

Concentration of ^{134}Cs + ^{137}Cs bonded to the organic fraction of sediments offshore Fukushima, Japan

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We conducted a sequential chemical extraction experiment for radiocesium (^{134}Cs + ^{137}Cs) using 21 surface sediment samples and two sets of size-fractionated surface sediment samples collected offshore Fukushima. Our results support earlier reports in this area that organic substances in marine sediments have an apparently higher preference for radiocesium than mineral substances. Observations suggest that mineral and organic substances in the marine sediments offshore Fukushima have the same order of preference for radiocesium, primarily because of the existence of ionic competitors in seawater. The apparent preference of radiocesium for organic material is greater because of the partial coverage of sediment mineral surfaces by organic substances. By using these relationships, we created a 2-D map of radiocesium concentration in sediment organic fraction in offshore-Fukushima region. Combining our results with existing monitoring data of marine benthos in offshore-Fukushima sediments, we estimated a transfer coefficient of radiocesium from sediment organic materials to benthic polychaetes as less than 0.03–0.008.

Keywords: Fukushima Dai-ichi Nuclear Power Plant accident, radiocesium, sediment, organic fraction, sequential extraction

INTRODUCTION

In the years that have passed since the 2011 Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, a dramatic decrease in radiocesium (^{134}Cs + ^{137}Cs) concentrations has been observed in both seawater and marine products in the oceanic region near Fukushima Prefecture. The radiocesium concentration in seawater in the continental margin areas offshore Fukushima reached over 100 Bq/kg immediately following the accident, but has decreased to less than 100 mBq/kg after 2012, and continues to decrease (Oikawa *et al.*, 2013; Kaeriyama *et al.*, 2014). During April–June 2011, 57.7% of marine fishery products caught offshore Fukushima contained radiocesium concentrations higher than the present standard limit for marine food in Japan (i.e., 100 Bq/kg-wet), but during October–December 2013, 98.3% of marine fishery products caught in the same region contained less than 100 Bq/kg-wet (JFA, 2014). In contrast to this significant decrease in radiocesium in water and marine organisms, marine sediment still contains significant levels of radiocesium. The concentration of radiocesium in surface

sediments still exceeds 500 Bq/kg-dry in a wide area of the continental margin shallower than 100 m offshore Fukushima Prefecture, and shows only a small decreasing trend since the time of the accident (Kusakabe *et al.*, 2013; Ambe *et al.*, 2014; Black and Buesseler, 2014). Since present seawater can only contaminate marine fish to the order of 10^0 Bq/kg-wet when considering a typical concentration factor of 1.0×10^2 (IAEA, 2004), marine sediment is the only potential source of radiocesium that can be transferred into the marine ecosystem of the present marine environment off Fukushima. Therefore, assessment of the efficiency of the transfer of radiocesium from sediment to the oceanic ecosystem is a key parameter for predicting future changes in pollution levels in marine products harvested in the Fukushima region.

The Fukushima Prefectural Fisheries Station has made a substantial effort to monitor the radiocesium concentration in benthos. Based on their data, the concentration of ^{137}Cs was less than 40 Bq/kg-wet for polychaetes and less than 20 Bq/kg-wet for crustaceans and mollusks in continental margin areas offshore Fukushima in the autumn of 2012 (Sohtome *et al.*, 2014). Considering that surface marine sediments in this region contain several hundred Bq/kg-dry of ^{137}Cs (Ambe *et al.*, 2014) and that benthic organisms, especially polychaetes, mainly obtain their food from sediments, we can estimate that the trans-

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Table 1. Specifications and measurement results of samples

Station No.	Sampling date	Latitude	Longitude	Bottom depth [m]	Median grain size [μm]	OF [%]	$C_{s_{\text{bulk}}}$ [Bq/kg-dry]	$C_{s_{\text{org}}}$ [Bq/kg-org-dry]	CR	IR [%]
S1	2012.7.11	36°20'	140°55'	257	142	0.8	49 ± 5.5	350	7	5.8
S2	2012.7.11	36°20'	140°50'	120	136	0.7	78 ± 6.6	1440	19	12.0
S3	2012.7.11	36°20'	140°45'	59	889	0.4	153 ± 9.7	2440	16	5.9
S4	2012.7.11	36°20'	140°40'	33	201	1.0	310 ± 20	1090	4	3.4
S20	2012.7.12	36°40'	141°10'	261	233	0.6	103 ± 6.3	520	5	3.0
S21	2012.7.12	36°40'	141°05'	144	265	0.5	60 ± 4.7	850	14	7.0
S22	2012.7.12	36°40'	141°00'	133	161	1.0	180 ± 13	1330	7	7.3
S23	2012.7.12	36°40'	140°55'	111	87	1.6	180 ± 14	960	6	8.9
S24	2012.7.12	36°40'	140°50'	70	116	1.0	270 ± 21	1300	5	4.9
S25	2012.7.12	36°40'	140°45'	33	no data	0.3	69 ± 5.9	490	7	2.4
S59	2012.7.13	37°05'	141°25'	177	247	0.6	83 ± 5.4	470	6	3.2
S60	2012.7.13	37°05'	141°20'	151	225	0.6	104 ± 6.3	2360	23	13.9
S61	2012.7.13	37°05'	141°15'	140	85	1.3	101 ± 7.6	600	6	7.4
S62	2012.7.13	37°05'	141°10'	120	87	1.6	440 ± 27	1200	3	4.5
S63	2012.7.12	37°05'	141°05'	72	158	1.6	690 ± 32	1840	3	4.2
S64	2012.7.12	37°05'	141°01'	25	167	0.9	910 ± 32	3120	3	3.2
S92	2012.7.15	37°40'	141°03.5'	24	118	1.0	710 ± 28	3390	5	4.9
S93	2012.7.15	37°40'	141°05'	28	407	0.2	82 ± 5.9	1270	16	4.0
S94	2012.7.15	37°40'	141°10'	37	723	0.1	31 ± 3.4	780	25	3.2
S95	2012.7.15	37°40'	141°15'	59	1240	0.1	47 ± 4.1	2330	50	5.0
S96	2012.7.15	37°40'	141°20'	100	146	0.7	230 ± 16	2080	9	6.5

Note: For definition of $C_{s_{\text{bulk}}}$, $C_{s_{\text{org}}}$, OF, CR and IR see the text.

fer coefficient of radiocesium from sediments to these organisms is of the order 10^{-1} (i.e., radiocesium is diluted through the sediment-feeding process of benthos). This is unusual because radiocesium generally tends to be concentrated, to a greater or lesser extent, when trophic level increases (IAEA, 2004). However, such a phenomenon can occur if a) some physiological excretion mechanism for radiocesium exists in the benthos or if b) the organic fraction of marine sediments contains a relatively lower amount of radiocesium compared to the sediment bulk concentration. The latter possibility can be assessed if information on the radiocesium distribution among various chemical components of marine sediments is available. However, such information is limited.

In terrestrial soils and freshwater sediments, radiocesium is known to be strongly bonded to clay minerals (e.g., Tsukada *et al.*, 2008; Detriche *et al.*, 2010; Qin *et al.*, 2012). However, Otosaka and Kobayashi (2012), by examining marine sediments collected 70 km south of FDNPP, found that approximately 20% of the ^{137}Cs was contained in the organic fraction, which accounted for only 5% (w/w) of the bulk sediment. This indicates that the organic fraction of marine sediment contains a radiocesium concentration four times higher than the bulk sediment and hence, that marine sediment has a unique sorbent preference for radiocesium. Unfor-

tunately, the findings of Otosaka and Kobayashi (2012) were based on data from only two cores, and the generalizability of their results within the marine environment remains unknown. The primary purpose of this study is to obtain information about the component-fractionated concentration of radiocesium in marine sediments with various chemical/morphological characteristics, as well as to obtain radiocesium concentration levels, and assess the findings of Otosaka and Kobayashi (2012). We also discuss the reason why radiocesium in marine sediment has such a weaker preference for mineral substances compared to terrestrial soils/sediments.

EXPERIMENTAL

Samples and pre-treatments

Sediment cores were collected from 113 stations gridded with a $5' \times 5'$ latitude/longitude interval within the continental margin offshore Fukushima in the summer of 2012 (Ambe *et al.*, 2014). In each core, sediments of the top 0–14 cm were separated into six layers, with a 0–1 cm layer designated as “surface sediment.” The bulk concentrations of ^{137}Cs and ^{134}Cs , median grain size, and combustion loss were then measured for each sample (see Ambe *et al.*, 2014 for details). From this sediment archive, we chose 21 surface samples along four transects

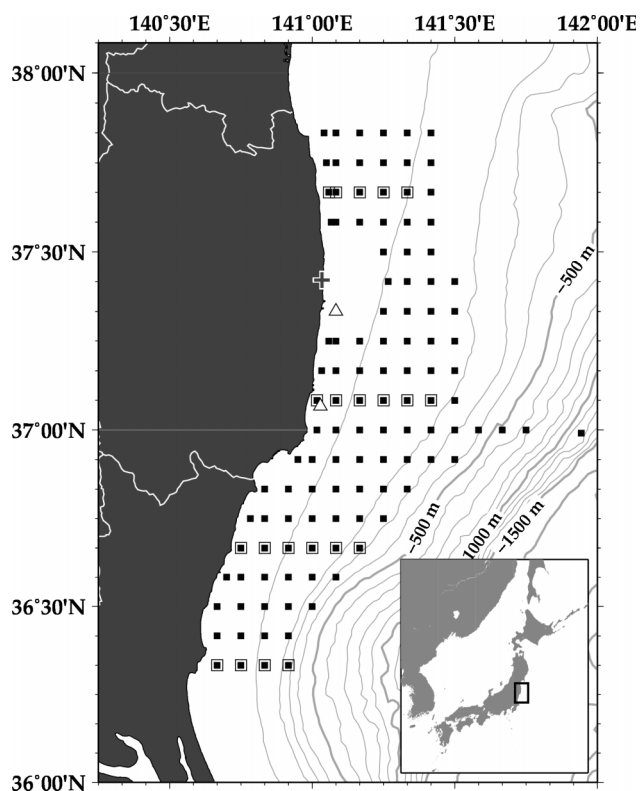


Fig. 1. Map showing the locations of the samples. Open squares represent the samples used for the bulk extraction experiment (Table 1), and open triangles represent those used for the size-fractionated extraction experiment (Table 2). Sampling stations of Ambe et al. (2014) are overlain with solid squares.

(Fig. 1) that cover a wide range of variation in latitude, bottom depth, grain size, and radiocesium concentration (Table 1). From each sample, a 30–60 g aliquot was sieved using a 2 mm mesh and then tested using the sequential extraction described in the next section. For most samples, all sediments passed through the 2 mm mesh sieve, but large, biologically formed fragments, such as shells and leaf wrecks, were removed by the sieve in some samples.

In addition to the aforementioned grid samples, two large-volume samples of surface sediment were collected at two stations on the Fukushima continental margin (Fig. 1, Table 2) to perform a size-fractionated sequential extraction experiment. At each station, 1600–1800 g of surface sediment (0–1 cm) was collected using multiple casts of a G.S.-type gravity corer (RIGO LTD., cat. no. 5174). The collected samples were dried at 80°C for several days and sieved through a 2 mm mesh to remove large components. The samples were then separated into five size fractions, 250–2000 μm , 150–250 μm , 106–150 μm , 75–106 μm , and <75 μm , by using mesh sieves. The relative amounts of each fraction, along with the bulk concentra-

Table 2. Specifications and measurement results of size-fractionated samples

Station No.	Sampling date	Latitude	Longitude	Bottom depth [m]	Size fraction	Relative amount of size fraction [%]	OF [%]	$C_{S_{\text{bulk}}}$ [Bq/kg-dry]	$C_{S_{\text{org}}}$ [Bq/kg-org-dry]	CR'
S116	2012.12.13	37°20'	141°05'	11	before fractionation	0.98	4.7	310 ± 11	2730	9
					0–75 μm	2.76	1.8	1910 ± 33	9210	5
					75–106 μm	14.32	1.5	228 ± 10	3320	15
					106–150 μm	68.37	7.9	198 ± 8.6	2830	14
					150–250 μm	13.55	1.7	260 ± 11	950	4
IWK-B	2012.12.12	37°04'	141°1.7'	30	before fractionation	3.70	1.2	209 ± 9.4	2970	14
					0–75 μm	16.78	1.7	330 ± 11	5400	16
					75–106 μm	54.07	2.0	194 ± 8.9	2680	14
					106–150 μm	23.75	1.7	169 ± 8.3	1870	11
					150–250 μm	1.70	1.8	260 ± 11	2580	10
								2650	10	

Note: For definition of $C_{S_{\text{bulk}}}$, $C_{S_{\text{org}}}$, OF and CR' see the text.

tions of ^{134}Cs and ^{137}Cs , are listed in Table 2. For fractionated samples with a total volume less than 20 g, all samples after subsampling of combustion loss were used for sequential extraction. For others, a 20 g aliquot was subsampled and used for sequential extraction.

Sequential extraction

The method by Tessier *et al.* (1979) was applied for sequential extraction. This scheme consists of four sequential extractions, which roughly represents a “surface-exchangeable” fraction, carbonate fraction, amorphous Fe-oxide fraction, and organic fraction. However, we did not use the measurement results of the fractions other than the organic fraction in the following discussions because the amounts of the other fractions are so small that the estimated radiocesium concentrations have >100% estimation error in most samples. In each extraction step, extractant was removed by centrifugation. The sediment samples were then dried and weighed carefully, and the amounts of ^{134}Cs and ^{137}Cs were measured for 10,000–40,000 s using a Ge detector (ORTEC GMX60-83) in the General Environmental Technos Co., Ltd. (KANSO) laboratory. The concentration of radiocesium in the extracted fractions was then estimated by dividing the amount of change in radiocesium content through each extraction step by that of sediment weight.

RESULTS

The estimated radiocesium concentration in the organic fraction ($C_{\text{S}_{\text{org}}}$) of each grid sample along with that of the bulk sediment ($C_{\text{S}_{\text{bulk}}}$) is listed in Table 1. The relative amount of the organic fraction (OF) in each sediment sample was also calculated from the decrease in sediment weight that occurred through the oxidant extraction step, and the results are listed in Table 1.

OF was within the range of 0.1–1.6% (w/w), which resembles the range generally observed in the continental margins of Japan (e.g., Mishima *et al.*, 1999; Kao *et al.*, 2001; Usui *et al.*, 2006). This result suggests that in our experiment, we appropriately extracted the organic contents of the samples. $C_{\text{S}_{\text{bulk}}}$ ranged from 31 to 910 Bq/kg-dry, and $C_{\text{S}_{\text{org}}}$ ranged from 345 to 3390 Bq/kg-org-dry. Concentration ratio (CR) and inventory ratio (IR) of the radiocesium in the organic fraction compared with that in the bulk sediment were then calculated by Eqs. (1) and (2), respectively:

$$\text{CR} = C_{\text{S}_{\text{org}}}/C_{\text{S}_{\text{bulk}}} \quad (1)$$

$$\text{IR} = (C_{\text{S}_{\text{org}}} \cdot \text{OF})/C_{\text{S}_{\text{bulk}}} \quad (2)$$

The CR showed a large variation from 3 to 50, suggesting that the radiocesium sorption preference to the or-

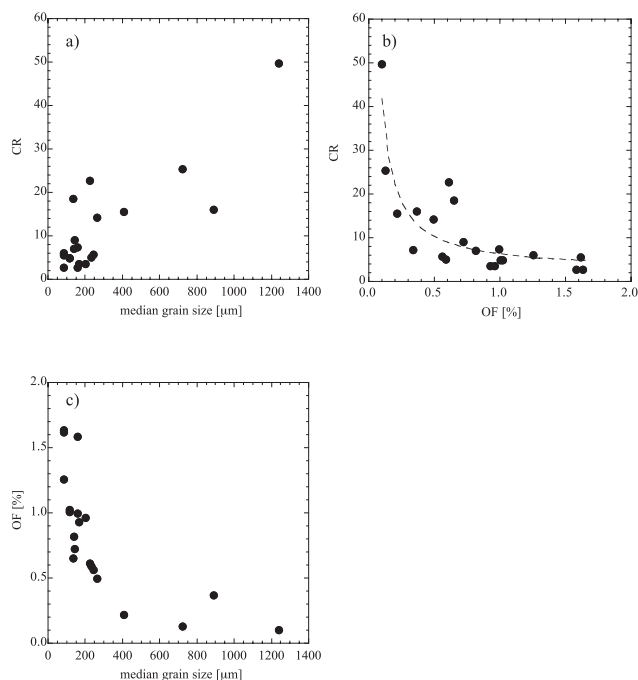


Fig. 2. Plot of a) CR vs. median grain size, b) CR vs. OF and c) OF vs. median grain size for 21 bulk samples. The dashed line in b) represents a regression curve with the following equation: $\text{CR} = (3.94 \pm 0.5)/\text{OF} + (2.4 \pm 1.7)$ [$n = 21$, $r^2 = 0.76$].

ganic fraction varies with sediment characteristics and/or environmental conditions. However, we can at least state from our measurement results that the radiocesium concentration in the organic fraction of marine sediment is always several times larger than that of the bulk sediment. Hence, the findings of Otosaka and Kobayashi (2012) are common along the continental margin near Fukushima. The IR, in contrast, showed values slightly lower than those obtained by Otosaka and Kobayashi (2012) in the northern Ibaraki Prefecture (*ca.* 20%).

The relationships among the CR, median grain size, and OF were investigated and the results are shown in Fig. 2. The CR is roughly in proportion to the median grain size (Fig. 2a) and in inverse proportion to the OF (Fig. 2b). The roughly inverse proportionality of the OF to the median grain size (Fig. 2c) suggests that either grain size or OF is the main controlling factor of the CR, but another parameter also has a certain relation with CR because of the inverse-proportional relation between these two parameters. However, we cannot determine which parameter is more important from these data alone. Therefore, we examined the radiocesium concentration in the organic fraction of size-fractionated marine sediments, in which the grain-size effect is eliminated from the CR variation.

$C_{\text{S}_{\text{org}}}$ and $C_{\text{S}_{\text{bulk}}}$ for each size fraction of the two ma-

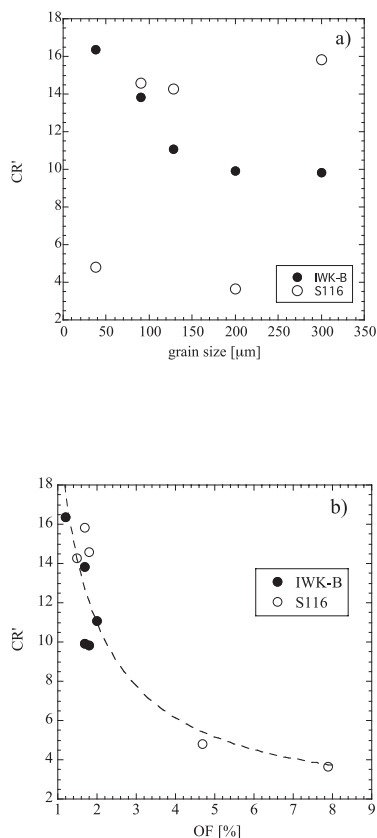


Fig. 3. Plots of a) CR' vs. grain size and b) CR' vs. OF for the size-fractionated samples. Open and solid circles represent data from Stations S116 and IWK-B, respectively. The dashed line in b) represents a regression curve of both S116 and IWK-B data. The regression equation is as follows: $CR' = (19.4 \pm 3.2)/OF + (1.3 \pm 1.8)$ [$n = 10$, $r^2 = 0.82$].

rine sediments (IWK-B and S116), along with OF, are listed in Table 2. For this experiment, we calculated the concentration ratio of Cs_{org} compared with Cs_{bulk} within the same size fraction (CR').

The grain size distributions of organic and mineral substances within the same size fraction were considered to be similar. CR' values should therefore show little variation among the size fractions assuming that the main contributor to the CR variation (Table 1) is median grain size. However, the observed CR' showed a large variation against grains size, ranging from 4 to 16 (Fig. 3a). Moreover, the observed CR' was inversely proportional to OF (Fig. 3b). These results indicate that the main contributor to CR and CR' may be OF rather than grain size.

DISCUSSION

Causes of high CR for radiocesium in marine sediment

Observed CR values for all sediment samples in this

study ranged from 3 to 50, clearly showing a general preference of radiocesium for the organic fraction of marine sediments. The CR' value of the pre-fractionation samples (same as CR for bulk samples) of S116 and IWK-B was also far higher than 1, *ca.* 9 and 14, respectively, and such large CR' values remained even after the size fraction (Table 2). This result indicates that the high CR value observed in Table 1 cannot be attributed to the different size distributions between the organic and inorganic fractions, such as a high-OF tendency in a fine-scale fraction.

Sediments and soils on land have a highly selective, non-exchangeable cesium adsorption capacity of up to 1×10^{-3} mol/kg-dry (*ca.* 4×10^{11} Bq/kg-dry for Cs-137), owing to the existence of frayed edge sites in illite crystals (e.g., Nakao *et al.*, 2012). However, in the marine environment, seawater contains 1×10^{-2} mol/L of potassium and 2×10^{-9} mol/L of stable cesium (e.g., Millero, 2013). Although cesium ions have a preference 1000 times higher than potassium ions on frayed edge sites (Kruyts and Delvaux, 2002), seven-order larger concentration of potassium than cesium in sea water make occupation ratio of cesium on frayed edge sites a five-thousandth of that of potassium. Expected non-exchangeable cesium adsorption capacity for marine sediments is thus in the order of 1×10^{-6} to 1×10^{-7} mol/kg-dry. In practice, Tagami and Uchida (2013) concluded that the stable cesium concentration in coastal sediments around Japan is in the range of 0.9×10^{-5} to 5.5×10^{-5} mol/kg-dry, by compiling historical measurement data. The value they obtained is about one order of magnitude higher than our theoretical estimation because a significant amount of their samples were collected from stations near the river mouth, where sediments may contain mineral particles that originated from land soils. Nevertheless, the results of Tagami and Uchida (2013) imply that stable isotopes occupy non-exchangeable adsorption sites effective for cesium in marine sediments. Newly supplied radiocesium from the accident can therefore, only be bound to non-selective, exchangeable sorption sites, for which the distribution coefficient of radiocesium is estimated as 300–4000 L/kg-dry (IAEA, 2004). Organic substances in marine sediment also have non-selective sorption sites for cesium (e.g., Rigol *et al.*, 2002; Kim *et al.*, 2006), but little is known about the distribution coefficient of cesium between marine organic material and seawater. Bunzl and Schimmack (1991) measured the distribution coefficient of cesium in organic-rich soils on land (Of and Oh horizons of Acrisols), and obtained a result of 100–2000 L/kg dry. Nakamaru *et al.* (2007), on the other hand, measured the distribution coefficient of cesium in various soils in Japan (Andsols, Cambisols, and Fluvisols) and obtained values from 360 to 4930 L/kg dry. Nakamaru *et al.* (2007) also found that the percentage of organic-bound Cs in soils

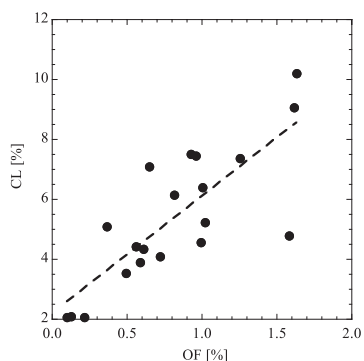


Fig. 4. Plot of CL against OF for 21 bulk samples. The dashed line shows linear regression.

was on the same order as soil organic carbon. These results indicate that the distribution coefficient of cesium for organic substances in soils is in the order of 10^2 – 10^3 L/kg-dry. The texture of organic substances differ significantly between marine sediments and land soil. In both media, humic substances constitute a major part of the organic fraction, but marine humic substances have higher carbohydrate and protein contents and richer saturated CC-bonds than terrestrial ones (e.g., Fookien and Liebezeit, 2000; Giovanela *et al.*, 2004). Despite such differences, we may be able to consider from the above information that mineral and organic substances in the marine sediments offshore Fukushima have the same order of preference for FDNPP-derived radiocesium. The apparent preference of radiocesium for organic material is further increased when organic substances cover the surface of mineral particles (e.g., Keil *et al.*, 1994; Mayer, 1994, 1999). Mayer (1999), for example, found that even 0.5% (w/w) of organic carbon can cover more than 10% of the total sediment surface area. In this case, with the assumption that organic and inorganic surfaces have the same preference for cesium, the observed CR of radiocesium becomes more than 20.

The results of our study showed an inversely proportional relationship between the CR (or CR') and the OF (Figs. 2b and 3b), indicating that the relative amount of organic-bound radiocesium compared with the total amount (IR) has a roughly constant value for a wide OF range. The factor of inverse proportionality between the CR and the OF obtained from the regression curve of Fig. 2b was 3.9 ± 0.5 , which roughly corresponds to the average value of the IR shown in Table 1. Such a situation can occur, if we assume the following: 1) Organic material has the same affinity with Cs as mineral material, but CR becomes higher because of the partial coverage of sediment surface area by organic matter. 2) The extent of organic coverage to sediment surface area is roughly con-

stant (ca. 3.9% of total surface) for a wide range of OF (i.e., the OF does not change the extent of coverage but thickness of organic film). The observational results of Mayer (1999) indicated that the extent of coverage of sediment mineral surface area by organic substances has a rather constant value (ca. 10–20% of total sediment surface area) in the wide range of organic loadings from 0.4 to 10% (w/w). Their results support that it is possible for our assumption to occur.

2-D mapping of organic-bound radiocesium in offshore-Fukushima surface sediment

To assess the potential ability of radiocesium transformation from sediment organic substances over the entire continental margin area offshore Fukushima, we applied a two-parameter regression fitting of the CR against median grain size and combustion loss. We chose combustion loss (CL) instead of the OF as an explanatory variable because the latter parameter was not measured in all of the samples in Ambe *et al.* (2014). Figure 4 shows the regression of the CL against the OF measured for 21 bulk samples used in this study. Linear regression showed that the observed CL values were larger than the OF by roughly four times, although the linearity of CL against OF was statistically significant ($r^2 = 0.63$). We applied the Japanese Industrial Standard method No. A1226, in which sediments are heated to 750°C for 1 hour, to obtain the CL. It is known that a part of calcite and dolomite within the sediment may emit their carbonate ion as CO_2 at temperatures greater than 700°C (e.g., Dean, 1974). The larger values of CL compared to OF values suggest that such reactions might have occurred during the combustion process in our experiment. Nevertheless, the observed linearity between CL and OF confirmed that our CL values can represent the relative magnitude of the OF of the samples.

By using the data from the 21 stations shown in Fig. 1, we obtained the following equation:

$$\text{CR} = 0.0255\mu + 20.08/\text{CL} - 0.69 \quad (r^2 = 0.736) \quad (3)$$

where μ represents the median grain size in μm . When only CL is used for regression, the equation becomes:

$$\text{CR} = 57.60/\text{CL} - 2.40 \quad (r^2 = 0.44) \quad (4)$$

The regression coefficient of Eq. (4) is significantly smaller than that of Fig. 2b ($r^2 = 0.76$) because of the medium regression coefficient of CL against OF (Fig. 4, $r^2 = 0.63$). As we discussed in the previous section, the primary controller of the CR is considered as the OF (and hence CL). However, we applied the two-parameter equation (Eq. (3)) instead of Eq. (4), to obtain as accurate a value as possible. Although both μ and CL are not per-

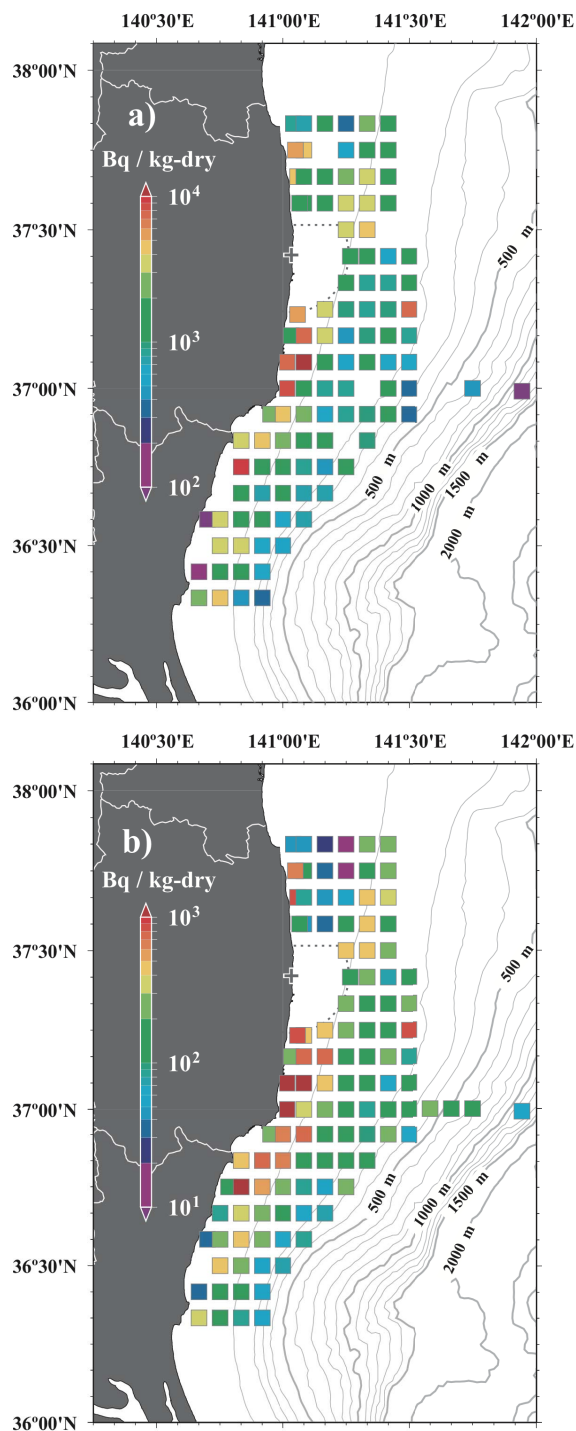


Fig. 5. Distribution of a) calculated Cs_{org} and b) observed Cs_{bulk} at the stations of Ambe *et al.* (2014). Note that the range of the color bar is 10 times higher in Cs_{org} than in Cs_{bulk} . The plus sign in each illustration represents the location of the Fukushima Dai-ichi Nuclear Power Plant.

fect representations of the OF and the values contain some errors, we can obtain a more reliable estimation of the CR by combining these two parameters.

We applied Eq. (3) to the 113 surface stations observed by Ambe *et al.* (2014), and multiplied the calculated CR values by Cs_{bulk} at each station to obtain Cs_{org} . The results are shown in Fig. 5a, along with the map of Cs_{bulk} obtained by Ambe *et al.* (2014) (Fig. 5b). In the area south of FDNPP, the distribution pattern of Cs_{bulk} roughly resembles that of Cs_{org} , although relatively high Cs_{org} values compared with Cs_{bulk} can be observed at stations located near the shore. The highest Cs_{org} value obtained in this area was 10,300 Bq/kg-org-dry, a value that is approximately 10 times larger than the highest Cs_{bulk} value (1240 Bq/kg-dry) observed by Ambe (2014). The typical range of Cs_{bulk} south of FDNPP was 2000–7000 Bq/kg-org-dry for the region shallower than 100 m, and 500–1500 Bq/kg-org-dry for the region with bottom depth ranging from 100 to 200 m. At stations north of FDNPP, Cs_{org} had medium concentrations (ca. 300–3600 Bq/kg-org-dry), even at stations where extremely low Cs_{bulk} values were observed (ca. 10–100 Bq/kg-dry; Ambe *et al.*, 2014). This is because the sediments in mid-depth area (ca. 30–100 m) north of FDNPP consist mainly of large particles with low organic matter content, which leads to high CR values from the calculation by Eq. (3). Contrastingly, both Cs_{bulk} and Cs_{org} maintained relatively low values at shallow stations south of 36°40' N and at continental slopes.

Why do benthos have low radiocesium concentrations despite high Cs_{org} ?

Our 2-D extrapolation suggests that the typical range of Cs_{org} for the area south of FDNPP and shallower than 100 m is 2000–7000 Bq/kg-org-dry. Since the typical water content of marine sediments in this area is approximately 30% (Ambe *et al.*, 2014), these values correspond to the range of 1,400–5,000 Bq/kg-org-wet. On the other hand, the radiocesium content of polychaetes in this area in the autumn of 2012 was less than 40 Bq/kg-wet (Sohtome *et al.*, 2014). The transfer coefficient of radiocesium for polychaetes in the process of feeding on benthic organic matter, if calculating on a carbon-to-carbon basis, thus becomes less than 0.03–0.008. Some portion of radiocesium sorbed onto mineral substances may also transfer to polychaetes as the distribution coefficient of radiocesium between seawater and the mineral fraction of the sediment becomes low in low-pH environments (e.g., Elprince, 1978), such as in the digestive tract of these organisms. Even with this possibility taken into account, it is definite that the transfer coefficient of radiocesium for polychaetes is less than 1. Such an extremely low transfer coefficient should have made a great contribution toward preventing radiocesium contamina-

tion of demersal fish via seafloor sediment in the continental margin area offshore Fukushima, but the physiological mechanism remains a subject for future study. Oceanic polychaetes and many other oceanic benthos are osmoconformers, organisms in which cations, such as potassium and cesium, can be easily exchanged between cells and the surrounding water (e.g., Schmidt-Nielsen, 1997). Radiocesium, therefore, might have been promptly emitted from their tissues after the rapid decrease in seawater radiocesium concentration.

CONCLUSION

Based on the multi-station sequential extraction experiment, we found that offshore Fukushima, organic substances have an apparently higher preference than mineral substances for radiocesium in marine sediments. The observed relationship between the ratio of Cs_{org} to Cs_{bulk} and organic content indicated that both organic and mineral substances have the same preference in sediments offshore Fukushima, primarily as the result of the existence of potassium and stable cesium in seawater, which occupied the frayed edge sites of marine sediments before the accident. The apparent preference of radiocesium for organic material increased due to the partial coverage of sediment mineral surfaces by organic substances. However, we should note that the high preference of radiocesium for organic material does not necessarily indicate higher cesium sorption onto the sediment with higher organic content. Ambe *et al.* (2014) found a positive correlation between Cs_{bulk} and organic matter content in sediments offshore Fukushima, but they pointed out that these sediments simultaneously showed a negative correlation between Cs_{bulk} and median sediment grain size. Since organic matter content and median grain size are negatively correlated in these sediments, they suggested that the primary controller of Cs_{bulk} may be median grain size, and that the observed relationship between Cs_{bulk} and organic content is a consequence. Saengkul *et al.* (2013) found a negative relationship between Cs_{bulk} and organic content in near-coastal sediments in the Gulf of Thailand, contrary to the case of our study area, which showed a positive relationship between the organic content and the percentage of silt in the sediment. These results indicate that Cs_{bulk} is controlled mainly by the grain size distribution and that organic matter content only changes the extent of the relative accumulation of radiocesium in the organic fraction compared with Cs_{bulk} .

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