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ORIGINAL ARTICLE

Radionuclide Concentrations in Benthic Invertebrates from Amchitka and Kiska Islands in the Aleutian Chain, Alaska

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Abstract Concentrations of 13 radionuclides (^{137}Cs, ^{129}I, ^{60}Co, ^{152}Eu, ^{90}Sr, ^{99}Tc, ^{241}Am, ^{238}Pu, ^{239,249}Pu, ^{234}U, ^{235}U, ^{236}U, ^{238}U were examined in seven species of invertebrates from Amchitka and Kiska Islands, in the Aleutian Chain of Alaska, using gamma spectroscopy, inductively coupled plasma mass spectroscopy, and alpha spectroscopy. Amchitka Island was the site of three underground nuclear test (1965–1971), and we tested the null hypotheses that there were no differences in radionuclide concentrations between Amchitka and the reference site (Kiska) and there were no differences among species. The only radionuclides where composite samples were above the Minimum Detectable Activity (MDA) were ^{137}Cs, ^{241}Am, ^{239,249}Pu, ^{234}U, ^{235}U, ^{236}U, and ^{238}U. Green sea urchin (*Strongylocentrotus polyacanthus*), giant chiton (*Cryptochiton stelleri*), plate limpets (*Tectura scutum*) and giant Pacific octopus (*Enteroctopus dofleini*) were only tested for ^{137}Cs; octopus was the only species with detectable levels of ^{137}Cs (0.262 ± 0.029 Bq/kg, wet weight). Only rock jingle (*Pododesmus macroschisma*), blue mussel (*Mytilus trossulus*) and horse mussel (*Modiolus modiolus*) were analyzed for the actinides. There were no interspecific differences in ^{241}Am and ^{239,240}Pu, and almost no samples above the MDA for ^{238}Pu and ^{236}U. Horse mussels had significantly higher concentrations of ^{234}U (0.844 ± 0.804 Bq/kg) and ^{238}U (0.730 ± 0.646) than the other species (both isotopes are naturally occurring). There were no differences in actinide concentrations between Amchitka and Kiska. In general, radionuclides in invertebrates from Amchitka were similar to those from uncontaminated sites in the Northern Hemisphere, and below those from the contaminated Irish Sea. There is a clear research need for authors to report the concentrations of radionuclides by species, rather than simply as 'shellfish', for comparative purposes in determining geographical patterns, understanding possible effects, and for estimating risk to humans from consuming different biota.

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1 Introduction

Radionuclides enter the environment from natural geologic sources and from fallout from historic nuclear weapons testing (Aarkrog, 2003; Duran, Povinec, Fowler, Airey, & Hong, 2004), from nuclear facility and submarine accidents (Baeza et al., 1994; Cooper et al., 1998; Livingston & Povinec, 2000; UNSCEAR, 2000; Sanchez-Cabeza & Molero, 2000; Amundsen et al., 2002; Aumento, Donne & Eroe, 2005), and from discarded nuclear wastes (Fisher et al., 1999; IAEA, 1999). Over 500 atmospheric nuclear weapons tests were conducted from 1945–1980, primarily in the Northern Hemisphere (UNSCEAR, 2000). 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). Some radionuclide concentrations, such as the plutonium found in mussels in some regions, however, are due mainly to upwelling of mid-depth waters from the Pacific Ocean, not to radioactive dumping in the region local (Farrington, Davis, Tripps, Phelphs, & Galloway, 1987). The potential for human health and environmental effects from consumption of radionuclides in marine organisms is clear, particularly for ^{137}Cs and ^{239}Pu (Moscati & Erdmann, 1974; Shenber, Elshamis, Elkikli, & Elayan, 1999).

Increasingly, the public, regulators, managers and policy makers are interested in assessing the health of ecosystems, the food chain, and human foods. Because of the importance of fish and shellfish consumption throughout the world, and the occurrence of atmospheric deposition of radionuclides into marine environments, a number of monitoring programs for radionuclides have been established in Asia (Duran et al., 2004), in the Sea of Japan (Togawa, Povinec, & Pettersson, 1999; JCAC, 2003, 2004), in the Irish Sea (RPII, 2003, 2004), in the French Mediterranean (Charmasson, Barker, Calmet, Pruchon, & Thebault, 1999), and in the Black Sea (Bologa, 2000). Other biomonitoring programs have been established to evaluate possible exposure from nuclear facility operations (Poon & Au, 2002; Shinohara, 2004), as well as exposure due to dumping by the Former Soviet Union (Togawa et al., 1999; Yamada, Aono, & Hirano, 1999; Lystsov, Murzin, & Nezhdanov, 1999; Matishov, Matishov, Anisimova, Dzhenyuk, & Zuev, 2001). Some of this biomonitoring is ongoing near decommissioned nuclear power plants and reprocessing plants (e.g., Sellafield in the United Kingdom, Sanchez-Cabeza & Molero, 2000; in Taiwan, Hung, Huang, & Shao, 1998). There is also interest in assessing radionuclide concentrations in organisms near nuclear waste facilities, including nuclear testing sites.

In this paper we examine the concentrations of a suite of radionuclides in several invertebrates in the marine environment around Amchitka Island in the Aleutian chain of Alaska. This paper reports on part of the biologic study carried out by the Consortium for Risk Evaluation with Stakeholder Participation (CRESP). CRESP is a multi-university, multi-disciplinary organization, assisting the U.S. Department of Energy (DOE) in pursuing remediation of nuclear wastes generated during the Cold War manufacture and testing of nuclear weapons. Even though the invertebrate community in the Aleutians is mostly inconspicuous, compared to other regions (O'Clair, 1977), some invertebrates could prove valuable as indicators of radionuclide exposure because they are relatively sedentary, and occupy different trophic groups. Moreover some are consumed as part of regional subsistence diets (APIA, 2002; Patrick, 2002; Hamrick & Smith, 2003).

We test the null hypotheses that 1) there were no interspecific differences in radionuclide concentrations, and 2) there were no differences in radionuclide concentrations between Amchitka and a reference site (Kiska Island). Amchitka was the site of three underground nuclear tests from 1965 to 1971. Although radionuclide concentrations were assessed in marine biota in the early 1970s (Merritt & Fuller, 1977), little testing has occurred since then, although Dasher et al. (2002) assessed radionuclide concentrations in terrestrial biota.

These data can be used to assess whether the subsistence foods and commercial fish are safe for consumption, to establish a baseline for future comparison, and to determine whether the concentrations found in fish and birds at Amchitka are similar to those found in other regions of the Northern Hemisphere. This work is part of a larger multi-disciplinary project by CRESP to provide the information to assure the protection of human health and the environment, and to provide a baseline for monitoring in the context of a long-term stewardship plan for Amchitka (Powers, Burger, Kosson, Gochfeld, & Barnes, 2005, Powers, Burger, Kosson, & Gochfeld, 2006; Burger, Gochfeld, Kosson, & Powers, 2006). Further, radionuclide concentrations in benthic organisms living at high latitudes have been poorly investigated (Matishov & Matishov, 2004), and the present study contributes to the overall knowledge of radionuclides in invertebrates from this region.

2 Study Sites and Methods

2.1 Study sites

Amchitka Island (51.5° N lat; 179° E long) and Kiska Island (about 120 km to the west, 177.5° E Long) in the western Aleutian Chain of Alaska, are part of the Alaska Maritime National Wildlife Refuge. The islands are bordered on the south by the North Pacific and on the north by the Bering Sea (Figure 1). The marine biological resources in the region are of high value in cultural (including subsistence), commercial, and ecological terms (NRC, 1996; Jewett, 2002). It is also one of the most seismically, tectonically and volcanically active regions of the world (Jacob, 1984; Page, Biswas, Lahr, & Pulpan, 1991; Eichelberger, Freymueller, Hill & Patrick, 2002; Jewett, 2002; Patrick, 2002).

Amchitka Island was a military base opposing the Japanese occupation of Kiska Island in World War II. In the 1960s Amchitka was chosen for underground nuclear tests over the objections of local people and foreign governments (O'Neill, 1994; Kohlhoff, 2002). The remoteness of Amchitka, the tectonic activity (which might 'hide' a nuclear test signature in seismic noise), and its proximity to the Soviet Union were factors in its selection (Kohlhoff, 2002). There were three nuclear tests in 1965, 1969, and 1971. *Cannikin* (about 5 Mt in 1971) was the last and largest U. S. underground test. The three Amchitka test shots accounted for about 16% of the total energy released from the U.S. underground testing program (Robbins, Makhijani, & Yih, 1991; Norris & Arkin, 1998; DOE, 2000). The relevant source term information is classified and unavailable to the public and the authors. The releases of radiation to the surface were not considered to pose serious health or ecological risks at the time (Seymour & Nelson, 1977; Faller & Farmer, 1998), and recent studies by Dasher et al. (2002) did not indicate any current surface contamination. The infrastructure on the island (buildings, roads, wells) was removed by the Department of Energy (DOE) during remediation of surface contamination in 2001. There is no current technology for

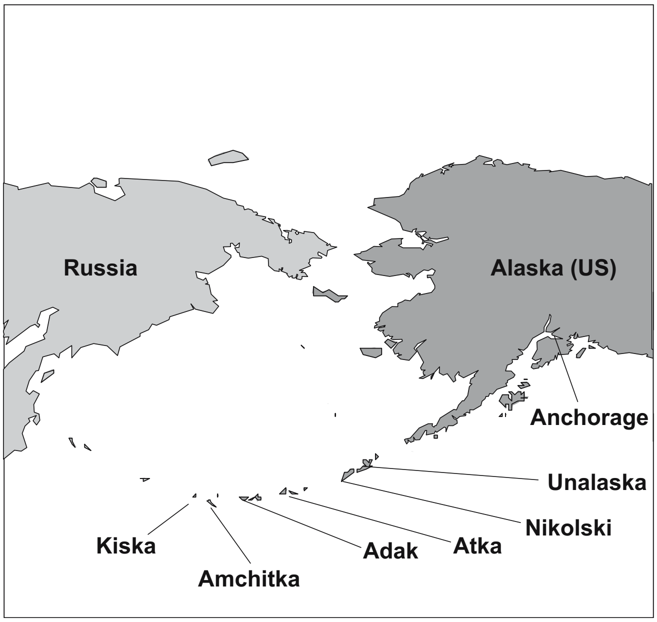

Figure 1 Map of Alaska showing locations of Aleut villages that took part in the research process, as well as Amchitka and Kiska Islands.

remediating the test cavities, to inactivate or entrap the radiation, or over the long term, to interdict its transport by ground water through fissures and porous rock to the sea.

The intense heat of the detonations at Amchitka presumably melted the rock, and trapped much of the radioactive material in a glass-like matrix. However, radionuclides were also distributed in the rubble-filled chimney, and the permanence of the vitrified residuals is unknown. As rainfall recharges the freshwater aquifer in the island's subsurface, radionuclides dissolved in the flowing groundwater could be carried through natural faults and fissures to the sea (DOE, 2002a). The DOE's groundwater model predicted that breakthrough might occur any time from 10 to 1,000 years after the blasts (DOE, 2002a), but their risk assessment models predicted no current or future risk to humans (DOE, 2002b). Since it is already several decades since the detonations, it is important to assess whether there has been any seepage and to examine the concentrations of radionuclides in biota as potential bioindicators for future biomonitoring. CRESP's studies (Powers et al., 2005) refined these estimates.

Kiska Island, the reference site, had similar intertidal and benthic communities (Burger et al., in press). It was occupied by the Japanese during World War II, and suffered intensive allied bombing, but never had any nuclear testing.

2.2 Protocol

Under appropriate state collecting permits, invertebrates were collected from the intertidal and subtidal zones of both Amchitka and Kiska Islands in a balanced design (where possible) from late June–July 2004. Species collected were green sea urchin (*Strongylocentrotus polyacanthus*), plate limpets (*Tectura scutum*), blue mussel (*Mytilus trossulus*), giant Pacific octopus (*Enteroctopus dofleini*), giant chiton (*Cryptochiton stelleri*), horse mussel (*Modiolus modiolus*) and rock jingle (*Pododesmus macroschisma*); all but the last three are subsistence foods of the Unangan (Aleut) peoples. Except for octopus, these were some of the commonest species available, and they had a wide distribution. These invertebrates represent different nodes on the food chain, including grazers (chitons, limpets, sea urchins), filter feeders (jingles, mussels), and predators (octopus). Further, they represent different marine zones, from intertidal (blue mussels, limpets), to benthic (giant chiton, rock jingles, horse mussels, giant Pacific octopus, sea urchins).

Sea urchins, plate limpets (called Chinese Hats by Aleut subsistence hunters), and blue mussels were collected by both the Aleut and scientist teams on our expedition, the other species were collected by divers along transects perpendicular to shore and adjacent to each of the three test sites and at Kiska. All specimens were tracked from field collection to their ultimate analytic destination with chain of custody forms. Our overall protocol was to collect enough sample material from each specific location to composite (at least five individuals) from a given site). Algae, fish and birds were also collected at the same time (Burger et al., submitted-a,b).

In the shipboard laboratory, all specimens were scanned with a handheld counter for gross alpha, gamma and beta. They were processed, measured (length, weight), cut into segments (octopus only), packaged and labelled. Samples were then immediately frozen for later analysis. Radionuclide analyses were conducted on composites containing five or more (depending on size) individuals each. Most composites were made up to 85 g, but some 1,000 g composites were analyzed for 72 h for ^{137}Cs to achieve appropriate MDAs. For 1,000 g samples, many more individuals were required per composite: Sea urchins (50–75 individuals), rock jingle (89–142 individuals), plate limpets (51–99 individuals), giant chitons (109 individuals), and blue mussel (115–229 individuals). All preparation and analytic work were performed in radio-clean laboratories, checked by daily wipe samples (all below MDA). Samples were homogenized in a radio-clean and metal-clean laboratory at Rutgers University, and subsequently analyzed for radionuclides at Vanderbilt University and Idaho National Laboratory (INL).

Our radionuclide analysis design was based on trophic group considerations, sample availability and quantity (Table I). Detailed analytic and quality assurance methods are published on the CRESP website www.cresp.org; Powers et al., 2005, 2006). 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}Pu), and uranium (^{234}U, ^{235}U, ^{236}U, ^{238}U). Analyses at Vanderbilt and Idaho National Laboratory provided inter-laboratory validation

Table I Number of composites of invertebrates collected at Amchitka and Kiska analyzed for radionuclides

Species	Cs 137	I-129	Co-60	Eu-152	Sr-90	Alpha[a]	Tc-99
Sea urchin	3 (4)	5	4	4	8		4
Rock jingle	3 (4)	4	4	4	4	21	3
Plate limpet	2						
Giant gumboot	1						
Blue mussel	2					9	
Horse mussel						8	
Giant Pacific octopus	4						

Shown for ^{137}Cs are 1,000 g samples (100 g samples). All others were composites of 100 g or less.

[a] Alpha analysis included the actinides ^{241}Am, ^{238}Pu, ^{239,240}Pu, ^{234}U, ^{235}U, ^{236}U, and ^{238}U.

(Powers et al., 2005). Gamma emitters (^{137}Cs, ^{152}Eu, ^{60}Co) were analyzed using gamma spectroscopy with high purity germanium detectors calibrated to the standard container geometry. ^{129}I was analyzed by low energy photon measurement. The beta emitter ^{90}Sr was analyzed by its daughter decay product, yttrium-90 (^{90}Y). Counts were adjusted for background counts, and the Minimum Detectable Activity (MDA) was ± 2 SD background. The actinides (uranium, plutonium, americium) were quantified using radiochemical techniques and alpha spectroscopy (average MDAs ranged from 0.052 for ^{241}Am to 0.102 for ^{235}U Bq/kg). All values are presented in Bq/kg, wet weight, both in 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 1,000 g samples for 72 h. MDAs for ^{137}Cs ranged from 5.57–6.25 Bq/kg for 100 g samples, and 0.18–0.36 Bq/kg for 1,000 g samples.

There were no values above the MDA for ^{129}I, ^{60}Co, ^{52}Eu, ^{90}Sr, ^{99}Tc, and ^{238}Pu, and these isotopes are not discussed further.

3 Results

3.1 Radiocesium

Only octopus had detectable concentrations of ^{137}Cs, all other invertebrates, even with 1,000 g composites, were below the MDA. Some composites had over 100 individuals (due to small body size). All four octopus samples had detectable ^{137}Cs concentrations (mean of 0.262 ± 0.029 Bq/kg, wet weight; maximum value of 0.302 Bq/kg). Octopus were only collected at Amchitka; none were found at Kiska.

Table II Comparison of actinide levels in invertebrates

Isotope	Source	Rock jingle	Blue mussel	Horse mussel	Chi square (*P* value)
Number of composites		21	9	8	
Am-241	A	0.021 ± 0.04	0.017 ± 0.004	0.016 ± 0.004	0.20 $P < 0.90$
Pu-239,240	A	0.024 ± 0.012	0.019 ± 0.004	0.022 ± 0.011	0.49 $P < 0.78$
U-234	N	0.446 ± 0.079	0.598 ± 0.194	0.844 ± 0.804	5.69 $P < 0.058$
U-235	N	0.015 ± 0.026	0.021 ± 0.014	0.030 ± 0.048	1.28 $P < 0.53$
U-236	A	(0.011)			
U-238	N	0.345 ± 0.071	0.558 ± 0.165	0.730 ± 0.646	16.3 $P < 0.003$[a]

Given are the means (± standard deviation, wet weight) in Bq/kg with the values plus half the MDA for those below the MDA. Where there are very few values above the MDA for an isotope, the actual values are given in parenthesis. For source, A = anthropogenic, and N = natural.

[a] This difference is due to one high outlier, suggesting that this difference is not biologically significant.

Table III Interisland comparison for actinides in rock jingles

Isotope	Range of reported values (from MDA to highest value)	Mean ± SD	Kruskal Wallis Chi Square	Number of detects (%)	Fisher Exact test *P* value
Am-241					
Amchitka	−0.047	0.02 ± 0.012	0.61 P = 0.44	5 of 15 (33 %)	*P* = 0.69
Kiska	−0.043	0.022 ± 0.012		2 of 6 (33 %)	
Pu 239,240					
Amchitka	−0.060	0.024 ± 0.013	0.01 *P* = 0.94	4 of 15 (27 %)	*P* = 0.57
Kiska	−0.038	0.023 ± 0.010		2 of 6 (33 %)	
U-234					
Amchitka	−0.583	0.439 ± 0.090	0.02 *P* = 0.88	15 of 15 (100%)	*P* = 1.0
Kiska	−0.493	0.462 ± 0.041		6 of 6 (100 %)	
U-235					
Amchitka	−0.048	0.023 ± 0.009	0.22 *P* = 0.64	1 of 15 (7%)	*P* = 0.18
Kiska	−0.064	0.028 ± 0.018		2 of 6 (33%)	
U-236					
Amchitka				0 of 15	*P* = 0.29
Kiska	(0.011)			1 of 6	
U-238					
Amchitka	−0.451	0.345 ± 0.70	0.001 *P* = 0.97	15 of 15 (100%)	*P* = 1.0
Kiska	−0.440	0.345 ± 0.078		6 of 6 (100%)	

Given is mean ± standard deviation (Bq/kg, wet weight computed with values above the MDA and half the MDA for non-detects). Amchitka n = 15, Kiska n = 6 composites.

3.2 Actinides

There were significant interspecific differences for ^{234}U and ^{238}U, both naturally occurring radionuclides (Table II). Horse mussels had significantly higher concentrations than the other species. There were no differences in the anthropogenic radionuclides, perhaps due to their relatively low concentrations.

There were no interisland differences in any actinides for rock jingles, the species with the largest number of composites (Table III). Similarly, there were no interisland differences in actinides for blue mussels (Table IV) or horse mussels (Table V). There were also no interisland differences in the percent of composites above the MDA for rock jingles and mussels. The concentrations are given by island for each species separately because these data will be used as the baseline for long-term biomonitoring at Amchitka (Burger et al., 2006), and to allow other geographical comparisons.

4 Discussion

4.1 Interspecific and interisland comparisons

Interspecific differences in concentrations of contaminants, including radionuclides, are usually due to differences in trophic concentration (Denton & Burdon-Jones, 1986; Jackson, 1991; Kasamatsu & Ishikawa, 1997; Watras et al., 1998; Wiener and Spry, 1996; Burger et al., 2001), size and age (Lange, Royals, & Connor, 1994; Burger et al., 2001; Pinho et al., 2002; Green & Knutzen, 2003), and habitat (Burger et al., 2002). In general, concentrations are higher in species that are larger, older, and in a higher trophic group. Trophic level relationships have also been reported for ^{137}Cs (Pentreath, 1973; Matishov & Matishov, 2004), although Dietz et al. (2000) did not find an increase in ^{137}Cs concentrations with increasing trophic level. At Amchitka, ^{137}Cs showed a trophic group relationship only in that octopus had

Table IV Interisland comparison for actinides in blue mussels

Isotope	Range of reported values	Mean ± SD	Kruskal Wallis Chi Square	Number of detects (%)	Fisher Exact test *P* value
Am-241					
Amchitka				0 of 6	*P* = 0.33
Kiska	(0.025)			1 of 3	
U-234					
Amchitka	–0.844	0.542 ± 0.170	1.07 *P* = 0.30	6 of 6	*P* = 1.0
Kiska	–0.949	0.710 ± 0.224		3 of 3	
U-235					
Amchitka	(0.045, 0.039)			2 of 6	*P* = 0.42
Kiska				0 of 3	
U-238					
Amchitka	–0.799	0.518 ± 0.150	1.07 *P* = 0.30	6 of 6	*P* = 1.0
Kiska	–0.844	0.623 ± 0.204		3 of 3	

Given is mean ± standard deviation (Bq/kg, wet weight (computed with values above the MDA and half the MDA for non-detects). Amchitka n = 6 composites, Kiska n = 3 composites. Where there is only one value, it is given in parentheses. The full suite of actinides were analyzed, but they are not given if all were below the MDA.

levels above the MDA, while other species did not. Octopus was the only predatory invertebrate examined, the other species examined were grazers and filter-feeders (O'Clair, 1977).

For the actinides, the only interspecific differences were in the naturally occurring ^{234}U and ^{238}U. These are the only isotopes with all composites above the MDA for blue mussel, horse mussel, and rock jingle. For both isotopes, horse mussel had the highest concentrations, and rock jingle had the lowest. We cannot account for this difference, since all three species are filter-feeders. It may be related to

Table V Interisland comparison for actinides in horse mussels

Isotope	Range of reported values	Mean ± SD	Kruskal Wallis Chi Square	Number of detects %	Fisher Exact test *P* value
Am-241					
Amchitka				0 of 6	*P* = 0.25
Kiska	0.021			1 of 2	
Pu 239,240					
Amchitka	0.048			1 of 6	*P* = 0.75
Kiska				0 of 2	
U-234					
Amchitka	–2.78	0.995 ± 0.890	1.78 *P* = 0.18	6 of 6	*P* = 1.0
Kiska	–0.468	0.390 ± 0.110		2 of 2	
U-235					
Amchitka	(0.142, 0.049)			2 of 6	*P* = 0.54
Kiska				0 of 2	
U-238					
Amchitka	–2.28	0.849 ± 0.715	1.78 *P* = 0.18	6 of 6	*P* = 1.0
Kiska	–0.466	0.371 ± 0.134		2 of 2	

Given is mean ± standard deviation (Bq/kg, wet weight (computed with values above the MDA and half the MDA for non-detects). Amchitka n = 6, Kiska n = 2 composites.

differential toxicokinetics (absorption, elimination and storage) among the three species.

It is also useful to briefly compare the concentrations in the invertebrates with those for algae and fish (Burger et al., unpublished data; Powers et al. 2005). There were no detectable concentrations of ^{137}Cs in algae, but there were interspecific differences in some radionuclides: *Ulva* had the highest concentrations of ^{241}Am, *Alaria fistulosa* had the highest concentrations of ^{239,240}Pu, and *Fucus* had the highest concentrations of ^{234}U, ^{235}U, and ^{238}U. However, concentrations of all radionuclides were generally low and near the MDA for all isotopes in algae. In contrast, there were significant differences in ^{137}Cs as a function of species, but not location for top predatory fish. Like the invertebrates, ^{234}U and ^{238}U, isotopes that are primarily natural in origin, had the highest actinide detection rates in fish, and there were no significant differences in mean concentrations between Amchitka and Kiska.

4.2 Geographical comparisons

Concentrations of radionuclides in seawater vary in different oceans; the seas with the highest concentrations of key radionuclides are the Irish Sea (^{239,249}Pu, ^{90}Sr, ^{137}Cs) and Baltic Sea (^{137}Cs, Livingston & Povinec, 2000). Similarly, concentrations of radionuclides vary in invertebrates. For example, Valette-Silver and Lauenstein (1995) reported on concentrations of radionuclides in bivalves collected along the coastal United States (but not from Alaska). They drew several conclusions: 1) ^{241}Am and ^{137}Cs concentrations were higher along the West coast of the U.S. compared to the East or Gulf coasts, 2) there were no significant locational differences for the other radionuclides, and 3) there was a significant decline in radionuclide concentrations in bivalves between the mid-1970s and the early 1990s.

4.2.1 Radiocesium

Comparative data for ^{137}Cs are available on some invertebrates (Table VI). Except for octopus, all concentrations in invertebrates at Amchitka were below the MDA; in the discussion below, we are thus comparing the MDAs at Amchitka with the reported values or MDAs from elsewhere. The concentrations in the invertebrates at Amchitka were similar to or below those from uncontaminated sites in the Northern Hemisphere, and were well below those from the Irish Sea. While the average concentration in non-Irish Sea Northern Hemisphere sites (Table VI) is 0.03 Bq/kg, some presumably uncontaminated sites have even higher concentrations. Concentrations for mollusks from the Asia-Pacific regional seas averaged 0.1 Bq/kg (Robison & Noshkin, 1999; Duran et al., 2004), although Ishikawa, Kagaya, and Saga (2004) reported values of 0.03 Bq/kg from the coast of Japan. From the Mediterranean Sea, ^{137}Cs concentrations averaged 0.10 Bq/kg generally (Sanchez-Cabeza & Molero, 2000), but averaged 1.23 Bq/kg from the

Table VI Comparison of ^{137}Cs levels in invertebrates

	Irish Sea[a]	Other sites[b]	Amchitka (2004)[c]
Sea Urchin		1.8	<MDA
Mollusks			
Mean level	3.98	0.03	<MDA
Range	<MDA – 16	<MDA – 0.41	0.09 – 0.60(MDAs)
Number of analyses	323	112	12/8
Octopus	None available	14.8[d]	0.262

Given are means (Bq/kg, wet weight). Octopus is not included in this table because comparative data were not found (see text for other data).

[a] Data from RPII (2003, 2004), CEFAS (2003, 2004), BNFL (2004). None available for sea urchin.

[b] Data from CEFAS (2003, 2004), RPII (2003, 2004), JCAC (2003, 2004), Hong Kong Observatory (2003). Sea urchin data (mean) from Matishov and Matishov (2004).

[c] For range, average MDAs are given since all composites were below the MDA. 12/8 refers to 12 1,000-g samples (72 h), and 8 100-g samples (24 h).

[d] Data from northern Italy (after Gallelli et al., 1997).

Thermaikos Gulf in Greece (Catski & Florou, 2006). The variation, even within the Mediterranean, suggests that site-specific information is essential, especially for species that are used for subsistence or fisheries.

Although some studies report on crustaceans, as well as mollusks (Duran et al., 2004), sea urchins are seldom examined (Matishov & Matishov, 2004). This may partly be due to technical difficulties. Digestion of sea urchins in this study ruined the platinum crucibles, and we could not use the concentrations for actinides. The other difficulty with reported values by group is that individual species cannot be compared around the world; this seems to be particularly a problem with 'shellfish', rather than with algae or fish.

There are relatively few data on contaminants in octopus, mainly because they are difficult to capture reliably, particularly from specific locations. Even for metals, there are few data, and studies report concentrations from only 5 (Anderson, 2003) to 12 individuals (Seixas, Bustamante, & Pierce, 2005). Gallelli, Panatto, Perdelli, and Pellegrino (1997), however, did measure ^{137}Cs in two species of octopus from the Ligurian Sea in northern Italy, and found means (±standard deviation) of 14.8 ± 10.9 and 10.6 ± 10.4 Bq/kg in 1987, and 2.5 ± 2.1 and 4.8 ± 4.3 Bq/kg in 1988. This may relate to time since the Chernobyl accident (April 26, 1986), and it is important to have more recent data. The mean ^{137}Cs concentrations in octopus from Amchitka were 0.26 ± 0.029 Bq/kg, much lower than those from the Ligurian Sea. Because of their ability to concentrate ^{137}Cs, Gallelli et al. (1997) suggested that they would be a good bioindicator. Yamada et al. (1999) examined two species of octopus from the Japanese coast and found low concentrations of ^{137}Cs (mean of 0.075 Bq/kg for two animals).

4.2.2 Actinides

There are also few comparative data on actinides for invertebrates, and often the data are presented only as concentration factors, and not as the concentrations (needed for comparisons among regions). Sanchez-Cabeza and Molero (2000) reported ^{241}Am concentrations of 0.004 Bq/kg in mussels from the Mediterranean Sea, and Marzano, Fiori, Jia, and Chiantore (2000) reported ^{241}Am concentrations of 0.002 Bq/kg in mussels from the Antarctic; both were much lower than those we found at Amchitka. Robison and Noshkin (1999) reported mean ^{241}Am values for mollusks at Enewetak Atoll (0.020 ± 0.005 Bq/kg, wet weight) that were similar to those we found at Amchitka. However, the concentrations at Bikini Atoll were higher (0.32 ± 0.29 Bq/kg, Robison & Noshkin, 1999). Some of these differences may be due to the year, since the Atoll data span the period from 1972 to 1991, and to their use as nuclear testing grounds (Robison & Noshkin, 1999). These analyses were based on sample sizes of 2–12 for all species of mollusks.

Robison and Noshkin (1999) reported mean ^{239,240}Pu for shellfish from Bikini and Enewetak Atolls in Micronesia (0.032 ± 0.024 Bq/kg, wet weight), which was similar to the concentrations at Amchitka. Skwarzec (1997) also reported mean concentrations of ^{239,240}Pu from the Baltic Sea (0.022 ± 0.001) that were very similar to those from Amchitka. However, Shinohara (2004) reported mean ^{239,240}Pu concentrations for shellfish (0.0036, maximum of 0.008 Bq/kg), which were lower than Amchitka, but similar to concentrations from the Mediterranean (Sanchez-Cabeza & Molero, 2000). Both of the previous papers presented means without listing the species, the number of detects/non-detects, or the MDAs. It is thus difficult to evaluate whether many values were above detection limits. This limits their use for comparative purposes, and for selection of bioindicators. Further, computing the dose to non-human or human consumers of shellfish from data without such parameters is difficult. The significance of exposure assessments for non-human biota based on empirical data with such data gaps may lead to faulty risk characterization (Avila et al., 2004). This lack of species-specific information for shellfish is even more problematic when computing human dose, which must relate directly to consumption patterns for different species (or types) of shellfish.

Few studies report the concentrations of the naturally occurring actinides. However, Rollo, Camplin, Allington, and Young (1992) analyzed a range of invertebrates and fish from the marine environment of the United Kingdom. Mean concentrations of ^{234}U in shellfish ranged from 1.4–5.6 Bq/kg (wet weight), ^{235}U ranged from 0.027–0.19 Bq/kg, and ^{238}U ranged from 1.2–5.5 Bq/kg (Rollo et al., 1992). The concentrations at Amchitka were all well below these concentrations, but higher than those from the coast of Japan (0.20 Bq/kg ^{238}U, Ishikawa et al., 2004). The concentrations of ^{238}U in bivalves from the Baltic Sea (average 0.69 Bq/kg, Skwarzec, 1997), were similar to those in bivalves from Amchitka. However, the concentrations of ^{238}U from the Mediterranean Sea (mean of 0.0002 Bq/kg, Sanchez-Cabeza & Molero, 2000), and Antarctic (mean of 0.001, Marzano et al., 2000), were well below those found at Amchitka. In sharp contrast, concentrations of ^{238}U from the eastern Black Sea averaged 35 Bq/kg, wet weight (Topcuoglu, Ergul, Baysal, Olmez, & Kut, 2003). Although uranium formed at the time of the Earth's creation about 4.7 billion years ago, it is not uniformly distributed over the surface. Concentrations tend to be higher in rocks of volcanic origin, for example, than in sedimentary rocks (USGS, 2006). Thus the contribution of surface erosion to uranium content of the sea would be higher in areas with relatively rich soils.

4.3 Use of invertebrates as bioindicators

Many invertebrates accumulate contaminants, such as radionuclides, because they are sedentary or not very mobile, are benthic, and graze or filter feed. They are ideal indicators because they can accumulate radionuclides many times above the concentrations in seawater (Matishov & Matishov, 2004). Where concentrations in seawater and sediment are very low, however, all analyses might well be below the MDA, as occurred in this study for several radionuclides (^{129}I, ^{60}Co, ^{52}Eu, ^{90}Sr, ^{99}Tc, ^{238}Pu), and even for ^{137}Cs (except for the predatory octopus). Further, in order to achieve low enough detection concentrations of ^{137}Cs in octopus, we had to use 1,000 g samples, counted for 72 h. While it is not difficult to obtain enough material from octopus for large composites, it is more difficult for smaller species, such as limpets, mussels, and sea urchins. And all of these were considered delicacies by the Aleut team members. These are commonly used subsistence foods (APIA, 2002; Patrick, 2002; Hamrick & Smith, 2003), especially if Aleuts are stranded overnight during hunting forays or when they first land on beaches to set up temporary camps.

Invertebrates seem to be particularly useful for actinides, however, where small sample sizes can be used. All of the composites were above the MDA for the naturally occurring ^{234}U and ^{238}U. Since there were interspecific differences for these isotopes (horse mussels had the highest concentrations), some species are better than others. That is, the species that accumulate the most, even in a region of relatively low concentrations, would be the best bioindicators.

Finally, for invertebrates to be useful as bioindicators, concentrations must be attributable to individual species. For example, there are thousands of species of benthic organism in the seas (Matishov & Matishov, 2004). Thus, there is a clear need for authors to report concentrations by species (along with MDAs and percent of samples above the MDA), and then group them by 'shellfish' if this is necessary for other purposes. While it is helpful to lump many species into a 'shellfish' category when examining overall trophic relationships (primary producers, herbivores, predators), it is not when examining specific risk to the individual species themselves, or to predators that eat them. People do not eat shellfish, they eat mussels, clams, or oysters. Risk can only be determined using contaminant levels in specific shellfish, and consumption rates of those individual species. That is, assessors determine the risk from consuming mussels or clams.

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