Distribution and Behavior of Radiocaesium in Water and Sediment of Retention Basins at Kashiwa City, Chiba Prefecture

Yujiro Ichikawa, Toshiaki Nakata, Tomohiro Inoue, Mayu Namekata, Eri Honda, Hideo Ishii, Akira Imura, Yoko Fujimura, Suekazu Naito, Tsutomu Tanaka and Yoshihiko Takahashi

Chiba Prefectural Environmental Research Center
1–8–8 Iwasakinishi, Ichihara-shi, Chiba Pref. 290–0046, Japan
y.ichkw17@pref.chiba.lg.jp

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According to the research conducted by the Government of Japan and Chiba Prefectural Government, the sediments of Lake Tega and its tributaries are still highly contaminated by radiocaesium (over 1,000 Bq/kg) derived from the Fukushima Daiichi Nuclear Power Plant. To investigate the possible sources that would transport and redistribute radiocaesium to the Lake Tega system, which is urbanized compared to Fukushima Prefecture, we have selected three retention basins, corresponding to upstream of Lake Tega, located in Kashiwa City, Chiba Prefecture. Temporal changes in concentrations of radiocaesium in water and sediment samples were determined during 2013–2014. The concentrations of radiocaesium in both samples tended to decrease during the sampling period, although some of the sediment samples were still high radiocaesium concentrations exceeding 10 kBq/kg. These observations suggest that the radiocaesium in the retention basins, which play an important role in supplying radiocaesium to the downstream area, is highly contaminated compared to the downstream area. Increments of the concentrations of radiocaesium in particulate form were observed from the water samples collected immediately after the passage of large typhoon Wipha. This result implies that heavy rainfall events may be playing an important role in the migration of existing radiocaesium from the catchment areas to the Lake Tega system.

Key Words: Fukushima Daiichi Nuclear Power Plant, retention basins, radiocaesium in water, radiocaesium in sediment, Kashiwa City, Chiba Prefecture

1. Introduction

On March 2011, the Great East Japan Earthquake and the following tsunami induced the accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP). A massive amount of radionuclides were released into the environment, causing serious widespread contamination throughout Eastern Japan. As a research project consigned from the Government of Japan, the Japan Atomic Energy Agency (JAEA) has played the central role for monitoring, and the assessment of the distribution of radioactive contaminants and their behavior in the environment, all of which have been proceeded. However, the target region of this project is mainly within the Fukushima Prefecture, and then there are a few monitoring data and/or knowledge on the behaviors of the FDNPP-derived radionuclides for other prefectures.

The Government of Japan and JAEA constructed detailed deposition density maps of the FDNPP-derived radionuclides. From the point of view of the longer-term influence of radioactive contamin-

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tion, the most important γ-ray emitting radionuclides derived from FDNPP are $^{134}\text{Cs}$ (half-life of 2.1 years) and $^{137}\text{Cs}$ (half-life of 30.2 years), and these two radioisotopes will hereafter be focused upon in this study. Such maps showed relatively high activities of radioisotopes ($^{134}\text{Cs} + ^{137}\text{Cs}$; 30–100kBq/m²) in a region of northwestern Chiba Prefecture at an initial stage of monitoring. This region includes the Intensive Contamination Survey Area (ICSA), where decontamination actions of municipalities are desired, because an estimated additional annual cumulative dose for individuals is between 1mSv and 20mSv. Eight municipalities are designated as ICSA in northwestern Chiba Prefecture. Among them, Kashiwa City, Matsudo City, Noda City, Nagareyama City, and Abiko City are within 20–40 km from the central part of Tokyo. The estimated total population of 5 cities sums up to 1,352,632 (accounting for approximately 21.8% of the total population of Chiba Prefecture), which indicates that these cities are densely-populated suburbs.

However, more than 4 years passed since the FDNPP accident, the concentrations of radioisotopes exceeding 1,000 Bq/kg have still been observed in the sediments of Lake Tega and its largest tributary, the Ohori River, which streams across Nagareyama City and Kashiwa City. These observations suggest that radioisotopes is supplied from the upstream sector of the Ohori River system. In order to protect residents in the densely-populated suburbs, more detailed understanding of the distribution of radioisotopes and their behavior in the urbanized area are needed. Therefore, it is important to elucidate the possible sources that would transport and redistribute radioisotopes to the Ohori River system.

In this paper, we describe the distribution and migration behaviors of radioisotopes in three types retention basins in Kashiwa City, which is upstream of the Ohori River and located in the urbanized area of the Kanto region, Japan.

2. Materials and experiments

2.1 Studied sites

Three types of retention basins at Kashiwa City, whose drainage water flows to the Ohori River, were selected as sampling sites. The overhead view map illustrated in Fig. 1 shows the locations of the sampling sites. As a matter of convenience, the respective basins will be named K1 basin, K2 basin, and K3 basin, hereafter in accordance to Fig. 1. Their geographical coordinates, capacity, and surface area are summarized in Table 1.

![Map indicating the location of three retention basins (K1 basin, K2 basin, and K3 basin) where the studies were conducted.](source)
Schematic diagrams of the retention basins and the sampling stations are drawn in Fig. 2. These basins are classified as on-site storage excavated basins, which are artificially constructed to avoid any flood damage to the downstream area. They are also designed not to exceed the planned maximum drainage flow, which prevents exciscation and submergence of the facilities. The K1 basin and the K2 basin consist of 3 inlet pipelines and 1 outlet pipeline. The K3 basin consists of 1 inlet pipeline and 1 outlet line.

Housings, parks, and forestlands are included in the surrounding environment of the K1 and K2 basins, whereas the environment around the K3 basin is commercially developed area.

2.2 Studied periods

Rainfall events may affect the transport and redistribution of existing radiocaesium from the catchment areas to the retention basins. In early summer, most parts of Japan get visited by a rainy season,

| Site   | Latitude            | Longitude           | Capacity (m³) | Surface Area (m²) |
|--------|---------------------|---------------------|---------------|-------------------|
| K1 basin | 35°53′56.46″N | 139°56′35.79″E | 99,660        | 31,641            |
| K2 basin | 35°53′40.40″N | 139°56′06.29″E | 136,157       | 48,948            |
| K3 basin | 35°52′52.12″N | 139°58′08.56″E | 15,511        | 1,270             |

Fig. 2  Diagrams indicating outlines of the respective basins and locations of the sampling stations. (a) K1 basin, (b) K2 basin, (c) K3 basin.
namely tsuyu. From statistical data provided by the Japan Meteorological Agency (JMA) in the Kanto region of Japan, tsuyu lasts from the beginning of June to the middle of July. Also, typhoons, which originate over the Northwest Pacific Ocean and are accompanied by strong winds and heavy rainfall, frequently hit Japan from July to October.

To understand the behavior of radiocaesium due to rainfall events, studied periods were planned according to Table 2. Studied periods were divided into 5 periods as follows: period 1, before tsuyu; period 2, after tsuyu; period 3, immediately after the passage of large typhoon Wipha; period 4, after typhoon season; and period 5, one year after period 4.

The precipitation data used in this study was obtained from Abiko meteorological observing station, which is approximately 15.7 km, 15.2 km, and 12.3 km from the K1, K2, and K3 basins, respectively. In 2013, JMA announced the starting date and ending date of tsuyu in the Kanto region as June 10 and July 6, respectively. The total amount of precipitation observed during tsuyu in 2013 was 182 mm. On October 16 2013, large typhoon Wipha struck the Kanto region accompanied by extremely heavy rainfall. The recorded precipitation amount was 196.5 mm in a single day. Study during period 3 was conducted 2 days after the passage of Wipha. The total amount of precipitation between periods 2 and 4, and between periods 4 and 5, was 843 mm and 1,464 mm, respectively. All of the sampling procedures, the details of which are described in the following sections, were conducted in basically calm weather.

2.3 Water sampling and treatment

Water was collected at fixed sampling stations as shown in Fig. 2. The sampling stations were located adjacent to the inlets and outlets of the respective basins. A central point was selected just for the K3 basin. In case the sampling stations were shallow and easily approachable, 5 L of surface water was gently collected by ladle to avoid scattering sediment lying on the basin bottom. At sampling stations that were not easily approached, 5 L of surface water was collected by means of a string-fixed bucket. All the water samples were sealed in a polyethylene container and stored in a refrigerator at 4°C until treatment.

Radioaesium in the water samples are divided into roughly two different types, dissolved and particulate forms. For the separation of dissolved and

| Period | Dates          | Instructions                              |
|--------|----------------|-------------------------------------------|
| Period 1 | May 15 and 23 2013 | Before tsuyu*                            |
| Period 2 | Aug. 26 and 28 2013 | After tsuyu                               |
| Period 3 | Oct. 18 2013     | 2 days after the passage of large typhoon Wipha |
| Period 4 | Dec. 2 and 4 2013 | After typhoon season                      |
| Period 5 | Nov. 4 and 5 2014 | Approximately 1 year after period 4       |

* The starting and ending date of tsuyu in 2013 were June 10 and July 6, respectively.
particulate forms, all water samples were filtered through a glass fiber filter (Toyo Roshi Kaisha Ltd., GS-25, φ47 mm) with a pore size of 1 μm. The water samples of period 5 were further filtered through membrane filters (Merck Millipore Ltd., φ47 mm) with pore sizes of 0.45 μm, 0.22 μm, and 0.1 μm, in turn. Filter papers were placed over silica gel in a desiccator cabinet at room temperature to dry. The mass concentrations (mg/L) of particulate substances trapped on the filters were measured. Thus the concentration of particulate radiocaesium was calculated on both a volume basis (mBq/L) and weight basis (kBq/kg-dry weight) for data analysis.

Filtrates were gradually concentrated by electric heater at a temperature of ≦80°C until the volume reached under 100 mL. Filter papers and concentrated filtrates were sealed in a 100 mL polypropylene container (U-8 hereafter) to measure the activity of radiocaesium with a germanium semiconductor (Ge) detector.

2.4 Sediment sampling and treatment

The sediment samples were collected at fixed sampling stations indicated in Fig. 2. An Ekman-dredge sampler or handy shovel was used for the sediment sampling, and the sediments from the top 3 cm were subjected to radioactivity measurements. They were enclosed in a polyethylene sealed bag and stored in a refrigerator at 4°C until treatment.

The sediments were screened up to a pore size of 2 mm by means of a wet sieving method. Thereafter, sediments that passed through a 2 mm screen were dried in an oven at 105°C for 24h. The dried sediments were used to measure the following: 1) carbon content (%); 2) ignition loss (%); 3) clay, silt, and sand content (%) to determine specific surface area; and 4) activity of radiocaesium (kBq/kg-dry weight).

Organic carbon content was quantified with a CN analyzer (Yanako Inc., MT-700). Ignition loss was calculated by the difference between the measured weight of the dried sediment treated in an oven at 600°C for 1 h and the initial weight. These two values were used to indicate the organic matter content of sediment.

To analyze the clay, silt, and sand content of sediment, the particle size composition of the sediments was measured using a laser diffraction particle size distribution analyzer (Horiba Ltd., LA-300). This analyzer can screen particles ranging in size from 0.1–600 μm. The particle size composition of the >600 μm fraction was obtained by sieving and weighing in advance of the analysis with the laser diffraction particle size distribution analyzer.

The same sediments that were used to measure the concentrations of radiocaesium was next sealed in the U-8 container and introduced to a Ge detector.

2.5 Radioactivity measurement

All the samples prepared for the measurement of the concentration of radiocaesium were analyzed with a Ge detector (Canberra Industries Inc., GC2520, P-type), which is coupled to a multi-channel analyzer (Canberra Industries Inc., DSA1000). The detector’s resolution was 1.75 keV, which is determined by the full width at half maximum of 60Co energy peak 1,332 keV, and the relative detection efficiency was 32.9%.

The γ-ray counting efficiency of the Ge detector was estimated by γ-ray counting efficiency curve which was constructed by measuring certified γ-ray source samples (prepared and supplied by Japan Chemical Analysis Center) of 109Cd, 57Co, 134Ce, 51Cr, 137Cs, 54Mn, 88Y, 59Fe, and 60Co at 5 different axial distances (1 cm up to 5 cm) from the surface of detector. These source samples were sealed in the U-8 container with known weight. The specific energy peaks for individual radionuclides were analyzed and quantified with Spectrum Explorer software (Canberra Industries Inc., version 1.72). Energy peaks of 605 keV and 662 keV were selected to
quantify $^{134}$Cs and $^{137}$Cs concentrations, respectively. The radioactive decay for $^{134}$Cs and $^{137}$Cs concentrations were corrected at each sampling date. Also, the cascade summing effect for $^{134}$Cs was corrected according to the software program.

### 3. Results

#### 3.1 Temporal variation of the concentration of radiocaesium in the water samples

Tables 3 and 4 show the observed concentrations of dissolved and particulate radiocaesium in the respective basins. Also the temporal variations of the dissolved and particulate $^{137}$Cs are shown in Fig. 3. The arithmetical means of activity ratios of $^{134}$Cs/$^{137}$Cs (decay-corrected to March 21, 2011) calculated from overall samples collected during this study were $1.04 \pm 0.03$ ($n=52$) and $1.03 \pm 0.06$ ($n=52$) for dissolved and particulate forms, respectively. The Kanto region was mainly contaminated by the FDNPP-derived radionuclides during March 21–23, 2011 and the activity ratios of $^{134}$Cs/$^{137}$Cs were nearly 1 at that time.\(^7\) This result indicates that most of the radiocaesium in the retention basins originated from the FDNPP accident.

#### Table 3 Concentration of dissolved radiocaesium (mBq/L)

| Sampling station | Period 1 | Period 2 | Period 3 | Period 4 | Period 5 |
|------------------|----------|----------|----------|----------|----------|
|                  | $^{134}$Cs $^{137}$Cs | $^{134}$Cs $^{137}$Cs | $^{134}$Cs $^{137}$Cs | $^{134}$Cs $^{137}$Cs | $^{134}$Cs $^{137}$Cs |
| 1:1              |          |          |          |          |          |
| 1:2              |          |          |          |          |          |
| 1:3              |          |          |          |          |          |
| 1:4              |          |          |          |          |          |
| 2:1              |          |          |          |          |          |
| 2:2              |          |          |          |          |          |
| 2:3              |          |          |          |          |          |
| 2:4              |          |          |          |          |          |
| 3:1              |          |          |          |          |          |
| 3:2              |          |          |          |          |          |
| 3:3              |          |          |          |          |          |
| 3:4              |          |          |          |          |          |

#### Table 4 Concentration of particulate radiocaesium

| Sample station | Concentration of dissolved radiocaesium (mBq/L) | Concentration of particulate radiocaesium (mg/L) |
|----------------|-----------------------------------------------|-----------------------------------------------|
| 1              | $^{134}$Cs $^{137}$Cs | $^{134}$Cs $^{137}$Cs |
| 2              | $^{134}$Cs $^{137}$Cs | $^{134}$Cs $^{137}$Cs |
| 3              | $^{134}$Cs $^{137}$Cs | $^{134}$Cs $^{137}$Cs |
| 4              | $^{134}$Cs $^{137}$Cs | $^{134}$Cs $^{137}$Cs |

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\(^7\) Not determined
Both concentrations of dissolved and particulate radiocaesium decreased gradually during the studied period (2013–2014). In addition, no correlations were found between the radiocaesium concentrations in the inlet and outlet.

### 3.2 Temporal variation of the concentration of radiocaesium in the sediment samples

Observed concentrations of radiocaesium in the sediment samples collected at the respective basins are shown in Table 5. The temporal variation of the concentration of $^{137}$Cs is shown in Fig. 4. The arithmetical mean of activity ratios of $^{134}$Cs/$^{137}$Cs (decay-corrected to March 21, 2011) calculated from overall samples collected during this study was $0.99 \pm 0.03$ ($n=48$).

During the studied period, the concentrations of radiocaesium in the sediments decreased; however, the concentrations of $^{134}$Cs + $^{137}$Cs exceeding 10 kBq/kg were measured in the sediments of every retention basin. Even during period 5, which was about 3.5 years after the FDNPP accident, some of the concentrations of $^{134}$Cs + $^{137}$Cs in sediment samples still exceeded 10 kBq/kg.

### 4. Discussion

#### 4.1 Behavior and distribution of $^{137}$Cs in the water samples

From Fig. 3, especially if the data from period 3 is excluded, the average portions of dissolved and particulate $^{137}$Cs to the total were 58% and 42%, respectively, which tend to show higher values in dissolved
form than particulate one. This result corresponds with observations of the Ohori River in calm weather as reported by Yamashita et al.8) However, from the research conducted at the Fukushima Prefecture side of the Abukuma River, opposite results were obtained.1) These differences could be attributed to differences in land use. Land use profiles show that most of Kashiwa City is residential (housing 53.2%, roads 1.5%),8) in contrast to Fukushima Prefecture, where 71% of the total area is forest.9) These profiles

| Sampling station | Period 1 | Period 2 | Period 5 |
|------------------|----------|----------|----------|
|                  | $^{137}$Cs | $^{137}$Cs+ $^{134}$Cs | $^{137}$Cs |
| 1-1              | 5.1±0.1  | 15.3±0.1 | 10.8±0.0 |
| 1-2              | 5.9±0.2  | 17.8±0.3 | 6.7±0.1  |
| 1-3              | 9.2±0.2  | 28.0±0.4 | 13.1±0.1 |
| 1-4              | 9.0±0.1  | 26.4±0.1 | 24.7±0.1 |
| 2-1              | 2.9±0.1  | 9.0±0.1  | 9.0±0.1  |
| 2-2              | 6.0±0.0  | 18.3±0.1 | 23.8±0.1 |
| 2-3              | 2.9±0.1  | 6.3±0.1  | 10.9±0.1 |
| 2-4              | 3.2±0.1  | 11.6±0.1 | 5.9±0.0  |
| 3-1              | 18.8±0.3 | 56.3±0.6 | 44.4±0.1 |
| 3-2              | 4.1±0.1  | 12.8±0.1 | 14.2±0.1 |
| 3-3              | 15.3±0.1 | 45.4±0.1 | 12.8±0.1 |

Table 5 Concentration of radiocaesium in the sediment samples (kBq/kg)

Fig. 4 Temporal variation of concentrations of $^{137}$Cs in the sediment samples collected at individual sampling stations. (a) K1 basin, (b) K2 basin, (c) K3 basin.
demonstrate that Kashiwa City is more urbanized. Such factors may affect the behavior and distribution of $^{137}$Cs in the water environment.

Also, it was unclear whether the portion of the particulate $^{137}$Cs in retention basin waters might change when filter papers with different pore sizes were used. Unlike the other periods, all of the water samples that were collected during period 5 were filtered through filter paper with different pore sizes. To confirm whether these differences occur because of the different pore sizes of the filter papers, the data for the particulate $^{137}$Cs were compared both on a volume basis and weight basis (Fig. 5). Although there were a few samples exhibiting higher values with pore sizes of $0.1 \mu m$ compared to $1 \mu m$, the differences of the portion of the particulate $^{137}$Cs due to the difference of the pore size were not found to be significant based on the $t$-test ($n=11$). These results indicate that the particulate $^{137}$Cs contained in particles with the pore size of $>1 \mu m$ rather than fine particles ($0.1–1 \mu m$) that we tested ($0.1 \mu m$ and $1 \mu m$).

Sakaguchi et al. \cite{10} filtered the water samples collected at Abukuma River and Kuchibuto River with pore sizes of $125 \mu m$, $64 \mu m$, $3 \mu m$, $0.45 \mu m$, and with an ultrafiltration procedure, and the maximum contribution of $^{137}$Cs was found in the $3–63 \mu m$ pore-size fraction. Presuming from previous findings \cite{10} and our result, the contribution of particulate radiocaesium in the $1–0.1 \mu m$ fraction is small compared to the $>1 \mu m$ fraction. Various studies conducted at Fukushima Prefecture were filtering water samples in the $0.45 \mu m$ pore size.\cite{1, 11–14} In spite of differences in the pore sizes of filter paper, our results regarding particulate $^{137}$Cs in fresh waters are comparable with those in previous studies.

From the data obtained during period 3, the portions of dissolved and particulate $^{137}$Cs to the total were approximately 50% and 50%, respectively, which indicates a different tendency compared to the other periods. The massic concentrations of the particulate $^{137}$Cs obtained through this study are illustrated in Fig. 6. Figure 6 clearly shows that the massic concentrations of the particulate $^{137}$Cs in the period 3 were relatively higher than that in other periods. The arithmetical mean of the massic concentrations of the particulate $^{137}$Cs at the K1 basin and K2 basin calculated for period 3 was 2.9 times higher than that of period 2.

Field study during period 3 was carried out 2 days after the passage of large typhoon Wipha. Therefore, the water level was high and the average flow velocity was about $11 \text{ m/s}$, which is faster than the flow velocity of the other periods ($1–8 \text{ m/s}$). Thus the conditions of each basin during period 3 were different from those in calm weather, and it is apparent that an extremely heavy rain event increased the particulate $^{137}$Cs in the retention basin waters. Considering these observations, it is conceivable that large quantities of $^{137}$Cs in the retention basins, either remaining in the catchment area or redistributed from the sediment, could be migrated during heavy rain events.

A similar observation was obtained at the Natui
River and the Same River: the particulate radio-caesium were 21–56% of total radiocaesium in the normal flow condition but were close to 100% after the passage of large typhoon Roke on September 2011. Therefore, the migration of radiocaesium from the catchment area or the redistribution of sediment can also occur during heavy rain events, regardless of land use.

4.2 Identification analysis of the sources of particulate $^{137}\text{Cs}$

Numerous studies have been conducted at various sites to identify the origins of suspended particulate matter in water, which is thought to be important in the migration of radionuclides. We have previously published the concentration of radiocaesium in the soil and the paved road dust both collected at Kashiwa City. The soil samples were collected as part of a related study that was conducted on the same schedule and at the same sites. If runoff from the soil surface is contributing to the composition of suspended particulate matter in the water environment, there should be a certain correlation between the two. However, the correlation between the average massic concentration of the particulate $^{137}\text{Cs}$ (kBq/kg) and the 0–1 cm surface soil layer $^{137}\text{Cs}$ (kBq/m$^2$) was weak ($R=0.12$).

In January 2013, we measured the paved road dust from Kashiwa-no-ha Park, which is located adjacent to the studied retention basins. At then, the concentration of $^{137}\text{Cs}$ in the paved road dust was in range of 5.3–56.6 kBq/kg, with an arithmetical mean value of 27.9 kBq/kg. The entire paved road of Kashiwa-no-ha Park was later decontaminated using a high-pressure water jet. However, much of the public paved road still has not been decontaminated. It should be noted that even if the amount of road dust was small, the runoff would tend to concentrate road dust in particular locations, such as in the retention basins. Yamashita et al. point out that the contribution of $^{137}\text{Cs}$ transported via road dust to $^{137}\text{Cs}$ in the Ohori River water accounted for 39% of the total $^{137}\text{Cs}$. Since the road dust in the catchment areas of each retention basin were not collected/measured during this studied period, we were not able to evaluate the relationship between the particulate $^{137}\text{Cs}$ and road dust $^{137}\text{Cs}$.

The concentrations of $^{137}\text{Cs}$ in particulate matter were plotted to that in sediments, and the diagram and correlation coefficient ($R$) are shown in Fig. 7. The relationship between the concentrations of $^{137}\text{Cs}$
in sediment and particulate matter indicates a moderate correlation ($R = 0.50$). Thus, it is likely that chemical properties of the particulate $^{137}$Cs are similar to $^{137}$Cs in sediment. From this result, we speculated that one of the origins of particulate matter in the water environment is resuspended sediment.

4.3 Highly-concentrated radiocaesium in the sediment samples of K3 basin

The sediments at St. 3-1, the only sampling station among the retention basins, where samples were collected from the central part of the basin, exhibited relatively high concentrations throughout the studied period, and showed little decrease. The concentrations of $^{134}$Cs + $^{137}$Cs in the sediments measured at St. 3-1 were as follows: period 1, 56 kBq/kg; period 2, 40 kBq/kg; period 4, 44 kBq/kg; and period 5, 31.5 kBq/kg. These observations are one order of magnitude higher than that of the Ohori River and Lake Tega at corresponding period. The inlet and outlet pipelines of the K3 basin are structurally located close, as shown in Fig. 2(c). From the specific structure of the K3 basin, the flow of water at St. 3-1 could be stagnant compared to other sampling stations. Therefore, the higher radiocaesium might be remained in the sediment of St. 3-1 due to poor mixing process.

These observations suggest that the remaining radiocaesium in the retention basins are highly concentrated compared to the downstream area, which means that they are playing an important role in supplying radiocaesium to the downstream area. As a result, continuous monitoring of radiocaesium in the retention basins is required of countermeasures for reducing radioactive contamination and for radiological protection of the public.

4.4 Relationship between sediment characteristics and concentration of $^{137}$Cs

He and Walling performed adsorption experiments of $^{137}$Cs to the different-size particles of mineral soil and sediment samples. They showed that soil particles with smaller sizes (greater specific surface area) increase the adsorption of $^{137}$Cs, with a relationship that can be approximated by a power function with an exponent coefficient of 0.65. To verify their hypothesis, we quantified the clay, silt, and sand content and estimated the specific surface areas of all of the sediment samples collected in the retention basins with the laser diffraction particle size distribution analyzer. The specific surface areas were estimated by assuming that all particles had a uniform spherical shape. However, in this study, no correlation was seen between the concentration of $^{137}$Cs and the specific surface area of the sediment samples (Fig. 8). The same result was confirmed for the soil samples that were collected in Kashiwa City. We make this assumption based on the specific type of soil that was sampled, namely Kanto Loam, which covers many part of Kanto region and has vast numbers of pores, many that are on the surface and are less than 5 nm in diameter. Due to this particular structure, this soil shows behavior that is different to other soil types. Therefore, in this study, the specific characteristics of Kanto Loam may be the cause for the contradictory result with the report of He and Walling. Other specialized skills and knowledge would be needed to understand the mechanism of the adsorption of $^{137}$Cs to Kanto Loam. Hence, it is an issue that will be considered
in the future. Also supporting our result, Fan et al.\textsuperscript{14}) collected sediment samples at Kuchibuto River and concluded that particle size (specific surface area) may not be the dominant factor in the fixation of $^{137}$Cs.

Correlation analysis between the concentration of $^{137}$Cs and index values for organic matter content in sediments was carried out as shown in Fig. 9. Good positive correlations between the concentration of $^{137}$Cs and both carbon content ($R=0.75$) and ignition loss ($R=0.56$) were observed. Similar observations were obtained in the sediment samples of the Ohori River\textsuperscript{19}) and Lake Tega.\textsuperscript{20}) These results suggest that organic matter is playing an important role in the accumulation of $^{137}$Cs. In a research report on the vertical distribution of $^{137}$Cs in forest soil located at Fukushima Prefecture and sampled on September 2013,\textsuperscript{21}) 35–77\% of the overall $^{137}$Cs deposited on the forest floor still remained in the top organic matter rich soil layer, where concentrations of the $^{137}$Cs were about one order of magnitude higher than the underlying mineral soil. This indicates that much of the $^{137}$Cs accumulated in the organic matter-rich soil were adsorbed without elution or infiltration. The chemical and physical mechanisms controlling the relationship between organic matter and the accumulation of $^{137}$Cs is a topic for future research.

4.5 Distribution coefficients determined in the retention basins of urbanized area

Radionuclides deposited on land surface as a result of various nuclear power station accidents, as well as a large number of nuclear weapon tests conducted between the 1940s and 1980s, are thought to be eventually transported to the water environment. Radionuclides travel through the water environment mainly as dissolved and/or particulate forms. To evaluate the migration behavior of radionuclides in water environment via these two forms, the solid–liquid distribution coefficient (Kd) has been used extensively.\textsuperscript{22}) The distribution coefficient (Kd) between dissolved and particulate $^{137}$Cs is defined by the formula (1):

\begin{equation}
K_d = \frac{C_d}{C_p}
\end{equation}
\[
K_d = \frac{C_{\text{Solid}}}{C_{\text{Liquid}}} \quad (1)
\]

Where \( C_{\text{Solid}} \) is the concentration of radionuclides in particulate matter (Bq/kg) and \( C_{\text{Liquid}} \) is the concentration of dissolved radionuclides (Bq/L). The distribution coefficient is determined on the assumption of equilibrium between dissolved and particulate radionuclides. The distribution coefficient is not a unique value because it varies according to changes of chemical and mineral composition in particulate matter. This is a useful tool for comparing the relative behavior of \(^{137}\text{Cs} \) in water environment with other studies conducted in Japan after the FDNPP accident.

Table 6 indicates the \( K_d \) values obtained in this study and those reported in other studies for comparison. The average \( K_d \) value for all calm weather periods (excluding data of period 3) was \( 8.2 \times 10^4 \text{L/kg} \). This value is the same order of magnitude as the value obtained in the Ohori River during calm weather (\( 3.2 \times 10^4 \text{L/kg} \)). This suggests that the structural differences between the river and the retention basin were small. On the other hand, the \( K_d \) values obtained at Fukushima Prefecture were one or two orders of magnitude larger than those at Kashiwa City. This difference could be attributed to differences in land characteristics between Fukuo-
shima Prefecture and Kashiwa City.

The Kd values observed at the K1 basin and the K2 basin were slightly higher during period 3 than during calm weather. As described in section 4·1, this was due to the increment of the concentration of particulate $^{137}$Cs caused by the influence of the typhoon.

The Kd values obtained at three retention basins increased with time. This suggests that the ratio of dissolved $^{137}$Cs to the total $^{137}$Cs in the water sample decreased faster than that of particulate $^{137}$Cs. Such a situation implicates further fixation of $^{137}$Cs to particulate matter with time.23)

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要 旨

千葉県柏市内の調整池における水・底泥中放射性セシウムの
分布状況と環境動態

市川有二郎†，中田利明，井上智博，行方真優，本田恵理，石井栄勇，飯村晃，
藤村栄子，内藤季和，田中勉，髙橋良彦

千葉県環境研究センター
290-0046 千葉県市原市岩崎西1-8-8
† y.ichkw17@pref.chiba.lg.jp

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手賀沼を流束とする柏市内の調整池の中水ならびに底泥中における放射性セシウムの分布状況
と環境動態を検証した。福島第一原子力発電所事故から約4年が経過した時期の底泥から10kBq/
kg以上の放射性セシウムが検出され、調整池は下流域に対する放射性セシウムの供給源の1つで
あることが裏付けられた。台風などの豪雨時において、水中放射性セシウムの懸濁態の存在割合
が平水時と比較して上昇することから、豪雨は放射性セシウムの下流域への流出に関与している
ことが示唆された。