21 (starting at 7 a.m. in Shinjuku). ^{131}I and other radioactive nuclides such as ^{132}Te, ^{134}Cs and ^{137}Cs present in the atmosphere in aerosol form, in gaseous form, or in both, were deposited to the ground by rain. The deposition of ^{131}I on the ground occurred not only by wet deposition but also by dry deposition (Morino *et al*., 2011).

(iii) A part of ^{131}I deposited on the ground with rain was trapped in soil. ^{131}I remaining in the surface water was effectively transported to Tokyo Bay (or to Pacific Ocean) via rivers in one week.

(iv) From March 22 to March 29, the ^{131}I concentration in river water greatly decreased. Considering that stable iodine is more abundant in river water than that in rain water (Fuge and Johnson, 1986; Kametani *et al*., 1992), the $^{131}I/^{127}I$ (specific activity of ^{131}I) in rain water must have also decreased by diluting the radioactive ^{131}I with stable iodine (^{127}I).

(v) From March 30, the decay-corrected specific activity of ^{131}I in the Edogawa River water (and the Arakawa River water) became constant. Probably, some ^{131}I remaining on the land was gradually brought to rivers with isotopic equilibration between ^{131}I and ^{127}I.

Temporal variations of ^{134}Cs and ^{137}Cs concentrations and $^{131}I/^{137}Cs$ activity ratios in water samples collected in late March to early April

Concentrations of ^{134}Cs and ^{137}Cs in water samples collected from the three rivers concerned in this study were in the level of their detection limits; ~2 Bq/L for 3600 s counting and ~0.5 Bq/L for 12 h counting. The highest concentration was observed to be 2.5 ± 0.2 Bq/L for ^{137}Cs in the Edogawa River water on March 31. An activity ratio of $^{134}Cs/^{137}Cs$ for this sample is approximately one, which is consistent with the values reported for radiocesium derived from the FDNPP accident (e.g., Kinoshita *et al*., 2011; Sakaguchi *et al*., 2012; Yamamoto *et al*., 2012), suggesting that ^{134}Cs and ^{137}Cs detected in river samples of this study originated from the FDNPP accident. These radiocesium nuclides must have become aerosols near the FDNPP accident site and transferred to the surrounding environment, which were then deposited on the ground as fallout. Daily measurements of fallout collected at Shinjuku indicated a sudden increase of ^{134}Cs and ^{137}Cs concentrations on March 20 and high concentrations of these nuclides until March 24, becoming similar to the case of ^{131}I (TMIPH, 2011a). However, as seen

in Table 1, the concentration change of ^{137}Cs for the river water from the Edogawa River is completely different from that for ^{131}I, implying that ^{137}Cs and ^{131}I once present together in aerosol behaved differently after the deposition to the ground as fallout with rain.

Following the radioactivity measurement of fallout sampled at Shinjuku, 83900 and 6310 Bq/m^2 were obtained for ^{131}I and ^{137}Cs, respectively, as total activity values for four days from March 20 at 9 a.m. to March 24 at 9 a.m. (TMIPH, 2011a). If (i) ^{137}Cs and ^{131}I behaved similarly during the transportation to river water and (ii) their concentration ratios in fallout from March 20 to 24 are not variable over the Tokyo Metropolis including Kanamachi, the ^{137}Cs concentration in the river water sampled on March 28 at Kanamachi would have been approximately 5 Bq/L, which is roughly an order of magnitude higher than the measured value (0.6 Bq/L). Apparently, ^{137}Cs was much less effectively transferred to the Edogawa River compared with ^{131}I. From these observations, we speculate the following concerning the behavior of radioactive ^{137}Cs and ^{134}Cs:

(i) Radiocesium nuclides were transported in particulates from the FDNPP accident site, and were deposited on the ground with rain from March 20 to March 22.

(ii) With these particulates deposited on the ground being transported to surface water either in the dissolved or particulate forms, ^{137}Cs can enter the river water system. However, ^{137}Cs can be effectively adsorbed on soil particles such as clay minerals, making ^{137}Cs remaining on the ground to a much larger degree than ^{131}I (e.g., Kato *et al*., 2012).

As shown in Tables 1 to 3, ^{134}Cs and ^{137}Cs concentrations peaked on March 31 (morning) for river waters from the Edogawa, the Arakawa, and even the Tamagawa Rivers. High concentrations of ^{134}Cs and ^{137}Cs were also observed on March 31 at two sampling sites (Fujiokaohashi Bridge and Mikunibashi Bridge) of the Watarasegawa River. Coincidentally, relatively high radioactivities of ^{134}Cs and ^{137}Cs were observed in the fallout recovered in Shinjuku. As discussed below, the radioactivity of ^{131}I was not proportionally high in the same fallout. These observations imply that aerosol containing ^{134}Cs and ^{137}Cs were rather widely dispersed over the Kanto Plain and deposited on the ground as fallout on March 30 to March 31 and that ^{134}Cs–^{137}Cs and ^{131}I were fractionated in such aerosol, with ^{131}I being depleted.

Following the systematic survey of radioactive nuclides dispersed by FDNPP accident including ^{131}I, ^{134}Cs and ^{137}Cs in soil samples (Kinoshita *et al*., 2011), ^{134}Cs and ^{137}Cs were similarly distributed to ^{131}I but their activity ratios represented by ^{131}I/^{137}Cs were not exactly identical. Daily measurements of fallout collected at Shinjuku indicated a sudden increase of ^{134}Cs and ^{137}Cs concentrations for March 19 to March 24, being similar to the case of ^{131}I (TMIPH, 2011a). The activity ratio of ^{131}I/^{137}Cs, however, changed with increase from 5.2 (=2900/560) for 24 h from March 20 at 9 a.m. to 109 (=36000/330) for 24 h from March 23 at 9 a.m. (numerical values given in fractions are in Bq/m^2/d). The ratio then decreased to approximately 1 to 10 in the period from March 24 to April 1. Apparently, ^{131}I and ^{137}Cs in fallout were largely fractionated.

Activity ratios of ^{131}I/^{137}Cs in river water samples are given in Tables 1, 2, and 4 (no such ratios are given in Table 3 because no meaningful values are available). Although large uncertainties (1σ) (propagated values based on uncertainties due to counting statistics of individual count rates) are attached to individual ratios, several trends can be observed. The change in the activity concentration of ^{137}Cs was much smaller than that of ^{131}I. Therefore, a similar case seen in the change of the activity concentration of ^{131}I can be confirmed; a drastic decrease of the ratio from March 28 to 30, followed by a steady value, and a larger decrease at Kanamachi on the Edogawa River compared with the change at Asaka on the Arakawa River. The largest ratio of 52 (±18) at Kanamachi on March 28 is an order of magnitude higher than the ratio (8.4) observed in fallout at Shinjuku collected on the same day. A less clear but similar trend can be seen in the comparison of ^{131}I/^{137}Cs activity ratios between the river water at Asaka and the fallout at Shinjuku. Notably, ^{131}I and ^{137}Cs were fractionated during the transportation from the falling site of aerosol to the sampling site of river water. Furthermore, ^{131}I is more effectively transferred into rivers than ^{137}Cs.

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