ISSN 1068-3739, Russian Meteorology and Hydrology, 2021, Vol. 46, No. 5, pp. 312–318. Allerton Press, Inc., 2021.
Russian Text The Author(s), 2021, published in Meteorologiya i Gidrologiya, 2021, No. 5, pp. 38–45.

Transformation of Radiocesium Speciation in Ponds at the Vicinity of Fukushima Dai-ichi Nuclear Power Plant and Dynamics of Its Distribution in Sediment–Water System

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Received June 9, 2020
Revised June 9, 2020
Accepted October 6, 2020

Abstract—The paper is concerned with the results of ^{137}Cs monitoring in the irrigation ponds of the Okuma town in the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) exclusion zone. The ^{137}Cs activity concentrations in the ponds appeared to be higher than those in the rivers and dam reservoirs in the region. The study has revealed a trend for a decline in ^{137}Cs activity concentrations, both particulate and dissolved. The rate of particulate ^{137}Cs decline was much higher than that of dissolved. The total distribution coefficient K_d (^{137}Cs) in the suspended sediment–water system in the studied ponds was decreasing in time with the rate constant of 0.12–0.18 year^{-1}. Assuming that the decrease in K_d is associated with decomposition of hot glassy particles, the time scale of ^{137}Cs leaching from them in these water bodies was estimated to be 5–8 years. These estimates are consistent with the findings of recent laboratory experiments on the subject. With respect to seasonal variations, the highest levels of dissolved ^{137}Cs in the studied ponds were observed from June to October as a function of specific pond and monitoring year. Based on data about ^{137}Cs speciation in the bottom sediment top layer of the ponds and its distri- bution in the sediment–water system, the exchangeable radiocesium interception potential $RIP^{ex}(K)$ for the ponds sediments was estimated to be 1650–2250 mg-eq/kg, which is within the range of values measured by laboratory studies.

DOI: 10.3103/S1068373921050058

Keywords: Radioactive cesium, Fukushima Dai-ichi NPP, ponds in the FDNPP exclusion zone, suspendent sediment–water system, exchangeable radiocesium interception potential

INTRODUCTION

The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident caused by the tsunami following the Great East Japan Earthquake in March 2011 resulted in the contamination of extensive territories on the Honshu Island (Japan) by radioactive cesium isotopes ^{134}Cs (half-life $T_{1/2}$ = 2.06 years) and ^{137}Cs ($T_{1/2}$ = 30.17 years), which rekindled the interest in the behavior of radiocesium in specific geoclimatic conditions of Japan. Radiocesium was deposited northwest of the NPP, forming a footprint about 20 km wide and 50–70 km long [9, 12, 18] and leading to the contamination of both terrestrial and water ecosystems. The initial ratio of cesium isotopes ^{134}Cs/^{137}Cs in the Fukushima fallout was about unity [12]. With time, the contribution of ^{134}Cs to radiation contamination was decreasing as compared to ^{137}Cs due to faster decay, and as of today the main radionuclide of dose significance is ^{137}Cs.

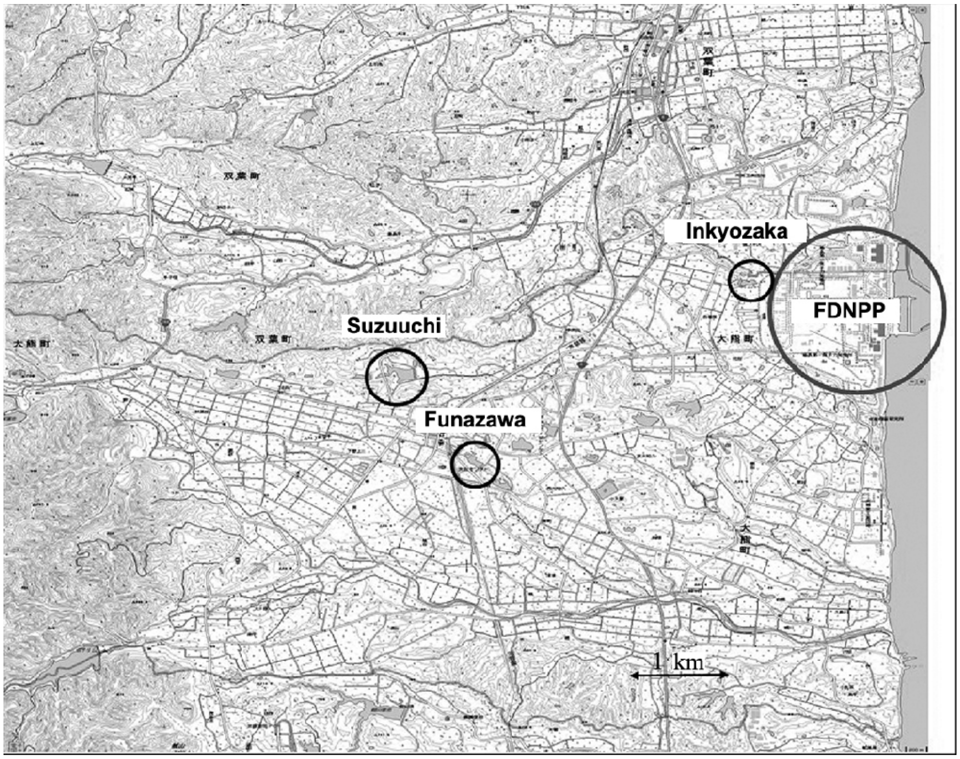

Fig. 1. Location of the studied ponds on the territory of Okuma town in the vicinity of the FDNPP.

After the Chernobyl NPP (ChNPP) accident, closed water bodies such as lakes, ponds and stagnant reservoirs were found to be more sensitive to radioactive contamination [3, 6, 16]. On the territory of Fukushima prefecture there are more than 3700 ponds of varying size, many of which are used for paddy water supply. Irrigation ponds were created over the course of centuries in Japan for rice cultivation. These ponds are also a concern because they are used for fishing and watering of agricultural fields, which can cause crop contamination [11, 24, 26].

The purpose of the present paper is to study the distribution of Fukushima-derived radiocesium and its dynamics in closed and semi-closed ponds of the FDNPP exclusion zone and to compare with the radionuclide behavior in similar objects in the ChNPP zone.

MATERIALS AND METHODS

The total of 137 small and medium-size ponds occur in the Okuma town in the FDNPP 10-km exclusion zone. Of them, three ponds Inkyozaka, Suzuuchi and Funazawa were selected as objects for study (Fig. 1). Here are the characteristics of the considered ponds in the FDNPP exclusion zone:

Pond	Inkyozaka	Suzuuchi	Funazawa
Northern latitude	37 25.499	37 24.950	37 24.363
Eastern longitude	141 01.05	140 58.791	140 59.173
Distance from FDNPP, km	0.24	3.75	3.5
^{137}Cs deposition, kBq/m^2	2250 1350	6850 1870	2370 495
Water surface area, m^2	6500	4100	10700
Maximum depth, m	2.0	1.0	2.5

The selected ponds represent the main pond types in the exclusion zone, where the ^{137}Cs deposition is varying from 2 to 7 MBq/m^2 [4, 21, 22]. Water sampling from the ponds was conducted once a month in 2015–2017, and at least once every two months in 2018–2019. Water samples of 4 l were collected from the surface layer and then filtered through 0.45 m membrane filters in laboratory. The activity concentrations of ^{134}Cs and ^{137}Cs were determined in filtrate and on suspended material separately by gamma spectrometry

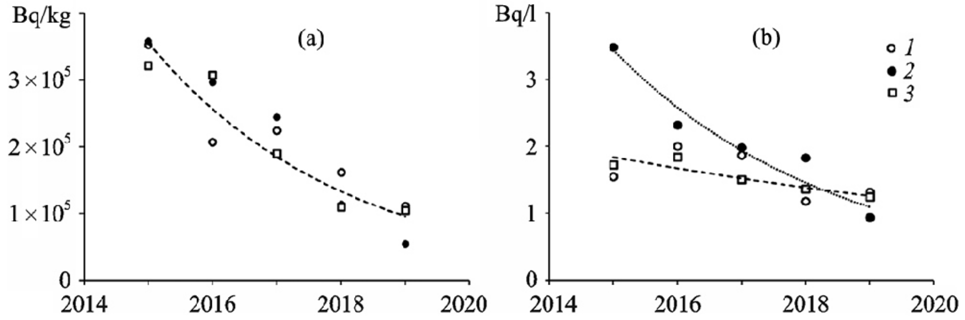

Fig. 2. Time dependence of the (a) particulate and (b) dissolved ^{137}Cs activity concentrations in the ponds of Okuma town in the vicinity of FDNPP (*1*) Inkyozaka, (*2*) Suzuuchi, (*3*) Funazawa.

Table 1. Kinetic characteristics of the ^{137}Cs activity concentration decline in the studied ponds of Okuma town

Pond	Suspended sediment			Solution		
	$_{\text{ef}}$	$_{\text{env}}$	$T_{1/2\ \text{env}}$, year	$_{\text{ef}}$	$_{\text{env}}$	$T_{1/2\ \text{env}}$, year
	year^{-1}			year^{-1}		
Inkyozaka	0.254	0.231	3.00	0.085	0.062	11.20
Suzuuchi	0.347	0.324	2.14	0.286	0.263	2.64
Funazawa	0.327	0.304	2.28	0.095	0.072	9.63

using the high-purity germanium detector (HPGe) CANBERRA GC3018. The concentrations of the main cations K^+, NH_4, Na^+, Ca^{2+}, and Mg^{2+} were determined by the ion chromatography using Thermo Scientific DIONEX 1100. The stable ^{133}Cs was determined by the inductively coupled plasma spectrometry ICP-MS (Perkin Elmer ELAN DRC 2).

RESULTS AND DISCUSSION

Figure 2 shows the time dependence of the mean annual particulate and dissolved ^{137}Cs activity concentrations from 2015 to 2019 in the ponds Inkyozaka, Suzuuchi, and Funazawa. In all three ponds, the dissolved and particulate ^{137}Cs activity concentrations were decreasing. Table 1 includes the quantitative characteristics of ^{137}Cs decline in the Okuma ponds. Here $_{\text{ef}}$ is the effective rate constant for decline in ^{137}Cs activity concentration; $_{\text{env}}$ is the rate constant for decline in ^{137}Cs activity concentration due to natural attenuation processes: $_{\text{env}} = {}_{\text{ef}} -$, where is the rate constant for ^{137}Cs decay equal to 0.023 year^{-1}; $T_{1/2\text{env}}$ is the ^{137}Cs concentration half-reduction due to natural attenuation processes.

It follows from Fig. 2 and Table 1 that the particulate ^{137}Cs activity concentration is declining faster than the dissolved ^{137}Cs. The partitioning of the radionuclide between the sediment and solution is described by the distribution coefficient K_{d} (l/kg) equal to the ratio of the particulate radionuclide activity concentration (Bq/kg) to its dissolved activity concentration (Bq/l) at equilibrium [15]. For research purposes the notions of the total distribution coefficient K_{d} and the exchangeable distribution coefficient K_{d}^{ex} are used [15]. K_{d} is equal to the ratio of the total activity concentration of the radionuclide in the solid phase to its activity concentration in solution, whereas K_{d}^{ex} is equal to the ratio of the exchangeable radionuclide activity concentration in the solid phase to its activity concentration in solution. The advantage of the exchangeable distribution coefficient is that its value can be calculated easily based on data about sorption capacity of the solid phase and water cation composition [2, 15].

A faster reduction in the particulate ^{137}Cs concentration, as compared to the dissolved one, is expected to result in a decrease of the K_{d} value for the given time period, which is corroborated by Fig. 3 showing time dependence of the ^{137}Cs total distribution coefficient in the ponds. The rate constants of K_{d} decline for the ponds under study are in the range 0.12–0.18 year^{-1}.

Table 2. The forms of existence and distribution of ^{137}Cs in the sediments–water system of Okuma ponds

Parameter	Inkyozaka	Suzuuchi	Funasawa
$_{\text{sed}}$, %	44 5	22.6 2.3	19.6 4.4
$_{\text{s}}$, %	3.6	0.4	3.0
K_d, l/kg	$(1.4\quad 0.7)\quad 10^5$	$(1.2\quad 0.1)\quad 10^5$	$(1.6\quad 0.8)\quad 10^5$
K_d^{ex}	$(5.5\quad 2.5)\quad 10^4$	$(2.9\quad 1.0)\quad 10^4$	$(3.3\quad 1.6)\quad 10^4$
$RIP^{ex}(K)$, mg-eq/kg	1650 800	2250 800	2250 1100

$_{\text{sed}}$ and $_{\text{s}}$ are the exchangeable form of ^{137}Cs in bottom sediments (the upper 3-cm layer) and soil of catchment (the upper 3-cm layer), respectively [20, 21].

Figure 4 illustrates seasonal variations of dissolved ^{137}Cs concentration in the FDNPP exclusion zone ponds for 2016 and 2017. Obviously, ^{137}Cs concentrations grow significantly in the summer–early autumn. In the ponds Inkyozaka and Funazawa the highest concentrations were observed from July to October, while in the pond Suzuuchi the highest concentration of ^{137}Cs occurred in June.

Table 2 includes results of determining the fraction of ^{137}Cs exchangeable forms in the 3-cm top sediment layer in comparison with similar data for soils on the catchments [7, 8, 22]. As seen, soils are characterized by extremely low values of exchangeable ^{137}Cs fraction, even compared with Chernobyl data. At the same time, the bottom sediment top layer formed as a result of sedimentation, is characterized by the high fraction of exchangeable ^{137}Cs (up to 40% in Inkyozaka).

The above data about ^{137}Cs speciation allow us to calculate the total (K_d) and exchangeable (K_d^{ex}) distribution coefficients [2] in Okuma ponds (Table 2), and on this basis to calculate Radiocesium Interception Potential RIP^{ex} in accordance with equation [5]:

$$RIP^{ex}(K) = K_d^{ex}(^{137}Cs)([K^+] + 5[NH_4])$$

where $[K^+]$ and $[NH_4]$ are potassium and ammonium concentrations in mg-eq/l.

On the territories contaminated after the Chernobyl accident, the values of the radiocesium distribution coefficient in the closed lakes and ponds were much lower than those reported for the large open lakes [3, 6, 16]. For example, in the closed Lake Svyatoe in the Bryansk oblast (Russia) in 1993–1995, 7–9 years after the Chernobyl accident K_d was $(3.5\quad 0.7)\quad 10^3$ l/kg [3]. Similar values of K_d ($8.1\quad 10^3$ l/kg) were reported for the closed lake Vorsee in Baden-Wurttemberg (Germany) in the same time period [16]. On the other hand, in large open Lake Constance (Germany/Austria/Switzerland) having high self-purification capacity with respect to radiocesium the $K_d(^{137}Cs)$, values were $(1–10)\quad 10^5$ l/kg [16], which is close to $K_d(^{137}Cs)$ in the FDNPP zone water bodies.

The K_d values in the studied ponds of Okuma ($>10^5$ l/kg) appeared to be about two orders of magnitude higher than those in closed lakes Svyatoe and Vorsee, and close to the values in Lake Constance [16], as well as the rivers and dam reservoirs of the FDNPP contaminated area [25]. In our view, this can be attributed to two reasons. The first is associated with the occurrence of insoluble glassy hot particles in the Fukushima fallout [13] and their higher content in the vicinity of the FDNPP. The second reason consists in the high binding ability of soils sediments in the Fukushima contaminated area (Table 2). The obtained high estimates of $RIP^{ex}(K)$ showing the ability of sediment particles to adsorb ^{137}Cs selectively are consistent with the values measured in laboratory experiments [17, 23] for the Fukushima area soils. The sediments of the studied ponds in terms of their ability to adsorb radiocesium selectively are even superior to those of Lake Constance in Europe [16].

The trend for a decline in the total distribution coefficient K_d reported for all three studied ponds (Fig. 3) can be due to ^{137}Cs leaching from glassy hot particles the proportion of which in the near zone of FDNPP can be as high as 30% [14]. In this respect, the rate constant of K_d decline in the ponds is a characteristic of weathering of glassy hot particles. Then the time scale of ^{137}Cs leaching from particles (the value reverse to the rate constant) is estimated to be 5–8 years. After the limiting stage of ^{137}Cs leaching from a glassy hot particle as a result of its decomposition or weathering, radiocesium gets fixed quite quickly by micaceous clay minerals with the time scale of 2–3 months. The derived estimates are in agreement with the outcomes of laboratory experiments to study radiocesium leaching from glassy hot particles in various salt solutions [19].

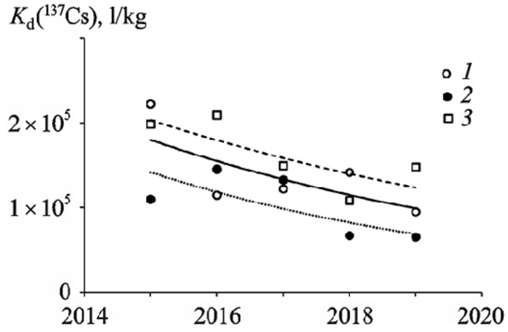

Fig. 3. Time dependence of the ^{137}Cs total distribution coefficient for the sediment–water system in the Okuma town ponds in the FDNPP exclusion zone: (*1*) Inkyozaka, (*2*) Suzuuchi, (*3*) Funazawa.

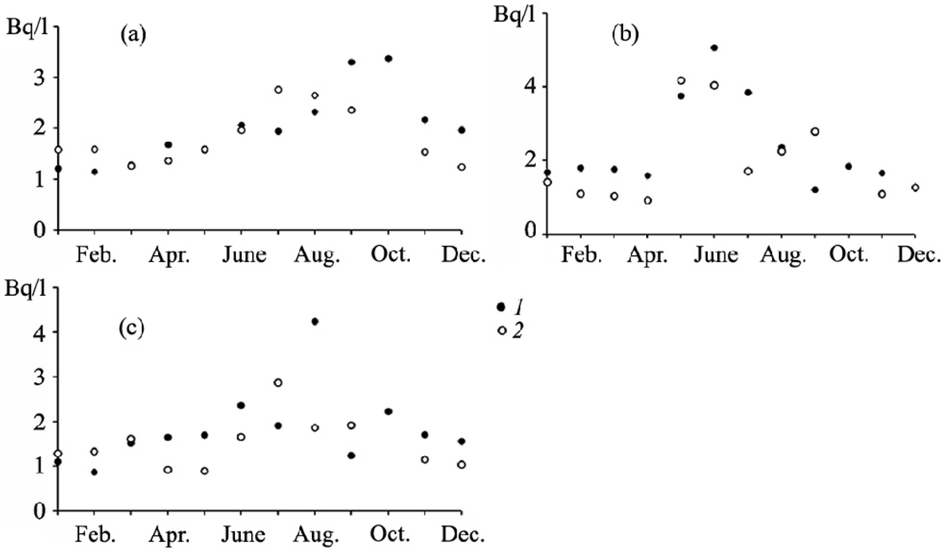

Fig. 4. Seasonality of the dissolved ^{137}Cs activity concentrations in the water of the Okuma town ponds of the FDNPP exclusion zone in (*1*) 2016 and (*2*) 2017: (a) Inkyozaka, (b) Suzuuchi, (c) Funazawa.

CONCLUSIONS

The studies of ^{137}Cs behavior in the irrigation ponds of the city Okuma in the vicinity of FDNPP conducted in 2015–2019 have demonstrated that the concentrations of this radionuclide in the ponds are higher than those in the rivers and dam reservoirs of the region. The highest levels of dissolved ^{137}Cs in the studied ponds were observed from June to October as a function of pond and monitoring year.

A trend for decline in both particulate and dissolved ^{137}Cs activity concentrations was revealed. The reduction rate of the particulate ^{137}Cs activity concentrations was notably higher than of the dissolved one.

This manifested itself in the trend for decline of the total distribution coefficient K_d(^{137}Cs) in the sediment–water system with the rate constant 0.12–0.18 year^{-1}. Assuming that the decrease in K_d is associated with decomposition of hot glassy particles, the time scale of ^{137}Cs leaching from them in the ponds under study is estimated to be 5–8 years. The obtained estimates are consistent with the findings of recent laboratory experiments on the subject.

In absolute magnitude, the value of K_d(^{137}Cs) in the studied ponds appeared to be much higher than the corresponding values in the closed lakes of the Chernobyl contaminated area and comparable to the values characteristic of the rivers and reservoirs of the FDNPP contaminated area.

Based on the data on the ^{137}Cs speciation in the bottom sediment top layer of the ponds and its distribution in the sediment–water system, the exchangeable radiocesium interception potential $RIP^{ex}(K)$ for the ponds sediments was estimated to be 1.7–2.3 g-eq/kg, which is within the range of values measured in laboratory.

FUNDING

The study was supported by the Japan Society for the Promotion of Science (JSPS), project number 18H03389.

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