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Current status and future control of cesium contamination in plants and algae in Fukushima

Radioactive cesium accumulation in seaweeds by the Fukushima 1 Nuclear Power Plant accident—two years' monitoring at Iwaki and its vicinity

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Abstract Accumulations of radionuclides in marine macroalgae (seaweeds) resulting from the Fukushima 1 Nuclear Power Plant (F1NPP) accident in March 2011 have been monitored for two years using high-purity germanium detectors. Algal specimens were collected seasonally by snorkeling at Nagasaki, Iwaki, Fukushima Prefecture (Pref.), Japan, ca. 50 km perimeter from the F1NPP. Additional collections were done at Soma, Hironocho, Hisanohama and Shioyazaki in Fukushima Pref. as well as at Chiba Pref. and Hyogo Pref. as controls. In May 2011, specimens of most macroalgal species showed ^{137}Cs levels greater than 3,000 Bq kg^{-1} at Shioyazaki and Nagasaki. The highest ^{137}Cs level recorded 7371.20 $\pm$ 173.95 Bq kg^{-1} in *Undaria pinnatifida* (Harvey) Suringar on 2 May 2011, whereas seawater collected at the same time at Shioyazaki and Nagasaki measured 8.41 $\pm$ 3.21 and 9.74 $\pm$ 3.43 Bq L^{-1}, respectively. The concentration factor of marine macroalgae was estimated to be ca. 8–50, depending on taxa and considering a weight ratio of wet/dry samples of ca. 10. ^{137}Cs level declined remarkably during the following 5–6 months. In contrast, the ^{137}Cs level remained rather stable during the following 12–16 months, and maintained the range of 10–110 Bq kg^{-1}. Contamination was still detectable in many samples in March 2013, 24 months after the most significant pollution.

Keywords ^{134}Cs · ^{137}Cs · F1NPP · ^{40}K · Radionuclide accumulation · Seaweeds

Fukushima Daiichi Nuclear Power Station is cited as Fukushima 1 Nuclear Power Plant in the present manuscript.

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Introduction

Radioactive contamination of the coastal seawater following the Fukushima 1 Nuclear Power Plant (F1NPP) accident was the most significant artificial radioactive liquid release into the sea ever known, on a short time and space scale basis (Buesseler et al. 2011; Linsley et al. 2005). The amount of radionuclides discharged from the F1NPP immediate after the accident was estimated to be 5–10 PBq to the atmosphere and 3–6 PBq to the sea, the latter being caused by direct leakages of the contaminated cooling water (Estournel et al. 2012; Kawamura et al. 2011; Miyazawa et al. 2012; Tsumune et al. 2012).

The seawater contamination has become severe by (1) a deposition from an atmospheric contamination plume, (2) direct, artificial releases of highly contaminated waters into the sea, and (3) the transport of radiopollutants into the sea by surface water leaching through contaminated soil or by river systems. The first case was most serious during mid-March 2011, and the second from late March to early April 2011. The contaminated seawater may be diluted or has been transported offshore by currents. Along the coast, however, a significant portion of the radioactive Cs and other radioactive substances are believed to have been absorbed by coastal organisms or bound to suspended particles, causing a deposition of radiopolluted sediments on the sea bottom. The concentration of radionuclides reached the maximum in mid-April 2011, but it thereafter declined exponentially. However, in addition to the inflow through the river systems, artificial release of radionuclides from F1NPP is reported to have continued at least until 2012 (Kanda 2013).

Among the radionuclides discharged by the accident, the effects of ^{134}Cs and ^{137}Cs are most serious because of their high levels and long decay periods. These radionuclides are considered to most substantially contribute to the contamination of marine organisms due to the contaminated seawater. Marine macroalgae (so-called seaweeds) grow by absorbing nutrient salts and other minerals directly from seawater at the surface of their thalli. Cs^+ is soluble in seawater, and algal cells are thought to absorb Cs^+ through certain K^+ transporters (Kanter et al. 2010; Zhu and Smolders 2000), resulting in Cs^+ accumulation within the cells.

The concentration factors (CFs) of marine macroalgae for ^{137}Cs were reported to vary considerably from species to species (Coughtrey and Thorne 1983; Pentreath 1976; Tateda and Koyanagi 1994), but IAEA (2004) recommended the value of 50. Marine macroalgae may also capture the ions in phycocolloids such as alginate, fucan, agar, and carrageenan, although the affinity of Cs^+ for these phycocolloids is not well-established (Morris et al. 1980). Marine macroalgae are primary producers in coastal ecosystems, and diverse benthic animals as well as fishes feed on them. Therefore, the radioactive contamination of marine macroalgae will be transferred to consumer animals through the food chain, and may become concentrated by biological accumulation.

Prior to the accident, the concentrations of ^{137}Cs in the surface water of the Pacific Ocean were in the range of 1–4 Bq m^{-3} (Ikeuchi 2003; Nakanishi et al. 2010; Povinec et al. 2004), which mainly resulted from global fallout due to atmospheric nuclear weapon tests. Tateda and Koyanagi (1994) have reported the background concentrations of ^{137}Cs in representative green, red and brown marine macroalgae, e.g. *Ulva pertusa* Kjellman, *Neodilsea yendoana* Tokida, *Saccharina religiosa* (Miyabe) C.E. Lane, C. Mayes, Druehl & G.W. Saunders (=*Laminaria religiosa* Miyabe), *Sargassum horneri* (Turner) C. Agardh, *S. thunbergii* (Mertens ex Roth) Kuntze, in the range of ca. 0.03–037 Bq kg wet weight^{-1}. Later, Morita et al. (2010) have compared the level of ^{90}Sr and ^{137}Cs in *Undaria pinnatifida* collected from three representative areas (Nagasaki Pref., Kanagawa Pref. and Niigata Pref.) in Japan and in *Saccharina longissima* (Miyabe) C.E. Lane, C. Mayes, Druehl & G.W. Saunders (=*Laminaria longissima* Miyabe) from Hokkaido during a 1998–2008 period and reported ^{137}Cs levels of 0.03–0.08 and 0.05–0.09 Bq kg wet weight^{-1} for *U. pinnatifida* and *S. longissima*, respectively. Morita et al. (2010) also compared concentrations of ^{90}Sr and ^{137}Cs in some additional kelp and *Sargassum* species and reported ^{137}Cs concentrations of 0.02–0.34 kg wet weight^{-1}.

Several agencies have conducted investigations to measure the radioactive quantities of major fishery organisms after the F1NPP accident. Some algal taxa have been included in the target species, e.g., *Pyropia* (*Porphyra*), *Saccharina* (*Laminaria*), *Eisenia*, *Gloiopeltis*, *Monostroma*, but the number of species was rather limited, and rather biased in respect to phylogenetic diversity, habitat, and life history (e.g., intertidal *vs*. subtidal, annual *vs*. perennial). In addition, measurement protocols were not standardized among the investigations, and the detection levels were sometimes rather high (e.g., $>$ 20 Bq kg^{-1} for fresh specimens: http://www.jfa.maff.go.jp/e/inspection/index.html) depending on the available facilities. Therefore, it was difficult to compare the data to clarify the general fate of radioactive contamination in marine macroalgae, and to discuss the differences in the CFs for radionuclides among the taxa.

In the present study, we aimed to monitor the level of radionuclides including ^{134}Cs, ^{137}Cs and ^{40}K based on a standardized protocol using high-purity germanium detectors and to undertake multi-seasonal sampling as much as possible.

Materials and methods

Specimens were seasonally collected by snorkeling at Nagasaki, Fukushima Pref., ca. 50 km distant from the F1NPP (Fig. 1). Additional specimens were collected at the following localities: Hironocho, Hisanohama, Shioyazaki and Soma (Fukushima Pref.); Iwanuma (Miyagi Pref.); Kamogawa and Katsuura (Chiba Pref.); and Iwaya (Hyogo Pref.) (Fig. 1). The list of the specimens is shown in Table 1 (measurements made at Kobe University) and Table 2 (measurements made at Iwaki Meisei University).

For the measurements at Kobe University, sorted specimens were identified based on their morphology, preliminarily air-dried overnight using a fan at room temperature, and then dried in an oven at 90–100 °C for 8 h. The weight ratios of wet/dry samples were roughly 10, and this value was used in the comparisons of literature data based on wet

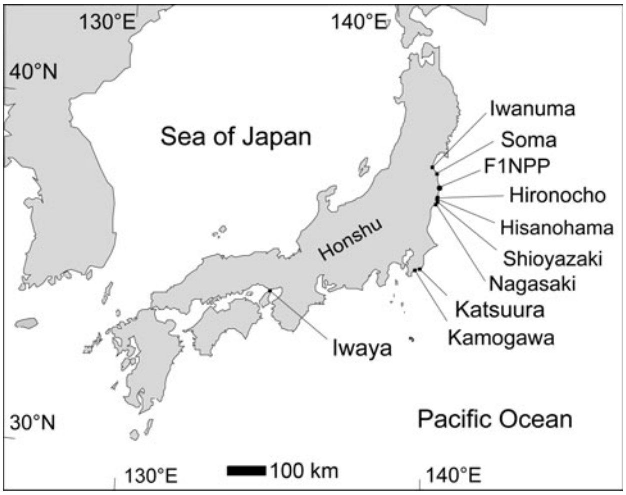

Fig. 1 Locations of the collection sites of the specimens investigated in the present study

samples, and concentration factors. Dried specimens were preliminarily broken into fragments manually, and powdered using a blender (Waring J-SPEC 7011BUJ, Conair Corp., Stamford, CT, USA). Powdered specimens were packed in U-8 sample cups (56 mm ø and 68 mm high) and used for measurement. The protocol of radioactivity measurements is described in Mimura et al. (2014). In Tables 1 and 2, "Error" is the overall error estimated by the analyzing software from systematic errors in the system together with the standard deviation of the counting. Seawaters collected at the coasts of Nagasaki and Shioyazaki in May 2011 were filled in U-8 sample cups and used for measurements without evaporation.

For the measurements at Iwaki Meisei University, sorted specimens were identified based on their morphology, washed with fresh water and pre-dried at room temperature. Dried specimens were desiccated in an oven at 60 °C for 48 h, powdered using a blender, and packed in U-8 sample cups. ^{134}Cs and ^{137}Cs were measured using a GEM40P4-76 germanium detector (Seiko EG & G, Tokyo, Japan) following the manufacturer's instructions.

The first collections (May 2011) of the macroalgal specimens were kept frozen at −20 °C until drying. However, this protocol resulted in some loss of radioactivity in meltwater (dripping). Accordingly, in order to assess the loss of radioactivity during thawing, some specimens (collected in July 2011) were either frozen before drying or directly dried, and the radioactivity was compared between these drying protocols. In addition, the radioactivity of the meltwater obtained from the thawed specimens was also measured.

Statistical problems

In some radioactivity measurements, such as for macroalgal samples, we could not obtain a sufficient amount of samples that allow us to ensure statistical reliance of the measurement. There was also a limitation in the total running time on a germanium detector. In other cases, the total volume of collected samples was not large enough to fill the U-8 sample cup, resulting in rather low detection signals. For all these experiments, only data are presented with notification that the precision of data was not statistically tested.

Results

The ^{134}Cs:^{137}Cs ratios of examined specimens were plotted according to time series (Fig. 2). The ratio reduced to ca. 0.6 after two years, which agreed well with the expected value by natural decay (0.79 after one year, and 0.64 after two years).

Results of the measurements at Kobe University (^{134}Cs, ^{137}Cs and ^{40}K) and Iwaki Meisei University (^{134}Cs and ^{137}Cs) are listed in Table 1 and Table 2, respectively. On 2 May and 6 May 2011 (Shioyazaki and Nagasaki, about 50 km from F1NPP), specimens of most macroalgal species, i.e., *Ulva pertusa*, *U. linza* Linnaeus, *Scytosiphon lomentaria* (Lyngbye) Link, *Eisenia bicyclis* (Kjellman) Setchell, *Undaria pinnatifida*, *Sargassum muticum* (Yendo) Fensholt, and *S. thunbergii*, showed ^{137}Cs levels greater than 3,000 Bq kg^{-1} (Figs. 3, 4). The highest ^{137}Cs level was 7,371.20 ± 173.95 Bq kg^{-1} in *Undaria pinnatifida* (Fig. 3; sample code k010 in Table 1). The ^{134}Cs:^{137}Cs ratio was ca. 0.97. Exceptionally, *Gloiopeltis furcata* (Postels & Ruprecht) J. Agardh showed a relatively lower value of 767.37 ± 32.90 Bq kg^{-1} (Fig. 4; sample code k014 in Table 1). Seawater collected on May 2 at Shioyazaki (Fig. 3) and Nagasaki (Fig. 4) measured 8.41 ± 3.21 and 9.74 ± 3.43 Bq L^{-1}, respectively. In control samples from Awaji Island in May 2011, ^{137}Cs levels were below the detectable level of 0.001–0.002 in *Undaria pinnatifida*, *Sargassum muticum*, and *Ulva pertusa* (Table 1).

^{137}Cs levels remarkably declined at Nagasaki within the following 5–6 months. In July 2011, ^{137}Cs ranged from ca. 300–600 Bq kg^{-1} (Fig. 5), declined to 40–200 Bq kg^{-1} in October 2011 (Fig. 6) and became lower than 100 Bq kg^{-1} in most samples in July 2012 (Fig. 7) and December 2012 (Fig. 8). However, ^{137}Cs was still detectable in the range of 10–100 Bq kg^{-1} in many samples in March 2013, 24 months after the date of highest records (Fig. 9).

The progression of ^{137}Cs levels differed in different taxa. In the green alga *Ulva pertusa*, which has very short lifetime (ephemeral species), ^{137}Cs levels clearly declined during the summer of 2011, but remained in the range of 10–110 Bq kg^{-1} from winter 2011 to spring 2013 (Fig. 10). ^{40}K was somewhat higher in May 2011 samples,

Table 1 List of samples examined at Kobe University, and the results of ^{134}Cs, ^{137}Cs and ^{40}K measurements for dried samples when not specially noted

Sample code	Collection date	Taxonomy			Locality	Note (sample preparation and nature of samples)	^{134}Cs			^{137}Cs			^{40}K		
		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
k001	9 July 2011	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Fertile thalli, directly dried	320.57	16.07	0.012	346.14	22.70	0.009	1,132.70	310.81	0.282
k002	2 May 2011	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Shioyazaki, Iwaki, Fukushima	Frozen	5,406.70	68.22	0.029	5557.90	93.10	0.020	2,494.30	375.95	0.309
k003	2 May 2011	Ulvophyceae	Ulvales	*Ulva pertusa*	Nagasaki, Iwaki, Fukushima	Frozen	3,009.50	97.10	0.054	2897.80	122.56	0.042	2,780.80	1013.30	0.942
k004	9 July 2011	Ulvophyceae	Ulvales	*Ulva pertusa*	Nagasaki, Iwaki, Fukushima	Directly dried	487.54	58.31	0.051	567.83	74.20	0.046	n.d.	n.d.	1.381
k005	2 May 2011	Phaeophyceae	Fucales	*Sargassum thunbergii*	Shioyazaki, Iwaki, Fukushima	Frozen	4,151.90	60.11	0.026	4239.90	81.19	0.020	2,507.50	384.80	0.315
k006	2 May 2011	Phaeophyceae	Ectocarpales *s.l.*	*Scytosiphon lomentaria*	Shioyazaki, Iwaki, Fukushima	Frozen	5,417.10	131.01	0.073	5672.50	172.27	0.041	n.d.	n.d.	1.013
k007	6 May 2011	Phaeophyceae	Ectocarpales *s.l.*	*Scytosiphon lomentaria*	Nagasaki, Iwaki, Fukushima	Frozen	4,544.90	69.54	0.031	4616.00	93.95	0.021	n.d.	n.d.	0.376
k008	2 May 2011	Phaeophyceae	Fucales	*Sargassum muticum*	Shioyazaki, Iwaki, Fukushima	Frozen	3,991.30	98.26	0.049	4282.40	132.04	0.038	1,981.80	808.57	0.760
k009	2 May 2011	Ulvophyceae	Ulvales	*Ulva pertusa*	Shioyazaki, Iwaki, Fukushima	Frozen	4,231.90	138.52	0.070	4340.20	182.55	0.062	n.d.	n.d.	1.409
k010	2 May 2011	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Shioyazaki, Iwaki, Fukushima	Frozen	7,433.50	133.33	0.060	7371.20	173.95	0.047	1,152.20	799.52	0.800
k011	9 July 2011	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Juvenile thalli, directly dried	529.92	48.56	0.160	569.08	60.43	0.142	n.d.	n.d.	3.396
k012	9 July 2011	Phaeophyceae	Fucales	*Sargassum horneri*	Nagasaki, Iwaki, Fukushima	Basal portion, directly dried	457.65	22.24	0.053	501.06	29.90	0.044	2,867.30	411.99	1.061
k013	2 May 2011	Ulvophyceae	Ulvales	*Ulva linza*	Shioyazaki, Iwaki, Fukushima	Frozen	5,516.50	128.99	0.073	5433.80	165.31	0.038	n.d.	n.d.	0.944
k014	6 May 2011	Rhodophyceae	Gigartinales	*Gloiopeltis furcata*	Nagasaki, Iwaki, Fukushima	Frozen	691.86	23.50	0.015	767.37	32.90	0.012	371.85	260.95	0.257
k015	6 May 2011	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Frozen	4,690.00	71.89	0.031	4810.30	96.65	0.023	2,957.80	462.11	0.383
k016	9 July 2011	Rhodophyceae	Gigartinales	*Grateloupia lanceolata*	Nagasaki, Iwaki, Fukushima	Directly dried	318.48	28.83	0.106	352.40	36.17	0.090	n.d.	n.d.	2.073
k017	9 July 2011	Ulvophyceae	Ulvales	*Ulva pertusa*	Nagasaki, Iwaki, Fukushima	Frozen	606.50	66.73	0.200	576.93	85.96	0.199	n.d.	n.d.	5.182
k018	9 July 2011	Phaeophyceae	Fucales	*Sargassum horneri*	Nagasaki, Iwaki, Fukushima	Tip portion, directly dried	340.77	21.65	0.018	321.15	26.71	0.016	4,783.70	525.79	1.280
k019	9 July 2011	Phaeophyceae	Laminariales	*Saccharina japonica*	Nagasaki, Iwaki, Fukushima	Directly dried	426.87	37.52	0.136	425.32	45.34	0.097	n.d.	n.d.	2.638
k020	9 July 2011	Phaeophyceae	Fucales	*Sargassum yamadae*	Nagasaki, Iwaki, Fukushima	Directly dried	499.62	25.71	0.064	498.74	33.71	0.061	1,871.40	446.75	1.295
k023	9 July 2011	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Fertile thalli, frozen	278.38	16.89	0.045	292.40	21.88	0.038	1,987.90	355.52	0.976

Table 1 continued

Sample code	Collection date	Taxonomy			Locality	Note (sample preparation and nature of samples)	^{134}Cs			^{137}Cs			^{40}K		
		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
k024	17 May 2011	Phaeophyceae	Fucales	*Sargassum muticum*	Iwaya, Awaji Isl., Hyogo	Directly dried	n.d.	0.00	0.002	n.d.	0.00	0.003	2,768.80	25.04	–
k025	17 May 2011	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Iwaya, Awaji Isl., Hyogo	Directly dried	n.d.	0.00	0.001	n.d.	0.00	0.001	1,841.80	34.15	0.029
k026	31 May 2011	Phaeophyceae	Ectocarpales *s.l.*	*Colpomenia sinuosa*	Iwaya, Awaji Isl., Hyogo	Directly dried	n.d.	0.00	0.001	n.d.	0.00	0.001	496.50	45.30	0.042
k027	31 May 2011	Ulvophyceae	Ulvales	*Ulva pertusa*	Iwaya, Awaji Isl., Hyogo	Directly dried	n.d.	0.00	0.001	n.d.	0.00	0.001	1,147.60	42.53	0.037
k028	30 May 2011	Phaeophyceae	Ectocarpales *s.l.*	*Scytosiphon lomentaria*	Iwaya, Awaji Isl., Hyogo	Directly dried	n.d.	0.00	0.007	n.d.	0.00	0.005	1,955.90	188.09	0.176
k029	6 September 2011	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Directly dried	681.86	20.68	0.043	773.22	30.83	0.034	2,115.40	278.58	0.703
k030	6 September 2011	Ulvophyceae	Ulvales	*Ulva pertusa*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.220	202.85	43.48	0.158	n.d.	n.d.	4.519
k031	6 September 2011	Phaeophyceae	Laminariales	*Saccharina japonica*	Nagasaki, Iwaki, Fukushima	Juvenile thallus, directly dried	193.70	25.07	0.097	230.31	36.71	0.086	4,214.10	860.98	2.389
k032	6 September 2011	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Old sporophyll, directly dried	255.02	31.38	0.121	224.34	36.26	0.108	n.d.	n.d.	2.716
k035	14 October 2011	Rhodophyceae	Gigartinales	*Hypnea asiatica*	Nagasaki, Iwaki, Fukushima	Directly dried	114.04	28.88	0.045	161.96	28.91	0.039	2,423.00	1144.60	1.108
k036	14 October 2011	Ulvophyceae	Ulvales	*Ulva pertusa*	Nagasaki, Iwaki, Fukushima	Directly dried	36.56	4.75	0.006	43.05	6.67	0.007	1,013.10	194.79	0.177
k037	14 October 2011	Phaeophyceae	Laminariales	*Saccharina japonica*	Nagasaki, Iwaki, Fukushima	Directly dried	57.64	4.20	0.005	67.41	6.09	0.005	1,120.30	158.44	0.137
k038	14 October 2011	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Directly dried	51.42	9.67	0.014	64.29	10.74	0.013	1,380.30	372.98	0.348
k039	14 October 2011	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Mature thalli, directly dried	105.99	4.78	0.004	114.00	6.66	0.004	995.04	126.90	0.109
k040	14 October 2011	Rhodophyceae	Gigartinales	*Neodilsea yendoana*	Nagasaki, Iwaki, Fukushima	Directly dried	53.92	6.21	0.009	68.46	8.43	0.007	1,550.90	264.19	0.233
k041	14 October 2011	Phaeophyceae	Fucales	*Sargassum yamadae*	Nagasaki, Iwaki, Fukushima	Directly dried	200.17	14.01	0.013	229.20	17.54	0.012	1,522.60	401.82	0.380
k042	14 October 2011	Rhodophyceae	Gigartinales	*Ahnfeltiopsis paradoxa*	Nagasaki, Iwaki, Fukushima	Directly dried	59.91	5.33	0.007	86.65	7.68	0.006	761.02	184.22	0.170
k043	14 October 2011	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Juvenile thalli, directly dried	59.57	6.33	0.008	69.15	8.25	0.008	1,742.10	259.79	0.226
k045	14 October 2011	Rhodophyceae	Ceramiales	*Chondria crassicaulis*	Nagasaki, Iwaki, Fukushima	Directly dried	121.40	7.23	0.006	111.81	9.18	0.008	2,772.70	244.74	0.192
k046	2 May 2011	Seawater			Shioyazaki, Iwaki, Fukushima	Seawater	9.54	2.25	0.004	8.41	3.21	0.003	n.d.	n.d.	0.096
k047	2 May 2011	Seawater			Nagasaki, Iwaki, Fukushima	Seawater	11.27	2.15	0.003	9.74	3.43	0.003	n.d.	n.d.	0.094

Table 1 continued

Sample code	Collection date	Taxonomy			Locality	Note (sample preparation and nature of samples)	^{134}Cs			^{137}Cs			^{40}K		
		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
k048	9 July 2011	Meltwater (dripping)			Nagasaki, Iwaki, Fukushima	From *Undaria pinnatifida*	39.48	3.10	0.003	42.21	4.05	0.003	340.45	97.78	0.091
k049	9 July 2011	Meltwater (dripping)			Nagasaki, Iwaki, Fukushima	From *Scytosiphon lomentaria*	344.71	8.63	0.005	383.09	12.01	0.005	n.d.	n.d.	0.120
k050	9 July 2011	Meltwater (dripping)			Shioyazaki, Iwaki, Fukushima	From *Sargassum thunbergii*	402.97	8.86	0.005	436.72	12.14	0.004	502.65	116.14	0.108
k051	9 July 2011	Meltwater (dripping)			Shioyazaki, Iwaki, Fukushima	From *Eisenia bicyclis*	697.87	11.19	0.006	809.50	16.20	0.005	440.83	113.32	0.106
k052	5 December 2011	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Juvenile thalli, directly dried	54.38	7.13	0.009	69.84	12.27	0.009	2,512.00	310.20	0.232
k053	5 December 2011	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Mature thalli, directly dried	96.84	8.52	0.007	106.11	12.00	0.010	2,258.50	275.40	0.210
k054	5 December 2011	Rhodophyceae	Gigartinales	*Neodilsea yendoana*	Nagasaki, Iwaki, Fukushima	Directly dried	45.30	7.30	0.010	40.58	13.32	0.012	1,082.40	281.80	0.249
k056	5 December 2011	Rhodophyceae	Corallinales	*Calliarthron* sp.	Nagasaki, Iwaki, Fukushima	Directly dried	25.56	5.17	0.007	34.10	8.95	0.007	n.d.	n.d.	0.197
k057	5 December 2011	Rhodophyceae	Ceramiales	*Dasya sessilis*	Nagasaki, Iwaki, Fukushima	Directly dried	219.43	22.28	0.022	234.59	29.33	0.021	4,101.20	705.21	0.592
k058	5 December 2011	Phaeophyceae	Fucales	*Sargassum yamadae*	Nagasaki, Iwaki, Fukushima	Directly dried	70.49	7.93	0.007	82.22	11.74	0.010	2,037.50	297.70	0.234
k059	5 December 2011	Rhodophyceae	Gigartinales	*Ahnfeltiopsis paradoxa*	Nagasaki, Iwaki, Fukushima	Directly dried	100.85	9.72	0.010	92.07	11.75	0.009	468.71	261.31	0.248
k060	5 December 2011	Phaeophyceae	Laminariales	*Saccharina japonica*	Nagasaki, Iwaki, Fukushima	Directly dried	67.14	7.07	0.007	78.82	10.21	0.008	1,793.10	259.95	0.209
k061	5 December 2011	Phaeophyceae	Laminariales	*Saccharina japonica*	Nagasaki, Iwaki, Fukushima	Directly dried	57.12	8.51	0.011	59.34	13.53	0.011	2,279.10	351.17	0.283
k062	5 December 2011	Phaeophyceae	Ralfsiales	*Analipus japonicus*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.048	197.20	41.70	0.042	n.d.	n.d.	1.285
k063	5 December 2011	Rhodophyceae	Gigartinales	*Gloiopeltis furcata*	Nagasaki, Iwaki, Fukushima	Directly dried	51.77	10.06	0.008	53.65	11.71	0.011	592.87	309.57	0.296
k064	7 March 2012	Phaeophyceae	Fucales	*Sargassum horneri*	Nagasaki, Iwaki, Fukushima	Directly dried	133.87	23.47	0.031	187.58	30.54	0.023	2,015.00	856.60	0.831
k065	7 March 2012	Rhodophyceae	Bangiales	*Bangia fuscopurpurea*	Nagasaki, Iwaki, Fukushima	Directly dried	160.51	8.98	0.009	226.22	13.80	0.009	936.70	256.60	0.240
k066	7 March 2012	Ulvophyceae	Ulvales	*Ulva linza*	Nagasaki, Iwaki, Fukushima	Directly dried	66.25	10.30	0.016	85.16	14.60	0.015	1,865.00	443.30	0.408
k067	7 March 2012	Phaeophyceae	Fucales	*Sargassum thunbergii*	Nagasaki, Iwaki, Fukushima	Directly dried	106.37	6.40	0.007	137.03	9.24	0.006	1,654.00	208.10	0.178
k068	7 March 2012	Phaeophyceae	Ectocarpales *s.l.*	*Scytosiphon lomentaria*	Nagasaki, Iwaki, Fukushima	Directly dried	82.48	7.86	0.008	115.59	10.55	0.009	2,442.00	285.50	0.237

Table 1 continued

Sample code	Collection date	Taxonomy			Locality	Note (sample preparation and nature of samples)	^{134}Cs			^{137}Cs			^{40}K		
		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
k069	7 March 2012	Rhodophyceae	Bangiales	*Pyropia yezoensis*	Nagasaki, Iwaki, Fukushima	Directly dried	24.47	4.78	0.008	51.06	9.61	0.008	1,195.00	252.30	0.230
k070	7 March 2012	Phaeophyceae	Ectocarpales *s.l.*	*Petalonia fascia*	Nagasaki, Iwaki, Fukushima	Directly dried	53.68	8.59	0.013	84.37	12.50	0.014	3,159.00	446.90	0.381
k071	7 March 2012	Ulvophyceae	Ulvales	*Monostroma nitidum*	Nagasaki, Iwaki, Fukushima	Directly dried	46.99	9.58	0.017	69.71	10.99	0.013	1,614.00	412.70	0.400
k072	7 March 2012	Phaeophyceae	Ralfsiales	*Analipus japonicus*	Nagasaki, Iwaki, Fukushima	Directly dried	76.48	5.71	0.006	105.77	8.00	0.005	1,064.00	184.90	0.164
k073	7 March 2012	Phaeophyceae	Ralfsiales	*Analipus japonicus*	Nagasaki, Iwaki, Fukushima	Directly dried	93.27	6.47	0.008	101.18	8.32	0.008	1,512.00	258.00	0.243
k074	7 March 2012	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Directly dried	33.69	4.58	0.005	62.91	6.44	0.005	1,642.00	178.60	0.148
k075	7 March 2012	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Directly dried	26.96	3.95	0.005	37.92	5.43	0.005	2,999.00	193.90	0.140
k076	9 April 2012	Phaeophyceae	Fucales	*Sargassum horneri*	Nagasaki, Iwaki, Fukushima	Directly dried	17.43	2.09	0.003	21.75	2.40	0.003	1,996.70	101.66	0.082
k077	9 April 2012	Ulvophyceae	Ulvales	*Ulva pertusa*	Nagasaki, Iwaki, Fukushima	Directly dried	42.63	4.35	0.006	60.71	5.65	0.006	862.87	172.48	0.164
k078	9 April 2012	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Directly dried	21.71	1.83	0.002	28.45	2.48	0.002	1,588.50	89.13	0.073
k079	9 April 2012	Phaeophyceae	Fucales	*Sargassum thunbergii*	Nagasaki, Iwaki, Fukushima	Directly dried	67.57	2.97	0.003	90.22	4.19	0.003	1,293.00	1016.60	0.088
k080	9 April 2012	Phaeophyceae	Desmarestiales	*Desmarestia ligulata*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.007	n.d.	0.00	0.007	1,743.30	203.49	0.186
k081	9 April 2012	Phaeophyceae	Ectocarpales *s.l.*	*Scytosiphon lomentaria*	Nagasaki, Iwaki, Fukushima	Directly dried	48.66	2.99	0.003	71.76	4.17	0.003	1,603.20	115.51	0.099
k082	9 April 2012	Rhodophyceae	Rhodymeniales	*Lomentaria hakodatensis*	Nagasaki, Iwaki, Fukushima	Directly dried	157.82	3.96	0.003	221.71	6.00	0.003	897.33	95.59	0.086
k083	9 April 2012	Rhodophyceae	Bangiales	*Pyropia yezoensis*	Nagasaki, Iwaki, Fukushima	Directly dried	38.64	3.26	0.004	49.65	3.92	0.004	967.23	130.02	0.120
k084	9 April 2012	Rhodophyceae	Ceramiales	*Pterosiphonia pinnulata*	Nagasaki, Iwaki, Fukushima	Directly dried	253.76	7.22	0.007	360.95	10.61	0.006	2,149.90	190.20	0.169
k085	9 April 2012	Rhodophyceae	Gigartinales	*Ahnfeltiopsis paradoxa*	Nagasaki, Iwaki, Fukushima	Directly dried	29.80	2.24	0.003	44.04	2.91	0.002	982.30	87.27	0.077
k086	9 April 2012	Phaeophyceae	Ectocarpales *s.l.*	*Colpomenia sinuosa*	Nagasaki, Iwaki, Fukushima	Directly dried	29.86	2.60	0.004	41.46	3.64	0.004	649.33	113.44	0.107
k087	9 April 2012	Phaeophyceae	Ectocarpales *s.l.*	*Colpomenia sinuosa*	Nagasaki, Iwaki, Fukushima	Directly dried	222.63	9.12	0.010	302.99	12.39	0.009	3,865.90	282.48	0.243
k088	9 April 2012	Phaeophyceae	Laminariales	*Saccharina japonica*	Nagasaki, Iwaki, Fukushima	Directly dried	10.05	1.89	0.003	15.28	2.59	0.003	1,988.00	103.71	0.084
k089	9 April 2012	Phaeophyceae	Ralfsiales	*Analipus japonicus*	Nagasaki, Iwaki, Fukushima	Directly dried	53.89	2.75	0.003	77.91	3.90	0.003	983.29	95.03	0.085

Table 1 continued

Sample code	Collection date	Taxonomy			Locality	Note (sample preparation and nature of samples)	^{134}Cs			^{137}Cs			^{40}K		
		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
k090	9 April 2012	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Directly dried	7.84	1.17	0.002	8.58	1.53	0.002	1,051.40	76.61	0.007
k091	9 April 2012	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Juvenile thalli, directly dried	8.30	2.89	0.005	18.21	2.96	0.004	1,905.00	142.12	0.123
k092	9 April 2012	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Sporophyll, directly dried	16.73	1.98	0.003	22.49	2.54	0.003	2,767.40	104.09	0.077
k093	10 April 2012	Phaeophyceae	Fucales	*Sargassum yamadae*	Yoshio, Katsuura, Chiba	Directly dried	n.d.	0.00	0.003	n.d.	0.00	0.003	1,184.00	102.98	0.091
k094	10 April 2012	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Yoshio, Katsuura, Chiba	Directly dried	n.d.	0.00	0.002	n.d.	0.00	0.002	1,745.10	93.14	0.076
k095	10 April 2012	Phaeophyceae	Dictyotales	*Padina arborescens*	Yoshio, Katsuura, Chiba	Directly dried	n.d.	0.00	0.006	n.d.	0.00	0.006	1,579.40	183.42	0.166
k096	10 April 2012	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Yoshio, Katsuura, Chiba	Directly dried	n.d.	0.00	0.003	n.d.	0.00	0.003	1,991.10	94.59	0.075
k097	10 April 2012	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Kamogawa, Chiba	Directly dried	n.d.	0.00	0.003	n.d.	0.00	0.003	1,652.30	95.39	0.079
k098	10 April 2012	Phaeophyceae	Laminariales	*Ecklonia cava*	Kamogawa, Chiba	Directly dried	n.d.	0.00	0.003	n.d.	0.00	0.003	2,714.60	119.12	0.092
k099	10 April 2012	Phaeophyceae	Fucales	*Sargassum fusiforme*	Kamogawa, Chiba	Directly dried	n.d.	0.00	0.002	n.d.	0.00	0.002	3,236.60	91.82	0.061
k100	10 April 2012	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Kamogawa, Chiba	Directly dried	n.d.	0.00	0.003	n.d.	0.00	0.003	1,922.90	103.42	0.084
k101	23 May 2012	Phaeophyceae	Fucales	*Sargassum horneri*	Terashima, Iwanuma, Miyagi	Directly dried	67.12	26.24	0.029	76.05	23.03	0.022	1,701.70	723.22	0.711
k102	23 May 2012	Ulvophyceae	Ulvales	*Ulva prolifera*	Terashima, Iwanuma, Miyagi	Directly dried	370.13	20.46	0.023	485.87	27.16	0.022	1,268.70	663.95	0.655
k103	23 May 2012	Phaeophyceae	Fucales	*Sargassum horneri*	Matsukawaura, Soma, Fukushima	Directly dried	55.55	2.55	0.003	81.23	3.75	0.003	1,619.50	97.97	0.081
k104	23 May 2012	Ulvophyceae	Ulvales	*Ulva pertusa*	Soma Port, Soma, Fukushima	Directly dried	n.d.	0.00	0.012	n.d.	0.00	0.011	449.78	329.17	0.329
k105	23 May 2012	Ulvophyceae	Ulvales	*Ulva pertusa*	Matsukawaura, Soma, Fukushima	Directly dried	21.80	3.16	0.004	26.03	3.99	0.004	1,015.10	157.70	0.148
k106	23 May 2012	Phaeophyceae	Desmarestiales	*Desmarestia ligulata*	Soma Port, Soma, Fukushima	Directly dried	n.d.	0.00	0.005	6.25	5.01	0.005	1,261.70	154.20	0.141
k107	23 May 2012	Phaeophyceae	Fucales	*Sargassum muticum*	Soma Port, Soma, Fukushima	Directly dried	n.d.	0.00	0.009	13.29	8.60	0.008	2,181.51	287.91	0.260
k108	23 May 2012	Rhodophyceae	Ceramiales	*Delesseria serrulata*	Soma Port, Soma, Fukushima	Directly dried	14.26	4.03	0.005	25.17	4.34	0.004	1,723.80	145.10	0.128
k109	9 April 2012	Ulvophyceae	Ulvales	*Monostroma nitidum*	Nagasaki, Iwaki, Fukushima	Directly dried	25.73	3.51	0.007	27.32	5.02	0.006	703.11	180.50	0.173

Table 1 continued

Sample code	Collection date	Taxonomy			Locality	Note (sample preparation and nature of samples)	^{134}Cs			^{137}Cs			^{40}K		
		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
k110	23 May 2012	Rhodophyceae	Gigartinales	*Grateloupia lanceolata*	Matsukawaura, Soma, Fukushima	Directly dried	12.96	3.27	0.006	n.d.	0.00	0.005	825.74	168.62	0.160
k111	23 May 2012	Rhodophyceae	Gigartinales	*Schizymenia dubyi*	Matsukawaura, Soma, Fukushima	Directly dried	n.d.	0.00	0.004	n.d.	0.00	0.003	721.71	108.01	0.101
k112	23 May 2012	Rhodophyceae	Gigartinales	*Schizymenia dubyi*	Matsukawaura, Soma, Fukushima	Directly dried	19.68	6.73	0.007	17.97	7.14	0.007	882.64	216.67	0.210
k113	23 May 2012	Rhodophyceae	Corallinales	*Gelidium elegans*	Matsukawaura, Soma, Fukushima	Directly dried	60.91	4.66	0.006	90.92	6.23	0.006	1,165.70	177.93	0.167
k115	23 May 2012	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Matsukawaura, Soma, Fukushima	Directly dried	49.35	2.50	0.003	54.70	3.31	0.003	1,428.50	99.27	0.008
k116	9 December 2012	Ulvophyceae	Ulvales	*Ulva pertusa*	Hirono, Futaba, Fukushima	Directly dried	36.73	3.03	0.004	54.46	4.98	0.004	0.00	0.00	0.209
k117	9 December 2012	Rhodophyceae	Gigartinales	*Chondrus giganteus*	Hirono, Futaba, Fukushima	Directly dried	13.61	1.84	0.002	22.89	2.22	0.002	0.00	0.00	0.132
k118	9 December 2012	Phaeophyceae	Ectocarpales *s.l.*	*Petalonia fascia*	Hirono, Futaba, Fukushima	Directly dried	33.09	2.67	0.003	48.98	3.81	0.004	0.00	0.00	0.200
k119	9 December 2012	Rhodophyceae	Gigartinales	*Neodilsea longissima*	Hirono, Futaba, Fukushima	Directly dried	13.66	2.15	0.002	n.d.	0.00	–	0.00	0.00	0.139
k122	10 December 2012	Phaeophyceae	Fucales	*Sargassum yamadae*	Nagasaki, Iwaki, Fukushima	Directly dried	17.05	1.79	0.002	22.02	2.63	0.003	1,299.00	88.13	0.075
k123	10 December 2012	Ulvophyceae	Ulvales	*Ulva pertusa*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.007	26.19	6.81	0.006	769.50	208.60	0.202
k124	10 December 2012	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.003	10.49	3.04	0.003	1,996.00	106.00	0.086
k125	10 December 2012	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Directly dried	19.62	3.96	0.004	21.56	4.08	0.005	2,457.00	160.10	0.135
k128	10 December 2012	Phaeophyceae	Dictyotales	*Spatoglossum pacificum*	Nagasaki, Iwaki, Fukushima	Directly dried	41.53	2.92	0.004	68.91	5.62	0.004	2,484.00	147.10	0.121
k130	10 December 2012	Rhodophyceae	Bangiales	*Pyropia yezoensis*	Nagasaki, Iwaki, Fukushima	Directly dried	12.21	2.80	0.004	26.86	3.84	0.005	657.28	148.70	0.143
k131	10 December 2012	Ulvophyceae	Ulvales	*Ulva prolifera*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.008	8.04	7.58	0.008	848.51	236.56	0.229
k132	10 December 2012	Ulvophyceae	Cladophorales	*Chaetomorpha moniligera*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.007	n.d.	0.00	–	3,770.30	247.20	0.214
k133	10 December 2012	Rhodophyceae	Gigartinales	*Chondrus ocellatus*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.003	6.68	3.58	0.004	677.03	112.57	0.106

Table 1 continued

Sample code	Collection date	Taxonomy			Locality	Note (sample preparation and nature of samples)	^{134}Cs			^{137}Cs			^{40}K		
		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
k134	10 December 2012	Rhodophyceae	Gigartinales	*Grateloupia turuturu*	Nagasaki, Iwaki, Fukushima	Directly dried	10.32	2.49	0.003	9.46	4.22	0.004	1,163.40	128.02	0.116
k135	10 December 2012	Rhodophyceae	Gigartinales	*Ahnfeltiopsis paradoxa*	Nagasaki, Iwaki, Fukushima	Directly dried	10.64	2.14	0.002	9.84	1.87	0.003	604.94	81.66	0.076
k138	10 December 2012	Rhodophyceae	Gigartinales	*Gloiopeltis furcata*	Nagasaki, Iwaki, Fukushima	Directly dried	64.95	8.00	0.014	92.33	12.25	0.013	5,190.10	427.40	0.376
k139	10 December 2012	Rhodophyceae	Gigartinales	*Grateloupia lanceolata*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.007	8.51	6.27	0.006	688.58	198.65	0.193
k140	10 December 2012	Rhodophyceae	Gigartinales	*Plocamium cartilagineum*	Nagasaki, Iwaki, Fukushima	Directly dried	22.16	3.92	0.007	59.69	6.30	0.006	514.62	206.47	0.204
k141	10 December 2012	Rhodophyceae	Ceramiales	*Chondria crassicaulis*	Nagasaki, Iwaki, Fukushima	Directly dried	41.82	4.62	0.005	47.81	5.45	0.006	3,859.40	213.11	0.174
k142	9 December 2012	Phaeophyceae	Fucales	*Sargassum thunbergii*	Hisanohama, Iwaki, Fukushima	Directly dried	345.96	5.95	0.004	592.67	98.32	0.004	1,401.50	112.60	0.098
k143	9 December 2012	Rhodophyceae	Gigartinales	*Chondrus giganteus*	Hisanohama, Iwaki, Fukushima	Directly dried	38.55	2.44	0.003	59.74	3.62	0.003	1,149.10	97.59	0.086
k144	9 December 2012	Rhodophyceae	Gigartinales	*Chondrus giganteus*	Hisanohama, Iwaki, Fukushima	Directly dried	18.33	3.51	0.004	36.82	3.47	0.004	1,257.70	122.81	0.110
k145	9 December 2012	Phaeophyceae	Fucales	*Sargassum muticum*	Hisanohama, Iwaki, Fukushima	Directly dried	123.97	4.21	0.004	209.89	6.76	0.004	1,503.30	1290.70	0.113
k146	9 December 2012	Phaeophyceae	Fucales	*Sargassum fusiforme*	Hisanohama, Iwaki, Fukushima	Directly dried	21.49	1.86	0.003	34.70	3.37	0.003	4,206.90	116.52	0.078
k147	9 December 2012	Phaeophyceae	Ectocarpales *s.l.*	*Colpomenia sinuosa*	Hisanohama, Iwaki, Fukushima	Directly dried	219.97	4.88	0.004	363.65	7.70	0.003	3,025.40	122.48	0.093
k148	9 December 2012	Phaeophyceae	Laminariales	*Saccharina japonica*	Hisanohama, Iwaki, Fukushima	Directly dried	33.55	2.71	0.003	60.87	35.82	0.003	3,479.60	118.90	0.086
k149	9 December 2012	Phaeophyceae	Laminariales	*Saccharina japonica*	Hisanohama, Iwaki, Fukushima	Directly dried	34.03	2.82	0.003	54.74	3.61	0.003	3,218.60	122.46	0.091
k150	9 December 2012	Rhodophyceae	Ceramiales	*Chondria crassicaulis*	Hisanohama, Iwaki, Fukushima	Directly dried	42.05	4.27	0.005	58.60	6.18	0.005	3,937.10	186.64	0.147
k151	16 March 2013	Phaeophyceae	Fucales	*Sargassum horneri*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.003	10.64	3.11	0.003	2,250.80	108.28	0.085
k152	16 March 2013	Phaeophyceae	Fucales	*Sargassum yamadae*	Nagasaki, Iwaki, Fukushima	Directly dried	9.69	2.30	0.005	7.91	4.56	0.004	840.94	140.74	0.132

Table 1 continued

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		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
k153	16 March 2013	Ulvophyceae	Ulvales	*Ulva pertusa*	Nagasaki, Iwaki, Fukushima	Directly dried	9.79	1.87	0.003	19.12	3.24	0.003	1,453.10	117.82	0.103
k154	16 March 2013	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00		7.18	1.84	0.003	1,274.50	91.32	0.079
k155	16 March 2013	Phaeophyceae	Fucales	*Sargassum thunbergii*	Nagasaki, Iwaki, Fukushima	Directly dried	13.92	2.35	0.003	20.90	2.88	0.003	1,738.20	124.22	0.106
k156	16 March 2013	Rhodophyceae	Gigartinales	*Ahnfeltiopsis flabelliformis*	Nagasaki, Iwaki, Fukushima	Directly dried	16.71	3.77	0.008	22.80	7.53	0.007	1,349.40	230.51	0.217
k158	16 March 2013	Rhodophyceae	Rhodymeniales	*Lomentaria hakodatensis*	Nagasaki, Iwaki, Fukushima	Directly dried	34.77	3.08	0.004	60.47	4.43	0.004	993.42	130.86	0.121
k159	16 March 2013	Phaeophyceae	Dictyotales	*Pachydictyon coriaceum*	Nagasaki, Iwaki, Fukushima	Directly dried	26.23	4.94	0.007	38.11	5.35	0.007	1,494.50	229.91	0.216
k160	16 March 2013	Ulvophyceae	Cladophorales	*Cladophora* sp.	Nagasaki, Iwaki, Fukushima	Directly dried	50.88	5.23	0.006	75.11	6.10	0.006	1,029.20	190.05	0.179
k161	16 March 2013	Ulvophyceae	Ulvales	*Ulva prolifera*	Nagasaki, Iwaki, Fukushima	Directly dried	44.66	4.58	0.006	67.05	5.66	0.006	1,218.00	182.82	0.172
k162	16 March 2013	Ulvophyceae	Cladophorales	*Chaetomorpha moniligera*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.004	10.23	3.07	0.004	3,397.30	168.28	0.134
k164	16 March 2013	Rhodophyceae	Gigartinales	*Ahnfeltiopsis paradoxa*	Nagasaki, Iwaki, Fukushima	Directly dried	11.31	2.13	0.002	16.85	2.18	0.002	902.53	85.45	0.076
k165	16 March 2013	Rhodophyceae	Gigartinales	*Gloiopeltis furcata*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.003	n.d.	0.00	0.003	534.66	105.28	0.100
k166	16 March 2013	Rhodophyceae	Gigartinales	*Grateloupia lanceolata*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.004	n.d.	0.00	0.004	601.26	125.92	0.120
k167	16 March 2013	Rhodophyceae	Gigartinales	*Grateloupia lanceolata*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.003	7.42	1.97	0.003	503.27	85.20	0.081
k169	16 March 2013	Rhodophyceae	Gigartinales	*Polyopes affinis*	Nagasaki, Iwaki, Fukushima	Directly dried	27.53	4.41	0.004	48.01	4.06	0.004	533.00	133.37	0.128
k170	16 March 2013	Rhodophyceae	Gigartinales	*Polyopes affinis*	Nagasaki, Iwaki, Fukushima	Directly dried	12.20	2.42	0.004	13.51	3.60	0.003	377.65	107.94	0.105
k171	16 March 2013	Phaeophyceae	Ralfsiales	*Analipus japonicus*	Nagasaki, Iwaki, Fukushima	Directly dried	13.12	2.58	0.006	36.35	5.03	0.005	931.90	180.17	0.172
k173	16 March 2013	Rhodophyceae	Ceramiales	*Chondria crassicaulis*	Nagasaki, Iwaki, Fukushima	Directly dried	12.14	2.48	0.004	15.77	3.92	0.004	4,277.60	152.07	0.110
k174	16 March 2013	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Directly dried	6.70	2.11	0.003	n.d.	0.00	0.029	1,352.80	100.01	0.086
k175	16 March 2013	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Directly dried	5.29	2.10	0.003	11.27	2.96	0.003	1,808.80	102.71	0.084
k176	16 March 2013	Ulvophyceae	Cladophorales	*Cladophora albida*	Nagasaki, Iwaki, Fukushima	Directly dried	62.13	5.30	0.006	113.19	8.75	0.006	1,573.60	205.12	0.190
k177	16 March 2013	Rhodophyceae	Gigartinales	*Mazzaella japonica*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0.00	0.004	8.24	2.33	0.004	771.82	109.99	0.103

Table 1 continued

Sample code	Collection date	Taxonomy			Locality	Note (sample preparation and nature of samples)	^{134}Cs			^{137}Cs			^{40}K		
		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
k178	16 March 2013	Ulvophyceae	Ulvales	*Ulva pertusa*	Nagasaki, Iwaki, Fukushima	Directly dried	32.05	3.56	0.005	60.48	5.13	0.005	1,362.20	161.87	0.148
k179	16 March 2013	Ulvophyceae	Ulvales	*Ulva pertusa*	Hirono, Futaba, Fukushima	Directly dried	26.06	4.60	0.007	58.17	5.74	0.006	1,016.70	201.46	0.192
k180	16 March 2013	Rhodophyceae	Gigartinales	*Chondrus giganteus*	Hirono, Futaba, Fukushima	Directly dried	8.03	2.00	0.003	13.81	3.09	0.003	537.88	90.53	0.086
k183	16 March 2013	Rhodophyceae	Bangiales	*Pyropia yezoensis*	Hirono, Futaba, Fukushima	Directly dried	n.d.	0.00	0.005	11.26	2.88	0.004	759.90	146.33	0.139
k184	16 March 2013	Ulvophyceae	Ulvales	*Ulva prolifera*	Hirono, Futaba, Fukushima	Directly dried	78.64	6.22	0.008	139.62	8.75	0.007	828.08	230.72	0.224
k185	16 March 2013	Phaeophyceae	Ectocarpales *s.l.*	*Petalonia fascia*	Hirono, Futaba, Fukushima	Directly dried	n.d.	0.00	0.005	21.69	3.26	0.004	1,394.70	138.48	0.125
k186	16 March 2013	Rhodophyceae	Gigartinales	*Neodilsea longissima*	Hirono, Futaba, Fukushima	Directly dried	7.08	2.59	0.003	14.36	3.25	0.003	1,756.70	113.02	0.096
k187	16 March 2013	Rhodophyceae	Gigartinales	*Gloiopeltis furcata*	Hirono, Futaba, Fukushima	Directly dried	14.63	2.89	0.005	18.17	3.36	0.005	437.27	148.11	0.145
k188	16 March 2013	Rhodophyceae	Gigartinales	*Gloiopeltis furcata*	Hirono, Futaba, Fukushima	Directly dried	9.04	2.42	0.005	24.57	3.71	0.005	542.12	143.03	0.138
k189	16 March 2013	Phaeophyceae	Ralfsiales	*Analipus japonicus*	Hirono, Futaba, Fukushima	Directly dried	22.10	2.30	0.003	36.36	3.02	0.003	1,113.90	1017.40	0.910
k190	16 March 2013	Rhodophyceae	Ceramiales	*Chondria crassicaulis*	Hirono, Futaba, Fukushima	Directly dried	29.82	2.84	0.003	49.95	4.25	0.003	3,051.80	126.98	0.097
k191	16 March 2013	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Hirono, Futaba, Fukushima	Directly dried	n.d.	0.00	0.003	11.93	2.67	0.002	1,313.60	89.82	0.077

Values of detection limit in Bq kg^{-1}. n.d. means not detectable (blow the detection limit). Error means overall error estimated by the analyzing software from systematic errors in the system together with the standard deviation of the counting

Table 2 List of samples examined at Iwaki Meisei University, and the results of ^{134}Cs and ^{137}Cs measurements

Sample code	Collection date	Taxonomy			Locality	Note (sample preparation and nature of samples)	^{134}Cs			^{137}Cs		
		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
IMU001	2 May 2011	Phaeophyceae	Dictyotales	*Spatoglossum pacificum*	Shioyazaki, Iwaki, Fukushima	Frozen	5,200	138	156	9,430	179	133
IMU002	2 May 2011	Phaeophyceae	Fucales	*Sargassum yamadae*	Shioyazaki, Iwaki, Fukushima	Frozen	1,210	25.2	25.8	1,980	31.5	19.9
IMU003	6 May 2011	Phaeophyceae	Laminariales	*Saccharina japonica*	Nagasaki, Iwaki, Fukushima	Frozen	3,910	116	144	6,020	138	108
IMU004	6 May 2011	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Frozen	3,670	96.5	107	5,980	119	89.2
IMU005	6 May 2011	Rhodophyceae	Gigartinales	*Grateloupia lanceolata*	Nagasaki, Iwaki, Fukushima	Frozen	2,920	123	159	4,390	146	144
IMU006	9 July 2011	Phaeophyceae	Dictyotales	*Dictyota dichotoma*	Nagasaki, Iwaki, Fukushima	Frozen	556	72.1	167	1,130	88.2	148
IMU007	9 July 2011	Phaeophyceae	Ectocarpales *s.l.*	*Colpomenia sinuosa*	Nagasaki, Iwaki, Fukushima	Frozen	720	163	446	1,390	189	434
IMU008	9 July 2011	Phaeophyceae	Ectocarpales *s.l.*	*Scytosiphon lomentaria*	Nagasaki, Iwaki, Fukushima	Frozen	590	53.4	98.5	1,080	68.3	101
IMU009	9 July 2011	Phaeophyceae	Desmarestiales	*Desmarestia ligulata*	Nagasaki, Iwaki, Fukushima	Frozen	154	7.70	11.3	255	9.98	11.5
IMU010	9 July 2011	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Frozen	676	16.0	21.0	1,240	21.1	17.5
IMU011	9 July 2011	Rhodophyceae	Gigartinales	*Chondrus ocellatus*	Nagasaki, Iwaki, Fukushima	Frozen	288	64.1	174	333	63.7	164
IMU012	9 July 2011	Rhodophyceae	Gigartinales	*Grateloupia sparsa*	Nagasaki, Iwaki, Fukushima	Frozen	569	50.0	102	1,030	58.8	81.1
IMU013	9 July 2011	Rhodophyceae	Gigartinales	*Ahnfeltiopsis paradoxa*	Nagasaki, Iwaki, Fukushima	Frozen	174	13.2	23.3	339	17.4	22.9
IMU014	9 July 2011	Rhodophyceae	Rhodymeniales	*Gastroclonium pacificum*	Nagasaki, Iwaki, Fukushima	Frozen	733	43.8	70.2	1,190	53.1	64.5
IMU015	14 October 2011	Phaeophyceae	Fucales	*Sargassum horneri*	Nagasaki, Iwaki, Fukushima	Frozen	267	62.3	172	470	66.2	147
IMU016	4 July 2012	Phaeophyceae	Ralfsiales	*Analipus japonicus*	Nagasaki, Iwaki, Fukushima	Directly dried	54.2	6.30	13.8	87.8	7.52	13.7
IMU017	4 July 2012	Phaeophyceae	Dictyotales	*Dictyota dichotoma*	Nagasaki, Iwaki, Fukushima	Directly dried	113	8.20	14.8	150	9.31	14.7
IMU018	4 July 2012	Phaeophyceae	Ectocarpales *s.l.*	*Leathesia difformis*	Nagasaki, Iwaki, Fukushima	Directly dried	128	10.4	20.7	213	12.2	18.5
IMU019	4 July 2012	Phaeophyceae	Ectocarpales *s.l.*	*Scytosiphon lomentaria*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0	51.0	56.3	15.8	45.7
IMU020	4 July 2012	Phaeophyceae	Ectocarpales *s.l.*	*Tinocladia crassa*	Nagasaki, Iwaki, Fukushima	Directly dried	27.3	8.31	24.4	38.7	9.62	27.0
IMU021	4 July 2012	Phaeophyceae	Fucales	*Sargassum confusum*	Nagasaki, Iwaki, Fukushima	Directly dried	15.5	3.80	10.6	30.0	4.20	9.20
IMU022	4 July 2012	Phaeophyceae	Fucales	*Sargassum horneri*	Nagasaki, Iwaki, Fukushima	Directly dried	17.7	3.75	10.3	37.4	4.75	11.1
IMU023	4 July 2012	Phaeophyceae	Fucales	*Sargassum thunbergii*	Nagasaki, Iwaki, Fukushima	Directly dried	14.3	2.81	7.59	31.3	3.60	8.00
IMU024	4 July 2012	Phaeophyceae	Laminariales	*Undaria pinnatifida*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0	10.8	23.0	4.25	11.2
IMU025	4 July 2012	Phaeophyceae	Laminariales	*Saccharina japonica*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0	10.8	23.7	3.92	9.39
IMU026	4 July 2012	Phaeophyceae	Laminariales	*Eisenia bicyclis*	Nagasaki, Iwaki, Fukushima	Directly dried	14.8	3.23	8.69	19.0	3.67	9.52
IMU027	4 July 2012	Ulvophyceae	Ulvales	*Ulva pertusa*	Nagasaki, Iwaki, Fukushima	Directly dried	15.3	3.76	10.5	23.7	4.32	11.0
IMU028	4 July 2012	Ulvophyceae	Cladophorales	*Chaetomorpha moniligera*	Nagasaki, Iwaki, Fukushima	Directly dried	17.1	3.99	11.0	26.5	4.93	12.7
IMU029	4 July 2012	Ulvophyceae	Codiales	*Codium lucasii*	Nagasaki, Iwaki, Fukushima	Directly dried	n.d.	0	48.0	109	18.3	4.50
IMU030	4 July 2012	Ulvophyceae	Bryopsidales	*Bryopsis plumosa*	Nagasaki, Iwaki, Fukushima	Directly dried	72.4	8.32	19.0	86.7	8.96	19.5
IMU031	4 July 2012	Rhodophyceae	Corallinales	*Corallina pilulifera*	Nagasaki, Iwaki, Fukushima	Directly dried	36.2	3.56	7.29	63.9	4.63	7.87
IMU032	4 July 2012	Rhodophyceae	Corallinales	*Calliarthron yessoense*	Nagasaki, Iwaki, Fukushima	Directly dried	14.7	3.53	9.82	22.1	3.83	9.45
IMU033	4 July 2012	Rhodophyceae	Gigartinales	*Chondracanthus intermedius*	Nagasaki, Iwaki, Fukushima	Directly dried	38.9	6.04	14.9	54.2	6.64	14.7
IMU034	4 July 2012	Rhodophyceae	Gigartinales	*Chondrus ocellatus*	Nagasaki, Iwaki, Fukushima	Directly dried	23.2	4.07	10.3	40.6	4.87	10.4
IMU035	4 July 2012	Rhodophyceae	Gigartinales	*Ahnfeltiopsis paradoxa*	Nagasaki, Iwaki, Fukushima	Directly dried	27.9	35.9	8.22	47.0	4.54	9.15
IMU036	4 July 2012	Rhodophyceae	Ceramiales	*Chondria crassicaulis*	Nagasaki, Iwaki, Fukushima	Directly dried	69.0	7.69	16.4	105	8.81	15.4
IMU037	4 July 2012	Rhodophyceae	Ceramiales	*Laurencia okamurae*	Nagasaki, Iwaki, Fukushima	Directly dried	129	7.29	11.3	239	9.59	9.84

Table 2 continued

Sample code	Collection date	Taxonomy			Locality	Note (sample preparation and nature of samples)	^{134}Cs			^{137}Cs		
		Class	Order	Species			Bq kg^{-1}	Error	Detect. limit	Bq kg^{-1}	Error	Detect. limit
IMU038	10 December 2012	Phaeophyceae	Fucales	*Sargassum horneri*	Nagasaki, Iwaki, Fukushima	Directly dried	16.3	4.69	13.6	n.d.	0	16.0
IMU039	10 December 2012	Phaeophyceae	Fucales	*Sargassum siliquastrum*	Nagasaki, Iwaki, Fukushima	Directly dried	60.5	7.04	16.2	118	8.37	13.1
IMU040	10 December 2012	Phaeophyceae	Laminariales	*Saccharina japonica*	Nagasaki, Iwaki, Fukushima	Directly dried	26.2	5.23	13.8	47.5	6.22	13.9

Values of detection limit in Bq kg^{-1}. n.d. means not detectable (blow the detection limit). Error means overall error estimated by the analyzing software from systematic errors in the system together with the standard deviation of the counting

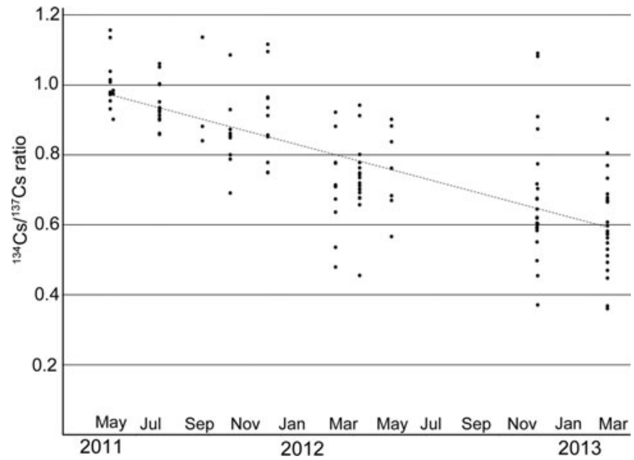


Fig. 2 Evolution of ^{134}Cs/^{137}Cs ratio of the specimens examined. Dotted line indicates the theoretical expected value by the spontaneous decay of ^{134}Cs and ^{137}Cs

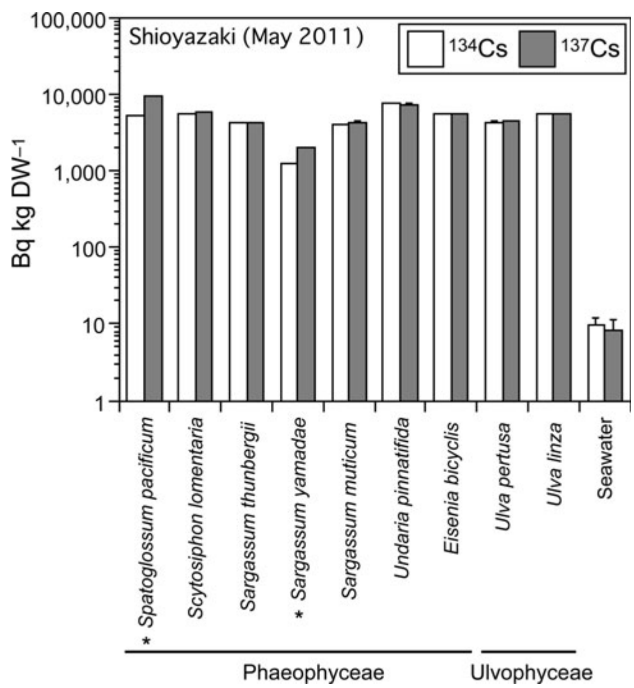


Fig. 3 Quantities of ^{134}Cs and ^{137}Cs of diverse marine macroalgae and seawater collected in May 2011 at Shioyazaki, Iwaki, Fukushima Pref., Japan. Measurement unit for seawater is Bq kg^{-1} (wet weight). *Asterisks* show data by the measurement at Iwaki Meisei University

but was rather constant in the other samples (ca. 1,000–1,500 Bq kg^{-1}). The annual brown algae *Undaria pinnatifida* (Fig. 11) and *Sargassum horneri* (data not shown) showed similar patterns of ^{137}Cs progression: they showed rapid decline in the first year, and continuously declined until March 2013. The ^{40}K level of *Undaria pinnatifida* of the specimens from Chiba Pref. and Hyogo Pref. were 1,840–1,990 Bq kg^{-1} (Fig. 11).

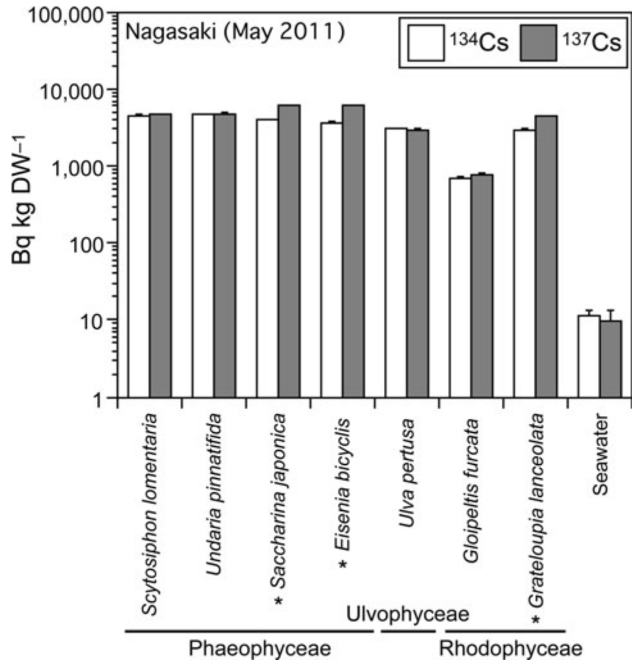


Fig. 4 Quantities of ^{134}Cs and ^{137}Cs of diverse marine macroalgae and seawater collected in May 2011 at Nagasaki, Iwaki, Fukushima Pref., Japan. Measurement unit for seawater is Bq kg^{-1} (wet weight). *Asterisks* show data by the measurement at Iwaki Meisei University

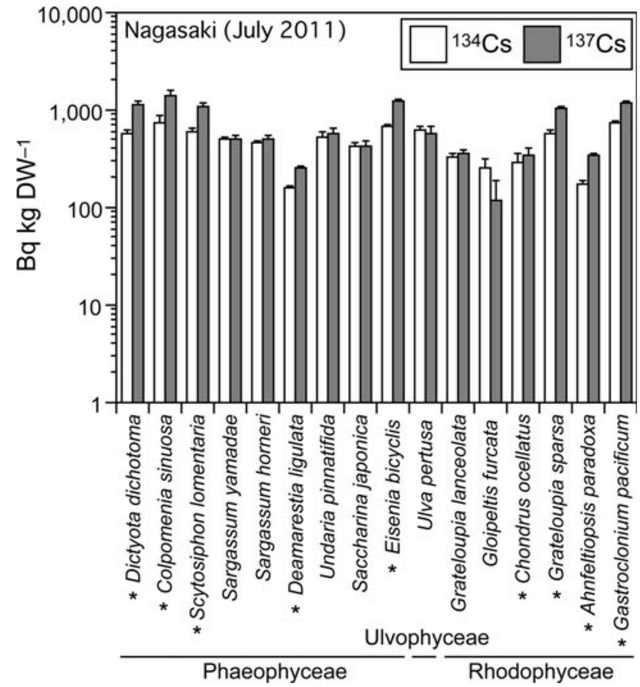


Fig. 5 Quantities of ^{134}Cs and ^{137}Cs of diverse marine macroalgae collected in July 2011 at Nagasaki, Iwaki, Fukushima Pref., Japan. *Asterisks* show data by the measurement at Iwaki Meisei University

Perennial brown algae such as *Eisenia bicyclis* (Fig. 12) and *Sargassum yamadae* (Fig. 13), which are important elements of algal beds in the area, also showed similar progressions in ^{137}Cs levels, although the level of ^{40}K (ca. 1,500–2,000 Bq kg^{-1}) was generally higher than in *Ulva pertusa* (Fig. 11). The ^{40}K levels of *Eisenia bicyclis* and *Sargassum yamadae* of the specimens from Chiba Pref. were 1,650–1,750 and 1,180 Bq kg^{-1}, respectively, and were comparable to those in Fukushima Pref. (Figs. 12, 13). Unfortunately, there were few species of red algae that could be collected seasonally. *Ahnfeltiopsis paradoxa* (Suringar) Masuda (Fig. 14) showed a clear decrease in ^{137}Cs levels in 2011, but it was also still in a detectable range of ca. 10 Bq kg^{-1} in March 2013. ^{40}K levels were in the range of 800–1,000 Bq kg^{-1}, which was comparable to *Ulva* (green alga) and considerably lower than *Undaria*, *Eisenia* and *Sargassum*) (brown algae).

Discussion

^{137}Cs concentrations in seawater off the eastern Japan coast prior to the accident were in the same order of magnitude as other surface oceanic waters, between 1 and 3 Bq m^{-3} for ^{137}Cs (Nakanishi et al. 2010). After the accident, measured concentrations in a 30 km perimeter around the plant exceeded 10 Bq L^{-1} (or 10,000,000 Bq m^{-3}) (Bailly du Bois et al. 2012).

The ^{137}Cs levels in seawater at the coast of Iwaki (Shioyazaki and Nagasaki) one month after the mass discharge of high concentration contaminated water in April measured ca. 8–10 Bq L^{-1} in our own measurements (Fig. 4). These values agree well with the report of Bailly du Bois et al. (2012). The coast around Shioyazaki and Nagasaki was estimated to have been exposed up to ca. 60 Bq L^{-1} for about two weeks (15 April–1 May 2011) according to the estimated geographical distribution of the ^{137}Cs in Bailly du Bois et al. (2012).

The influence of the radioactive liquid effluents escaping directly from the nuclear power plant was particularly significant from 26 March to 8 April 2011 in the vicinity of the plant. Concentrations measured after 10 April 2011 declined in the vicinity of the plant. The perennial brown alga *Eisenia bicyclis* also showed a similar pattern, but in the red alga *Ahnfeltiopsis paradoxa* the levels stayed about the same during December 2012 and March 2013. In contrast, the concentration of ^{40}K was relatively stable throughout the period, in accordance with the assumption that it is not of anthropogenic origin because the levels were comparable to those of the specimens from Chiba Pref. and Hyogo Pref.

Given that the seawater ^{137}Cs levels were in the range of 10–60 Bq L^{-1} at Shioyazaki and Nagasaki in April 2011 [according to our data and those by Bailly du Bois et al.

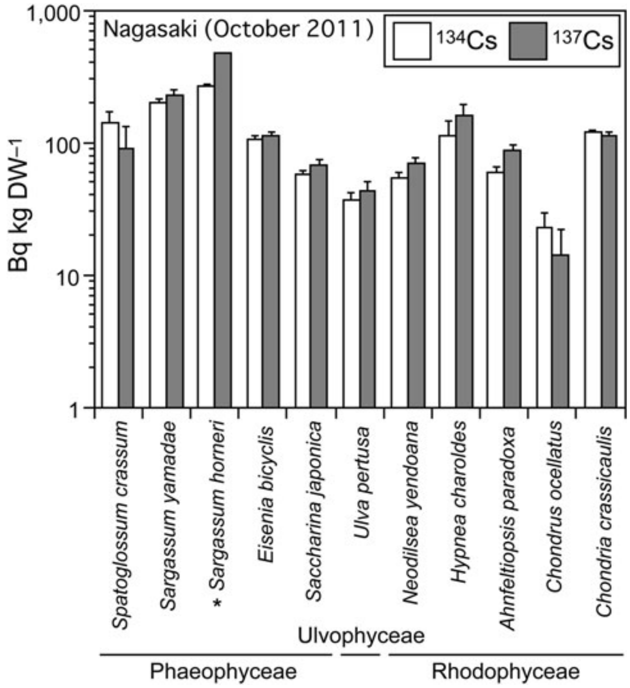


Fig. 6 Quantities of ^{134}Cs and ^{137}Cs of diverse marine macroalgae collected in October 2011 at Nagasaki, Iwaki, Fukushima Pref., Japan. *Asterisks* show data by the measurement at Iwaki Meisei University

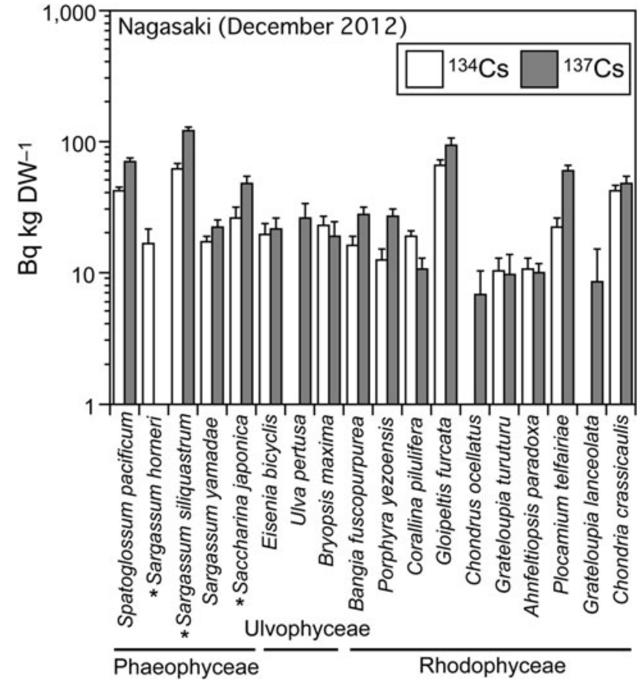


Fig. 8 Quantities of ^{134}Cs and ^{137}Cs of diverse marine macroalgae and seawater collected in December 2012 at Nagasaki, Iwaki, Fukushima Pref., Japan. *Asterisks* show data by the measurement at Iwaki Meisei University

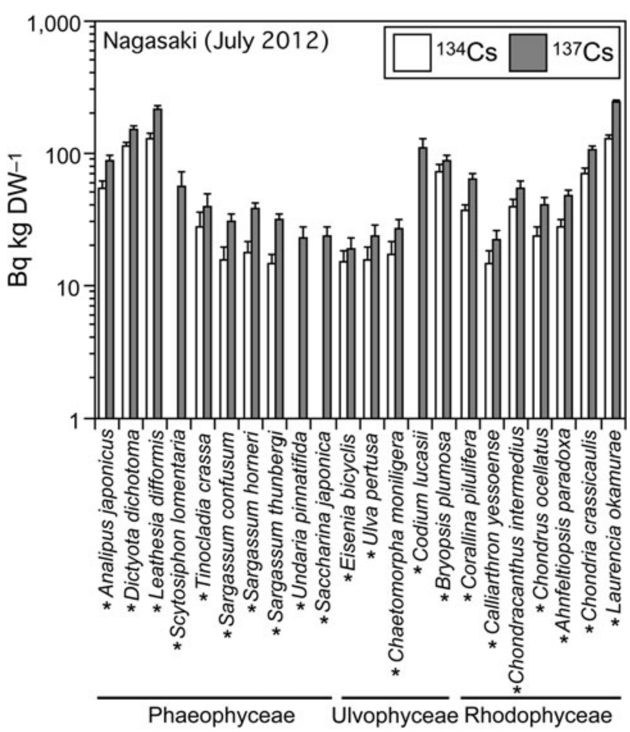


Fig. 7 Quantities of ^{134}Cs and ^{137}Cs of diverse marine macroalgae collected in July 2012 at Nagasaki, Iwaki, Fukushima Pref., Japan. *Asterisks* show data by the measurement at Iwaki Meisei University

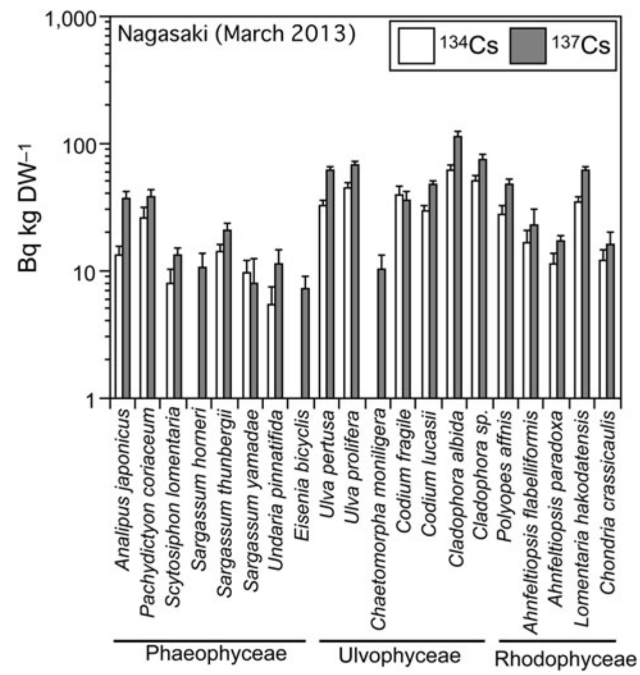


Fig. 9 Quantities of ^{134}Cs and ^{137}Cs of diverse marine macroalgae and seawater collected in March 2013 at Nagasaki, Iwaki, Fukushima Pref., Japan

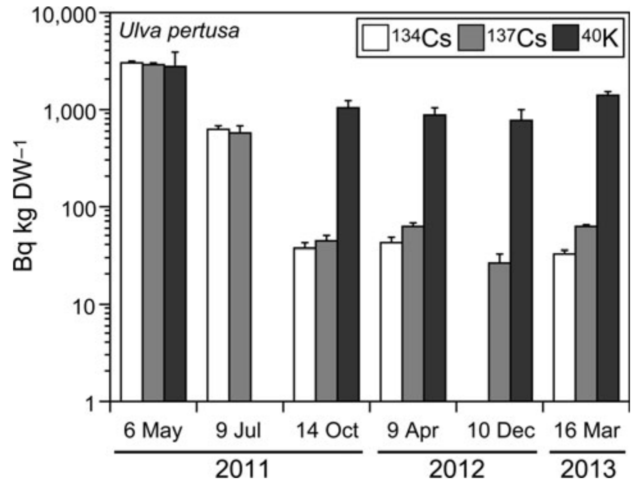


Fig. 10 Evolution of ^{134}Cs, ^{137}Cs and ^{40}K quantities in the annual green alga *Ulva pertusa* at Nagasaki, Iwaki, Fukushima Pref., Japan

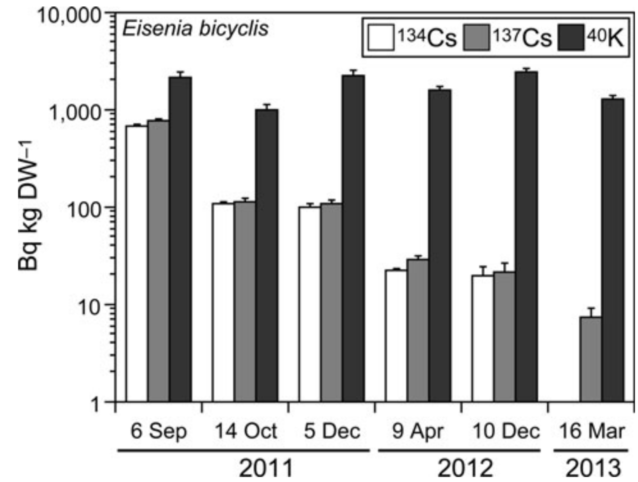


Fig. 12 Evolution of ^{134}Cs, ^{137}Cs and ^{40}K quantities in the perennial brown alga *Eisenia bicyclis* at Nagasaki, Iwaki, Fukushima Pref., Japan

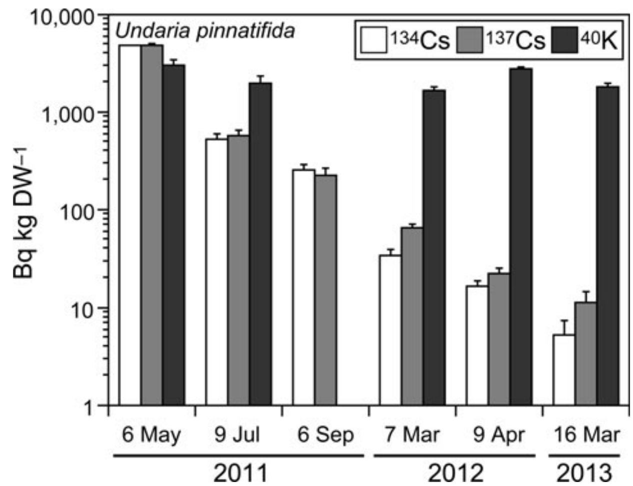


Fig. 11 Evolution of ^{134}Cs, ^{137}Cs and ^{40}K quantities in the annual brown alga *Undaria pinnatifida* at Nagasaki, Iwaki, Fukushima Pref., Japan

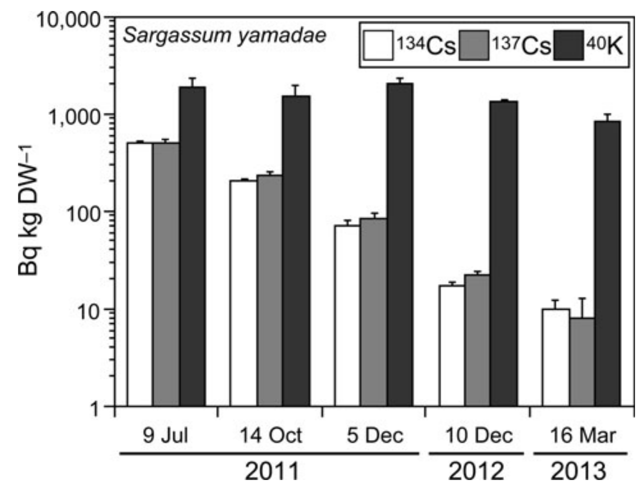


Fig. 13 Evolution of ^{134}Cs, ^{137}Cs and ^{40}K quantities in the perennial brown alga *Sargassum yamadae* at Nagasaki, Iwaki, Fukushima Pref., Japan

(2012)], and that seaweeds collected in May 2011 contained an average ^{137}Cs level of ca. 5,000 Bq kg^{-1}, the seaweeds are considered to have accumulated ^{137}Cs in their tissues by 8–50 folds compared to their environment (80–500 folds in dry samples, given the wet/dry mass ratio of 10). These CF values must be certainly underestimated because some part (or a large part) of the algal tissue of the specimens collected in early May 2011 (2 May and 6 May 2011) had already been grown up before the exposure to the highly radiopolluted seawater. In general, the growth rate of seaweeds differs depending on the species; even in rather fast growing species, such as *Ulva* spp. and *Undaria pinnatifida*, their growth period is longer than one month; and many species grow up in 3–4 months.

The above CF values of ca. 8–50 agree well with the known CF values of ca. 30–50 in marine macroalgae (IAEA 2004). Considering the underestimation in our data as mentioned above, macroalgal CF may be somewhat higher than the IAEA standard. This could be caused if the local concentration of ^{137}Cs in polluted seawater during the seaweed growth period (e.g., during later April 2011) were higher than that on the collection dates after May 2011.

The ^{137}Cs levels of the specimens directly dried after collection (air-dried and then heated; e.g., k001, k004 in Table 1), and those frozen before drying (frozen, thawed, air-dried and then heated; e.g., k017, k023) were roughly comparable. Meltwater from the frozen algal tissues (k048–

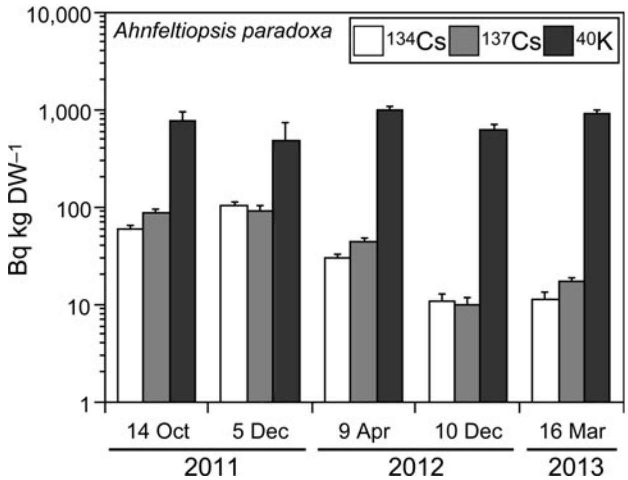


Fig. 14 Evolution of ^{134}Cs, ^{137}Cs and ^{40}K quantities in the perennial red alga *Ahnfeltiopsis paradoxa* at Nagasaki, Iwaki, Fukushima Pref., Japan

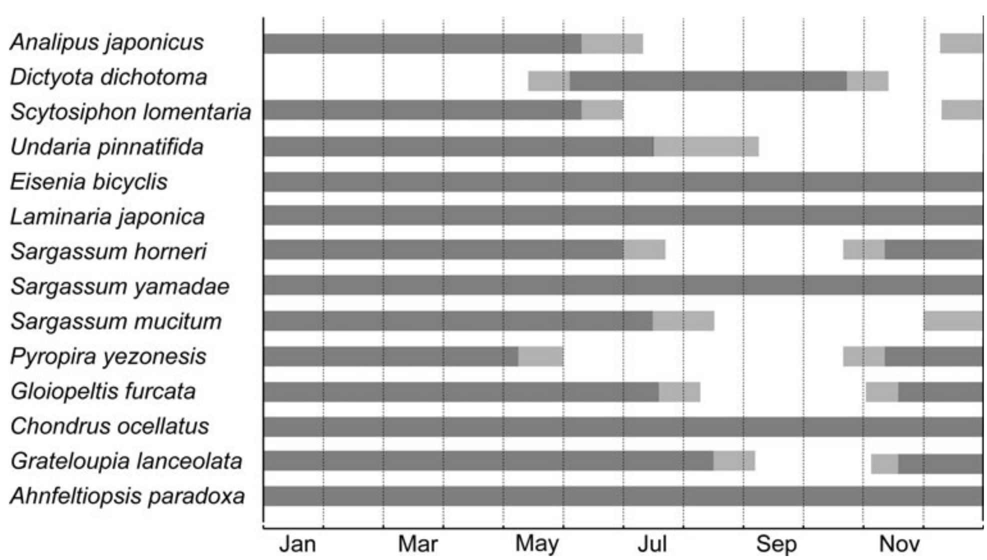
k051) contained significantly higher concentrations of ^{137}Cs compared to the thawed tissue, although the levels were rather variable depending on the taxa (ca. 40 Bq L^{-1} in *Undaria pinnatifida* to ca. 800 Bq L^{-1} in *Eisenia bicyclis*). The ^{137}Cs concentrations in meltwater was much higher than that in ambient seawater, which was below the detection level of ca. 1 Bq L^{-1} in early July 2011, and was comparable to that in fresh algal tissues, which was estimated from the present measurements of directly dried specimens. Thus, we concluded that the measurements based on dried specimens after the freezing process may underestimate the ^{137}Cs concentrations in fresh algal tissues.

We showed that the radioactive Cs levels in marine macroalgae decline rather rapidly during the summer of 2011. This may be ascribed to the fact that the marine macroalgae grow and turnover rapidly during this period. The growth periods of a number of annual taxa [e.g., *Scytosiphon lomentaria*, *Monostroma nitidum* Wittrock, *Ulva* spp., *Pyropia* (*Porphyra*) spp.] are less than six months irrespective of their life history patterns (see below; Fig. 15). Many taxa have life histories with alternation of heteromorphic sporophytes and gametophytes [e.g., *Undaria pinnatifida*, *Scytosiphon lomentaria*, *Monostroma nitidum*, *Pyropia yezoensis* (Roth) C. Agardh] and their macroscopic generations disappear during summer and autumn (Bold and Wynne 1985; Graham and Wilcox 2000; Hori 1993, 1994). In these cases, even though the thallus tissues had absorbed a large amount of radionuclides during development (i.e., late March–April 2011), they were replaced by new tissues, resulting in lower or null radionuclide concentrations before summer and autumn 2011.

Some of the taxa with heteromorphic life history have perennial macroscopic thalli, and these thalli persist during summer (e.g., *Eisenia bicyclis*, *Saccharina japonica*). However, they show intercalary growth with a growth zone in the transitional zone between blade and stipe, and the older parts of the thalli (blade) become gradually lost from the tip within several months (Bold and Wynne 1985). Many others, e.g., *Ulva* spp., *Dictyota dichotoma* (Hudson) J.V. Lamouroux, *Spatoglossum crissum* J. Tanaka, *Chondrus* spp., *Gloiopeltis furcate*, and *Ahnfeltiopsis paradoxa*, have life histories with alternation between isomorphic macroscopic generations. However, the growth period of a generation is generally less than 6 months as mentioned above.

A few taxa, e.g., *Codium* spp. and *Sargassum* spp., lack alternation of generations in their life history. Among them, *Sargassum horneri* is a winter-spring annual, and *S. yamadae* is a perennial species growing during a spring–summer term. Therefore, most of the macroalgae (and the tissues constituting their thalli) collected later than autumn 2011 grew by absorbing ambient nutrients including Cs

Fig. 15 Schematic presentation of growth period (phenology) of representative marine macroalgae at Iwaki and its vicinity



after a rapid decline of the radionuclide concentrations in seawater. However, it is noteworthy that the levels of ^{134}Cs as well as ^{137}Cs have remained more or less stable since winter 2012, and were still detectable in spring 2013 in most marine macroalgae (ca. 8–140 Bq kg^{-1} for ^{137}Cs in dried specimens). Considering the CF values of Cs in marine macroalgae, i.e., 8–50 in our study and 50 in the IAEA report, the ^{137}Cs level in ambient seawater of marine macroalgae habitats was considered to have retained at the minimum the level of ca. 0.02–0.3 Bq L^{-1} (based on CF 50). The detection limit of ^{134}Cs and ^{137}Cs in seawater differs remarkably depending on analytical facilities: ca. 0.0008 Bq L^{-1}, http://www1.kaiho.mlit.go.jp/KANKYO/press/press20121106.pdf; 0.025 Bq L^{-1}, http://www.env.go.jp/guide/budget/h25/h25-gaiyo/011.pdf; and 1.2 Bq L^{-1}, http://radioactivity.nsr.go.jp/ja/contents/8000/7638/24/278_i_0531.pdf. The concentration in seawater (0.02–0.3 Bq L^{-1}) is critical for the direct detection of radionuclides in some analytical facilities.

It has been suggested that marine macroalgal metal loads can be used as markers to track the geographical distributions of the metal concentrations in coastal seawaters, e.g., *Ulva* spp. (Caliceti et al. 2002; Haritonidis and Malea 1999), *Undaria pinnatifida* (Yamada et al. 2007). Similarly, the bio-monitoring of coastal ^{137}Cs using seaweeds must be very useful because marine macroalgae in general are shown to avidly accumulate ^{137}Cs in their tissues (CF = ca. 8–50), whereas they grow rather rapidly and, hence, turnover rapidly, so that they exert no influence of bioconcentration through the food chain.

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