



Radionuclides in marine macroalgae from Amchitka and Kiska Islands in the Aleutians: establishing a baseline for future biomonitoring

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Abstract

Levels of radionuclides in seven species of marine brown algae and *Ulva* were determined to establish a baseline for the Northern Pacific Ocean/Bering Sea (Aleutian Islands). There were differences in levels among algal species and locations (Amchitka Island vs Kiska Island). No values were above the minimum detectable activity (MDA) level for ¹³⁷Cs, ¹²⁹I, ⁶⁰Co, ¹⁵²Eu, ⁹⁰Sr, and ⁹⁹Tc. There were interspecific differences in some radionuclides: *Ulva lactuca* (= *Ulva fenestrata*) had the highest levels of ²⁴¹Am, *Alaria fistulosa* had the highest levels of ^{239,240}Pu, and *Fucus distichus* (= *Fucus gardneri*) had the highest levels of ²³⁴U, ²³⁵U, and ²³⁸U. However, levels of all radionuclides were generally low and near the MDA for all isotopes. Although Amchitka Island had higher levels of ^{239,240}Pu than Kiska, the differences were very small and not significant biologically. The data indicate that algae can be useful bioindicators of actinides

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because they accumulate them at very low environmental levels, allowing them to provide early warning of any potential seepage of radionuclides into the marine environment. Further, the data indicate that some species (the intertidal *Fucus*) are better accumulators than others, and these should be used as bioindicators in future monitoring schemes.

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1. Introduction

Radiation protection and assessment of radionuclides in humans and the environment are important areas of public interest and public policy. Values of sustainability, transparency, and public participation are key elements of the public's view of environmental problems (Omenn, 2001), and their management. These values all require an understanding of baseline radionuclide levels in media and biota before risk and safety can be evaluated. Baselines for radionuclides should be available for any site where radionuclide wastes are stored and maintained, or where residual wastes remain. If the baseline data are low, they can serve to assure the public that the foods they eat from the region are safe, and that the ecosystem is not impaired. Baseline data are essential for future biomonitoring to assess the status and trends of radionuclides within a given ecosystem. Determining baselines is particularly important for nuclear waste sites or those where nuclear tests were detonated or nuclear accidents occurred.

Anthropogenic radioactivity comes from fallout from atmospheric explosions since 1945 and from emissions produced by nuclear and radioactive facilities (Baeza et al., 1994; Cooper et al., 1998), discarded nuclear wastes (Fisher et al., 1999), as well as other nuclear accidents (Aumento et al., 2005). The disposal of large quantities of radioactive wastes in the Arctic Seas by the former Soviet Union has prompted interest in radionuclides in the Bering Sea ecosystem (Fisher et al., 1999).

When the Cold War ended, the United States Department of Energy (DOE) was faced with a shift in priority from weapons development and production to the environmental management of the "legacy wastes" remaining from over 40 years of nuclear activities. The DOE's nuclear weapons complex has about 5000 facilities located at 16 major sites, and more than 100 smaller sites (Crowley and Ahearne, 2002). Over a hundred of the DOE sites contain chemical and radiological wastes generated by the production of nuclear weapons (DOE, 2000). The DOE's environmental management task averaged about \$6 billion a year in the 1990s, and represents 20% of the world's environmental remediation market (Sink and Frank, 1996). Some DOE sites cannot be cleaned up, but are scheduled for closure with the nuclear wastes in place. These include Amchitka Island which was the site of three underground nuclear test shots from 1965 to 1971. Surface contamination has been cleaned up, and the three underground cavities, 700–2300 m below the surface, are considered secure. Stewardship responsibility will be transferred to DOE's Office of Legacy Management in late 2006. Significant quantities of radionuclides remain in the test shot cavities, either incorporated into solid phases (e.g., amorphous and crystalline) formed during subsurface cooling after the explosion, adsorbed onto natural mineral surfaces (e.g. clay minerals) or dissolved in the pore-water. Thus, the potential exists for radionuclides from the shot cavities to slowly migrate with groundwater to the marine environment.

This potential for transport necessitates the establishment of baseline radionuclide levels in the surrounding marine environment and long-term stewardship.

The purpose of this study was to examine the levels of radionuclides in eight species of marine algae collected from Amchitka Island and Kiska Island (the reference site) in the Aleutians, Alaska. We tested the null hypothesis that there were no differences in radionuclide levels among species and between islands. We were particularly interested in establishing a baseline for future comparison, and in determining whether the levels found at Amchitka were similar to those found in other uncontaminated regions of the Northern Hemisphere. This work is part of a larger multi-disciplinary project by the Consortium for Risk Evaluation with Stakeholder Participation (CRESP) to provide the information to assure the protection of human health and the environment, and to provide a baseline for future monitoring of Amchitka (Burger et al., 2005, 2006a; Powers et al., 2005, 2006).

2. Materials and methods

2.1. Study sites

Marine macroalgae were collected from the marine waters around Amchitka Island (Rat Island Group; 51°N lat; 179°E long) and Kiska Island (51°N lat; 177°E long) in the Aleutian Chain in the Northern Pacific (Fig. 1). Only Amchitka Island experienced underground nuclear tests, and Kiska was used as a reference site. Both islands also saw military activity during World War II. Although the collection areas on the two islands were approximately 150 km apart, algae are sedentary and do not migrate appreciably. Kiska was selected after extensive consultation with stakeholders. Both islands are tundra-like, without trees. The rolling hills and mountains contain primarily grasses. The marine benthic resources around Kiska Island have not been described extensively. However, Kiska Island contains many of the same terrestrial and benthic environments as Amchitka (Burger et al., 2006a,b), with many marine species in

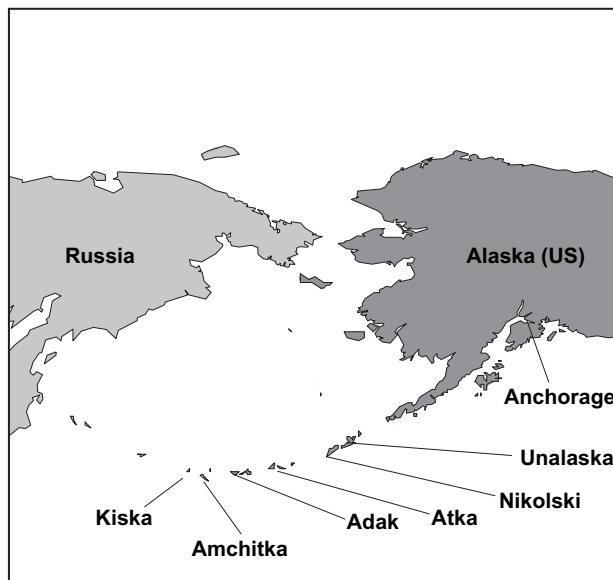


Fig. 1. Map showing the location of Amchitka and Kiska Islands in the Aleutian chain of Alaska.

common. Both islands are bordered on the south by the North Pacific and on the north by the Bering Sea (Fig. 1).

2.2. Site history and responsibility

Amchitka Island (Fig. 1), the scene of three underground nuclear tests in 1965, 1969 and 1971, remains the responsibility of DOE for protection of human health and the environment from adverse impacts as a consequence of the nuclear tests. Amchitka Island is unusual among DOE-contaminated sites because of its remoteness, depth of the contamination, and importance of its ecological resources and seafood productivity of the surrounding marine ecosystem (Burger et al., 2005; Merritt and Fuller, 1977). It is believed that most of the radioactive material from the Amchitka test shots is trapped in the vitreous matrix created by the intense heat of the blast, and is therefore permanently immobilized, but this is an assumption and DOE's models indicate that breakthrough into the sea will eventually occur (DOE, 2002a,b). Moreover, this information is classified and unavailable to stakeholders. While the DOE detonated above-ground tests on other remote oceanic islands, Amchitka is the only one where underground tests were made, making it far more difficult to assess and technically impossible to remove the residual radiation.

Amchitka Island was designated a wildlife preserve in 1913, but was released for military activity during World War II (Kohlhoff, 2002). Today it is part of the Alaska Maritime National Wildlife Refuge system under the aegis of the U.S. Fish & Wildlife Service. At the time of the underground nuclear test shots, there was considerable controversy about testing at Amchitka because of the potential health risks to the local Aleuts, the serious damage to the marine ecosystem, and the possible generation of tsunami activity (Greenpeace, 1996; Kohlhoff, 2002). Although there was some release of radiation to the surface, the leaks were not considered to pose serious health risks at the time (Faller and Farmer, 1998; Seymour and Nelson, 1977). The controversy about radionuclide contamination continued (Kohlhoff, 2002), with increasing concern about the possibility of subsurface transport of radionuclides from the three cavities to the marine environment (DOE, 1997), particularly in light of it being one of the most active and dynamic subduction zones on earth (Eichelberger et al., 2002).

Kiska Island did not experience any underground nuclear tests, but both the Japanese and later the U.S. occupied the island during World War II. It has not been occupied since that time.

2.3. Protocol

Under appropriate state permits, algae were collected from both Amchitka and Kiska Islands from late June to July 2004. Species collected were *Ulva lactuca*, *Fucus distichus*, *Alaria nana*, *Alaria fistulosa*, *Laminaria bongardiana*, *Laminaria saccharina*, *Laminaria yezoensis* and *Cymathere triplicata*. Taxonomic genus and species treatments vary. The *Ulva* are sometimes separated as *Ulva fenestrata*, the *Fucus* as *Fucus gardneri*, and *C. triplicata* was formerly considered in *Laminaria*. These were some of the commonest species available, have a wide distribution, and are also reported as subsistence foods of the Unangan (Aleut) people. For the purposes of analysis, due to small samples sizes, we combined the *Laminaria* and *Cymathere* species because there were no significant differences in radionuclide levels among them. All specimens were tracked from field collection to their ultimate destination with chain of custody forms. Our overall protocol was to collect five algae plants from a given site for compositing. Benthic invertebrates, fish and birds were collected at the same time (Burger et al., 2006c, in press).

In the shipboard laboratory, algae were scanned with a handheld counter for gross alpha, gamma and beta radiation levels; no radiation over background was encountered in any sample. Algae were processed, measured (length), cut into segments, packaged and labelled. Some of the *Alaria* and *Laminaria* were several meters long, requiring the sampling of small parts of their blades. We removed samples from the middle of the blade (for *Alaria* and *Laminaria*) and from the middle of the thallus for *Ulva* and *Fucus*. Samples were then immediately frozen for later analysis. Radionuclide analyses were conducted on composites of five individuals each.

Samples were washed vigorously with tapwater to remove both the seawater and any external biota, and were rinsed again with deionized water before processing. To replicate culinary practices, we did not use solvents or detergents to wash samples. Thus radionuclides adsorbed to the mucus surface would be included in our results. Samples were then homogenized in a radio-clean (confirmed by wipe samples) and metal-clean laboratory at Rutgers University, and subsequently analyzed for radionuclides at Vanderbilt University and Idaho National Laboratory (INL). Wipe samples on shipboard and laboratory tabletops and laboratory blenders were below MDA.

Our radionuclide analysis design was based on sample availability and quantity. Detailed analytic and quality assurance methods are published on the CRESO website (INL, 2004; Powers et al., 2005, 2006). Homogenized kelp samples spiked with the target radionuclides were analyzed in each batch. We analyzed radioactive cesium (^{137}Cs), iodine (^{129}I), cobalt (^{60}Co), europium (^{152}Eu), strontium (^{90}Sr), technetium (^{99}Tc), americium (^{241}Am), plutonium (^{238}Pu , $^{239+240}\text{Pu}$), and uranium (^{234}U , ^{235}U , ^{236}U , ^{238}U). Analyses at Vanderbilt and Idaho National Laboratory provided inter-laboratory validation (Powers et al., 2005). Counts were adjusted for background counts, and the minimum detectable activity (MDA) was ± 2 SD background, and back extrapolated to date of collection.

Gamma emitters (^{137}Cs , ^{152}Eu , ^{60}Co) were analyzed using gamma spectroscopy with high purity germanium detectors calibrated to the standard container geometry. ^{99}Tc was “trapped” on an Eichrom TEVA resin to preconcentrate the analyte and remove potential interferences. Samples were stabilized with ammonia, dried and ashed, and then oxidized with nitric acid and hydrogen peroxide, prior to measurement by inductively coupled plasma-mass spectrometry (ICP-MS). The method uses rhenium spikes as a recovery surrogate. For iodine, samples were not extracted. After calibration, ^{129}I was analyzed by low energy photon measurement.

Solid phase extraction was used for selective separation of americium, strontium, plutonium, and uranium using a serial configuration of TRU and TEVA columns. The beta emitter ^{90}Sr was separated by sulphate precipitation, and analyzed by its daughter decay product $^{90}\text{yttrium}$. Actinides (uranium, plutonium, americium) were analyzed radiochemically using co-precipitation with neodymium fluoride (INL, 2002, 2004), followed by alpha spectroscopy.

All values were analyzed on wet weight, and are presented in Bq/kg (both for our samples and literature data). Initially for gamma emitters, we counted 100 g samples for 24 h, but all results were below the MDA. Thus to enhance sensitivity, we also analyzed 1000 g samples for 72 h. MDAs for individual samples for ^{137}Cs ranged from 5.57 to 6.25 Bq/kg for 100 g samples, and 0.18–0.36 Bq/kg for 1000 g samples. Mean MDAs were as follows: ^{60}Co (0.16 Bq/kg), ^{90}Sr (2.15 Bq/kg), ^{99}Tc (0.09 Bq/kg), ^{129}I (0.43 Bq/kg), ^{137}Cs (0.257 Bq/kg), ^{152}Eu (0.27 Bq/kg), ^{241}Am (0.27 Bq/kg), ^{234}U (0.042 Bq/kg), ^{235}U (0.051 Bq/kg), ^{236}U (0.036 Bq/kg), ^{238}U (0.043 Bq/kg), ^{238}Pu (0.032 Bq/kg), $^{239,240}\text{Pu}$ (0.046 Bq/kg).

2.4. Statistical analysis

We used Kruskal–Wallis non-parametric one-way analysis of variance (generating a χ^2 statistic) to examine differences among species, islands and the three test regions on Amchitka (SAS, 1995). To compute means for a given isotope, we used all values above the MDA, and for those below the MDA we used half of the MDA.

3. Results

Levels of all radionuclides were generally low and near the MDA for all isotopes, except the naturally occurring ^{234}U and ^{238}U . Radionuclides differed in levels; there were no values above the minimum detectable activity (MDA) level for ^{137}Cs ($N = 10$ for 1000 g and $N = 12$ for 100 g samples), ^{129}I ($N = 12$), ^{60}Co ($N = 12$), ^{152}Eu ($N = 12$), ^{90}Sr ($N = 12$), and ^{99}Tc ($N = 12$). There were detectable levels of most actinides in the algae examined, with the levels

Table 1
Levels of actinides in algae from Amchitka and Kiska

Isotope	<i>Ulva</i> ^a N = 12	<i>Fucus</i> ^b N = 14	<i>Alaria nana</i> N = 21	<i>Alaria fistulosa</i> N = 19	<i>Laminaria</i> ^c N = 18	Kruskal–Wallis χ^2 , <i>p</i> value
Am-241 A	(0.075)	(0.035, 0.023)	(0.024, 0.033)			27.6, <i>p</i> < 0.0001
Pu-238 A	(0.024, 0.123)			(0.015)		
Pu-239,240 A		0.031 ± 0.017	0.031 ± 0.018	0.051 ± 0.05	0.020 ± 0.023	28.9, <i>p</i> < 0.0001
U-234 N	0.317 ± 0.121	3.124 ± 1.09	0.986 ± 0.518	1.005 ± 0.557	0.446 ± 0.209	52.3, <i>p</i> < 0.0001
U-235 N		0.147 ± 0.052	0.015 ± 0.015	0.052 ± 0.042	0.044 ± 0.041	43.7, <i>p</i> < 0.0001
U-236 A		(0.044)		(0.022, 0.016)		25.5, <i>p</i> < 0.0001
U-238 N	0.246 ± 0.137	2.72 ± 0.953	0.843 ± 0.437	0.906 ± 0.484	0.431 ± 0.167	55.2, <i>p</i> < 0.0001

Given is the mean (±standard deviation, wet weight) in Bq/kg (values above MDA + half the MDA for those samples falling below the MDA). Where there were few values above the MDA for an isotope, those are listed in parentheses (no statistical test was performed). A = primarily anthropogenic, N = primarily natural.

^a *Ulva lactuca*, sometimes separated as *Ulva fenestrata*.

^b *Fucus distichus*, sometimes separated as *Fucus gardneri*.

^c Includes *Cymathere triplicata*.

of the naturally occurring ²³⁴U and ²³⁸U being higher than the levels of americium and plutonium isotopes (Table 1).

We tested the hypothesis that there were no interspecific differences in actinide levels (Table 1), and found that there were interspecific differences in levels of ²⁴¹Am, ²³⁹⁺²⁴⁰Pu, ²³⁴U, ²³⁵U, and ²³⁸U. *Ulva* had the highest levels of ²⁴¹Am, *A. fistulosa* had the highest levels of ²³⁹⁺²⁴⁰Pu, and *Fucus* had the highest levels of ²³⁴U, ²³⁵U, and ²³⁸U. The uranium isotopes were an order of magnitude higher in *Fucus* compared to the other algae (*Fucus* is intertidal).

Since Amchitka Island was the site of three underground nuclear tests, we wanted to assess whether there was a difference between Amchitka and Kiska (the reference site); thus we tested the hypothesis that there were no differences between the two islands (Table 2). When all algae are considered together, there were no significant differences in the percent of detects for Amchitka and Kiska. Only ²³⁹⁺²⁴⁰Pu showed an island difference, with levels slightly higher (*p* < 0.055) than at Kiska (Table 2).

We then tested the hypothesis that there were no significant differences in actinide levels among the three test shots (Table 3). There were no significant differences in the percent of analyses above the MDA, or in the mean MDAs, for the three test shots (Table 3). We were unable to examine ratio between ²³⁸Pu and ^{239,240}Pu because very few samples had both values above the MDA.

4. Discussion

4.1. Inter-island and interspecific differences

The only inter-island difference was in the mean levels of ²³⁹⁺²⁴⁰Pu from Amchitka. Since the levels of ²³⁹⁺²⁴⁰Pu were low, and the differences were small, they are unlikely to be important biologically, and do not indicate that there is any seepage from the underground nuclear test shots at Amchitka. They do, however, provide a baseline to evaluate any potential seepage in the future. That is, the groundwater models developed by the DOE (2002a) as well as the CRESP geophysical data (Powers et al., 2005) indicated that it was not a matter of if the seepage would occur, but when it would occur. The time frame of release is expected to be

Table 2
Comparison of actinide levels between Amchitka and Kiska for algae

Isotope	Range of reported values	Mean \pm SD	Number of detects (%)
Am-241			
Amchitka	<0–0.035	0.015 \pm 0.008	3 of 57 (5.3%)
Kiska	<0–0.075	0.016 \pm 0.013	2 of 27 (7.41%)
χ^2		0.0 (0.98)	0.15 (0.70)
Pu-239,240			
Amchitka	<0–0.207	0.036 \pm 0.034	14 of 57 (24.6%)
Kiska	<0–0.089	0.023 \pm 0.016	3 of 27 (11.1%)
χ^2		3.69 (0.055)	2.05 (0.15)
U-234			
Amchitka	0.080–4.82	1.168 \pm 1.029	57 of 57 (100%)
Kiska	0.117–5.11	1.067 \pm 1.248	27 of 27 (100%)
χ^2		0.94 (0.33)	0 (>0.99)
U-235			
Amchitka	<0–0.198	0.055 \pm 0.054	25 of 57 (46.9%)
Kiska	<0–0.254	0.042 \pm 0.066	9 of 27 (33.3%)
χ^2		1.57 (0.21)	0.84 (0.36)
U-236			
Amchitka	<0–0.044	0.002 \pm 0.008	3 of 57 (5.3%)
Kiska	<0–0.019	0 \pm 0.004	0 of 27 (0%)
χ^2		0.25 (0.61)	1.47 (0.22)
U-238			
Amchitka	0.077–4.37	1.042 \pm 0.914	57 of 57 (100%)
Kiska	0.058–4.47	0.910 \pm 1.056	27 of 27 (100%)
χ^2		1.39 (0.23)	0 (>0.99)

The means (Bq/kg, wet weight) were calculated using the values above the MDA + half the values above the MDA for values below the MDA. The mean values are compared using the non-parametric Kruskal–Wallis one-way analysis of variance and the proportion of detects is compared using a 2×2 contingency table. Both tests yield a χ^2 value. There were 57 algae analyses for Amchitka and 27 for Kiska.

several hundred years; there are considerable differences in the predicted time of release depending on the models used. Thus it was important to establish baseline radionuclide levels in biota, including algae for future evaluation.

There were significant interspecific differences in the levels of ^{241}Am , $^{239+240}\text{Pu}$, ^{234}U , ^{235}U , and ^{238}U ; of these, the uranium isotopes are naturally occurring, and the others are anthropogenic. For most isotopes *Fucus* had the highest levels, and sometimes were an order of magnitude higher than the other algae species (i.e. ^{234}U , ^{235}U and ^{238}U). Other authors have also reported that *Fucus* is a good accumulator of radionuclides, which contributes to its being useful for removing radionuclides from ecosystems (bioremediation) and as a bioindicator of radionuclide exposure (Douville et al., 2004; Lindahl et al., 2003, 2005; Nawakowski et al., 2004; Wallberg and Moberg, 2002). Interspecific differences in accumulation of radionuclides have been noted for elsewhere, such as the Baltic Sea (Strezov et al., 1996).

Although there are few temporal data from Amchitka Island, Dasher et al. (2002) (also pers comm 8 February 2006) reported that the five samples of *Fucus* they tested for $^{239+240}\text{Pu}$ ranged from 0.04 to 0.07 Bq/kg dry weight (equal to about 0.008–0.01 Bq/kg wet weight). The levels from Amchitka in 2004 ranged from below the MDA to 0.21 Bq/kg (ww), averaging about 0.04 Bq/kg across species. The small sample sizes preclude drawing a conclusion.

Table 3
Plutonium levels, Pu-239,240 (Bq/kg) wet weight, at the three Amchitka test shots

	Milrow	Long Shot	Cannikin	Kruskal–Wallis χ^2 , p value
Total analyzed	18	22	11	
Number > MDA	7	3	4	0.37, $p < 0.83$
MDA for kelp (Bq/kg)	0.044 ± 0.032	0.050 ± 0.025	0.045 ± 0.046	4.04, $p < 0.13$
Actual values > MDA and number of samples analyzed				
<i>Alaria fistulosa</i>	0.207; 0.080 ($N = 4$)	0.131; 0.103 ($N = 8$)	0.041 ($N = 3$)	
<i>Alaria nana</i>	All below MDA; mean of 0.082 ($N = 3$)	All below MDA; mean of 0.064 ($N = 6$)	0.043; 0.035 ($N = 4$)	
<i>Fucus distichus</i>	0.056; 0.052; 0.047; 0.044 ($N = 5$)	0.059 ($N = 5$)	($N = 0$)	
<i>Laminaria/Cymathere</i>	0.073 ($N = 6$)	All below MDA; mean of 0.036 ($N = 3$)	0.063 ($N = 4$)	

Shown are the levels of Pu-239,240 for Kelp at the three test shots. Table indicates no significant differences in either the percent of values above the MDA, or in the mean MDAs at each site.

4.2. Geographic comparisons

The levels of several radionuclides were below the MDA for algae collected at Amchitka and Kiska. In algae at Amchitka, all samples of ^{137}Cs were below the MDAs (which ranged from 0.18 to 0.36 Bq/kg). ^{137}Cs levels for algae from other Northern Hemisphere sites averaged 0.2 Bq/kg, except for the Irish Sea, which averaged 1.97 Bq/kg (Friedlander et al., 2005). These data were computed from several sources (BNFL, 2002–2004; CEFAS, 2003, 2004; Gafvert et al., 2003, 2004; JCAC, 2003, 2004; RPII, 2003, 2004), which have on-going monitoring programs of a range of radionuclides. The Amchitka/Kiska algae levels for ^{137}Cs are thus in agreement with data reported for other Northern Hemisphere locations thought to be uncontaminated, and were below the levels found in algae in the Irish Sea. Of the algae examined for the Irish Sea, *Fucus* had the highest levels (2.17 Bq/kg for ^{137}Cs). Since we also examined *Fucus*, and all levels were below the MDA, we are confident that the levels from Amchitka are well below other sites.

There are relatively few data for actinide levels for kelp, except for the Irish Sea, where Sellafield has conducted regular biomonitoring because of releases from its nuclear reprocessing facility (Cooper et al., 1998), especially of plutonium (Ryan et al., 1999). Below we compare the levels first for the anthropogenic radionuclides, and then the naturally occurring ones. The highest level for ^{241}Am from the present study was 0.59 Bq/kg, compared to a mean of 4.03 Bq/kg for several algae in the Irish Sea; the highest level (for *A. fistulosa*) for $^{239+240}\text{Pu}$ was 0.21 Bq/kg in the present study, compared to a mean of 8.4 Bq/kg for the Irish Sea; the highest level (*Fucus*) for ^{236}U was 0.044 Bq/kg in the present study (there are no comparative data for the Irish Sea). Thus, the maximum levels at Amchitka/Kiska for the anthropogenic radionuclides were at least an order of magnitude lower than the mean levels were for algae in the Irish Sea. The $^{239+240}\text{Pu}$ levels for *Fucus* from Baffin Bay (mean of 0.05 Bq/kg) and Norway (mean of 0.01 Bq/kg) are similar to those found at Amchitka and Kiska (Powers et al., 2005),

and presumably represent background levels. The levels for algae from the Red Sea (mean of 0.33–1.32 Bq/kg) were, however, higher than those found at Amchitka (Sam et al., 1998).

The comparisons are reversed, however, for the naturally occurring radionuclides. The highest average level for ^{234}U was 3.12 Bq/kg (*Fucus*) in the present study, compared to only 0.32 Bq/kg for the Irish Sea; the highest average level for ^{235}U was 0.147 Bq/kg (*Fucus*) in the present study, compared to only 0.02 Bq/kg for the Irish Sea; the highest average level for ^{238}U was 2.72 (*Fucus*) in the present study, compared to only 0.46 Bq/kg for the Irish Sea algae (MARINA, 2004).

Taken together, the comparison indicates that the levels of the anthropogenic radionuclides are at least an order of magnitude higher in algae from the Irish Sea compared with those collected at Amchitka and Kiska; the reverse is true for the naturally occurring uranium isotopes. The former conclusion is not surprising since the Irish Sea is contaminated with radionuclides from the Sellafield nuclear reprocessing facility (Cooper et al., 1998). These differences are not due to water temperature or latitude, or to species (since the comparisons were for the *F. distichus* in both studies; Fisher et al., 1999). Others have noted that naturally occurring radionuclides often contribute more to the effective human dose from marine foods (Robison and Noshkin, 1999), and this would probably be the case with the Amchitka/Kiska data since the naturally occurring radionuclides consistently had a higher percentage of levels above the MDA than did the anthropogenic ones.

Although uranium formed at the time of the Earth's formation about 4.7 billion years ago, it is not uniformly distributed over the surface. Concentrations tend to be higher in rocks of volcanic origin than in sedimentary rocks. Countries of Western Europe are generally uranium poor compared to North America, Australia and parts of South America (USGS, 2006). Thus the contribution of surface erosion to uranium content of the sea would be higher from the relatively rich soils of North America, than in Western Europe (Manuel et al., 1998). Whether such differences could explain an order of magnitude difference of naturally occurring uranium in biota from Amchitka and Kiska than elsewhere requires further study.

4.3. Use of algae as a bioindicator

Algae, especially kelp, have the ability to accumulate radionuclides (Bojanowski and Pempkowiak, 1977; Georgescu, 1978; Sam et al., 1998), both within the cells of the plant, and as physical adsorption to the surface which is not removed by chemical cleaning (Buyanov, 1973; Nakamura et al., 1979). The accumulation of radionuclides, such as ^{137}Cs and ^{90}Sr , depends on salinity, pH, and calcium levels, as well as environmental levels of these radionuclides (Bojanowski and Pempkowiak, 1977; Marchyulenene, 1978).

Algae have been used to examine exposure around industries using nuclear operations (Hung et al., 1998; Noshkin et al., 1981), and often accumulate higher levels of radionuclides than other organisms (Bonotto et al., 1981). *Fucus* and other seaweeds are frequently used as bioindicators of radionuclides in marine environments (Ben and Bonotto, 1991; FASSET, 2004). The concentrations of radionuclides in kelp more closely reflect the seawater rather than sediment levels (Ueda et al., 1985). The accumulation of radionuclides is proportional to the external concentrations, but the concentration factor can be as high as 1000 (Lattera and Bernhard, 1970), which makes them useful for bioremediation in some circumstances (Miskovic et al., 1992; Roach and Ashley, 1990). Some caution should be used, however, since the holdfast may have integrated radionuclides over several years, while the blades appear to reflect uptake only during the last few weeks, at least for *Fucus* studied by Hurtgen et al.

(1988). Algae are useful as bioindicators because they are accumulators and because they are common and widely distributed in many coastal environments (Al-Masri et al., 2003; Hung et al., 1998). *F. distichus* or its close relatives, are found along most north temperate coasts.

The data from this study indicate that marine algae from Amchitka were useful bioindicators for actinides in this environment, but not for other radionuclides, such as ^{137}Cs , ^{129}I , ^{60}Co , ^{152}Eu , ^{90}Sr , and ^{99}Tc . The differences in levels of the actinides among species suggest that some species are better accumulators than others, and could serve as bioindicators. Further, since the algae examined in this study lived in the same marine environment, differences among species found in this study indicate that *Fucus* is the highest bioaccumulator and would be the most useful as a bioindicator for the examined radionuclides.

Examination of radionuclide levels in algae is useful not only as a bioindicator of ecosystem contamination, but of potential human exposure since many species of kelp and other marine algae are subsistence foods for Alaskans (Garza, 2005), and for peoples elsewhere in the world (Phaneuf et al., 1999; Sharp et al., 1988; van Netten et al., 2000). In Wales, algae are an ingredient in laverbread, and the levels of ^{137}Cs can be as high as 0.20 Bq/kg (CEFAS, 2004). In Japan, most of the plutonium in the diet comes from the ingestion of algae (Hisamatsu et al., 1986), and they use algae as a bioindicator of potential radiological exposure from re-processing plants (Douville et al., 2004; Shinohara, 2004). Further, *Fucus* is also important as a human food, and is even sold in tablet form (van Netten et al., 2000).

5. Conclusions

The data collected on radionuclide levels in marine algae at Amchitka and Kiska Islands in 2004 enabled us to draw the following conclusions: (1) there were no levels above the MDA for ^{137}Cs , ^{129}I , ^{60}Co , ^{152}Eu , ^{90}Sr , and ^{99}Tc , (2) there were interspecific differences in ^{241}Am , $^{239+240}\text{Pu}$, ^{234}U , ^{235}U , and ^{238}U , (3) *Ulva* had the highest levels of ^{241}Am , *A. fistulosa* had the highest levels of $^{239+240}\text{Pu}$, and *Fucus* had the highest levels of ^{234}U , ^{235}U , and ^{238}U , (4) levels of all radionuclides were generally low and near the MDA for all isotopes, and (5) Amchitka Island has showed higher levels of $^{239+240}\text{Pu}$ than Kiska, but the differences were very small and probably not significant biologically. The data indicate that algae are useful bioindicators because they accumulate some radionuclides, even at very low environmental levels. The higher levels of naturally occurring uranium, compared to those found in algae from the contaminated Irish Sea, are intriguing and bears further examination.

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