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# Temporal trends of $^{137}$Cs activity concentration in pond waters in the vicinity of Fukushima Dai-ichi nuclear power plant

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**Abstract.** Closed and semi-closed water bodies, such as lakes and ponds, are important water resources in Fukushima area and they are the most sensitive environments to radioactive contamination after the Fukushima Dai-ichi nuclear power plant accident. Wakiyama et al. (2017) investigated $^{137}$Cs activity in water and bottom sediment in four ponds; Suzuuchi (SU), Funasawa (FS), Inkyozaka (IZ), and Kashiramori (KM), within 10 km zone from the FDNPP during 2015–2016. This study follows up their observation to address longer time trends of $^{137}$Cs activity concentration in pond waters and to show speciation of $^{137}$Cs in soil and bottom sediment. Mean total $^{137}$Cs activity concentration in water ranged from 2.5 to 29 Bq L$^{-1}$. There was not found steady trend in the activity concentration of total and particulate $^{137}$Cs for four ponds. The concentration of dissolved $^{137}$Cs was usually low in winter during the entire observation period in all four ponds. A tendency to a decrease in the $^{137}$Cs activity concentration in suspended sediments was found for four ponds and the decreasing rate constants, including radiological decay, on SU, FS, IZ and KM were 0.33, 0.53, 0.29 and 0.25 yr$^{-1}$, respectively. The results of sequential extractions of soil and bottom sediment samples showed higher proportion of bioavailable $^{137}$Cs, i.e., exchangeable and organic bound $^{137}$Cs, in bottom sediment than in the soil.

## 1 Introduction

Closed and semi-closed water bodies, such as lakes and ponds, are the most sensitive environments to radionuclide contamination. More than 3700 individual irrigation ponds of different sizes are located in Fukushima Prefecture, which are radiologically contaminated after the Fukushima Dai-ichi nuclear power plant (FDNPP) accident. The Tohoku Regional Agricultural Administration Office of the Ministry of Agriculture, Forestry and Fishery (MAFF) and the Fukushima Prefectural government (MAFF and Fukushima prefecture, 2015) investigated 2679 ponds in Fukushima Prefecture and reported their water and sediment contamination levels.

The long-term behaviour of radiocesium in a closed and semi-enclosed water system affected by the Chernobyl accident can serve as a basis for long-term prediction of changes in environmental radioactive contamination in Fukushima. For example, Putyrskaya et al. (2009) presented the results of more than ten years of observation on the behaviour of $^{137}$Cs in several lakes affected by the Chernobyl accident and elaborated a model calculation of temporal variations in the concentration of $^{137}$Cs activity concentration of the lake water. However, the meteorological and geographical conditions in these areas are different from those in the Fukushima area, and the applicability of these results requires verification (Konoplev et al., 2016).

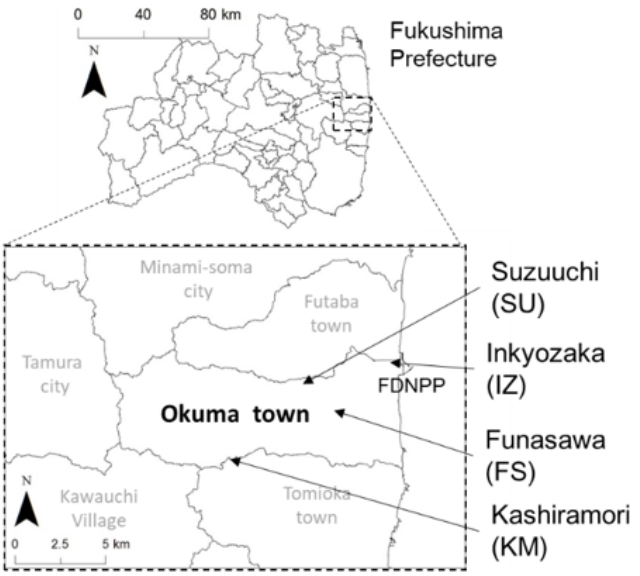


**Figure 1.** Location of study sites in Fukushima Prefecture and in Okuma town.

Wakiyama et al. (2017) demonstrated radiological contaminations in four ponds in the vicinity of FDNPP based on one-year observation. Although they suggested seasonal variation of dissolved $^{137}$Cs activity concentration of water, long-term trends remained unclear due to short period of their observations. This study shows longer temporal trends of $^{137}$Cs activity concentration in pond waters, as a continuation of observations undertaken by Wakiyama et al. (2017). In addition, we evaluated contamination of bottom sediment and its speciation.

## 2 Materials and method

### 2.1 Study site

Selected irrigation ponds of Suzuuchi (SU), Funasawa (FS), Inkyozaka (IZ) and Kashiramori (KM) are located in the city of Okuma within a radius of 10 km from FDNPP (Fig. 1). The surface area of these ponds of SU, FS, IZ and KM was 4100, 10 700, 6500 and 8100 $m^2$, respectively, and ambient dose equivalent rates measured on March 2014 around SU, FS, IZ and KM were 24, 6.0, 11 and 3.3 $\mu Sv\,h^{-1}$, respectively (Wakiyama et al., 2017). The average annual precipitation and the average annual air temperature in this region were 1493 mm and 12.3 °C, respectively, according to Namie weather station data from 1976 to 2015.

### 2.2 Sampling and analyses of pond water

We had collected 2 L of water samples from the four ponds approximately once a month since August 2015 to August 2017. Then, water samples were filtered using 0.45 µm-pore cellulose acetate membrane filter units in the laboratory. Suspended sediment (SS) concentrations in water (SSC, $kg\,L^{-1}$) were quantified by dividing weight of SS by volume of filtered water. Suspended sediments and filtrates were measured with Ge detectors to quantify the $^{137}$Cs activity concentration in SS ($C_{Cs\text{-}SS}$, $Bq\,kg^{-1}$) and dissolved $^{137}$Cs activity concentration ($C_{Cs\text{-}dis}$, $Bq\,L^{-1}$), respectively. Measurements of $^{137}$Cs activity were done with standard electrode coaxial Ge detectors (CANBERRA GC4020, Canberra, USA) with relative efficiency of 42.6 % at the Institute of Environmental Radioactivity, Fukushima University. Measurements were taken for 90 000 s for $^{137}$Cs activity concentrations in SSs and 240 000 s for dissolved $^{137}$Cs activity concentrations to ensure minimal statistical errors, which were $< 5$ % and $< 10$ %, respectively.

Particulate $^{137}$Cs activity concentration in water ($C_{Cs\text{-}par}$, $Bq\,L^{-1}$), total $^{137}$Cs activity concentration in water ($C_{Cs\text{-}tot}$, $Bq\,L^{-1}$), and the apparent distribution coefficient ($K_d$, $L\,kg^{-1}$), were calculated by following equations.

$$C_{Cs\text{-}par} = C_{Cs\text{-}SS} \times SSC \quad (1)$$

$$C_{Cs\text{-}tot} = C_{Cs\text{-}par} + C_{Cs\text{-}dis} \quad (2)$$

$$K_d = C_{Cs\text{-}SS} \div C_{Cs\text{-}dis} \quad (3)$$

### 2.3 Sampling and analyses of soil and bottom sediment

Soil and bottom sediment samples were collected in June 2016. Soils at a depth of 30 cm using a liner core sampler DIK-110C (DAIKI, Japan) with a plastic cylinder insert 5 cm in diameter. In laboratory, surface soils, to the depths of 3–5 cm, were cut for every 1–2 cm, and then remained subsurface soils were cut by 3–5 cm. Each soil sample was dried at 50 °C for at least three days, then ground and homogenized in a mortar. Three cores of bottom sediment were obtained using 4 cm diameter polyvinyl core tubing in each pond. The bottom sediment cores were taken from a boat and the sampling depth ranged from 33 to 56 cm. Bottom sediments to the depth of 10 cm were cut for interval of 2.5–5 cm, and subsurface bottom sediments were cut by 5–10 cm.

Each bottom sediment sample was dried at 50 °C for at least three days, then ground and homogenized in a mortar. The homogenised samples of both soil and bottom sediments were packed into 100 mL containers and measured to determine its $^{137}$Cs activity concentration ($Bq\,kg^{-1}$). The measurements were done with standard electrode coaxial Ge detectors (CANBERRA GC4020, Canberra, USA) with relative efficiency of 42.6 % at the Institute of Environmental Radioactivity, Fukushima University. Measurements were taken up to 85 600 s to ensure a minimal statistical error of $< 5$ % for almost samples. Eight samples taken from the deep portion of the bottom sediment had low $^{137}$Cs activity concentrations, at a minimum of 1.83 $Bq\,kg^{-1}$, and high relative error, at 12.8 % maximum.

**Table 1.** Summary of $^{137}$Cs activity concentration in pond waters.

| | SU | | | FS | | | IZ | | | KM | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | *n* | Mean | S.D* | *n* | Mean | SD* | *n* | Mean | S.D* | *n* | Mean | SD* |
| Dissolved $^{137}$Cs activity concentration ($Bq\,L^{-1}$) | 24 | 2.5 | 1.4 | 21 | 1.7 | 0.76 | 24 | 1.9 | 0.64 | 12 | 0.25 | 0.085 |
| Particulate $^{137}$Cs activity concentration ($Bq\,L^{-1}$) | 24 | 26 | 29 | 24 | 9.5 | 11 | 24 | 7.2 | 9.1 | 24 | 1.3 | 1.5 |
| Total $^{137}$Cs activity concentration ($Bq\,L^{-1}$) | 24 | 30 | 22 | 21 | 12 | 7.2 | 24 | 9.2 | 3.8 | 12 | 1.5 | 1.1 |
| $^{137}$Cs activity concentration in SS ($kBq\,kg^{-1}$) | 24 | 290 | 95 | 24 | 270 | 120 | 24 | 250 | 110 | 24 | 130 | 86 |
| Apparent distribution coefficient, $K_d$ ($10^5\,L\,kg^{-1}$) | 24 | 1.4 | 0.45 | 21 | 2.0 | 0.93 | 24 | 1.4 | 0.75 | 12 | 6.8 | 6.2 |

* SD means standard deviation ($1\sigma$).

The proportion of $^{137}$Cs in the exchangeable fraction, the organic fraction and the fraction strongly bound to the particles in the soil and bottom sediment were evaluated by a sequential extraction. Soil samples for quantifying the proportion were prepared by mixing up 0–1 and 1–2 cm of soil samples which are described in Sects. 2–3. The soil samples were passed through 2 mm-mesh sieves. Bottom sediment for quantifying the proportion were additionally taken by an Ekman-Berge sampler on June 2016, simultaneous to bottom the core sampling. The bottom samples were dried at 50 °C.

The exchangeable fraction of the soil and bottom sediment were extracted with 1 M $CH_3COONH_4$ and the extract was measured for $^{137}$Cs activity. Subsequently, organic matter was decomposed with mixture of 5 % of $H_2O_2$ solution and nitric acid. The extract was measured for $^{137}$Cs activity. The measurements were done with same Ge detectors for soil and bottom sediment samples. Measurements were taken up to 53 000 s to ensure a minimal statistical error of $< 5$ %. Only for the organic-bound fraction of $^{137}$Cs in bottom sediment from IZ, relative error was 14.6 % because of low $^{137}$Cs activity.

## 3 Results and discussion

### 3.1 $^{137}$Cs activity concentrations in pond water

The observation results are presented in Table 1. Particulate $^{137}$Cs accounted for 73 %–88 % of total $^{137}$Cs, on average. The mean apparent $K_d$ value ranged from $1.4 \times 10^5$ to $6.0 \times 10^5\,L\,kg^{-1}$. These values were higher than those observed in the Chernobyl area by at least one order of magnitude, as previous authors suggested (e.g., Tsukada and Ohse, 2016). These results show a stronger affinity of $^{137}$Cs for sediment particles in Fukushima.

Figure 2 shows temporal variations in total, particulate and dissolved $^{137}$Cs activity concentration in pond water. Both total and particulate $^{137}$Cs activity concentrations varied significantly throughout observation, and the temporal variations were similar. These similar variations were attributed to high proportions of particulate $^{137}$Cs activity concentration, as shown in Table 1. In contrast, the concentration of dissolved $^{137}$Cs was usually low in winter during the entire observation period in all ponds. These results confirm that the seasonal cycle exists on dissolved $^{137}$Cs in ponds, as it was previously found in the Chernobyl-affected area (e.g., Putyrskaya et al., 2009).

Decreasing trends of $^{137}$Cs activity concentration in SSs were found for all ponds (Fig. 3). The temporal trend of $^{137}$Cs activity concentration in SS was expressed by the following equation,

$$C_{\text{Cs-SS}}(t) = C_{\text{Cs-SS},0} \exp(-\lambda t) \quad (4)$$

where $t$ is the elapsed time since the accident (year), $C_{\text{Cs-SS}}(t)$ is $^{137}$Cs activity concentration in SSs at time $t$ ($Bq\,kg^{-1}$), $C_{Cs-SS,0}$ is initial $^{137}$Cs activity concentration in SSs at $t = 0$ ($Bq\,kg^{-1}$) and $\lambda$ is rate constant including radiological decay ($yr^{-1}$). The rate constant on SU, FS, IZ and KM were 0.33, 0.52, 0.29 and 0.25 $yr^{-1}$, respectively. The corresponding periods of half-reduction for SU, FS, IZ

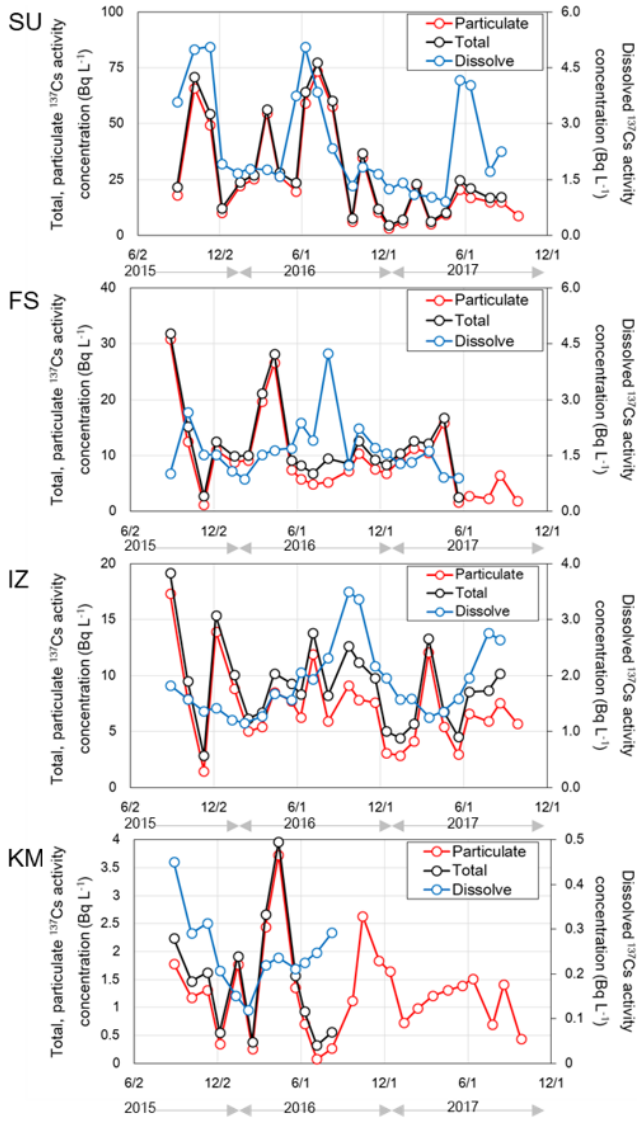


**Figure 2.** Temporal variations in total, particulate, dissolved $^{137}$Cs activity concentration in the different ponds.

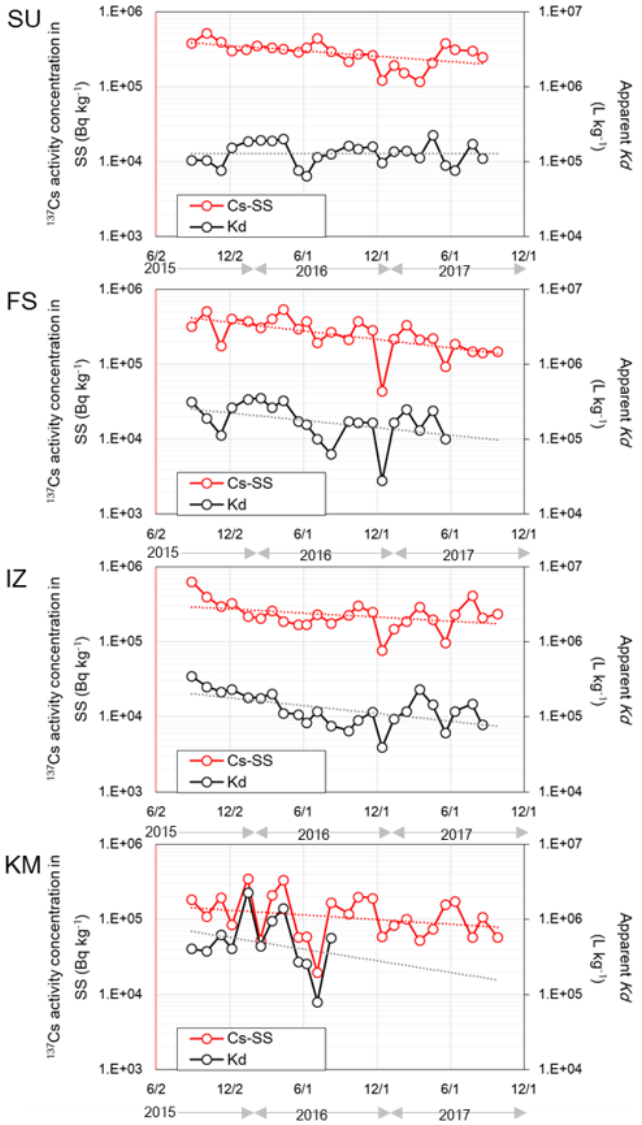


**Figure 3.** Temporal variations in $^{137}$Cs activity concentration of suspended sediment (Cs-SS) and apparent distribution coefficient $K_d$ in the different ponds.

and KM were 2.1, 1.3, 2.4 and 2.8 years, respectively. Since $^{137}$Cs activity concentration in SS was more variable than that dissolved $^{137}$Cs activity concentration, the temporal variation in apparent $K_d$ seemed to coincide with changes in the $^{137}$Cs activity concentration in SS.

## 3.2 $^{137}$Cs in soil and bottom sediment

The vertical distributions of $^{137}$Cs activity concentration in soil and bottom sediment are relatively similar for the all study sites (Fig. 4). As indicated by Wakiyama et al. (2017), the mean $^{137}$Cs inventory in the soil around SU, FS, IZ and KM were 6.8, 4.6, 2.9 and 0.42 MBq m$^{-2}$, respectively. Similarly, mean $^{137}$Cs inventory in bottom sediment in SU, FS, IZ and KM were 13, 8.9, 1.6 and 1.1 MBq m$^{-2}$, respectively. The $^{137}$Cs activity concentration both in the soils and bottom sediments decreased with depth.

The depth at which the soil contains 90 % of the inventory of $^{137}$Cs in the soil around the SU, FS, IZ and KM were 5, 7.6, 9.7 and 6.7 cm, respectively. The depth at which the soil contains 90 % of the inventory of $^{137}$Cs in bottom sediment on SU, FS, IZ and KM were 30, 17, 8.3 and 12.5 cm, respectively. Deeper penetration of $^{137}$Cs into bottom sediments than in the soil were found with the exception of IZ.

At least 95 % of $^{137}$Cs in soil was strongly bound to sediment particle in soil, while the $^{137}$Cs content in bottom sediment ranged from 42 % to 72 % (Fig. 5). The low proportions of $^{137}$Cs strongly bound with particle in bottom sedi-

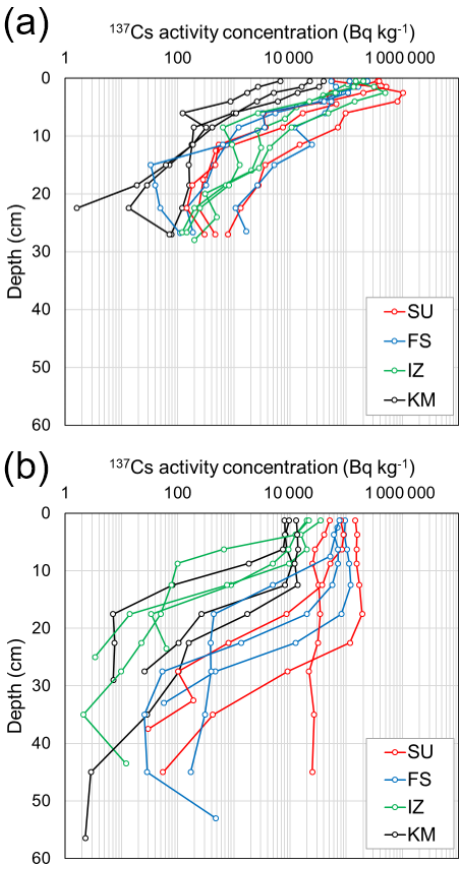


**Figure 4.** Vertical distribution of $^{137}$Cs activity concentration in soil **(a)** and bottom sediment **(b)**.

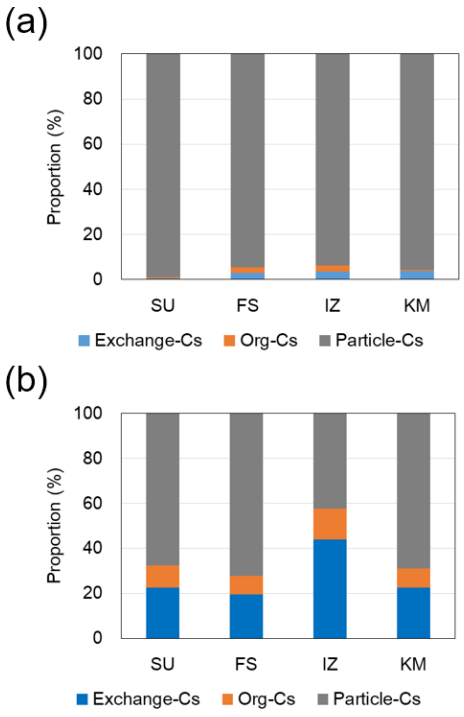


**Figure 5.** Proportion of exchangeable, organic and particle bound fractions of $^{137}$Cs in soil of 0–2 cm depth **(a)** and bottom sediment by an Ekman–Berge sampler **(b)**.

ment, suggest that the bioavailability of $^{137}$Cs was increased in bottom sediments.

Putyrskaya et al. (2009) demonstrated that percentage of exchangeable $^{137}$Cs was 5.7 % in average in 2006, based on sequential extraction of bottom sediment taken in lakes affected during the Chernobyl accident. Although the depth and water flow regime of ponds, located near Chernobyl, differed from ours, a high proportion of $^{137}$Cs in the exchangeable and organic fractions in Fukushima study sites suggested that the bioavailability of $^{137}$Cs in Okuma town ponds is still high.

These mobilities may have led to the vertical migration of $^{137}$Cs and controlled the fluctuation in dissolved $^{137}$Cs activity concentration. The effect of biological activities on $^{137}$Cs behaviour should be high in the spotlight.

## 4 Summary

We investigated the behaviour of $^{137}$Cs in four small ponds around Fukushima Dai-ichi nuclear power plant as a following-up of observation by Wakiyama et al. (2017). Continuous monitoring of $^{137}$Cs activity concentration in water confirmed the presence of a seasonal cycle of dissolved $^{137}$Cs activity concentration and newly revealed tendencies to a decrease trends in $^{137}$Cs activity concentration in suspended sediment for all ponds.

In addition, the sequential extractions of sediment samples showed a higher proportion of $^{137}$Cs in the exchangeable and organic fractions in bottom sediments than in soils and this indicates an increase in the bioavailability of $^{137}$Cs in bottom sediment. The effect of biological activity should be tested to better understand $^{137}$Cs activity in ponds.

**Data availability.** The data can be accessed by request to authors.

**Author contributions.** YW: Sampling, analyses and discussion as corresponding author. AK: Sampling, sample treatment and discussion. TW: Sampling, and discussion. TT: Sample analyses, and discussion. YI: Sampling, analyses, and discussion. KN: Sampling, discussion, and facilitating observation. IB: Sampling, sample treatment, and discussion.

**Competing interests.** The authors declare that they have no conflict of interest.

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