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Radioactive cesium concentrations in coastal suspended matter after the Fukushima nuclear accident

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ABSTRACT

Radioactive cesium concentrations in the suspended matter of the coastal waters around the Fukushima Daiichi Nuclear Power Plant (FDNPP) were investigated between January 2014 and August 2015. The concentrations of radioactive cesium in the suspended matter were two orders higher in magnitude than those determined in the sediment. In addition, we discovered highly radioactive Cs particles in the suspended matter using autoradiography. The geometrical average radioactivity of particles was estimated to be 0.6 Bq at maximum and 0.2 Bq on average. The contribution ratio of highly radioactive Cs particles to each sample ranged from 13 to 54%, and was 36% on average. A major part of the radioactive Cs concentration in the suspended matter around the FDNPP was strongly influenced by the highly radioactive particles. The subsequent resuspension of highly radioactive Cs particles has been suggested as a possible reason for the delay in radioactive Cs depuration from benthic biota.

1. Introduction

A nuclear accident occurred at the Fukushima Daiichi Nuclear Power Plant (FDNPP) after the Great East Japan Earthquake and tsunami on March 11, 2011. A large amount of radioactive cesium (Cs) was released into the atmosphere and the ocean from the FDNPP as a result of venting operations and hydrogen explosions (Chino et al., 2011; Winiarek et al., 2012; Buesseler et al., 2017). After the FDNPP accident, the accident-derived radioactive Cs has been detected in the atmosphere, lithosphere, biosphere, and hydrosphere (e.g., Chino et al., 2011; Yasunari et al., 2011; Buesseler et al., 2012; Yoshida and Kanda, 2012; Nagao et al., 2013). While the majority of the accident-derived radioactive Cs in surface waters adjacent to the FDNPP reportedly decreased by over four orders of magnitude from April 2011 to March 2015 (Aoyama et al., 2016; Buesseler et al., 2017), the concentrations of radioactive Cs in coastal sediment have decreased more slowly than those in seawater and demonstrate large fluctuations (Otosaka et al., 2014). Recently, a significant number of radioactive particles containing the higher ^{134}Cs and ^{137}Cs, which are radioactive by up to several dozen Bq per particle, were found in the soil, plant, and air dust samples (Adachi et al., 2013; Itoh et al., 2014; Satou et al., 2016). These samples contained water-insoluble spherical Cs-bearing (Adachi et al., 2013) and/or non-spherical heterogeneous Cs-bearing particles (Satou et al., 2016; Furuki et al., 2017; Higaki et al., 2017). This suggests that radioactive Cs may be preserved in soil for a long duration, primarily because of its insoluble characteristics. However, the pulse input of radioactive Cs associated with suspended matter takes place from the land to coastal ocean during heavy rains (Nagao et al., 2013). Therefore, it is highly likely that radioactive Cs-bearing particles would accumulate in the ocean, and be deposited in the coastal sediment.

Considering the radioactive Cs contamination of fish, the concentration of radioactive Cs in Japanese rockfish (*Sebastes cheni*) and flatfish (*Microstomus achne*) has been decreasing slower than in other fish species (Japan Fisheries Agency, 2016). The persistent elevation of radioactive Cs in certain species is likely due to their sediment and suspended matter-associated food chains (Johansen et al., 2015). The *S.*

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cheni and the *M. achne* prey on benthic crustacea and polychaeta that feed mainly on sediment and suspended organic matter (Igarashi and Sohtome, 2013; Matsumoto et al., 2015). Furthermore, resuspension of suspended matter increases with decreasing particle size (Biasi et al., 2001). In contrast, the concentration of radioactive Cs reportedly increased with the decreasing particle size (Tanaka et al., 2013). Therefore, suspended matter cycles may be an important mechanism to explain the persevering radioactive Cs pollution in the coastal environment. However, to the best of our knowledge, radioactive Cs concentrations in suspended matter have not been evaluated to date.

In this study, we investigated the radioactive Cs concentrations in the suspended matter of coastal waters and observed radioactive particles containing the higher ^{134}Cs and ^{137}Cs using autoradiography. Subsequently, we estimated the relative contributions of the higher ^{134}Cs and ^{137}Cs particles to the marine environment.

2. Materials and methods

2.1. Sample collection

Monthly or bi-monthly observations were carried out at the Kuma River estuary by the fishing vessel Koumei-maru between January 2014 and June 2015 (Station A; Fig. 1). Coastal observations were carried out by the research vessel Umitaka-maru of the Tokyo University of Marine Science and Technology between May 11 and May 18, 2015, and the Oshoro-maru of the Hokkaido University between July 26 and August 7, 2015 (Station B-I; Fig. 1). Suspended matter samples for radioactive Cs measurement were collected by in situ filtration with large-volume pumps (WTS-LV, McLane Research Laboratories, Inc., Falmouth, MA). The samples were filtered through filters with 142 mm diameter and 0.45 μm pore-size (PVDF, Durapore membrane filter, Merck Ltd., Tokyo, Japan). Suspended matter samples were collected from the bottom water and only station I collected the samples at the surface water (Fig. 1). The filters were wrapped in aluminum foil and stored at −25 °C.

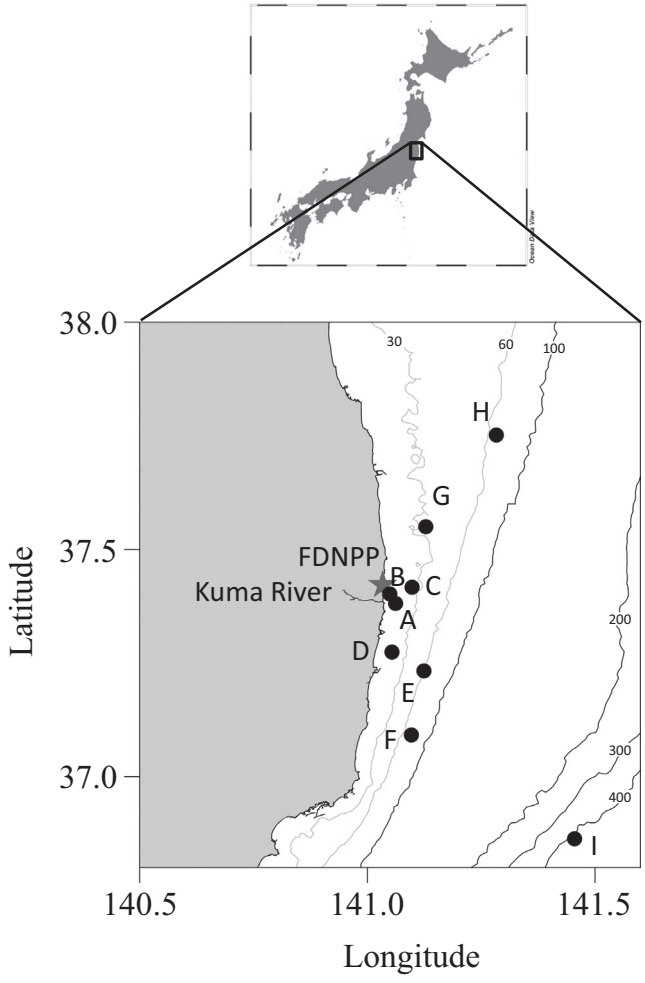

Fig. 1. Map of study site. Locations of sampling sites and FDNPP are indicated by black circles and gray star, respectively.

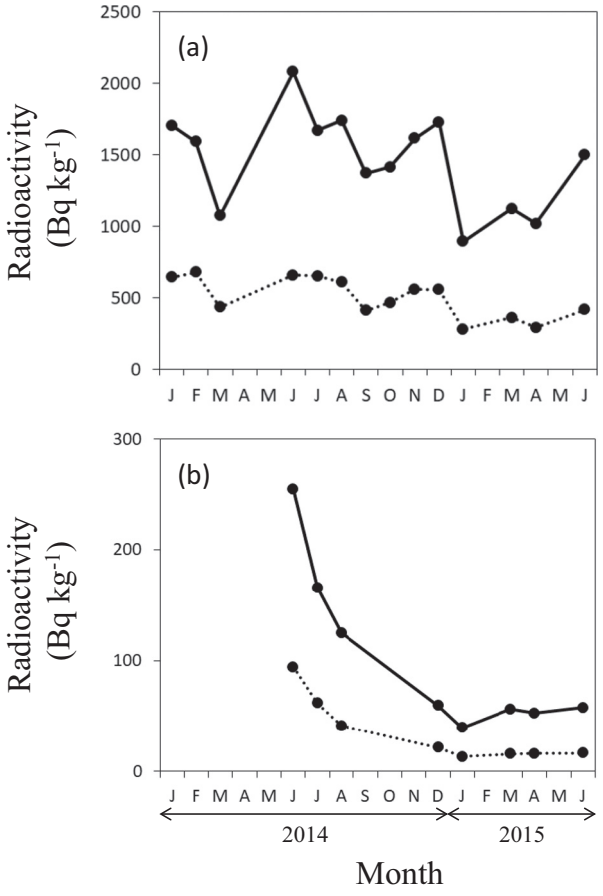

Fig. 2. Temporal variations of (a) suspended matter ^{134}Cs (dashed line) and ^{137}Cs (solid line) concentrations (Bq kg^{-1}) and (b) sedimentary ^{134}Cs (dashed line) and ^{137}Cs (solid line) concentrations (Bq kg^{-1}) at the Kuma river estuary (station A). The decay correction of concentration for ^{134}Cs and ^{137}Cs was done at each sampling date.

Surface sediment was collected at each station to compare the Cs concentration with that of suspended matter. Surface sediment samples were collected using a simple core sampler (Rigo Co., Ltd., Tokyo, Japan) at the Kuma River estuary (Station A) and a multiple-core sampler (Rigo Co., Ltd., Tokyo, Japan) at other stations (Station B–I). The sediment core samples were cut into 0–1 cm sections, put into polyethylene bags, and stored at −25 °C.

2.2. Sample analysis

2.2.1. Radioactivity measurement

The suspended matter samples were removed from the filters using a plastic spatula. Subsequently, the samples were dried at 60 °C, and filled in Teflon tubes. The specific gamma-rays of ^{134}Cs (604 and 795 keV) and ^{137}Cs (661 keV) were measured using a Ge detector. The radioactive Cs concentrations of suspended matter samples were measured using a gamma-ray spectrometer with a well-type Ge detector (Canberra EGPC 250-P21, Meriden, CT; 2.3 keV/1.33 MeV resolution and 20% relative efficiency). The sediment samples were freeze-dried