

## Concentrations and biological half-life of radioactive cesium in epigeic earthworms after the Fukushima Dai-ichi Nuclear Power Plant accident



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### A B S T R A C T

To understand the long-term behavior of radiocesium in the biological processes of a forest ecosystem, its concentration in Japanese epigeic earthworms (Megascolocidae), litter, and soil, and the ambient dose equivalent rates, were investigated after the Fukushima Dai-ichi Nuclear Power Plant accident. The metabolism of radiocesium in the earthworms was also investigated in the laboratory, and its biological half-life ( $T_b$ ) was estimated. The concentration of  $^{137}\text{Cs}$  in the habitat soil and litter changed from 2014 to 2016, with levels in the litter going from 44.9 Bq/g dw (in 2014) to 45.3 Bq/g dw (2015) and 10.7 Bq/g dw (2016); in soil, these values were 9.79 Bq/g dw, 7.14 Bq/g dw and 18.0 Bq/g dw, respectively. By contrast, no significant changes were observed in the concentrations in the earthworms, which were 4.87 Bq/g fw, 5.30 Bq/g fw and 4.67 Bq/g fw in 2014, 2015, and 2016, respectively. The ambient dose equivalent rates at the sampling site declined significantly over these three years, going from 2.15  $\mu\text{Sv/h}$  to 1.68  $\mu\text{Sv/h}$  and 1.35  $\mu\text{Sv/h}$ , mostly corresponding to physical decay of radiocesium. The majority (95%) of the  $^{137}\text{Cs}$  in the earthworms, observed via autoradiography, was concentrated primarily in the intestine. The clearance of  $^{137}\text{Cs}$  from the earthworms was described by dual exponential functions: the half-life in the rapid loss due to gut clearance was 0.10 days and a second slower loss due to physiological clearance was 27.4 days.

### 1. Introduction

The nuclear accident at the Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant (hereafter referred to as FDNPP) in March 2011 caused a serious release of radionuclides into the environment. Fukushima Prefecture is covered with 975,000 ha of forests, which accounts for approximately 71% of the total land area of the prefecture (Fukushima Prefectural Government, 2017). As a result, most of the radionuclides were captured in forest ecosystems (Hashimoto et al., 2012).

Radiocesium has long-term influence on the forest ecosystems due to its relatively long half-life ( $^{134}\text{Cs}$ : 2.06 y,  $^{137}\text{Cs}$ : 30.1 y). After the accident, radiocesium was deposited directly on the floor of the forest (Koarashi et al., 2012) and accumulated further via throughfall, litterfall, and stemflow processes (Kato et al., 2012; Teramaga et al., 2014; Loffredo et al., 2014). The annual vertical migration of  $^{137}\text{Cs}$  to a depth of 10 cm in the soil only accounts for 0.1% of the total  $^{137}\text{Cs}$  inventory

(Nakanishi et al., 2014). As observed after the Chernobyl accident (Tikhomirov and Shcheglov, 1994; McGee et al., 2000), a large part of the radiocesium accumulates on the soil surface, including in the litter, in the long term. The radiocesium circulates in the forest ecosystem via biological cycles such as reabsorption by trees, retention by fungi, and direct ingestion by organisms. Therefore, it could be bioavailable for long-term transfer, for example, the detritus food web. The latter has been suggested to be a primary pathway of radiocesium (Murakami et al., 2014; Tanaka et al., 2016; Ishii et al., 2017), which means that detritivores are important as long-term carriers of radiocesium in a forest ecosystem.

Earthworms contribute to the disturbance of soil as ecosystem engineers (Lavelle et al., 1997) and are an important food resource for higher trophic consumers such as arthropods, birds, mammals, etc. It has been suggested that detritivores, including earthworms, supply radiocesium from the highly contaminated forest floor to consumers through the food web (Murakami et al., 2014). Earthworms can,

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therefore, contribute to the circulation of radiocesium in the soil and its transfer to organisms through the food web.

The results of a field survey conducted after the Chernobyl accident, to assess the effects of radiation on non-human biota, demonstrated catastrophic effects in soil invertebrate communities (including earthworms) (IAEA, 2006). Soil invertebrates had higher absorbed dose rates than other animals because their habitats were highly contaminated. This decreased the population densities and species diversity of soil invertebrates with an absorbed dose higher than approximately 30 Gy; the population densities of earthworms, in particular, decreased due to their high radio-sensitivity during the egg and juvenile stages (Krivolutzkii et al., 1992). This is why the International Commission on Radiological Protection selected the earthworm as a reference animal to assess the effects of radiation on the environment (ICRP, 2008).

For these reasons, earthworms are an important species for predicting the long-term environmental behavior of radiocesium and assessing the effects of radiation on non-human species. However, only a few field studies have been conducted on earthworms after the FDNPP accident. Moreover, there are a lack of studies on the biokinetics of radiocesium on the family Megascolecidae, which includes most of the species in Japan (Ishizuka, 1999).

Epigeic earthworms that live in the litter layer and the soil surface layer are an important ecological group discerning the behavior of radiocesium, which mostly accumulates in the ground surface layer. Hasegawa et al. (2013) conducted a field study on different species of epigeic earthworms in Fukushima and reported that earthworms from ecological groups of epigeic have similar radiocesium concentrations because of their similar habitats and physiological characteristics. In the present study, therefore, we focused on epigeic earthworms at family level to understand the behavior of radiocesium in a forest ecosystem.

The objectives of this study are to understand the temporal changes in the radiocesium concentration in earthworms from the mountainous forests of Fukushima and to examine the biokinetics of radiocesium in these earthworms. We conducted a field survey in Fukushima and compared the chronological changes in radiocesium in earthworms, litter, soil, and the ambient dose equivalent rates from 2014 to 2016. We also observed the distribution of radiocesium in the earthworms' bodies using autoradiography and determined the radiocesium concentrations in their body wall muscles, guts, and the other organs. Finally, we conducted clearance experiments of  $^{137}\text{Cs}$  on the earthworm to estimate its biological half-life.

## 2. Materials and methods

### 2.1. Sampling site

The sampling site is an area of relatively high contamination located 40.1 km northwest of the FDNPP (latitude:  $37^{\circ}41'35''$  N, longitude:  $140^{\circ}44'08''$  E; Fig. 1). The initial deposition density of total radiocesium ( $^{134}\text{Cs} + ^{137}\text{Cs}$ ) at the sampling site estimated from airborne monitoring survey was  $1\text{--}3\text{ MBq/m}^2$  (MEXT, 2011). The landscape is hilly and mountainous area, with agricultural fields and residential areas surrounded by mountainous forests. Residents were not permitted to live in this area during the study period from 2014 to 2016. The sampling site is a mixed forest dominated by deciduous broad-leaved trees and had not been decontaminated at the time of sampling. It had a radius of about 30 m (an area of ca.  $2800\text{ m}^2$ ), and the survey was conducted at the same site throughout the three years of the study.

### 2.2. Measurement and estimation of ambient dose equivalent rates

Ambient dose equivalent rates were measured using a NaI scintillation survey meter (TCS171, Hitachi, Ltd., Japan) which calibrated by Hitachi Aloka Medical, Ltd. on November 18, 2011. The calibration was carried out with the replacing method of JIS Z 4511–2005, using the

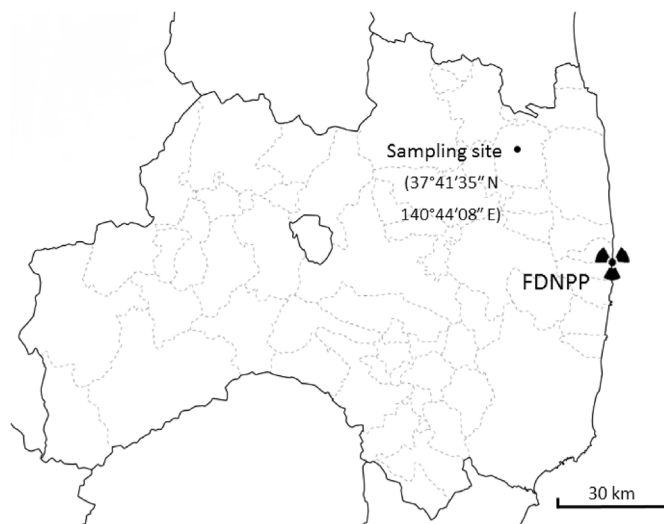


Fig. 1. Map of the sampling site and the Fukushima Dai-ichi Nuclear Power Plant (FDNPP).

working standard instruments (JCSS) irradiation apparatus. The measurements were conducted multiple points on the sampling site, 1 m above the ground and at least 20 m apart. To estimate these weathering of radiocesium from the sampling site, the ambient dose equivalent rates were compared to the estimated ambient dose equivalent rates from the physical half-life of radiocesium. For physical decay correction of ambient dose equivalent rates, the contribution ratio of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  to the ambient dose equivalent rates were calculated using a conversion factor (IAEA, 2000) which assumed that the radiocesium was distributed uniformly on the ground. This value was applied with the  $^{134}\text{Cs}/^{137}\text{Cs}$  activity ratio set to 1 from 2011.

### 2.3. Sampling and pretreatment of soil, litter, and earthworm

The soil was collected once at depths of 0–5 cm using a core sampler (50 mm in diameter, 51 mm high), and litter was sampled once in  $25\text{ cm} \times 25\text{ cm}$  area of the forest floor. The number of soil and litter samples taken in 2014, 2015 and 2016 were 1, 3, and 3, respectively. After being oven dried at  $105^{\circ}\text{C}$ , the soil was screened through a 2 mm square mesh, and the litter was homogenized in a blender. The epigeic earthworms were collected by hand from the litter layer and soil surface ( $\leq 5\text{ cm}$ ) from August to September. There were 1, 5, and 5 earthworm in five samples taken from 2014, 2015, and 2016, respectively; thus, 5, 25, and 25 individuals were collected, respectively, in those years. An additional 75 earthworms were collected in 2016 for the clearance experiments. The earthworms were identified as belonging into the family Megascolecidae from the external shape of their bodies, particularly the clitellum. In addition, the internal shapes were observed after the dissection of a few individuals. To measure the  $^{137}\text{Cs}$  accumulation in each part of the body, 25 earthworms were dissected and the intestines, body wall muscles, and other organs were separated. The  $^{137}\text{Cs}$  concentrations in the earthworms were determined from data including gut contents, since predators would consume the entire body of the earthworm (Sheppard et al., 1997); this method is therefore appropriate for understanding how radiocesium is transferred through the food web.

### 2.4. Measurement of $^{137}\text{Cs}$ in the samples

The soil and litter samples and the earthworms were packed into plastic containers (U-8: 47 mm diameter, 60 mm in height). The retention curve of  $^{137}\text{Cs}$  in the earthworms, which were packed individually in plastic petri dishes (60 mm in diameter, 15 mm high),

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