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Natural and Fukushima-derived radioactivity in macroalgae and mussels along the Japanese shoreline

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Abstract

Following the failure of the nuclear power plant in Fukushima Prefecture in March 2011, peer-reviewed publications describing radioactivity levels in organisms inhabiting coastal environments are scarce. This paper reports on elevated levels of ^{134}Cs and ^{137}Cs in macroalgae and mussels (up to $\sim 800 \text{ Bq kg}^{-1}$ dry wt.) in June 2011. Cs concentrations in biota sampled in early June 2011 were higher in areas south of Fukushima than sampled in the last third of the month north of Fukushima. Radioactivity from $^{134+137}\text{Cs}$ in organisms south of Fukushima were comparable to or lower than that from the naturally occurring ^{40}K in the same samples. While ^{210}Pb and ^{210}Po concentrations were generally lower than these other radionuclides, ^{210}Po as an α -emitter is more significant from a radiological viewpoint than γ -emitters as it can inflict greater biological damage. By applying known bioconcentration factors of Cs in biota, measured biota concentrations of Cs were also used to estimate Cs concentrations in coastal seawater to be in the range of 10^2 – 10^3 Bq m^{-3} . These estimates show that 3 months after the accident and maximal release of radioactive Cs, levels of Cs persisted in coastal waters, although at levels that were two orders of magnitude lower than at the time of release. These June coastal seawater Cs levels were four orders of magnitude above Cs concentrations off Japan prior to the Fukushima disaster.

1 Introduction

Following the devastation caused by the earthquake and tsunami on 11 March 2011, the Japanese public was left with anxiety regarding radioactivity release from the damaged Fukushima Daiichi nuclear power plant (NPP). Radioactivity released onto land led to contamination in vegetable produce, dairy, and meat products, and similarly release into the coastal ocean led to contamination of seafood. Recognizing public concerns, the Ministry of Agriculture, Forestry and Fisheries (MAFF) initiated a monitoring program that set allowable radioactivity levels in seafood to 500 Bq kg^{-1} wet weight

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(wet wt., later lowered to 100 Bq kg^{-1} wet weight) for radioactive cesium (i.e., sum of ^{134}Cs and ^{137}Cs) (MAFF). Post-Fukushima academic studies have either attempted to predict consequences of the fallout for human and animal health (Kryshev and Sazykina, 2011; Garnier-Laplace et al., 2011; Murakami and Oki, 2012) or focused on estimating the quantities of released radioactivity and their atmospheric and oceanic dissipation (Buesseler et al., 2011, 2012; Tsumune et al., 2011; Yoshida and Kanda, 2012; Masson et al., 2011). However, peer-reviewed studies of radioactive concentrations in post-Fukushima marine animals and plants remain surprisingly scarce. Apart from detailed radioactivity measurements in small fish and crustaceans collected offshore during an international survey in June 2011 (Buesseler et al., 2012), peer-reviewed reports of radioactivity in economically important coastal organisms (e.g., macroalgae, bivalves) are missing. However, TEPCO, MEXT and MAFF have used the internet as an outlet for continuous publication of radioactivity measurements in sediments, water and biota. While there is undeniably a value to a stream of data published on the internet webpage, this approach is missing key information on methodology and therefore missing quality assurance (MEXT; MAFF; TEPCO).

The goal of this study was to quantify June 2011 levels of Fukushima-derived γ -emitting cesium isotopes (i.e. ^{134}Cs , ^{137}Cs) in mussels and macroalgae, collected from coastal locations north and south of the Fukushima NPP (Fig. 1). A second goal was to put these levels in the context of naturally occurring radioactivity due to γ and β -emitting potassium (^{40}K and ^{210}Pb) and α -emitting polonium (^{210}Po) isotopes present in these organisms, which is directly relevant to assessing any increased radioactive dose for seafood consumers.

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2 Material and methods

2.1 Sample collection

Samples of green (*Ulva* sp.) and brown (species were not identified) macroalgae as well as mussels (*Mytilus galloprovincialis*; average shell length 45.0 ± 3.8 mm) were collected by hand along several Pacific beaches south (2 June 2011) and north of Fukushima NPP (21 June 2011) (Fig. 1). Collection times were different for the northern and southern locations due to logistical capabilities. Southern sampling sites, i.e., Isohara Port (36.81° N, 140.76° E), Nakoso beach and port (36.86° N, 140.79° E and 36.83° N, 140.80° E, respectively), and Cape Misaki (36.94° N, 140.91° E) were approximately 70 km south of Fukushima NPP, while northern sites Odonohama (38.28° N, 141.05° E) and Shichigama (38.29° N, 141.07° E) beaches, were approximately 30 km north of Fukushima NPP. Samples were sealed in plastic bags and shipped to Stony Brook University in New York in coolers with dry ice (-50° C).

2.2 Sample preparation

In the laboratory, mussel shells were separated from soft tissue, since only the latter was assumed to have accumulated radioactive Cs (Ke et al., 2000). Mussel tissue and macroalgae were placed into individual plastic containers and stored at -80° C for 24 h prior to freeze drying. All samples were then placed inside fast-freeze flasks and connected to a Labconco FreeZone Benchtop Freeze Dry System that was cooled to -52° C, and pressurized to 0.12 mbar (Buessler et al., 2012). After freeze drying, samples were ground with a ceramic mortar and pestle to obtain homogeneously distributed samples that varied in dry weight (dry wt.) from 3.1 to 79.1 g. Samples were stored in straight-side polypropylene 120-mL jars (Nalgene), which had been efficiency-calibrated to their specific geometry.

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2.3 ^{134}Cs , ^{137}Cs and ^{40}K analysis

We used a planar, low energy germanium detector (Canberra model GLP 3830) and Genie 2000 software (Canberra) to measure and analyze the spectrum of gamma emissions in our samples. Average counting time was 27 h, and the average counting uncertainties were 4 % for ^{134}Cs and ^{137}Cs , and 8 % for ^{40}K . The detector efficiency was determined as previously described by Buessler et al. (2012) for individual radioisotopes with varying geometries as follows: 1.7–3.5 % for ^{134}Cs (at 604.7 keV peak), 1.6–3.2 % for ^{137}Cs (at 661.7 keV peak), and 0.8–1.7 % for ^{40}K (at 1461 keV peak). Calculations of ^{134}Cs , ^{137}Cs , and ^{40}K concentrations in the samples assumed negligible self-absorption of gamma rays for these isotopes, given that they all emit at energies > 200 keV (Hurtado et al., 2007). Correction for coincidence summing was omitted as a low efficiency planar detector was used for sample analyses. Measured ^{134}Cs and ^{137}Cs were decay-corrected to the date of sample collection (taking into consideration their half lives of 2.07 and 30.08 yr, respectively).

2.4 Analyses of ^{210}Pb and ^{210}Po

Small sub-samples (0.5–1.0 g) were spiked with known amounts of ^{209}Po (0.07 Bq) and stable Pb (0.1 mg) as internal tracers of the radiochemical analysis. After the addition of tracers, sub-samples were acid-digested at 80–90 °C on a hot plate for 10–12 h using 65 % HNO_3 (30–40 mL) and a few drops of H_2O_2 . Digested samples were filtered (Albet Quantitative Filter Paper ashless hardened DP 1507), non-digested residuals discarded, and samples reconstituted to 80 mL of 1N HCl after evaporation to dryness. Samples were heated to 70 °C and ascorbic acid added to reduce Fe(III). The polonium isotopes were plated onto silver disks at 70 °C for 8 h while stirring. Polonium emissions were counted by alpha particle spectrometry using silicon surface barrier and PIPS detectors (EG & Ortec and Canberra). Upon plating and to assure the complete removal of polonium, the solutions were converted to 9 M HCl and passed through ion exchange columns (resin type Dowex 1 × 8, 100–200 mesh). Aliquots of each sample were taken

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before and after this step to determine the chemical yield for Pb by atomic absorption spectrometry. The solutions were re-spiked with ^{209}Po and stored for several months to attain sufficient ingrowth of ^{210}Po from ^{210}Pb . After 4–6 months, polonium isotopes were plated again onto silver disks and counted. Activities of both ^{210}Pb and ^{210}Po at the time of sample collection were calculated after appropriate in-growth and decay corrections (Church et al., 2013).

3 Results and discussion

3.1 Cesium in macroalgae and mussels

All samples of post-Fukushima mussels and macroalgae contained ^{134}Cs (range: 51 to 393 Bq kg $^{-1}$ dry wt.) and ^{137}Cs (range: 60–463 Bq kg $^{-1}$ dry wt., Table 1). Measured ^{137}Cs concentrations in mussels and macroalgae prior to the Fukushima accident (background) were two to three orders of magnitude lower. For example, brown macroalgae collected off the Japanese coast in 2007–2008 had ^{137}Cs concentrations of (0.038 to 0.058 Bq kg $^{-1}$ ww, or 0.14–0.21 Bq kg $^{-1}$ dw assuming wet/dry ratio of 0.27; Morita et al., 2010; Carvalho, 2011), and mussels (*Mytilus edulis*) collected from the southwestern Korean Sea in 1988 had 0.04 Bq kg $^{-1}$ dry wt. In the case of the shorter lived ^{134}Cs isotope (half-life = 2.07 yr), no other sources than the Fukushima accident could have produced the detected ^{134}Cs levels in mussels and algae. In addition, the ratio of ^{134}Cs : ^{137}Cs in our macroalgae samples was approximately 1 (mean \pm SD = 0.85 \pm 0.003 and 0.88 \pm 0.10 for the northern and southern sampling locations, respectively), consistent with ratios found in marine plankton (Buesseler et al., 2012) and similar to measured concentrations in the water following the radioactive discharge from the Fukushima NPP (Buesseler et al., 2011; MEXT; Aoyama et al., 2012).

Furthermore, our analyses revealed consistently lower radiocesium levels (i.e., the sum of ^{137}Cs and ^{134}Cs) in samples from northern locations than those in the southern locations. However, even in the more contaminated southern locations none of the

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5 samples exceeded the 500 Bq kg^{-1} wet wt. safety limit. Differences in Cs contamination between the two regions were greatest for mussels. *M. galloprovincialis* from the site north of the Fukushima NPP contained 7.5 times less radiocesium than southern samples (north: mean of 110 Bq kg^{-1} dry wt.; south: mean of 822 Bq kg^{-1} dry wt.). Green macroalgae from northern locations contained on average 141 Bq kg^{-1} dry wt. of radiocesium, which was 5.7 times less than in the southern sample (804 Bq kg^{-1} dry wt.), while brown macroalgae from the south contained approximately twice as much radiocesium as northern samples. Given that Cs uptake by macroalgae is proportional to the Cs concentrations in water (Gutknecht, 1965), we conclude that radiocesium levels were indeed higher in coastal waters south of Fukushima than in waters north of Fukushima at the time of sampling, likely due to the 19 day lag between sampling at these locations. Indeed, MEXT-online reports show a $\sim 30\%$ decline in ^{137}Cs seawater concentration at Dai-ichi within this time interval (i.e., 2–21 June 2011). Some of the difference in ^{137}Cs concentrations in biota collected at these two time points (i.e. the ^{137}Cs concentration difference between biota collected north on 21 June 2011 vs. those collected south on 2 June 2011) can be accounted for by the general decline of radioactivity at its discharge. Any further difference could be accounted for by the southward shore current along the Pacific coast of Honshu (Buesseler et al., 2012).

20 Cs concentrations in biota can be combined with published concentration factors (CF, i.e., the wet tissue to water activity ratio (L kg^{-1})) to estimate Cs concentrations in the water, assuming that the organisms were in equilibrium with ambient water (IAEA, 2004). Given that the biological half-life of Cs in green and brown macroalgae, and in mussels are 5–8 and 8 days, respectively (Gutknecht, 1965; Dahlgaard, 1980), it would appear that the assumption of equilibrium was met.

25 While CFs might be useful in approximating the concentration levels in the water, it is important to realize that these approximations come with an increased chance of error. For example, CFs of ^{137}Cs in various brown macroalgae varied up to 100% and in mollusks up to 67% in a study following radioactive waste disposal in Arctic seas (Fisher et al., 1999). The variation of CF values of ^{137}Cs in green mussels has been

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also related to body size, water pumping rate, and salinity (Ke et al., 2000). Due to observed variability of ^{137}Cs CFs in algae and mussels, which however is smaller for an individual species, estimates of ^{137}Cs concentrations in seawater based on literature CFs are therefore expected to fall within 4-fold of directly measured values.

By applying our measured Cs concentrations in mussels and using a Cs wet wt. CF for mussels as 60 (IAEA, 2004), we estimate total ambient radiocesium levels in northern and southern coastal sampling sites in June 2011 to have ranged from $2.9 \times 10^2 \text{ Bq m}^{-3}$ to $22.1 \times 10^2 \text{ Bq m}^{-3}$, respectively (Table 2). Estimates for seawater ^{134}Cs concentrations using our mussel concentrations of ^{134}Cs were 1.4×10^2 north of Fukushima NPP and 9.9×10^2 – $10.5 \times 10^2 \text{ Bq m}^{-3}$ south of Fukushima NPP (Table 2). Reported CF values for green and brown algae are 60 and 34, respectively (IAEA, 2004). Using these CFs and a dry to wet weight ratio of 0.18 for green algae and 0.27 for brown algae (Carvalho, 2011), we estimate seawater ^{134}Cs concentrations of 1.8×10^2 – $2.0 \times 10^2 \text{ Bq m}^{-3}$ north of Fukushima and $11 \times 10^2 \text{ Bq m}^{-3}$ south of Fukushima based on green algae measurements (Table 2). Brown algae-based measurements estimated ^{134}Cs concentrations of 6.4×10^2 – $10.1 \times 10^2 \text{ Bq m}^{-3}$ north of Fukushima and $19 \times 10^2 \text{ Bq m}^{-3}$ south of Fukushima (Table 2). Green algae-based estimates of ^{137}Cs in seawater north of Fukushima are 2.2×10^2 – $2.5 \times 10^2 \text{ Bq m}^{-3}$ and $13.1 \times 10^2 \text{ Bq m}^{-3}$ south of Fukushima, while brown algae-based estimates of ^{134}Cs in seawater north of Fukushima are 6.9×10^2 – $12.1 \times 10^2 \text{ Bq m}^{-3}$ and $22.4 \times 10^2 \text{ Bq m}^{-3}$ south of Fukushima (Table 2). These estimated ^{137}Cs concentrations in coastal seawater are generally about 2 orders of magnitude lower than the maximum ^{137}Cs concentrations recorded by TEPCO and presented by MEXT in mid April at 30 km offshore Fukushima ($\sim 200\,000 \text{ Bq m}^{-3}$; Buessler et al., 2011). It is practically impossible to relate our lower estimates at northern locations to those reported by MEXT, because ^{137}Cs detection limits declared in these reports were high (i.e., 6 Bq L^{-1} or $6 \times 10^3 \text{ Bq m}^{-3}$), therefore any Cs at concentrations lower than $6 \times 10^3 \text{ Bq m}^{-3}$ would not have been detected. ^{137}Cs concentrations predicted by us for the southern location on 2 June 2011 are at least 7-fold lower than $15 \times 10^3 \text{ Bq m}^{-3}$ reported online

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for seawater collected on 2 June 2011 near our sampling site (MEXT). ^{134}Cs concentrations measured in coastal seawater Onahama (Aoyama et al., 2012) near the southern sampling site in the present study, show up to 3-fold higher than the maximum CF-estimated values in the present study (Table 2). This agreement between our CF-estimation of seawater Cs and an actual measurement is consistent with an up to 4-fold error (see above).

3.2 ^{40}K , ^{210}Pb and ^{210}Po in macroalgae and mussels

Naturally occurring radioisotopes ^{40}K , ^{210}Pb and ^{210}Po were detected for all samples. Highest concentrations of ^{40}K were present in brown macroalgae (i.e., 842–1369 Bq kg⁻¹ dry wt.; Table 1), lower in green macroalgae (i.e. 426–511 Bq kg⁻¹ dry wt.; Table 1), and lowest mussel soft tissue (300–353 Bq kg⁻¹ dry wt.; Table 1). Concentrations of ^{210}Pb varied by an order of magnitude for brown algae (0.9 ± 0.10 – 8.3 ± 0.40 Bq kg⁻¹ dry wt.; Table 1), but were similar for green algae (5.1 ± 0.36 – 8.8 ± 0.48 Bq kg⁻¹ dry wt.; Table 1). ^{210}Pb in mussels varied by 6-fold (Table 1). Concentrations of ^{210}Po were similar among brown algae samples and ranged from 10 to 30 Bq kg⁻¹ dry wt. with an average of 19.5 ± 7.4 Bq kg⁻¹ dry wt. (Table 1), but they were variable among the green algae and mussels. ^{210}Po concentration in green algae ranged from 18.1 to 272 Bq kg⁻¹ dry wt., and from 14.0 to 300 Bq kg⁻¹ dry wt. for mussels.

The variability in the ^{40}K and ^{210}Po levels for brown macroalgae (Table 1) is not unusual (Thompson et al., 1982; Uğur et al., 2002; Goddard and Jupp, 2001). Thompson et al. (1982) showed that for just one species of a brown macroalga, *Fucus vesiculosus*, collected from the Irish Sea, ^{40}K concentrations varied 2-fold. Another study showed that ^{40}K concentrations can vary within algal divisions (Goddard and Jupp, 2001). In that study ^{40}K concentrations showed a 44-fold variation and ranged from 100–4400 Bq kg⁻¹ dry wt. in green algae, and 125–5400 Bq kg⁻¹ dry wt. in brown algae (Goddard and Jupp, 2001). Published ^{210}Po concentrations for green algae are scarce but one study reported ^{210}Po concentration of 15 Bq kg⁻¹ dry wt., which falls within the

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range of ^{210}Po concentration in green algae in this study. As in the present study, variability for ^{210}Po concentrations in mussels has been reported. For example, ^{210}Po concentrations showed 26-fold (52–1300 Bq kg⁻¹ dry wt.) variation for *Mytilus galloprovincialis* from various coastal locations of the Aegean sea (Uğur et al., 2002). Wildgust et al. (1998) suggested that variation of ^{210}Po concentrations in organisms could be linked to environmental conditions such as salinity or temperature, and to biological parameters such as the reproductive cycle phase.

From a human-health perspective, it is important to assess levels of natural radionuclides such as ^{40}K and ^{210}Po as these radionuclides are always present in seafood. Because ^{40}K is the dominant γ -emitter in the ocean and in marine biota, its evaluation as a potential radioactive dose contributor is important. ^{210}Po releases α -particles with its decay, and α emissions are 20 times more damaging to cells than γ and β emissions (Eckerman et al., 2012). In fact ^{210}Po has been shown as the radionuclide producing by far the greatest radioactive dose in marine biota, as it also is the greatest radioactive dose contributor to seafood consumers (Aarkrog et al., 1997). Therefore even though its radioactivity is lower, doses from the naturally occurring ^{210}Po in biota, including those from the Japanese shore (Table 1), can be of greater radiological importance than $^{134+137}\text{Cs}$.

4 Summary

This is the first report in the peer-reviewed literature on the levels of anthropogenic and natural radioactivity in the biota from coastal Japan following the Fukushima NPP failure. At the time of sample collection in June 2011, concentrations of Fukushima-derived ^{137}Cs and ^{134}Cs were elevated, and when combined (i.e. $^{134+137}\text{Cs}$) they did not exceed the 500 Bq kg⁻¹ wet wt. recommended safety level for seafood consumption. Biota concentrations of Cs were higher in the southern locations than in the north, likely due to the 19-day lag between sampling and the southward flow of coastal currents (Buesseler et al., 2012). Concentrations of individual (i.e., ^{210}Po and ^{40}K) natural

radionuclides showed levels that were 20-fold lower or 5-fold higher than those of $^{134+137}\text{Cs}$. A concentration factor based approach estimated that radioactive Cs concentrations in coastal waters at the northern location in the second half of June could range from 2.9×10^2 – $22.2 \times 10^2 \text{ Bq m}^{-3}$, and at the southern locations earlier on was either similar or up to 14-fold higher.

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Table 1. Dry weight based concentrations of ^{134}Cs , ^{137}Cs , ^{40}K and ^{210}Po in macroalgae and mussels sampled in June 2011 along the east coast of Japan. Activities were decay-corrected to the date of sample collection, except for ^{40}K given its very long half life (1.25×10^9 yr). σ – is the propagated counting error (Bq kg^{-1}); σ^* – results from the propagated counting error associated with measurement of ^{134}Cs and ^{137}Cs .

Sample type	NPP*	Location	Lat °N	Long °E	^{134}Cs Bq kg^{-1}	$\pm\sigma$	^{137}Cs Bq kg^{-1}	$\pm\sigma$	$^{134}\text{Cs}; ^{137}\text{Cs}$ $\pm\sigma^*$	^{210}Po Bq kg^{-1}	$\pm\sigma$	^{210}Pb Bq kg^{-1}	$\pm\sigma$	^{40}K Bq kg^{-1}	$\pm\sigma$
Brown algae	N	Shichigahama Beach	38.29	141.07	108.2	5.1	102.7	5.1	1.05 0.07	16.3	3.2	1.7	0.22	842	55.4
		Odonohama Beach	38.28	141.05	80.2	5.4	86.9	5.8	0.92 0.09	29.6	3.6	4.6	0.26	880	67.9
	S	Nakoso Port			124.1	6.2	143.2	6.6	0.87 0.06	10.3	2.2	0.9	0.10	1189	73.1
					126.7	4.4	152.3	4.8	0.83 0.04	23.7	4.4	8.3	0.40	1369	56
Green algae	N	Shichigahama Beach			238.7	4.7	282.2	5.1	0.85 0.02	17.8	3.2	2.2	0.15	1296	44
					66	5	84.3	4.9	0.78 0.07	18.1	2.7	5.1	0.36	511	51.6
	S	Odonohama Beach			59.6	2.2	72.7	2.4	0.82 0.04	115	6.9	5.4	0.28	509	25.5
					36.94	140.91	369.1	5.1	434.5	5.6	0.85 0.02	272	21.7	8.8	0.48
Mussels	N	Cape Misaki			51.3	2.1	59	2.2	0.87 0.05	14	2.9	1.3	0.11	353	21.1
					36.94	140.91	392.6	5.6	463.1	6.1	0.85 0.02	28.8	3.6	4.5	0.27
	S	Odonohama Beach			372.1	8.8	417	9.1	0.89 0.03	300	21	7.7	0.77	334	36.3

* North (N) or south (S) relative to the Fukushima nuclear power plant (NPP).

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Table 2. Estimates of ^{134}Cs and ^{137}Cs concentrations in coastal seawater at the locations north and south of the Fukushima NPP based on concentration factors (CFs; L kg^{-1}) derived from the literature (IAEA, 2004). Ranges are provided based on the maximum and minimum concentration values for the biota.

Sample type	NPP ^a	Dry/wet conversion factor	CF	Biota				Seawater			
				^{134}Cs		^{137}Cs		^{134}Cs		^{137}Cs	
				Bq kg ⁻¹ dry wt				× 10 ² Bq m ⁻³			
Brown algae	N	0.27	34	80	127	87	152	6.4	10.1	6.9	12.1
Green algae		0.18	60	60	66	73	84	1.8	2.0	2.2	2.5
Mussels		0.16	60	51 ^b		59 ^b			1.4 ^b		1.6 ^b
Brown algae	S	0.27	34	239 ^b		282 ^b		19.0 ^b		22.4 ^b	
Green algae		0.18	60	369 ^b		435 ^b		11.1 ^b		13.1 ^b	
Mussels		0.16	60	372	393	417	435	9.9	10.5	11.1	11.6

^a Indicates one value instead of a range when concentrations were available for only one sample.

^b North (N) or south (S) relative to the Fukushima nuclear power plant (NPP).

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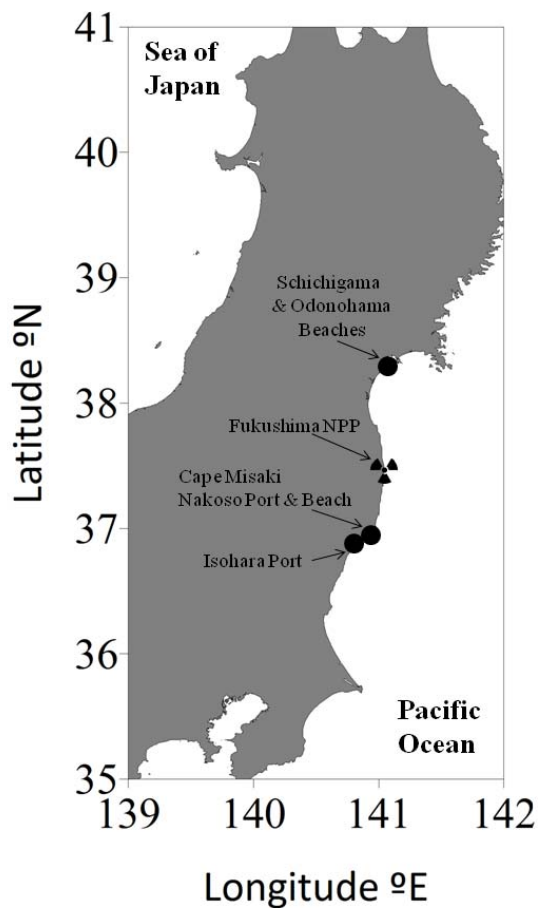


Fig. 1. Sampling locations for mussels and macroalgae along the east coast of Honshu Island of Japan.

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