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# Cumulative history recorded in the depth distribution of radiocesium in sediments deposited on a sandbar

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ABSTRACT

We collected sediments deposited on a sandbar from the surface to 20 cm in depth in the Abukuma River to clarify the history of radiocesium derived from the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. We analyzed the $^{137}$Cs concentration in the sediments from size-fractioned samples as well as bulk samples. The depth distribution of $^{137}$Cs showed the highest concentration in the deepest sediment layer (18–20 cm) studied, which indicates that sediments with a lower $^{137}$Cs concentration were transported and deposited on sediments having a higher $^{137}$Cs concentration. At the same time, the depth distribution suggests a decrease in radioactivity in provenance areas of the sediments. Analysis of the size-fractioned sediments indicated that the three sediment layers at 4–6 cm, 16–18 cm and 18–20 cm intervals had similar size distribution of $^{137}$Cs and grain size composition although the concentration levels of $^{137}$Cs were different according to their bulk concentrations. The size distribution of $^{137}$Cs also supported the possibility that the decrease in $^{137}$Cs concentration in bulk sediments above 18 cm is due to a decrease in the level of radioactivity in the catchment area. A comparison of the size distribution of $^{137}$Cs between the sediment layers above and below 18 cm suggested that the $^{137}$Cs concentration in the transported fine sediment particles decreased more with time than the $^{137}$Cs concentration in the coarse particles, reflecting the selective transport of the finer particles. The results of this study demonstrated that sediment layers deposited on a sandbar retained the cumulative history of the fluvial transport of radiocesium after the FDNPP accident.



## 1. Introduction

Since the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011, many researchers have acquired knowledge on the distribution of radionuclides, and particularly radiocesium ($^{134}$Cs and $^{137}$Cs), in the environment, covering aerosols, soils, sediments, forests, rivers, and the Pacific Ocean (e.g., Kaneyasu et al., 2012; Kato et al., 2012; Kozai et al., 2012; Ohno et al., 2012; Tanaka et al., 2012, 2013a,b,c, 2014, 2015; Adachi et al., 2013; Matsunaga et al., 2013; Kumamoto et al., 2014; Kanasashi et al., 2015; Nagao et al., 2015; Nagakawa et al., 2015; Sakaguchi et al., 2015). Most of the radiocesium deposited on the surface of the land remained within a depth of 5 cm from the surface (Kato et al., 2012; Tanaka et al., 2012; Ohno et al., 2012; Matsunaga et al., 2013).

It is important to understand the long-term migration of radiocesium in the environment, which has changed its initial distribution and caused secondary contamination. While the vertical migration of radiocesium fixed on the surface of the land would proceed slowly with time (Smith and Beresford, 2005), migration horizontally at surface of soil and sediment is much more rapid (Kinouchi et al., 2015). Radiocesium fixed in the surface soils and sediments in basins is eroded and is flushed into rivers during heavy rainfall events from surface runoff (Bonnett, 1990; Matsunaga et al., 1991; Fukuyama et al., 2005). Recent works have demonstrated that particulate matter is the main carrier of radiocesium in the rivers in Fukushima (Ueda et al., 2013; Nagao et al., 2015; Sakaguchi et al., 2015). The dominance of particulate radiocesium clearly reflects its strong fixation in clay minerals

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(Cremers et al., 1988; Vidal et al., 1995; Qin et al., 2012; Fan et al., 2014).

Radiocesium-bearing particles are transported from their provenances to downstream areas. Such transported sediments are often found to be deposited on sandbars. The concentration levels of radiocesium in transported sediments are controlled by that in the catchment area. The modes of physical movement in the suspended state or along a riverbed by rolling, sliding, and saltation can influence the mineralogy and chemical composition of the sediments though size sorting in rivers (Garzanti et al., 2010, 2011). It is well known that fine grain size fractions in sediments and soils have higher radiocesium concentrations (Livens and Baxter, 1988; Cundy and Croudace, 1995; Spezzano, 2005; Tsukada et al., 2008; Tanaka et al., 2013a, 2015). Therefore, the grain size composition can influence the total radioactivity of sediment. The higher contribution of fine-grained sediments will increase the radiocesium concentration in bulk sediments. However, analysis of bulk sediment samples does not provide data to distinguish the two factors controlling radiocesium concentration, i.e. grain size composition of sediments and radioactivity in their provenance areas. In this context, the size distribution of radiocesium is an important factor that needs to be considered to obtain a better understanding of radiocesium in deposited sediments.

Because rivers are dynamic systems, deposited sediments can be resuspended and transported further downstream when the water level rises after heavy rainfall. Therefore, it is not clear whether sediments remain on the sandbars or not during subsequent flooding events after deposition. The depth distribution of radiocesium in sediment on a sandbar can provide us with the history of the transport of radiocesium-bearing particles. In particular, as noted above, analysis of size-fractioned sediments allows us to discuss what factors control and change the concentration of radiocesium in sediment layers. With this aim in mind, we investigated the depth distribution of radiocesium in bulk sediments as well as in size-fractioned sediments.

## 2. Samples and methods

In this study, we collected sediment samples at the Kuroiwa site in the Abukuma River on June 27, 2013 (Fig. 1). The Abukuma River, the largest river in Fukushima Prefecture, flows into the Pacific Ocean through Miyagi Prefecture near the river mouth. The catchment area of the Abukuma River is 5390 $km^2$. The Kuroiwa site is located in the midstream region of the catchment area (N37°43′36″, E140°28′22″). The sediment samples were collected on a sandbar located on the right-hand riverside toward the downstream side. The sandbar is not submerged except for when the river level rises, indicating that the sediments were transported from upstream areas during heavy rainfall followed by flooding. The rainy season occurs every year in Japan, usually around mid-June to late July in Fukushima. In addition, typhoons often occur in August and September, and some of these impact the Japanese islands. Bank revetments protect both riversides at the Kuroiwa site; therefore, there is only a small direct supply of particulate matter from the surrounding areas compared with fluvial material transport from upstream areas. To study the depth distribution of radiocesium, sectioned sediment samples taken at 2 cm intervals over a depth of 0–20 cm were collected using a scraper plate over a sampling area of 450 $cm^2$ (15 cm × 30 cm) (Loughran et al., 2002; Kato et al., 2012).

The collected sediments were dried at room temperature before measurement of the radioactivity. Sediment samples collected at depth intervals of 4–6 cm, 16–18 cm, and 18–20 cm were separated into different grain size fractions, following the procedure described in Tanaka et al. (2015). Grains larger than silt size (>63 μm) were separated into five fractions: granules (>2000 μm), very coarse sand (850–2000 μm), coarse sand (500–850 μm), medium sand (250–500 μm), fine sand (125–250 μm), and very fine sand (63–125 μm) using a wet sieving method. Then, grains smaller than the silt size were further separated into the following size fractions: 40–63 μm, 20–40 μm, 10–20 μm, 2–10 μm, and <2 μm respectively, by sedimentation according to Stokes' law (Livens and Baxter, 1988; Cundy and Croudace, 1995; Clifton et al., 1999). As in our previous works (Tanaka et al., 2014, 2015), we did not remove any organic material from the sediment samples in order to keep samples in the original conditions in the environment. Therefore, potential aggregates composed of minerals and organic material could not be separated precisely (Phillips et al., 2000; Droppo, 2001).

The bulk and size fractioned sediment samples were homogenized before measurement of the radiocesium content using γ-ray spectrometry. The coarse grain samples were ground using an agate mortar into a fine powder before measurement. Samples with a small mass were loaded into a cylindrical polystyrene container with an inner diameter of 2.0 cm and height of 4.5 cm. A second container with an inner diameter of 5.0 cm and height of 6.8 cm was used for samples with larger mass.

The radioactivity was measured using γ-ray spectrometry employing a planar type Ge semiconductor detector (CANBERRA, GC4018/7915-30/ULB-GC) connected to a multi-channel analyzer (Tanaka et al., 2013b, 2015). The γ-ray count emitted by the $^{137}Cs$ in the samples was monitored at 662 keV. Then, the count rate was converted into a radioactivity value based on a calibration of the detection efficiency on the geometry of a sample in the same container using the IAEA reference material IAEA-444. All the data were obtained along with a standard deviation from the counting statistics within 10% of the data value. All the activities were decay-corrected to the sampling date of June 27, 2013. Then, the activity concentration in Bq/kg units was calculated based on the dry weight of the sample.

## 3. Results and discussion

### 3.1. Depth distribution of radiocesium in the bulk sediments

The analytical results of the $^{137}Cs$ concentration in the bulk sediment samples are given in Table S1. The depth distribution of $^{137}Cs$ showed the concentration range between 170 and 350 Bq/kg in sediment layers above 16 cm (Fig. 2, Table S1). The $^{137}Cs$ concentration increased abruptly below a depth of 16 cm. The deepest layer of the 18–20 cm interval showed the highest $^{137}Cs$ concentration of 3100 Bq/kg, which was an order of magnitude higher than the $^{137}Cs$ concentration above 16 cm. The depth distribution of $^{137}Cs$ in the bulk sediment showed an opposite trend to that observed in undisturbed soils, which show the highest concentration at the surface that decreases with depth (Kato et al., 2012; Ohno et al., 2012; Tanaka et al., 2012). Kato et al. (2012) reported 9400 Bq/kg of $^{137}Cs$ concentration in surface (0–0.5 cm) soil collected in Kawamata Town on April 28, 2011 (decay-corrected to the sampling date). Tanaka et al. (2012) also reported that $^{137}Cs$ concentrations in surface soils at 0–2.5 cm depth were 4400 and 6200 Bq/kg at Koriyama City and 420 Bq/kg at Yabuki Town, which were decay-corrected to the sampling date of April 13, 2011. The three sites at Kawamata Town, Koriyama City and Yabuki Town are located 20, 35 and 70 km, respectively, at upstream side from the Kuroiwa site in the Abukuma catchment area. The surface soils at the Kawamata and Koriyama sites showed higher $^{137}Cs$ concentrations than the deepest layer at the Kuroiwa site even after decay-correction to June 27, 2013. However, the Yabuki site had lower $^{137}Cs$ concentration in surface soil due to the initial low deposition density of $^{137}Cs$.