

# In situ measurement of $^{134}\text{Cs}$ and $^{137}\text{Cs}$ in seabed using underwater $\gamma$ -spectrometry systems: application in surveys following the Fukushima Dai-ichi Nuclear Power Plant accident

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Received: 31 August 2014 / Published online: 2 November 2014  
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**Abstract** Two underwater  $\gamma$ -spectrometric systems incorporating NaI(Tl) detectors (the seabed mooring system and the towing system) were used to measure radiocesium in seabed that was released due to the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident and assess the accuracy of in situ measurements on seabed. We also applied a simple calculating method for seabed radiocesium by separating the  $\gamma$ -spectra based on four contributions ( $^{134}\text{Cs}$  +  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ , U-series, and Th-series radionuclides). The concentration of  $^{134}\text{Cs}$  +  $^{137}\text{Cs}$  in sediments at two sites in Sendai Bay were calculated to be between 10–250 Bq/kg-dry through our method with a short integration time (1–5 min).

**Keywords** In situ  $\gamma$ -spectrometry · Radiocesium · Marine sediment · Fukushima Dai-ichi Nuclear Power Plant accident

## Introduction

The accident at the TEPCO's Fukushima Dai-ichi Nuclear Power Plant (FDNPP) on March 11, 2011, resulted in the widespread release of large amounts of volatile radionuclides such as  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , into the environment, particularly over the Pacific Ocean-side of eastern Japan [1]. As regards the marine environment, although concentrations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in seawater are known to have decreased rapidly from tens Bq/L to several mBq/L [2], decrease in the concentration of these radionuclides has occurred only slowly in marine sediment off the coast of Fukushima and

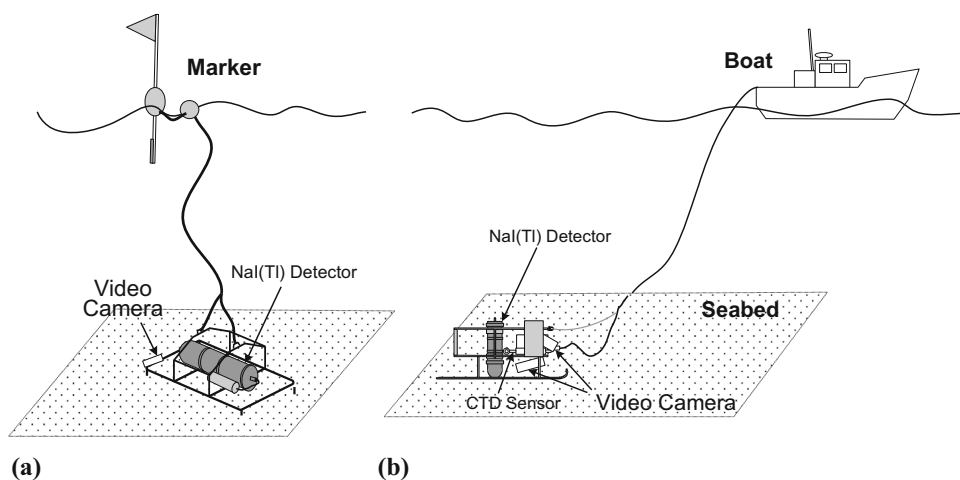
neighboring prefectures. Accordingly, these concentrations remain several orders of magnitude higher around the FDNPP than before the accident [3, 4]. Furthermore, radiocesium in marine sediments is heterogeneously distributed spatially, and its distribution has been observed to vary temporally. In order to clarify the transport mechanisms of radionuclides, particularly in the benthic environment, by the long-term influence of the FDNPP accident, it is necessary to investigate their distribution in the bottom sediment in detail. In situ  $\gamma$ -spectrometry has been shown to be a suitable method for this purpose, owing to the spatial continuity of acquired  $\gamma$ -ray data; this technique has already been applied in the area around the FDNPP [5, 6]. To improve the applicability of in situ  $\gamma$ -spectrometry in such settings, it is essential to investigate measurement stability and accuracy, particularly because the attenuation and scattering of  $\gamma$ -rays by sediment and seawater are extremely complex. However, examination of the stability or accuracy of determination of radionuclides concentration in seabed using this method had not been adequate, because the main purpose of employing this system in the previous studies was to survey the distribution of radionuclides in seabed as differences in concentration by place. In the present study, we assess the accuracy of  $\gamma$ -ray data obtained from underwater  $\gamma$ -spectrometry by conducting a field test in a marine area affected by the FDNPP accident, and employ a simple and rapid method to calculate radiocesium concentrations derived from the FDNPP accident in sediments using data with short integration times.

## Instruments and methods

We set up two underwater  $\gamma$ -spectrometric systems, the towing system and the seabed mooring system, to obtain

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**Fig. 1** Illustration of field operation of seabed  $\gamma$ -spectrometry systems.  
**a** Seabed mooring system,  
**b** Towing system



in situ measurement data on the seabed (Fig. 1). The mooring system (Fig. 1a) logged  $\gamma$ -ray spectra with a 1-min interval at a fixed station, whereas the towing system (Fig. 1b) monitored the  $\gamma$ -ray spectra on the seabed in real time with seabed images using underwater camera, and information on depth, temperature, and salinity. Both the portable systems (15–20 kg) could be easily lifted and retrieved manually in a small boat. Each system had an underwater  $\gamma$ -ray detector with a 3''  $\phi$  spherical NaI(Tl) crystal and 256 ch pulse height analyzer stored in a pressure vessel composed of Delrin<sup>®</sup> resin with a thickness of 10–11 mm. Background  $\gamma$ -spectra of these detectors have been obtained previously [7]. These systems were tested at two sites with different sediment properties in Sendai Bay (37–38 m water depth), approximately 80 km north of the FDNPP in September 2012 (Fig. 2). In order to examine the detection limit of underwater  $\gamma$ -spectrometric systems, continuous  $\gamma$ -ray data were obtained for approximately 2 h at each site using the seabed mooring system, with the NaI(Tl) detector at fixed position relative to the seabed. Moreover, when the towing system is used, it is difficult to rule out the change of efficiency of the NaI(Tl) detector by distance between the NaI(Tl) detector and the seabed. This effect was checked by towing over 300–400 m at a speed of  $\sim 1$  knot at each site (Fig. 2). Sediment samples were also collected by dragging a plastic cup along the seabed, and sediment characteristics for each site were examined.

The efficiency of the NaI(Tl) detector in the measurement of radionuclides in sediments and seawater has been estimated for several sediment types by the Monte Carlo simulation using the EGS5 code [8] in conjunction with a 3D geometric model (Fig. 3), based on average values of bulk density, water content, and elemental composition of continental shelf and slope sediments in the North Pacific [5]. In particular, simulations using the EGS5 code have been conducted for  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ , U-series ( $^{214}\text{Pb}$ ,

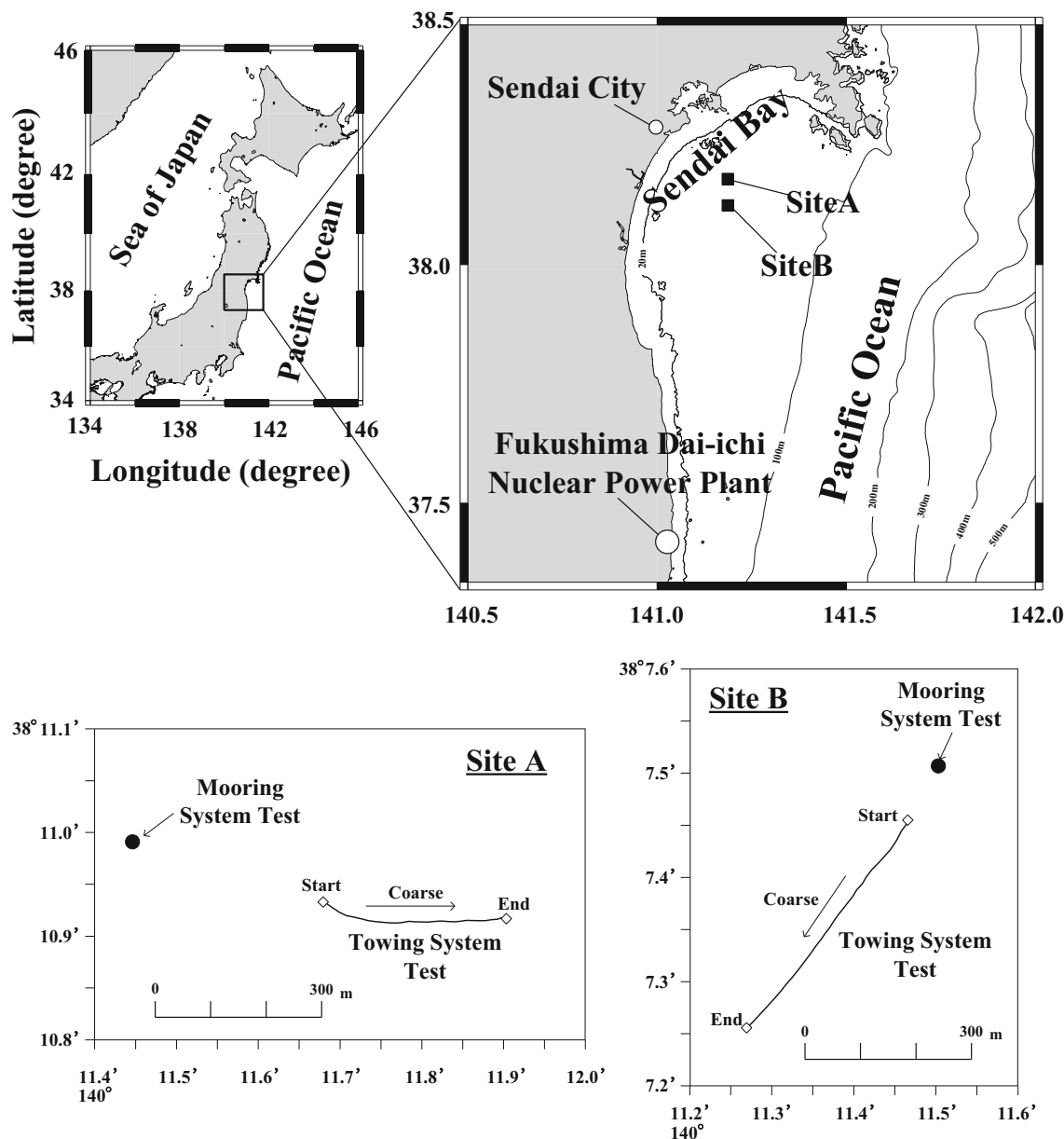
$^{214}\text{Bi}$ ,  $^{226}\text{Ra}$ ,  $^{234}\text{Th}$ ), and Th-series ( $^{208}\text{Tl}$ ,  $^{212}\text{Pb}$ ,  $^{212}\text{Bi}$ ,  $^{224}\text{Ra}$ ,  $^{228}\text{Ac}$ ) nuclides with  $10^8$  runs for each nuclide. The vertical distributions of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  radionuclides were assumed to be homogeneous in the sediment surface, and the distributions of  $^{40}\text{K}$ , U-series, and Th-series nuclides were assumed to be homogeneous across the entire bulk of the sediment.

## Results and discussion

### Efficiency of $\gamma$ -counting for radiocesium in sediments

Conversion factors for estimating  $^{137}\text{Cs}$  concentrations (Bq/g-wet, Bq/g-dry, and Bq/cm<sup>3</sup>) in sediments were calculated using the EGS5 code, based on the count rate of the photopeak (662 keV) using eight kind of sediments with different properties; with bulk density and water content in the ranges 1.4–2.0 g/cm<sup>3</sup> and 20–55 wt%, respectively [5]. These conversion factors varied 1.4 times in Bq/g-wet and 2.5 times in Bq/g-dry, respectively, mainly corresponding to changes in bulk density and water content. In contrast, the conversion factors for volumetric activity (Bq/cm<sup>3</sup>) remained nearly constant regardless of the bulk density and water content of sediments. Information regarding sediment properties can be obtained with a comparatively small uncertainty using volumetric concentration even if it is not available directly.

We plotted the conversion factor for  $^{137}\text{Cs}$  in sediment against the distance between the detector and the sediment (Fig. 4). About 90 % of the photopeak counts for 662 keV in sediment were based on the  $\gamma$ -ray of  $^{137}\text{Cs}$  within a distance of 15 cm from the detector. Our results demonstrate that the conversion factor varies by  $\sim 3$  times in response to changes with distance and by  $\sim 2$  times in response to changes in the vertical distribution of  $^{137}\text{Cs}$  in



**Fig. 2** Location of field testing sites (A and B) for in situ measurement

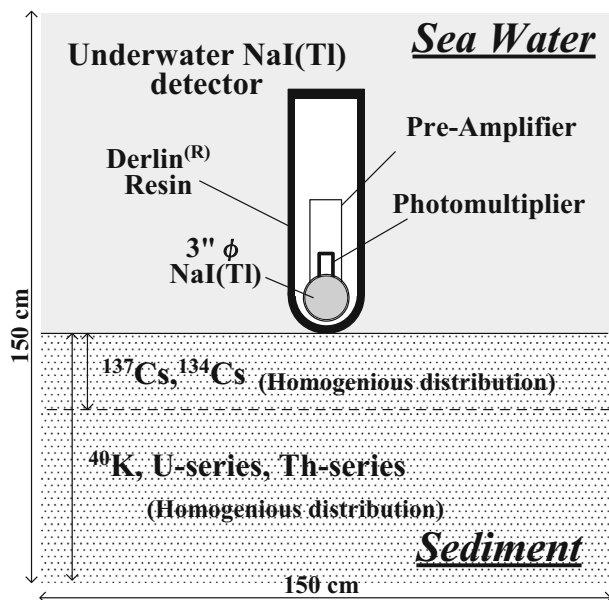
the sediment. As demonstrated by images from an underwater video camera (Fig. 5), the distance between the detector and the sediment surface varied by several centimeters, resulting in the sinking of the system in the muddy sediment and inclination due to the unevenness of the seafloor.

Uncertainty resulting from these variations in distance between detector and seabed can be corrected roughly using video from the underwater camera. If the underwater detector and seabed are irradiated with a marker line such as a laser beam and clear pictures are taken, it will be possible to correct the distance variation with higher certainty using image analysis method for every frame of the

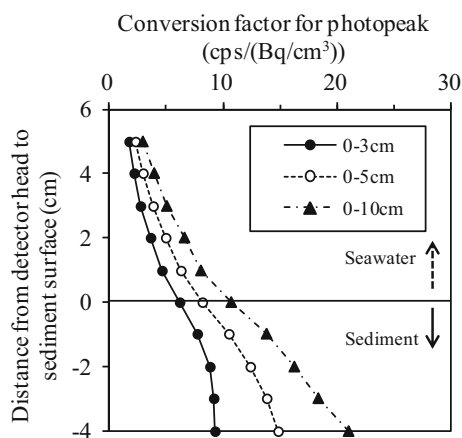
video. However, for high-precision estimations of  $^{137}\text{Cs}$  concentrations, the vertical distribution of  $^{137}\text{Cs}$  must be examined using conventional  $\gamma$ -spectrometry of sediment core samples.

Detection limits and precision

We now discuss the detection limits and repeatability error obtained using long time measurement data at a fixed position by the seabed mooring system. Gamma-ray spectra obtained using the seabed mooring system at Site A and Site B in Sendai Bay are illustrated in Fig. 6, with the detector background measured with Pb-shield in an

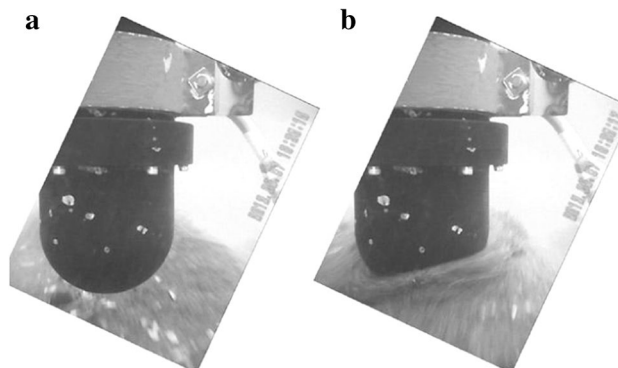


**Fig. 3** 3D geometric model for calculating efficiency using the EGS5 code. The NaI(Tl) crystal was positioned in the center of the calculation area (cubical area of  $150 \times 150 \times 150$  cm)

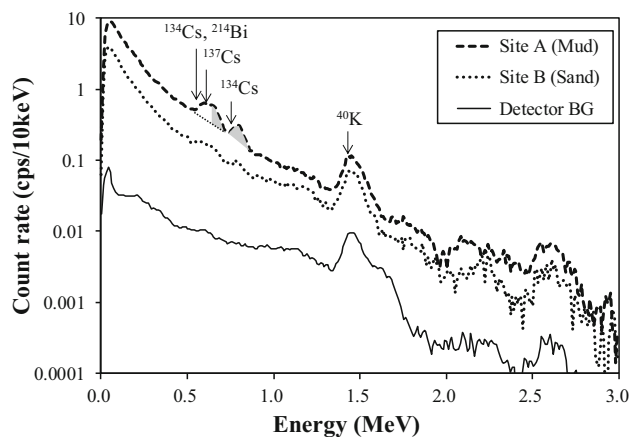


**Fig. 4** Conversion factor for  $^{137}\text{Cs}$  (662 keV) in sediment versus distance from detector to sediment and vertical distribution of  $^{137}\text{Cs}$  in sediment

underground laboratory as a Ref. [7]. The concentrations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in sediments were calculated to be 0.09 and 0.14 at Site A (a muddy site) and 0.03 Bq/cm<sup>3</sup> and below the detectable limit (i.e., <0.05 Bq/cm<sup>3</sup>) at Site B (a sandy site) based on the photopeak counts of the 796 and 662 keV  $\gamma$ -rays, respectively. Here, it was assumed that  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  are distributed homogeneously in the top 5 cm of the sediment, and  $\gamma$ -ray efficiencies were corrected using the distance between the detector and the sediment surface estimated by underwater images. The converted concentrations were 90 and 150 Bq/kg-dry at Site A and



**Fig. 5** Underwater camera images of the **a** contact and **b** detachment of the NaI(Tl) detector head with/from the sediment surface during towing on the seabed at Site B



**Fig. 6**  $\gamma$ -spectrum measured by the seabed mooring system in Sendai Bay with the background of the underwater NaI(Tl) detector for Ref. [7]. Shaded areas on the  $\gamma$ -spectrum indicate energy ranges used to calculate photopeak counts for 662 and 796 keV, respectively

30 Bq/kg-dry and below the detectable limit (i.e., <50 Bq/kg-dry) at Site B, calculated based on the bulk density and water content of sediment samples. These concentrations are within the same range as previous data obtained from sediments from Sendai Bay using conventional  $\gamma$ -spectrometry (several to several hundred Bq/kg-dry for both  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) [4]. Using the method that employs the photopeak counts of the 662 and 796 keV  $\gamma$ -rays, the detectable limits of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in sediments were found to be 50 and 70 Bq/kg-dry, respectively, for  $\gamma$ -spectra with integration times of 10 min. The integration times required to detect 100 Bq/kg-dry of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were 3 and 4 min, respectively. The detection limits for each integration time were defined as  $3\sigma$  of the repeatability error, which was calculated using repeat measurements for each integration time. Based on the photopeak counts of  $\gamma$ -spectra measured by the towing system and the

conversion factors corrected for the average distance between the detector and the seabed (as observed by underwater camera),  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations in sediments were measured to be 90 and 120 Bq/kg-dry, respectively, at Site A and below the detectable limit (i.e., <40 and <60 Bq/kg-dry, respectively) at Site B.

Application of a simple calculation method for the FDNPP-derived radiocesium

We applied a simple and rapid calculation method based on the following features of  $\gamma$ -ray emitters derived from the FDNPP accident on the seabed: (1) the  $^{134}\text{Cs}/^{137}\text{Cs}$  activity ratio was constant ( $\sim 1$ ; decay corrected with respect to March 2011 [4]); (2) only  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were detectable by the in situ  $\gamma$ -ray detector [4]; (3) the effects of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in seawater were negligible; and (4) the influence of cosmic rays was ignored. The measured total  $\gamma$ -spectra were separated into  $^{134}\text{Cs} + ^{137}\text{Cs}$ ,  $^{40}\text{K}$ , and U-series and Th-series radionuclides. Thus, the count rates  $u_1$ – $u_4$  for these four energy ranges can be described by the following equations.

$$\text{(Range 1: 0.45–0.9MeV)} u_1 = f_{\text{Cs}} C_{\text{Cs}1} + f_{\text{K}} C_{\text{K}1} + f_{\text{U}} C_{\text{U}1} + f_{\text{Th}} C_{\text{Th}1}$$

$$\text{(Range 2: 0.9–1.6MeV)} u_2 = f_{\text{Cs}} C_{\text{Cs}2} + f_{\text{K}} C_{\text{K}} + f_{\text{U}} C_{\text{U}2} + f_{\text{Th}} C_{\text{Th}2}$$

$$\text{(Range 3: 1.6–2.3MeV)} u_3 = f_{\text{U}} C_{\text{U}3} + f_{\text{Th}} C_{\text{Th}3}$$

$$\text{(Range 4: 2.3–3.0MeV)} u_4 = f_{\text{Th}} C_{\text{Th}4}$$

$C_{\text{Cs}1-2}$ ,  $C_{\text{K}1-2}$ ,  $C_{\text{U}1-3}$ , and  $C_{\text{Th}1-4}$  represent the count rates of the standard spectra for each energy range, whereas  $f_{\text{Cs}}$ ,  $f_{\text{K}}$ ,  $f_{\text{U}}$ , and  $f_{\text{Th}}$  represent the contribution factors of the associated components. We also corrected for the contribution of  $^{40}\text{K}$  in seawater (12 Bq/L at 34 psu) and the detector background. The contributions of each component were calculated according to a  $4 \times 4$  matrix method.  $^{134}\text{Cs} + ^{137}\text{Cs}$  as count rate of  $\gamma$ -spectra were given as  $f_{\text{Cs}}(C_{\text{Cs}1} + C_{\text{Cs}2})$ . The concentration of  $^{134}\text{Cs} + ^{137}\text{Cs}$  in sediment was converted from  $^{134}\text{Cs} + ^{137}\text{Cs}$  contributions based on count rates using a conversion factor calculated by Monte Carlo simulation; the lateral variations in  $^{134}\text{Cs} + ^{137}\text{Cs}$  concentration in sediments at sites A and B calculated according to this method are illustrated in Fig. 7. The concentrations at sites A and B were calculated using spectra integrated for 1 and 5 min, respectively. Measurement errors at sites A and B with 1 and 5 min integrated spectra, respectively, were about 30 % as  $1\sigma$  of repeatability errors calculated from data obtained by the seabed

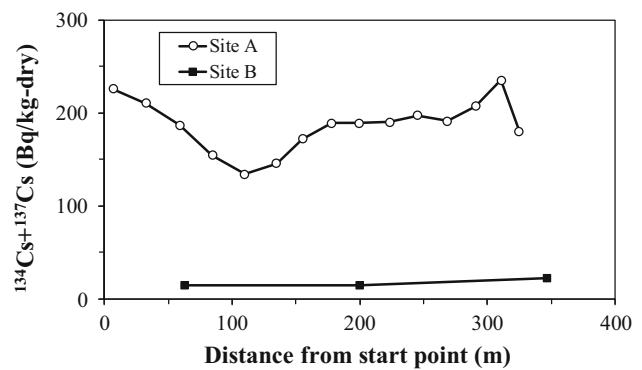


Fig. 7 Spatial variation of  $^{134}\text{Cs} + ^{137}\text{Cs}$  concentration in sediment calculated according to a  $4 \times 4$  matrix method. The concentrations for sites A and B were calculated using spectra integrated for 1 and 5 min, respectively. Radiocesium was assumed to be distributed primarily within the top 0–5 cm of the sediment

mooring system at these two sites. Although this method requires data with a wide energy range covering  $\gamma$ -rays from natural radionuclides, it facilitates the estimation of more detailed lateral variations in  $^{134}\text{Cs} + ^{137}\text{Cs}$  concentrations using  $\gamma$ -spectra than traditional methods and requires only short integration times.

**Acknowledgments** We thank Dr. M. Okano for his advice regarding procedures for field-testing of underwater systems and our data analysis methods. We also thank Prof. S. Nagao for the suggestion to conduct field tests of the seabed mooring system. This work was supported by a Grant-in-Aid from the Watanabe Memorial Foundation for the Advancement of Technology.

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