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The effects of sediment transport on temporal variation in radiocesium concentrations in very shallow water off the southern coast of Fukushima, Japan

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ABSTRACT

We studied the very shallow coastal water off Iwaki City, Fukushima Prefecture, and investigated: (1) temporal variation in ^{137}Cs concentrations; (2) particle-size distribution of sediments; and (3) the effect on variation by waves, component-fractionated concentration of radiocesium and mineral composition at three sampling stations (Yotsukura, Ena rocky reef and Ena sandy station). There was a decline in ^{137}Cs concentrations in sediment samples at all sampling stations between 425 and 1173 days after the accident. All stations had fluctuations in ^{137}Cs concentrations between 425 and 800 days. At Ena sandy station and Ena rocky reef stations the declines in ^{137}Cs concentrations slowed from about 800 days after the accident. Fluctuations in particle median diameters were seen, as well as in ^{137}Cs concentrations. At Yotsukura, where the fluctuation in median diameter was small, a constant decrease in ^{137}Cs concentrations was observed. We considered that bioturbation may contribute the constant decrease. At Ena sandy station, where the fluctuation of the median diameter was large, the fluctuation in ^{137}Cs concentrations was also large. The movement of sediments was evaluated by the Shields parameter, and results indicated that at any station where the sediment was moved more frequently, the fluctuation in ^{137}Cs concentrations was also large. The highly contaminated small particles moved from our stations due to wave action between 425 and 800 days after the accident. The remaining relatively large particles might contribute to the slowing down in reduction of ^{137}Cs concentrations from 800 days after the accident. However, the ^{137}Cs concentrations in sediments in very shallow water off the southern coast of Fukushima may continue to decline over time.

1. Introduction

The Great East Japan Earthquake and tsunami that struck on 11 March 2011 caused a serious accident at the Tokyo Electric Power Company (TEPCO) Fukushima Dai-ichi Nuclear Power Plant (FDNPP) (IAEA, 2011). By February 2012, 7.07 PBq of radiocesium (3.52 PBq of ^{134}Cs and 3.55 PBq of ^{137}Cs) were estimated to have been directly released into the ocean from the port of FDNPP (Kobayashi et al., 2013). In addition, the inputs to the coast of Fukushima Prefecture, including both direct release and atmospheric input, have been estimated to be 11–27 PBq (Bailly du Bois et al., 2012; Charette et al., 2013; Rypina et al., 2013). Those radiocesium were mainly diffused around the southern coast of the FDNPP after the accident (Aoyama et al., 2012; Tsumune et al., 2012). Since radiocesium in seawater was rapidly diluted after diffusion (Aoyama et al., 2013a, b), its concentrations off the coast of Fukushima Prefecture decreased exponentially during the first year (Kusakabe et al., 2013; Kusakabe, 2014; Oikawa et al., 2013; Buesseler et al., 2011). The concentrations in surface water at Tomioka, Fukushima, then reached 16.4 mBq/L in March 2016 (Aoyama et al., 2017). The concentrations of offshore water also ranged 0.9–8 mBq/L in January 2015 (Takata et al., 2016).

The released radiocesium exists as cations in seawater, contaminated suspended solids and precipitated to the seabed (Kanda, 2014). In June 2011, a high concentration (maximum 1020 Bq/kg) of radiocesium was detected after the accident in sediments of the northern coast of Ibaraki (Otosaka and Kobayashi, 2013). Although the

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concentration of radiocesium in sediments has declined steadily (Kusakabe et al., 2017), in February 2016, concentrations were still higher at 0.8–141 Bq/kg than the pre-accident level (Kusakabe et al., 2017), and long-term retention of radiocesium remains a concern (Kusakabe, 2014).

In addition, the food web of the coastal area contains microalgae (i.e., organic matter) adhering to sediment particles as a primary producer, and polychaetes, crustaceans, echinoderms and mollusks as primary consumers (Sohtome et al., 2014). Immediately after the accident, radiocesium was detected in these benthic species (Arakawa et al., 2015). Because these benthos were prey species of some demersal fish (Kasamatsu and Ishikawa, 1997), it was suggested that there is a possibility of radiocesium transfer from benthos to demersal fish contributing the slow decline in radiocesium concentrations in these demersal fish (Bezhenar et al., 2016; Buesseler, 2012, Buesseler et al., 2017; Johansen et al., 2015; Tateda et al., 2013, 2015, 2016; Wang et al., 2016). It was reported that organic matter actually attached to the sediments has an apparently high selectivity for radiocesium (Ono et al., 2015). Sohtome et al. (2014) described that the food web transfer of radiocesium through ingestion of benthos (especially Polychaeta and Malacostraca had slow decreasing trends) around the sediments is a necessary prerequisite to explain the above decreasing trend in demersal fish. When considering the possibility of radiocesium transfer to marine biota from sediments in coastal areas (especially in very shallow water), it is essential to understand the spatio-temporal variation in radiocesium concentrations in sediments of very shallow water.

In very shallow water, sediment particles are moved by bed load transport as well as suspended load transport (van Rijn, 1984). Then, to understand the spatio-temporal variation in radiocesium concentrations in surface sediments, it is important to evaluate the effect of sediment transport including bed load transport. Sediment transport containing radiocesium due to suspended load off the coast of Fukushima Prefecture after the accident was reported using sediment trap for offshore research (Honda et al., 2013; Buesseler et al., 2015).

However, information about the sediment transport of very shallow water is limited. In this report, we focused on the very shallow coastal water off Iwaki City, Fukushima Prefecture, and investigated: (1) temporal variation in radiocesium concentrations in surface sediments; (2) particle-size distribution of surface sediments; and (3) the effect of waves on the variation in radiocesium concentration, component-fractionated concentration and mineral composition.

2. Materials and methods

2.1. Field sampling

Sediment samples were collected nine times over 2 years from 2012 to 2014 (May, July, October, December 2012; February, May, October 2013; January, May 2014) in the very shallow water off Iwaki City, Fukushima Prefecture. Three stations several meters from the shore were selected: Yotsukura station (37.119°N, 141.010°E; bottom depth 1 m); Ena rocky station (36.954°N, 140.951°E; bottom depth 5 m) and Ena sandy station (36.955°N, 140.944°E; bottom depth 5 m) (Fig. 1). Three sediment samples were collected at each site, with the exception of the first sampling with single sample at each site. No sediment samples were collected in October 2013 and May 2014 at Ena sandy station as construction work was being carried out there. The distance from the FDNPP is about 35 km to Yotsukura station and about 50 km to Ena rocky and sandy station. To investigate the effect of waves on the variation and/or fluctuation in radiocesium concentration in surface sediment, the sediment sample in the sandy station was collected quantitatively (50 cm length × 10.5 cm width × 3 cm depth) using a plastic scoop by SCUBA diving. In the rocky station, sediments on rock were collected into plastic bags by an air-lift method, using air from a SCUBA tank (Fig. 2). A wavemeter was located in the eastern part of Onahama Port (36.918°N, 140.922°E; depth 23.8 m) (Fig. 1).

2.2. Radiocesium counting

Collected sediments were brought to a laboratory and dried for 5 h at 105 °C, then passed through a 2 mm mesh to remove gravel and shells (MEXT, 2004). Sediments were then shaken for 5 h at 200 rpm in a horizontal turn sieve (SKH-01, AS ONE, inc., Osaka) and classified into four particle-size classes (< 125 μm, 125–250 μm, 250–500 μm and 500–2000 μm). Dried and classified samples were placed in 100 mL plastic containers (U-8, AS ONE, inc., Osaka) and the γ-ray spectrum was measured by a Ge semiconductor detector (GEM20-70, SEIKO EG&G CO., LTD, Tokyo; less than 2keV/1.33 MeV of resolution). An efficiency calibration of the detector was made with volume radioactivity standard gamma sources (MX033U8PP, Japan Radioisotope Association, Tokyo). Measurements were given in 7200 s. Concentrations of radiocesium were represented as Bq/kg-dry (MEXT, 2004) and were decay-corrected to the date of collection. The relative uncertainty for ^{137}Cs activity was in the range of 8.2–15% (38–129 g) for sediment samples (k = 2).

2.3. Fractionation of ^{137}Cs

Otosaka and Kobayashi (2013) separated and defined three types of fraction of ^{137}Cs in sediments: ion-exchangeable, organic-bound and residue. The ion-exchangeable fraction is a form that couples ^{137}Cs and sediments electrically, and is exchangeable for cations such as K^+ in seawater. The organic-bound fraction is radiocesium contained in organic matter attached to sediment particles, and the residue is likely to be bound to minerals in sediment particles. Following Otosaka and Kobayashi (2013), each fraction was extracted and radiocesium concentrations were measured by Ge semiconductor detector as follows.

First, radiocesium concentrations of size-classified samples were measured with a Ge semiconductor detector (sample 1, all three types of fraction mentioned above). Second, the ion-exchangeable fraction was extracted by adding 1 M ammonium acetate to the powdered sample 1 (10 ml/g) and rotated for 6 h at room temperature (25 ± 1 °C). The extraction liquid was removed by filtration through a 0.45-μm membrane filter. Half of the residue was rinsed and dried for 5 h at 105 °C, and used for radiocesium counting (sample 2: organic-bound fraction and residue). To reduce the measurement error at this step, the counting time was set to 72,000 s. To the remaining half of the residue 10% hydrogen peroxide was added and reacted for 6 h at room temperature. The extraction liquid was then removed and the residue after extraction was dried as in the above process for sample 1, and the radiocesium concentrations were measured (sample 3: only residue). The radiocesium concentrations of each fraction were calculated by subtraction of concentrations of sample 1 from sample 2 and sample 2 from sample 3, respectively.

2.4. Measurement of particle-size distribution and identification of mineral composition

The particle-size distribution (0.375–2000 μm) of sediments was measured using laser diffraction/scattering particle-size analyzers (LS 200, Beckman Coulter, Inc., Tokyo). Calculation of median diameter (D_m) was based on particle-size distribution.

Sawhney (1970) reported that radiocesium was generally selectively adsorbed to 2:1-type layered silicate mineral such as mica. The structural analysis of crystalline material and measurement of mineral content for samples from Ena rocky station were carried out by X-ray diffraction methods. We initially assumed that high concentration sediment accumulated in the interstices of the rock. Therefore, we chose Ena rocky station.

2.5. Calculation of Shields parameter using wave data

The effect of external forces (waves or currents) on sediments was

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