Spatiotemporal distribution of radioactive cesium released from Fukushima Daiichi Nuclear Power Station in the sediment of Tokyo Bay, Japan

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The spatial and temporal distribution of ^{134}Cs and ^{137}Cs released from Fukushima Daiichi Nuclear Power Station in the Tokyo Bay sediments were investigated. The total radioactivity of ^{134}Cs and ^{137}Cs detected in the Tokyo Bay sediment ranged from 240 to 870 Bq/kg-dry in the estuary of Arakawa River, but the activities detected in other sites were about 90 Bq/kg-dry or less. These results suggested that radioactive cesium, which precipitated to the ground, was carried to the river along with clay particles by rainfall and transported to the estuary. The vertical distribution of radioactive cesium showed that it invaded deeper than estimated based on the accumulation rate of the sediment. It was described that the vertical distribution of radioactive cesium was affected by physical mixing of sediments by tidal current, flood, and bioturbation of benthos.

Key Words : *Fukushima Daiichi Nuclear Power Station Accident, Radioactive Pollution, Tokyo Bay, Sediment, Radioactive Cesium*

1. Introduction

Fukushima Daiichi Nuclear Power Station suffered serious damage by massive tsunami on March 11, 2011, releasing a large amount of radioactive fission products, including ^{90}Sr ^{129}Te, ^{129m}Te, ^{132}Te, ^{131}I, ^{132}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{139}Pu, ^{140}Pu, and ^{140}Ba [1)]. It has been estimated that about 1.5×10^{17} Bq of ^{131}I and about 1.3×10^{16} Bq of ^{137}Cs were released to the atmosphere [2)]. The radioactive nuclides such as ^{134}Cs and ^{137}Cs were precipitated onto ground from atmosphere by rain fall on March 21, 2011 [3)], and absorbed on clay minerals in the soil due to ion exchange [4)]. As a result, the metropolitan area and the Kanto district received the serious radioactive pollution by the Fukushima Daiichi Nuclear Power Station accident. The area of Tokyo Bay is about 1000 km^2 and its average water depth is about 17 m [5)]. Tokyo Bay is an enclosed inner bay in the southern Kanto district, and surrounded by Tokyo, Kanagawa, and Chiba Prefectures. Consequently, Tokyo Bay receives anthropogenic contamination mainly through river discharge as a final sink of the metropolitan area and the Kanto district. The major rivers flowing into Tokyo Bay are the Arakawa River, the Tamagawa River, and the Edogawa River [5)]. The Edogawa River watershed includes Kashiwa City which is highly radioactive polluted area [6)]. Therefore, the sediment of Tokyo Bay may have received radioactive contamination from the Fukushima Daiichi Nuclear Power Station accident. In order to clarify the behavior of the radioactive pollution of Tokyo Bay, the spatial and temporal distribution of the radioactive cesium in the sediment of Tokyo Bay was investigated. The data presented herein represent the first measurements of the radioactive contamination of the sediment of Tokyo Bay.

2. Sample and Analytical Procedure

(1) Sample collection

The sampling points in Tokyo Bay are shown in Figure 1. The sediment cores (sites A, B, C, and D) were obtained by divers using an acrylic corer (10 cm diameter, 100 cm length) on August 20, 2011. The lengths of the core samples obtained from sites A, B, C, and D were 34, 50, 70, and 50 cm, respectively. The surface sediments were collected with an Ekman-Birge sampler on October 1 and 2, 2011, and December 3, 4 and 5, 2011. The surface samples were obtained at the subcores (5 cm diameter, 15 cm length) from the Ekman-Birge sampler.

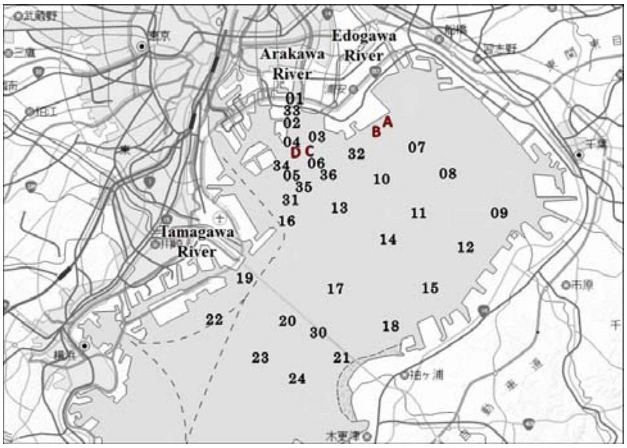

Figure 1. Map showing the sampling sites in Tokyo Bay. This map located from 35° 22' N to 35° 43' N and 139° 33' E and 140° 08' E. The map was provided by the Geographical Survey Institute, Japan.

(2) Analytical procedure

The core samples were cut into slices of 0.5, 1, and 2 cm in thickness, and the surface samples were homogenized to a 5 cm depth from the seabed. To measure ^{134}Cs and ^{137}Cs, the sediment samples were dried at 60 °C to a constant weight, pulverized with an agate mortar, and packed into a plastic container (6 cm diameter, 1.5 cm thickness). The radioactivity of ^{134}Cs (605 keV) and ^{137}Cs (662 keV) was determined by gamma-ray spectrometry using a HPGe detector (ORTEC, LO-AX/30P) connected with a 4096ch MCA. The activities of ^{134}Cs and ^{137}Cs in the sediment samples were decay-corrected to the sampling date. The detection efficiency of the detector and the geometrical efficiency of the sample volume were corrected using NIST Environmental Radioactivity Standard SRM 4350B (river sediment), SRM 4354 (freshwater lake sediment), and SRM 4357 (a blend of ocean sediment collected off the coast of Sellafield and in Chesapeake Bay). The detection limit of ^{134}Cs and ^{137}Cs in the dry sediment samples was estimated to be within 0.5 Bq/kg [7].

3. Results and Discussion

(1) Special distribution of radioactive cesium in the surface sediment of Tokyo Bay

The spatial distribution of the activities of $^{134+137}$Cs in the surface sediments (0-5cm depth layer) in Tokyo Bay on October 1 and 2, 2011 is shown in Figure 2. The activities of $^{134+137}$Cs at the estuary of the Arakawa River and the Tamagawa River ranged from 240 to 870, and 90 to 110 Bq/kg-dry, respectively. But the activities from the

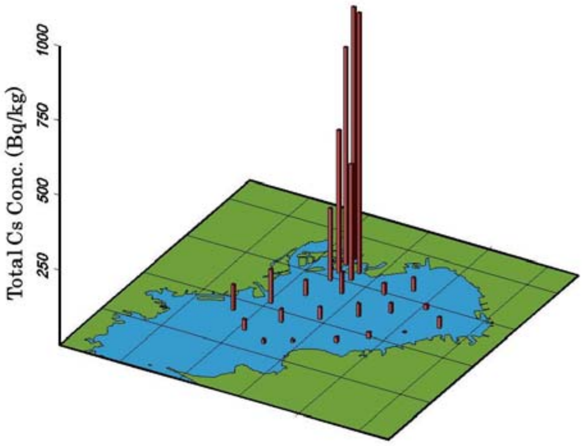

Figure 2. Spatial distribution of radioactive cesium in the sediments of Tokyo Bay collected on October 1 and 2, 2011 [8].

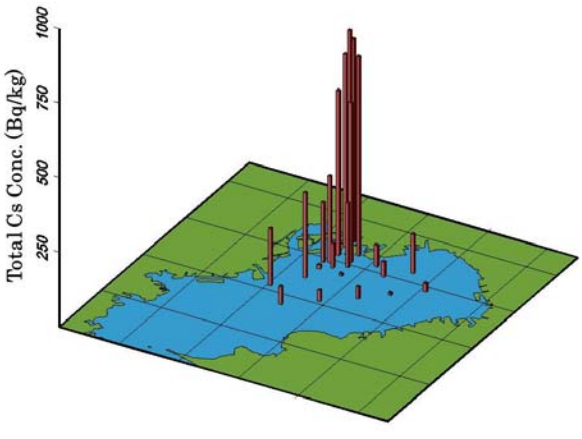

Figure 3. Spatial distribution of radioactive cesium in the sediments of Tokyo Bay collected on December 3, 4 and 5, 2011 [8].

other sites were about 90 Bq/kg-dry or less. The highest activities of $^{134+137}$Cs were observed in site 03 at the Arakawa River estuary. The radioactive cesium in the sediments of Tokyo Bay was determined to have resulted from this accident because the short half-life nuclide ^{134}Cs was detected. The spatial distribution of radioactive cesium in Tokyo Bay was indicated that the radioactive cesium immediately deposited after flowing into the river mouth. The results also suggested that the radioactive cesium, which precipitated to the ground, was carried to the river along with the clay particles by rainfall and transported to the estuary.

The spatial distributions of $^{134+137}$Cs on December 3, 4, and 5, 2011 are shown in Figure 3. The activities of $^{134+137}$Cs at the Arakawa River estuary ranged from 290-680 Bq/kg-dry. The activities exhibited a slightly decreasing tendency, but the contamination area was expanded.

(2) Vertical distribution of radioactive cesium in the sediment sample of Tokyo Bay

The vertical distributions of the activities of $^{134+137}$Cs in the sediment cores in Tokyo Bay collected on August. 20, 2011 are shown in Figure 4. The vertical distribution of $^{134+137}$Cs showed a high activities in the deep than surface

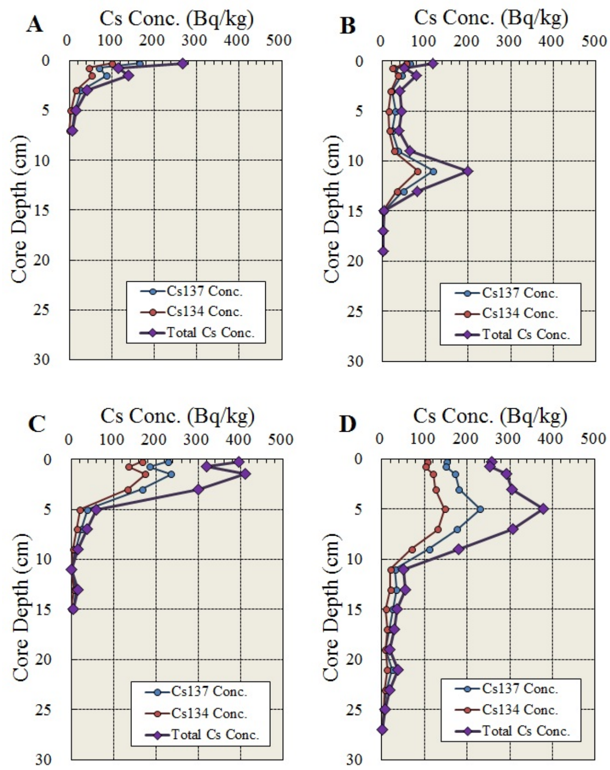

Figure 4. Vertical distributions of radioactive cesium in sediments around Sites A, B, C and D in the Tokyo Bay collected on August 20, 2011.

and it invaded deeper than estimated based on the accumulation rate of the sediment [5] (expect for the A core). For the B and D cores, there were the broad peaks at 10 and 5 cm respectively, and then the activities decreases with the depth. It was estimated that the vertical distribution of radioactive cesium was affected by physical mixing of sediments by tidal current, flood, and bioturbation of benthos [9].

The inventories of radioactive cesium in the A, B, C, and D cores are 600, 5000, 9500, and 18300 Bq/m^2, respectively. It is necessary to use the inventory value in order to assess the radioactive pollution of the sediment.

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