Chapter 5 Radiocesium Concentrations in the Organic Fraction of Sea Sediments

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Abstract Sequential chemical extraction of radiocesium was performed on 22 surface sediment samples to assess radiocesium concentration in the organic fraction of sea sediments (Csorg). Our results showed that Csorg of sea sediments was significantly larger than that of bulk sediments (Cs_{bulk}). The concentration factor of radiocesium in organic fraction against the bulk concentration (CF) varied from 3 to 50 off the Fukushima continental margin and showed a proportional relationship with median grain size and an inversely proportional relationship with organic content (OC) of the sediment. By using these relationships, the regression equation of Csorg based on median grain size, organic content, and Cs_{bulk} was determined to construct a two-dimensional (2-D) distribution of Cs_{ore} along the continental margin off the Fukushima region. The resultant map showed that the continental margin north of Fukushima Dai-ichi Nuclear Power Plant (FNPP) had moderate Csorg values despite very low Csbulk. On the other hand, sediments sampled at the mouth of Abukuma River showed extremely low CF, which might have been caused by the existence of river-derived sediment particles.

Keywords Sediment • Radiocesium • Organic fraction

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5.1 Introduction

In the assessment of radiocesium transportation from sea sediments to a marine demersal ecosystem, information is required not only on the concentration but also on biological ingestibility of sea sediment radiocesium. Although IAEA has provided a standard concentration factor of radiocesium from sea sediments in marine organisms (e.g., 1×10^2 for fish; IAEA 2004), its actual value may vary according to sediment properties such as grain size and chemical composition. Radiocesium concentration in the organic fraction of sediments (Cs_{org}) is an important factor because the transport of radiocesium from sediment to demersal ecosystem occurs primarily through the feeding/ingestion of carbon sediments by benthos. With regard to the FNPP accident, a large amount of data is available on the spatiotemporal distribution of radiocesium concentration in sea sediments (Cs_{bulk}) off the Fukushima Prefecture (Otosaka and Kobayashi 2012; Kusakabe et al. 2013; Otosaka and Kato 2014; Ambe et al. 2014). Unfortunately, insufficient data are available on the spatiotemporal distribution of Cs_{arg} . To address this issue, we conducted sequential chemical leaching experiments for 21 sea sediments sampled in July 2012 at $5' \times 5'$ grid stations off Fukushima Prefecture (Ambe et al. 2014; see Fig. 5.1 for station map) to measure Cs_{org} of these sediments. For details of sampling stations and experimental procedures, see Ono et al. (2015).

5.2 Cs_{org} and Its Relationship with Cs_{bulk}

Estimated radiocesium concentrations in organic fraction (Cs_{org}) and bulk sediment (Cs_{bulk}) in 21 grid samples are listed in Table 5.1. Cs_{bulk} ranged from 31 to 910 Bq/kg-dry and Cs_{org} ranged from 345 to 3,390 Bq/kg-org-dry. Concentration factor (CF) and inventory ratio (IR) of radiocesium in organic fraction against bulk sediment were then calculated by the following equation:

$$CF = Cs_{org} / Cs_{bulk}$$
(5.1)

$$IR = (Cs_{org} \cdot OC) / Cs_{bulk}$$
(5.2)

where OC represents the organic content of the sediment (Table 5.1).

CF values vary from 3 to 50, clearly illustrating that radiocesium concentration in the organic fraction of sea sediments is always several times larger than that of bulk sediment in areas off Fukushima. Despite these high CF values, IR showed relatively low values, ranging from 2.4 % to 13.9 %, reflecting low organic content in open ocean sediments.

Land sediments and soils have highly selective, nonexchangeable cesium adsorption capacity, up to 1×10^{-11} mol/kg-dry, because of the frayed edge sites in illite particles (Nakao et al. 2012). In marine environments, however, such nonexchangeable adsorption sites are occupied by stable cesium (~2×10⁻⁹ mol/l in seawater) and potassium (~1×10⁻² mol/l in seawater). Newly supplied radiocesium from the accident, therefore, can only be bound to nonselective, exchangeable sorption sites, with the distribution coefficient of radiocesium estimated to be



Fig. 5.1 Map of the location of the samples used in this study. *Thick gray line* denotes Abukuma River (only lower reaches are shown). *Open squares* denote the samples used for the bulk extraction experiment (Table 5.1), and *open triangles* denote the location of the off-Abukuma station (Table 5.2). Sampling stations of Ambe et al. (2014) are overlaid as *solid squares*

300–4,000 l/kg-dry (IAEA 2004). Organic substances in the sediments also have nonselective sorption sites for cesium, but so far little is known about the distribution coefficient of cesium between marine organic matter and seawater. On land, several observations have indicated that the distribution coefficient of cesium for organic substances in soils is of the order of 10^2 – 10^3 l/kg-dry (Bunzl and Schimmack 1991; Nakamaru et al. 2007). If we assume that marine organic substances have the same distribution coefficient of cesium as land soils, we can consider that mineral and organic substances in the off-Fukushima sediments have the same order of preference as FNPP-derived radiocesium. The apparent preference of radiocesium in organic substances (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 total sediment surface area. In this case, with the assumption that organic carbon and mineral surfaces to more than 20.

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				Bottom	Median grain		$\mathbf{C}_{\mathbf{S}_{\mathrm{bulk}}}$	$C_{S_{\mathrm{org}}}$		
Station no.	Sampling date	Latitude [N]	Longitude [E]	depth (m)	size (µm)	OC (%)	(Bq/kg-dry)	[Bq/kg-org-dry]	CF	IR [%]
S1	2012.7.11	36° 20'	140° 55'	257	142	0.8	49 ± 5.5	350	٢	5.8
S2	2012.7.11	36° 20′	$140^{\circ} 50'$	120	136	0.7	78±6.6	1,440	19	12.0
S3	2012.7.11	36° 20′	140° 45'	59	889	0.4	153 ± 9.7	2,440	16	5.9
S4	2012.7.11	36° 20′	$140^{\circ} 40'$	33	201	1.0	310 ± 20	1,090	4	3.4
S20	2012.7.12	36° 40′	141° 10'	261	233	0.6	103 ± 6.3	520	S	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	1,330	٢	7.3
S23	2012.7.12	36° 40′	140° 55'	111	87	1.6	180 ± 14	096	9	8.9
S24	2012.7.12	36° 40′	140° 50'	70	116	1.0	270±21	1,300	S	4.9
S25	2012.7.12	36° 40′	140° 45'	33	no data	0.3	69 ± 5.9	490	٢	2.4
S59	2012.7.13	37° 05'	141° 25'	177	247	0.6	83±5.4	470	9	3.2
S60	2012.7.13	37° 05′	141° 20'	151	225	0.6	104 ± 6.3	2,360	23	13.9
S61	2012.7.13	37° 05'	141° 15'	140	85	1.3	101 ± 7.6	600	9	7.4
S62	2012.7.13	37° 05′	141° 10'	120	87	1.6	440±27	1,200	m	4.5
S63	2012.7.12	37° 05'	141° 05'	72	158	1.6	690±32	1,840	e	4.2
S64	2012.7.12	37° 05′	141° 01'	25	167	0.9	910±32	3,120	m	3.2
S92	2012.7.15	37° 40′	141° 03.5′	24	118	1.0	710±28	3,390	S	4.9
S93	2012.7.15	37° 40′	141° 05'	28	407	0.2	82±5.9	1,270	16	4.0
S94	2012.7.15	37° 40'	$141^{\circ} 10'$	37	723	0.1	31 ± 3.4	780	25	3.2
S95	2012.7.15	37° 40′	141° 15'	59	1,240	0.1	47±4.1	2,330	50	5.0
S96	2012.7.15	37° 40'	141° 20'	100	146	0.7	230 ± 16	2,080	6	6.5
All data are re Note: For defi	produced from O	no et al. (2015) Cs _{org} , OC, CF, and	l IR, see the text							

f grid samples
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measurement re
Specifications and
Table 5.1

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5.3 Horizontal Distribution of Cs_{org} in off-Fukushima Continental Margin

CF is roughly proportional to median grain size and inversely proportional to OC (Fig. 5.2), suggesting that either or both of these properties are the main control factors of CF, although detailed analysis by Ono et al. (2015) concluded that OC is a major control factor and median grain size is minor. Using this information, we applied dual-parameter regression, appropriate for CF, against median grain size and combustion loss as follows:

$$CF = 0.0255\mu + 20.08 / IL - 0.69 (r^{2} = 0.736, \rho < 0.01)$$
(5.3)

where μ and IL represent median grain size in μ m (micrometers) and ignition loss in percentage, respectively. We chose IL instead of OC as an explanatory variable because the latter parameter was not measured for all samples reported by Ambe et al. (2014). Although IL somewhat overestimated the actual OC, we confirmed the linearity of IL against OC before the derivation of Eq. (5.3). We applied this equation to 113 surface stations observed by Ambe et al. (2014), and the calculated CF was multiplied by Cs_{bulk} in each station (Fig. 4.3 in Chap. 4) to obtain Cs_{org}. The results are shown in Fig. 5.3. A high Cs_{org} band exists just offshore south of FNPP, within which the highest Cs_{org} value of 10,300 Bq/kg-org-dry was obtained. In this area, the typical range of Cs_{bulk} south of FNPP was 2,000–7,000 Bq/kg-org-dry for the area with a bottom depth shallower than 100 m, and 500–1,500 Bq/kg-org-dry





Fig. 5.3 Distribution of calculated Cs_{org} (organic cesium) in the off-Fukushima continental margin area. Contours are drawn from the Cs_{org} value estimated by Eq. (5.3) for each station of Ambe et al. (2014)

for the area with bottom depth ranging from 100 to 200 m. In the station north of FNPP, Cs_{org} showed medium concentrations (~300–3,600 Bq/kg-org-dry) for the area with a bottom depth shallower than 100 m, and Cs_{bulk} values were extremely low (~10–100 Bq/kg-dry; see Ambe et al. 2014 and previous chapter); this is because the sediments of the mid-depth area (~30–100 m) north of FNPP consist mainly of large particles with low organic carbon content, which, using Eq. 5.3, leads to very high CF values. This result implies that the potential effect of sea sediment radioce-sium on benthos would not be too different between the area south of FNPP with a bottom depth shallower than 100 m, despite a significant Cs_{bulk} difference between these areas. Wada et al. (2013) detected similar radiocesium level of demersal fishes between these two areas after 2012. These findings suggest that Cs_{org} can be used as an indicator of the potential effect of sediment radiocesium on the demersal ecosystem.

				Bottom	Median		Cs _{bulk}	Csorg (Bq/		
Station	Sampling	Latitude	Longitude	depth	grain	OC	(Bq/	kg-org-		IR
no.	date	[N]	[E]	(m)	size (µm)	(%)	kg-dry)	dry)	CF	(%)
ABK-A	2013.8.22	38° 2.4′	$140^\circ\ 56.4'$	13	No data	16	$5,600 \pm 75$	7,882	1.4	23

Table 5.2 Specifications and measurement results of off-Abukuma patch

5.4 Cs_{org} and CF in off-Abukuma River Sediments

As the sediments described in the former sections are sampled from the continental margin, organic materials contained in these sediments are thought to be produced in the ocean. However, sediments in some local areas such as river mouths contain lithogenic particles, which were produced within freshwater or on land and then transported to the seafloor after the FNPP accident. For such sediments, CF can be considerably low because the nonexchangeable adsorption sites of mineral particles were not occupied by stable cesium or potassium at the time of the accident. To assess the CF value for such sediments, we performed additional Cs_{org} measurements for sediments taken from the local high radiocesium patch recently discovered by the Nuclear Regulation Office (NRA 2014), located just outside of the Abukuma River mouth, with a horizontal scale of about 900×400 m width.

Differing from the foregoing grid samples, Cs_{org} in the off-Abukuma patch showed significantly low CF values (~1.4; Table 5.2), possibly because of the significantly high OC value in the sample. Hence, a high-OC sediment tends to have a low CF value (Fig. 5.2). Another reason might be that the sediments in this patch contain a significant amount of lithogenic particles derived from the Abukuma River (Yamashiki et al. 2014). Although the observed Cs_{bulk} in this patch is the highest among the oceanic stations we observed, a low CF in the sediments causes the Cs_{org} value to be at the same level as the average value of off-Fukushima sediments. The monitoring results for marine products for the off-Miyagi prefecture region did not detect any local increase in the occurrence of high-Cs fishes in off-Abukuma regions (JFA 2014), despite the existence of a high-Cs patch in sediments. A significantly low CF in the off-Abukuma sediment patch may explain these observation results. Again, our results showed that not only Cs_{bulk} but also Cs_{org} are essential for accurately assessing the potential effect of sediment radiocesium on the demersal ecosystem in each region.

5.5 Summary

Our study clarifies that radiocesium concentration in the organic fraction of sea sediments is always larger than that in the organic fraction of bulk sediments. This result indicates that the transport efficiency of radiocesium from the organic fraction of sediments to the marine benthos is extremely low, because the radiocesium concentration in marine benthos is of the order of 10^1 Bq/kg-wet (see Chap. 7). The details of the physiological mechanism that results in such low transport efficiency is an important topic for future study.

Based on Cs_{org} , we assessed that the sediments in the off-Fukushima continental margin north of the FNPP have moderate potential to transport radiocesium to benthic ecosystems, despite the low Cs_{bulk} observed in this region. However, sediments off Abukuma River have less potential to transport radiocesium than the level inferred from its Cs_{bulk} value.

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