The ecological half-life of radiocesium in surficial bottom sediments of five ponds in Fukushima based on in situ measurements with plastic scintillation fibers†

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Plastic scintillation fibers (PSFs) have been instrumental in in situ surface contamination surveys post the Fukushima Daichi Nuclear Power Plant accident. Their deployment to monitoring bottom sediments in aquatic environments provides the spatial extent of contamination over wide areas compared to discrete points as provided traditionally by sediment sampling. This study evaluated the wide area ecological half-life (T_{eco}) of radiocesium concentration for surface sediments of five ponds in Fukushima using PSFs, monitoring data generated between 2013 and 2019. The least squares’ regression method was employed to evaluate the T_{eco}. Four ponds had a T_{eco} ranging from 3.0 ± 0.3 years to 11.4 ± 2.3 years. A forest-catchment pond exhibited a relatively long T_{eco} of 41.6 ± 55 years. Local variation in the T_{eco} appears to be influenced by sedimentation as we demonstrated larger values for areas showing potential sedimentation in the forest catchment pond. This study demonstrates the importance of wide area in situ monitoring techniques, such as PSF, in providing an overview of the spatial-temporal trends of radiocesium in bottom sediments and confirms the importance of forests as secondary contaminant sources to their drainage.

Environmental significance

In aquatic environments, most radiocesium is bound to sediments. Characterizing radiocesium concentration in surface sediments is, therefore, critical for assessing the associated risk to the ecosystem. However, due to labor and cost associated with sediment underwater sampling and subsequent radioactivity analysis, spatial distribution studies/monitoring for radiocesium in the waterbed are limited. Consequently, long-term impacts are defined mainly for discrete points. In situ techniques may potentially solve this problem and this study employed plastic scintillation fibers to monitor wide areas of five agricultural ponds in Fukushima and subsequently evaluated the ecological half-life of radiocesium concentration in the surface sediments. This provides practical parameters for the impact of the radionuclide in these media and their surrounding catchment and drainage.

Introduction

Radiocesium, 137Cs (half-life 30.2 years) and 134Cs (half-life 2.1 years), were the most significant long-lived artificial radionuclides released from Fukushima-Daiichi Nuclear Power Plants (FDNPP) in 2011. The total amounts released were approximately 20 PBq.1 Radiocesium, particularly 137Cs, is a long-term radiological concern because of its long half-life and strong

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Most sediment studies of radiocesium in aquatic environments focus on the vertical profiles: their distribution; their temporal vertical shifts; and on modelling radiocesium migration processes. Earlier vertical profiles for agricultural ponds in Fukushima had peaks within the surface layer (0–5 cm) with a sharp decrease in concentration beyond that layer. In more recent profiles, though dependent on sampling location, radiocesium concentration appears to peak below the surface layer with a more gradual fallout beyond the peaks suggesting a downward migration over time. Very few studies document the temporal changes of radiocesium in surface sediments and so far in Fukushima, no known studies have attempted to evaluate such changes in agricultural ponds. However, evaluating the decreasing rate of radiocesium in bottom sediments is important because sediments account for the largest proportion of the total radiocesium concentration. Specifically, surface sediments are important because they make radiocesium available for biological circulation and redistribution via dissolution, resuspension, erosion, and hydraulic flushing.

Temporal changes in radiocesium concentration can be quantified by the effective half-life which accounts for both physical decay and ecological processes or the ecological half-life which excludes physical decay. Most documented
effective or ecological half-lives of radiocesium in either soil or sediments were evaluated from sediment samples thereby providing values for discrete points. Sediment sampling underwater is labour-intensive, long, and becomes expensive for wide area coverage. Consequently, limited or small sample sizes are characteristic of many sediment studies, which prevents observation of and accountability for spatial heterogeneities across wide areas.

In situ measurement techniques offer an alternative or complement to conventional sampling for wide area spatial distribution under water. Examples of their deployment for radiocesium waterbed surveys include the Fangataufa and Mururoa lagoons which were nuclear weapon test sites; the Irish sea in the vicinity of the Sellafield Reprocessing Facility and off the coast of FDNPP in Japan. A clear observation is that these studies have primarily focused on marine environments whose underlying hydrodynamics are different from shallow lentic water bodies such as agricultural ponds. With Fukushima’s abundance of agricultural ponds, the Japan Atomic Energy Agency (JAEA) developed plastic scintillation fiber (PSF) detector technology for in situ monitoring of their bottom sediments. This technique enables observation of spatial variation in contamination over wide areas. Good agreement was observed between radiocesium concentrations estimated by the PSF and mean radiocesium concentration of sediment cores averaged to a 10 cm depth thereby validating the results.

The aim of this study was to evaluate the ecological half-life of radiocesium over wide areas of pond surface sediments based on in situ measurements with PSF. Typical ecological half-life estimations focus on surface sediments based on grab samples or the top layers (up to 10 cm depth) of sediments. The effective depth of PSF considered in this study is 10 cm based on the thickness of sediment core samples used to calibrate the PSF concentration since it started being applied. Therefore, the temporal changes reflect what is occurring in this layer of sediments and can provide some useful radioecological information. This is not a radiocesium migration study but rather a study of the overview of the spatial-temporal trends of radiocesium concentration over wide areas of bottom sediments based on in situ measurements with PSF. It is the first attempt to evaluate the ecological half-life over wide areas of bottom sediments, to use in situ measurements in bottom sediments to estimate the ecological half-life, and to study the ecological half-life in bottom sediments of ponds. The ecological half-life is evaluated in the context provided by Prohl et al., where all processes contributing to a decline in radioactivity, excluding physical decay and human actions such as decontamination (herein, only within the pond), are integrated. It lumps together all potential contributors, within the bulk upper 10 cm surface-sediment layer.

### Experimental

#### Study sites

We selected five ponds located in areas affected by radioactive fallout from the FDNPP accident. Fig. 1 shows the location and satellite images of the ponds and their catchment and Table 1 summarizes their characteristics. Pond P1 is located in a mountainous region with its catchment consisting entirely of the forest. Its average depth is 1.1 m while its catchment area is 303 000 m². P2 has a mixed catchment consisting of forest (50%), cultivated land (40%), and infrastructure (10%). Its catchment is 117 000 m² and its depth averages 2.1 m. The catchments of P3, P4 and P5 were difficult to isolate as they are mainly determined by the sewage system due to their location in residential areas. Their depths average 0.8 m, 1.7 m, and 4.0 m respectively. P5 lies within a 10 km radius of the FDNPP while

#### Table 1 Description and characteristics of the investigated ponds and their catchment

| Characteristic | P1 | P2 | P3 | P4 | P5 |
|----------------|----|----|----|----|----|
| Town/local govt | Iitate | Motomiya | Motomiya | Koriyama | Futaba |
| Area designation | SDA | ICSA | ICSA | ICSA | SDA |
| Latitude | 37.701801 | 37.513944 | 37.500787 | 37.376372 | 37.348583 |
| Longitude | 140.687407 | 140.478589 | 140.422833 | 140.346835 | 141.013197 |
| Elevation (m) | 498 | 299 | 252 | 250 | 29 |
| Distance from FDNPP (km) | 44 | 52 | 56 | 63 | 8.5 |
| Surface area (m²) | 2320 | 5350 | 858 | 4770 | 5420 |
| Catchment (m²) | 303 000 | 117 000 | — | — | — |
| Catchment : surface ratio | 131 | 22 | — | — | — |
| Depth (m) | Maximum | 3.2 | 3.4 | 1.2 | 2.3 | 5.9 |
| Mean | 1.1 | 2.1 | 0.8 | 1.7 | 4.0 |
| Minimum | 0.2 | 0.7 | 0.4 | 1.4 | 0.9 |
| Catchment land use (%) | Forest | 100 | 50 | — | — | — |
| Cultivation (paddies, etc.) | — | 40 | — | — | — |
| Infrastructure (road, building, etc.) | — | 10 | — | — | — |

* Ministry of the Environment designation for decontamination works; SDA: special decontamination areas; ICSA: intensive contamination survey areas. * Calculated by ArcGIS, ESRI, Co., Ltd., USA.
the others are located between 44 and 63 km away from the FDNPP. P2 has three main inlets while the other ponds have two.

**Measurement campaigns**

Sediment core sampling and in situ measurements with PSF were conducted annually between 2013 and 2019, with approximately one year intervals between sampling campaigns. The specific monitoring dates for each measurement campaign are detailed in Table 2. Ponds P3, P4, and P5 underwent decontamination, by dredging, either before or after the measurement campaigns covered in this paper. P3 was decontaminated in 2013 and 2018 while P4 and P5 were decontaminated in 2018, after the measurement campaigns. For P3, only the data collected between the first and second decontamination were used in the analysis.

**In situ measurement system and procedure**

The flexible 20 m long PSF is submersible in water and uses real-time kinematic global positioning systems to provide the positional count rate, in counts per second (cps), for every 20 cm-length of cable with a counting time of 100 seconds. The PSF used in this study is an optical fiber, SCSF-3HF (Kuraray Co. Ltd., Japan) consisting of a polystyrene core sequentially clad with polymethylmethacrylate and fluorinated polymers. The diameter and length of the fiber are 2 mm and 20 m respectively. Ten PSFs were gathered into one vinyl tube, and photomultiplier tubes (PMTs) were placed at both ends of the optical fiber. Hereon, PSF refers to PMT-coupled bundled PSFs.

Fig. 2 shows a schematic of the PSF measurement system, adopted from Sanada et al. The radiation counts are measured by placing the PSF at the bottom of the ponds where radioactive emissions, mostly gamma rays but also beta particles, produce directional scintillation light in the fiber. A time-to-amplitude converter outputs a positional total count spectrum to a computer based on the arrival time difference of the scintillations at each PMT. Positional resolution and the linearity between source and the peak channel are checked by 137Cs point sources as described by Nohtomi et al. Routine calibration before and after deployment uses radiocesium-spiked concrete calibration pads in a water tank. The conversion factor from the total count rate to the activity concentration was obtained by correlation with ex situ measurements of sediment core samples. This process and its results are explained in the next section. The minimum detection limit for radiocesium in ponds was 61 Bq kg\(^{-1}\). The PSF measurements were taken linearly as shown in Fig. 3 with a spacing of 5 m between the lines. Approximately the same positions and lines were used during each campaign.

Each pond was divided into grids, the concentration in each mesh was averaged and the mean value was allocated for that mesh. The number of data points used for temporal analysis were determined by the campaign that produced the least data points. This corresponds to the smallest areas on radiation maps which were generated through interpolation by the Kriging method using a geographic information system software, ArC GIS (ESRI Co, Ltd., USA).

**Conversion factor**

There are many ways of obtaining conversion factors, CF, from the count rate (cps) to the activity concentration for in situ waterbed detectors. These include the Monte-Carlo simulation, the use of standard calibration sources with a fitting function, and comparing with the concentration of sediments sampled from the same media where the in situ measurements are taken. This study employed comparison with laboratory-measured sediment activity concentration. Examples of studies that also employed this kind of calibration are and earlier PSF studies.

| Pond | Year | 2013 | 2014 | 2015 | 2016 | 2017 | 2018 | 2019 |
|------|------|------|------|------|------|------|------|------|
| P1   | 2013 | 7–11 Aug | 28 Aug–14 Sep | 1–6 Sep | 12-Dec | 12–13 Sep | 27–28 Aug |
| P2   | 31 Jul–2 Aug | 14–15 Oct | 24–26 Sep | 4–5 Mar | 13–14 Oct | 1–4 Sep |
| P3   | 11–13 Apr | 24–25 Sep | 3–7 Sep | 2–5 Sep | 4–6 Sep |
| P4   | 28–31 Oct | 20–24 Aug | 26 Aug–16 Sep | 11-Dec | 7–8 Aug |
| P5   | 28–31 Oct | 20–24 Aug | 26 Aug–16 Sep | 11-Dec | 7–8 Aug |
A total of 81 sediment cores were collected from the ponds at points coinciding with PSF measurement lines. Cores were collected using an HR type core sampler (Rigo, Co. Ltd., Japan) after which they were sectioned at 5 cm intervals and each section was analysed for radiocesium concentration (Bq kg\text{wet}^{-1}), on a wet basis, using a p-type high-purity germanium detector of 18% relative efficiency (GEM20-70, Ortec, Seiko EG\&G, USA) coupled to a multichannel analyser (MCA 7, Ortec, Seiko EG\&G, USA). The average water content of these sediment samples (0–10 cm) was 60.2% (Min: 52.0%; Max: 81.0%). Standard gamma sources were used to calibrate the spectrometer: $^{241}$Am, $^{109}$Cd and $^{152}$Eu (EG-CUSTOM, Isotope Products Laboratories, USA); $^{210}$Pb and $^{137}$Cs (EG-CUSTOM, Isotope Products Laboratories, USA). PSF are a total count system, hence the measured $^{137}$Cs and $^{134}$Cs concentrations for the top two layers (totalling 10 cm depth) were summed and averaged, then compared with the count rate obtained by the PSF. Fig. 4a shows the comparison between the radiocesium concentration of sediment core samples and the counts obtained by the PSF measurement system (a) and distribution of conversion factors (b). Data set and the median (309 cps (Bq kg\text{wet}^{-1})^{-1}) of their frequency distribution (Fig. 4b) was considered appropriate and applied in this study. The standard deviation of the CF, 165 cps (Bq kg\text{wet}^{-1})^{-1}, accounts for 53% of the median value. Uncertainty may arise from: (a) imprecise PSF-coincident core sampling points due to possible instability of boats during sampling; (b) variation in water content and radiocesium concentration depth profile of the core; (c) the detection mechanism of PSF because it does not distinguish the type and energy of the detected radiation, which is predominantly gamma rays but can include beta particles; and (d) inefficient detection of gamma rays at the deeper end of the depth captured by PSF, which is up to 10 cm. Despite these potential sources of uncertainty, there was a strong agreement ($R^2 = 0.81$, Fig. 4a).

To minimize point (a), care was taken to collect core samples within 20 cm to 2 m of PSF lines. Assuming unchanging water content of the sediments, points (b) to (d) would have a negligible effect on the temporal trend within the 10 cm surface sediment layer because measurement lines were maintained in each campaign so that differences would serve to reflect changes occurring within the surface layer captured by PSF.
Estimation of the ecological half-life

In this study, temporal analysis was based on the premise of exponential decay using a first-order kinetic equation\textsuperscript{20} that assumes that the rate of decrease of radiocesium concentration does not change within the time scale we dealt with. The measured PSF concentration was first corrected for physical decay of $^{134}$Cs (half-life = 2.1 years) and $^{137}$Cs based on the number of half-lives, $N_{\text{HL}}$, exhausted at measurement time from 15 March 2011. The concentration of $^{137}$Cs ($A_{137}$) was evaluated from the ratio of its $^{137}N_{\text{HL}}$ to that combined with $^{134}Cs_{\text{total}}N_{\text{HL}}$. The concentration of $^{137}$Cs was converted from the count rate of the PSF with eqn (1):

$$A_{137} = C_{\text{CF}} \frac{\exp(-\lambda_{137} t)}{\exp(-\lambda_{137} t) + \exp(-\lambda_{134} t)}$$  \hspace{1cm} (1)

where $C_{\text{CF}}$ is count rate of PSF, CF is the conversion factor evaluated in the preceding section and $t$ is the time elapsed (year) from the main radioactive fallout date (15 March 2011). The nuclide ratio of $^{137}$Cs and $^{134}$Cs was one at the time of the accident. To ascertain this approach, the relative deviation of $A_{137}$ to that of core samples was calculated as described in ref. 38 and their distribution is shown in Fig. 5. The mean and median are very close to zero, indicating a good fit. On average, $A_{137}$ deviated from the radiocesium concentration of core samples by 38% with a median of 2.2%. The normalized mean square error (NMSE) was 0.1006. The $^{137}$Cs concentrations were used with eqn (2) to determine their ecological half-life, $T_{\text{eco}}$.

$$A_{t} = A_{0} \exp\left(-\frac{\ln 2}{T_{\text{eco}}} t\right)$$  \hspace{1cm} (2)

where $A_{0}$ and $A_{t}$ are the $^{137}$Cs concentration initially and after time, $t$, from the main radioactive fallout date. The parameters $A_{0}$ and $T_{\text{eco}}$ were evaluated by the method of least-squares where the absolute value of the slope is $\ln 2/T_{\text{eco}}$. With this treatment, the half-life, $T_{\text{eco}}$, only depicts the ecological component which in this study, partly includes an element catchment decontamination.
Results and discussion

Spatial heterogeneity of radiocesium concentration

PSF measurements over whole ponds show spatial heterogeneity in radiocesium concentration throughout the monitoring period reported in this study. These spatial variations are illustrated through the radiation maps (Fig. 6 and S1†), where hotter colours indicate higher concentrations and colder colours show lower concentrations, the spread of the mesh data points (Fig. 7) and large standard deviations,²⁰ in the radiocesium concentrations. The 2015 mean (and standard deviation) for P1 to P5 were 2.3 (2.9) × 10³ Bq kg⁻¹ wet, 3.4 × 10³ (6.8 × 10³) Bq kg⁻¹ wet, 1.4 × 10⁴ (6.9 × 10⁴) Bq kg⁻¹ wet, 6.4 × 10³ (1.9 × 10⁴) Bq kg⁻¹ wet, and 1.3 × 10⁴ (4.9 × 10⁴) Bq kg⁻¹ wet respectively. These PSF-generated radiation maps and quantitative data provide good information on the horizontal distribution on the ponds' waterbed which can aid in the selection of sites for studies requiring core-sampling, thereby addressing uncertainties associated with the patterns of sediment deposition,²¹ common to radiocesium accumulation studies. For core samples, the average depth at which 90% of radioactivity occurred was 11.5 ± 4.4 cm. This histogram for this distribution is shown in the ESI (Fig. S2†).

There is a tendency for higher concentration of radiocesium in deeper areas (Fig. 6a–e) in the ponds, similar to the observation for the Chernobyl-derived radiocesium inventory in Lakes Päijäne²⁰ and Høysjøen⁴⁸ in Finland and Norway respectively. These observations of enhanced sediment (and, consequently, radiocesium) accumulation in deeper parts of the waterbed were defined as sediment focusing. This effect was also observed with FDNPP-derived radiocesium on the waterbed of the Ogi reservoir²⁴ and the Ukedo river estuary.⁴² Thornton et al.⁴⁰ observed patterns of elevated radiocesium concentration at bases of protrusions in the seabed near FDNPP using a towed gamma spectrometer. Our results reveal the occurrence of similar phenomena in ponds. For P1 (Fig. 6a–i) and P2 (Fig. 6b) these deeper and higher concentration areas are at the outlets while the inlets have lower concentrations. Ohnuma and Ishii¹⁸ found coarser low-concentration sediments prevailing at inlets and finer radiocesium-rich sediments dominating at the outlets of ponds. Funaki et al.³ observed a similar trend in sediment size distribution between the inlet and outlet of the Ogaki dam. The finer sediments dominating at outlets was attributed to lower settling velocities which transport them further than the heavier and courser sediments.⁴ This particle size dependence may reflect our results.

Temporal changes in radiocesium concentration

Fig. 7 shows the PSF mesh data, the overall mean values and the exponential fitting line for the mean values. The radiocesium concentration in all the ponds declines, albeit at different rates, with P1 having a long Teco; P2 and P3 having short Teco; and P4 and P5 having intermediate Teco (Table 3). Since this study accounted for the physical decay in the analysis, this means the effective half-life in all the ponds is shorter than the physical half-life. The likely main contributors to the declining trend include (a) hydraulic flushing out of sediments and (b) downward migration of radiocesium through sedimentation and diffusion-advective transport. Hydraulic flushing may strongly affect shallow ponds (for example P1, P3 and P4) because they can be easily agitated by strong winds,¹⁷,⁴³ heavy rains and typhoons;¹⁵ and ponds of large catchment-to-surface-area ratios which yield low radiocesium trap efficiency so that radiocesium-rich fine sediments are flushed out. Yoshimura et al.⁴² observed such hydraulic flushing in some Fukushima ponds of low trap efficiency. Sedimentation is more likely to produce a larger ecological half-life, as inferred by Juranova et al.²⁰ and Baltas et al.²¹

Table 3  Summary of the initial 137Cs concentration (a0) and ecological half-lives (Teco) for the five ponds estimated from measurements with PSF

| Pond | a0 (Bq kg⁻¹) | 95% confidence level | Teco (years) | 95% confidence level |
|------|--------------|----------------------|-------------|---------------------|
| P1   | 2.6 × 10³    | (2.2–3.0 × 10³)      | 41.6        | (–14.0 to 97.1)     |
| P2   | 6.3 × 10³    | (6.0–6.5 × 10³)      | 4.2         | (3.9–4.5)           |
| P3   | 3.0 × 10⁴    | (2.8–3.2 × 10⁴)      | 3.0         | (2.70–3.3)          |
| P4   | 7.3 × 10³    | (6.8–7.7 × 10³)      | 11.4        | (9.1–13.7)          |
| P5   | 1.6 × 10⁴    | (1.5–1.6 × 10⁴)      | 9.7         | (8.3–11.1)          |
et al.\(^{19}\) because it adds to the radioesium concentration. This may explain P1’s long \(T_{\text{eco}}\), \(42 \pm 55\) years, where the radiation maps (Fig. 6a) show increased radioesium concentration for 2016 (Fig. 6a(iii)) and 2017 (Fig. 6a(iv)) compared to 2015 (Fig. 6a(ii)) in an area near the top left inlet.

The observed apparent sedimentation in P1 seems contradictory to the expectation of low soil erosion from forest catchments.\(^{7,45}\)

However, findings by Walling and Quingping,\(^{46}\) Yoshimura et al.\(^{48}\) and Taniguchi et al.\(^{7}\) show that sediments eroded from forested or, generally, uncultivated catchments tend to be radioesium-rich. This is the most probable explanation for the observation in P1. Sometimes sedimentation may result in an overall increase in concentration so that the \(T_{\text{eco}}\) takes on a positive value. This is captured within the large standard deviation of P1’s \(T_{\text{eco}}\). On the other hand, radioesium concentration is relatively smaller in sediments eroded from cultivated\(^{48}\) and/or decontaminated\(^{47}\) catchments even though erosion rates are higher therein. Therefore, comparable quantities of sediments entering P2–P5 may have relatively lower radioesium concentration than in P1. This may also explain the observed \(T_{\text{eco}}\). Decontamination efforts in Fukushima’s Special Decontamination Areas (SDA) and Intensive Contamination Survey Areas (ICSAs) prioritise living spaces including infrastructure, farmlands and only portions of forests near areas frequented by people.\(^{48}\) Whole area decontamination was completed on 19 March, 2018 in ICSA and on 31 March 2017 and 4 March 2020 in Mitate and Futaba respectively.\(^{49}\)

To observe the local variability in the \(T_{\text{eco}}\) across P1, a grid by grid analysis was conducted. Each 5 m by 5 m grid had six data points, one for each campaign. The grids had been categorized according to the evaluated \(T_{\text{eco}}\) with category A of a \(T_{\text{eco}}\) less than 5 years, category B \(T_{\text{eco}}\) between 5 and 10 years, category C \(T_{\text{eco}}\) between 10 and 100 years, and category D \(T_{\text{eco}}\) over 100 years. No negative \(T_{\text{eco}}\) was observed. Fig. 8 shows the distribution of the \(T_{\text{eco}}\) in P1, color-coded for each category. The grids were 19% category A, 16% category B, 15% category C and 50% category D. Most category D grids coincide with areas displaying possible sedimentation such as the top-left inlet, extending to a portion of the central area, and the deepest parts of the pond at the outlet, including the hotspot region (Fig. 6a). Juranova et al.\(^{20}\) interpreting a long \(137\text{Cs}\) \(T_{\text{eco}}\) (370 ± 450 years) of sediment sampled at the Otava Pisek tributary of Vltava river in The Czech Republic, suggested an effective blocking of \(137\text{Cs}\) removal by ecological processes as a result of sediment aging in combination with \(137\text{Cs}\) influx from the catchment. Fukushima et al.\(^{17}\) observed a monotonic increase in the FDNPP-derived radioesium inventory at the centres of four Japanese lakes, which was attributed to catchment input or lateral sediment transport within the lakes. Similarly, the inter-grid \(T_{\text{eco}}\) variations may be related to levels of sedimentation. They may also be associated with granulometry, mineralogy, and geophysical chemistry, which are outside the scope of the present study. Even though this technique does not provide the exact ecological processes, an overview of what is occurring has been obtained, which can optimize sediment sampling for specific migration studies.

The \(T_{\text{eco}}\) for P2 to P5 range from 3.0 to 11.4 years (Table 3). P3 was decontaminated prior to the first measurement campaign and has the shortest \(T_{\text{eco}}\) but it is presently unclear if this is responsible for the observed \(T_{\text{eco}}\). No correlation was observed between the \(T_{\text{eco}}\) and the pond characteristics among P3–P5.

While a short \(T_{\text{eco}}\) such as for P2 to P5, may be considered a form of spontaneous decontamination, recontamination concerns may arise for areas onto which the ponds irrigate or overflow if hydraulic flushing is the dominant process. On the other hand, the longer \(T_{\text{eco}}\) for example in P1, generates concerns regarding long-term secondary supply of radioesium from the catchment. The \(137\text{Cs}\) average concentration for 52 Finnish lakes in 1992 was 27.4% of that in 1987,\(^{50}\) equivalent to a \(T_{\text{eco}}\) of 2.93 years.

It was not possible to make a statistical comparison among the ponds by catchment type and dominant land use due to limitations in the available data. However, the results can be used as case studies on the applicability of PSF to overview the relative distribution of radioactivity and quantify the temporal trends over wide areas from averaged data. Additionally, ponds are a type of lentic system characterized by mostly static waters unless agitated by winds or rains. There are no documented studies on the radioesium \(T_{\text{eco}}\) for bottom sediments of ponds associated with fallout from nuclear tests and the accidents in Chernobyl and Fukushima. Also, lentic system studies have focused on larger lakes with radioesium migration as the dominant research theme. Thus, this study provides novel case studies for (a) the \(T_{\text{eco}}\) of radioesium in sediments of ponds in Fukushima, (b) evaluating the \(T_{\text{eco}}\) of radioesium over wide areas of bottom sediments and (c) evaluating the \(T_{\text{eco}}\) of radioesium in the bottom sediments using in situ measurement.
with PSF. The $T_{\text{eco}}$ is an apparent value including both removal or downward migration of sediment and additional sedimentation. However, this apparent value can represent the real temporal change of radiocesium concentration in surface sediment making it a practical radioecological parameter.

**Conclusions**

In this study, the spatial-temporal changes in radiocesium concentration of bottom sediments over wide pond areas were quantified using data obtained by *in situ* radioactivity monitoring with plastic scintillation fibers (PSF). The decreasing trend of radiocesium was quantified by the ecological half-life which ranged from 3.0 ± 0.3 to 11.4 ± 2.3 years across three ponds draining residential areas, and was 4.2 ± 0.3 years in a mixed-catchment pond and 41.6 ± 55.5 years in a forest-catchment pond. PSF-generated spatial distribution patterns showed apparent sediment focusing in the ponds and, over time, catchment-derived radiocesium input for the forest catchment pond. Detailed grid-by-grid analysis for the forest-catchment pond demonstrated longer ecological half-lives across areas of potential sedimentation and sediment focusing. Thus, the ecological half-life is likely influenced by sedimentation and sediment focusing. Though specific ecological processes are not provided by the PSF technique, a useful overview is provided which can help focus and optimize the design of detailed study. The study also confirmed that forest catchments are potential secondary sources of contamination for their drainage basins. This ecological half-life is an apparent value that can represent the real rate of temporal change of radiocesium concentration in surface sediments.

**Conflicts of interest**

There are no conflicts to declare.

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