Paper

RADIONUCLIDES IN MARINE FISHES AND BIRDS FROM AMCHITKA AND KISKA ISLANDS IN THE ALEUTIANS: ESTABLISHING A BASELINE

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Abstract—**Amchitka Island (51° N lat, 179° E long) was the site of three underground nuclear tests from 1965–1971. There have been no substantive studies of radionuclides in marine fishes and birds in the area since the mid-1970's. In this study, levels of ^{60}Co, ^{52}Eu, ^{90}Sr, ^{99}Tc, ^{129}I, ^{137}Cs, and the actinides (^{241}Am, ^{238}Pu, ^{239,240}Pu, ^{234}U, ^{235}U, ^{236}U, and ^{238}U) were studied in ten marine fish species (including Pacific Cod *Gadus macrocephalus* and Pacific Halibut *Hippoglossus stenolepis*) and five marine bird species (including Glaucous-winged Gulls *Larus glaucescens*, Tufted Puffins *Fratercula cirrhata*, and Common Eider Ducks *Somateria mollissima)* from Amchitka. The same species were collected at a reference site, Kiska Island (52° N lat; 177° E long), about 130 km west of Amchitka. Each sample was a composite of edible muscle from five or more individual fish or birds of similar size (±15%) from the same sampling station. The null hypotheses of no differences among species or between Amchitka and Kiska were tested. Most analytic results were below the minimum detectable activity (MDA), even when 1,000 g sizes and 72 h counting times were used. The only radionuclides detected above the MDA were ^{137}Cs, ^{241}Am, ^{239,240}Pu, ^{234}U, ^{235}U, and ^{238}U. There were significant differences in ^{137}Cs as a function of species, but not location, for top predatory fishes. Of the fishes, eight of ten species had ^{137}Cs values above the MDA for some samples; only one bird, Glaucous-winged Gull, had ^{137}Cs values above the MDA. The highest concentrations of ^{137}Cs were in Dolly Varden [*Salvelinus malma*, 0.780 (Bq kg^{-1} wet weight)] and Pacific Cod (0.602 Bq kg^{-1}). In aggregate for any actinides, 73 of 234 (31%) composites for fish were above the MDA, compared to only 3 of 98 (3%) for birds. ^{234}U and ^{238}U, radionuclides that are primarily natural in origin, were routinely detected in these biological samples, but there were no significant differences in mean concentrations between Amchitka and Kiska. The concentrations of all radionuclides examined at Amchitka are similar to those of other uncontaminated Northern Hemisphere sites, and are lower than those reported for fishes and birds from the Irish Sea in the vicinity of the Sellafield nuclear reprocessing facility, an area with known contamination.**

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Key words: biological indicators; ^{137}Cs; plutonium; radioactivity, environmental

INTRODUCTION

The management of radioactive wastes and the protection of humans and the environment from residual wastes and nuclear accidents is an important element of radiation protection and public policy. Information on radionuclide concentrations in organisms at different nodes on the food chain, and in the foods that are eaten by people, is thus essential to understanding and quantifying potential risks from population exposures to these radionuclides. While models are useful in predicting what concentrations might be expected in different biota compartments in marine ecosystems (Kryshev et al. 2001; Matishov et al. 2001; Hakanson 2005), measurements of actual concentrations in biota and foods consumed are clearly more directly useful in predicting intake rates and ultimately doses, particularly when the public is included in determining what species and foods are tested (Burger et al. 2005). Further, using concentration factors to estimate concentrations in fishes and shellfish is not as direct as providing the data, and in any case, has proven not to be predictive for Arctic fishes (and thus presumably for subarctic fishes of similar behavior; Sazykina 1998).

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Amchitka Island was the site of three underground nuclear tests from 1965 to 1971, and although considerable biological and radiological work was conducted in the early 1970's (Merritt and Fuller 1977), there has been little evaluation of radionuclides in marine biota since then. In a primarily terrestrial study, Dasher et al. (2002) used the brown algae (*Fucus*) collected from Amchitka shores as a bioindicator of ^{240}Pu/^{239}Pu ratios and reported that the ratios were above the value of 0.18 indicative of global fallout (Krey et al. 1976; Buesseler and Halverson 1987), but noted that because of small sample sizes and potential analytical issues, more research was necessary. Amchitka has posed a challenge for the U.S. Department of Energy (DOE), state and federal regulators, and other stakeholders including the Aleuts and other people living in the Aleutian Islands. People were primarily concerned about the concentrations of radionuclides and mercury in marine biota and subsistence foods. Controversy also existed at the time of the tests because of concerns about the disruptive role of earthquakes and other geologic events on the nuclear residue remaining in the underground test cavities (Greenpeace 1996; U.S. DOE 1997, 2000; Kohlhoff 2002; Eichelberger et al. 2002).

The aims of the present study were to examine the concentrations of a wide range of radionuclides in fishes and birds in the marine ecosystem surrounding Amchitka Island, and to compare them to a reference site (Kiska Island). Specific objectives were to determine whether there were inter-specific (differences between species) and inter-island differences in radionuclide concentrations, and to determine the relative concentrations among radionuclides in biota from this region. These data can be used to examine seafood safety, to determine whether the concentrations found in fishes and birds at Amchitka were similar to those found in other regions of the Northern Hemisphere, and to establish a baseline for future monitoring. This work is part of a larger multidisciplinary 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 monitoring in the context of a long-term stewardship plan for Amchitka (Powers et al. 2005, 2006; Burger et al. 2006a). While these data are critical for the development of long-term protection and surveillance on Amchitka Island, they can also be used to provide baseline radionuclide data for fishes and birds in this region of the Northern Pacific Ocean and Bering Sea.

Although the human population density in the immediate vicinity is relatively low, the Bering Sea ecosystem provides a large percentage of the fish and shellfish for commercial sale in the United States and elsewhere (AFSC 2003). Understanding baseline concentrations is particularly important for the Bering Sea region, where there is intense commercial fishing. Dutch Harbor in the Aleutians, the port for commercial fish in the Bering Sea, had the highest tonnage of fish landings in the world in 2003, and provides 17% of Alaska's $811 million fish landings (2.3 million metric tons of fish; NOAA 2004). Over 90% of the world's fish catch comes from 10% of the world's oceans (including the Bering Sea; Waldichuk 1974). Thus, having baseline data from the Aleutians is important, even without any interest in Amchitka as an underground nuclear test site. Many of the fish examined are relatively sedentary (i.e., greenling, sole, rockfish), others regularly move short distances (i.e., cod simply move seasonally inshore and offshore along the same continental shelf region; Simenstad et al. 1977). Likewise, the seabirds are mobile, but resident year-round in the Aleutians, remaining within the same general area (White et al. 1977).

MATERIALS AND METHODS

Study sites

Amchitka Island (Fig. 1, 51° N lat; 179° E long), in the Aleutian chain of Alaska, is part of the Alaska Maritime National Wildlife Refuge system under the aegis of the U.S. Fish & Wildlife Service (U.S. FWS) and contains important ecological resources (Merritt and Fuller 1977; Burger et al. 2005). Amchitka Island is the only island where underground tests were conducted, making it difficult to assess and technically impossible to remove the residual radionuclide levels. It is unusual

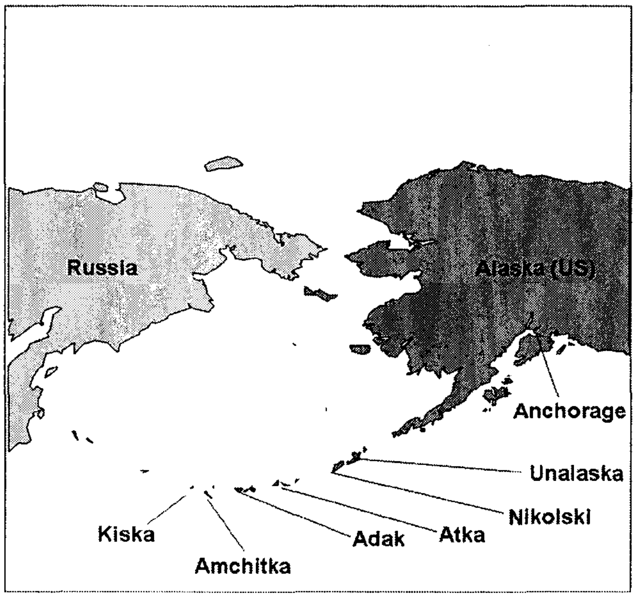

Fig. 1. Map of Amchitka and Kiska Islands in the Aleutian Chain of Alaska.

among DOE-contaminated sites because of its combination of remoteness, depth below ground surface of the contamination, and the importance of its ecological resources and seafood productivity that could be at risk if there were significant seepage of radionuclides from the test cavities to the marine environment. It is believed that most of the radioactive material from the Amchitka tests is trapped in the vitreous matrix created by the intense heat of the blasts, and is therefore permanently immobilized. This, however, is only an assumption, and some model results indicate that breakthrough of radionuclides into the marine environment will eventually occur (U.S. DOE 2002a and b; Powers et al. 2005). Data on radionuclide residues and conditions in the cavities remain classified.

After extensive consultation with the U.S. FWS and the Alaska Department of Environmental Conservation (ADEC), Kiska Island (51° N lat; 177° E long) was selected as the reference site. The reference had to be far enough away to be uninfluenced by Amchitka, yet close enough to share its geologic and biologic features. Both islands are bordered on the south by the North Pacific and on the north by the Bering Sea (Fig. 1). Kiska Island contains many of the same terrestrial and benthic environments and species composition as Amchitka (Burger et al. 2006b, c). The collection sites on Kiska and Amchitka were at least 130 km apart, a sufficient distance that most fish and birds remained within the vicinity of each island. Because of the ice free waters of the region, many of the sea birds do not migrate away from the area in the winter, but are residents in waters around their breeding island; many of the fish are sedentary (rock greenling, sole, rockfish) or move from inshore to offshore seasonally (cod) (Simenstad et al. 1977). Although it did not experience any underground nuclear tests, Kiska Island was occupied by both the Japanese and later the U.S. military during World War II, and Amchitka was a military base at the time.

Protocol

Fishes and birds were collected under appropriate federal and state permits from both Amchitka and Kiska Islands from late June–July 2004. Dolly Varden were only collected at Amchitka. Species collected, shown with their scientific names in Table 1, had a wide distribution, are reported as subsistence foods of the Unangan (Aleut) people, and some of the fishes are commercially important for the region. All specimens were tracked from field collection to their ultimate analytic destinations with chain-of-custody forms. Our overall protocol was to collect the same number of individuals of each species from near each of Amchitka's three test areas (*Long Shot, Milrow, Cannikin*) and from Kiska. Fishes were collected by Aleut fishermen using rods and reels, by the diving team using spears, and by a fisheries biologist on a National Oceanographic and Atmospheric Administration (NOAA) vessel by trawling. Adult birds were shot by the Aleut members of our team, and bird eggs and fledglings were collected from nesting colonies by hand (Burger et al. 2005; Jewett et al. 2006). The diving team consisted of four divers who worked in pairs at depths ranging from 5 to 27 m. The NOAA trawl was part of a standardized biennial fish survey in the Aleutians. Marine algae and invertebrates were collected at the same time (Burger et al. 2006b, d).

Fishes and birds were scanned on deck with a Ludlum Model 44-10 NaI gamma and a Ludlum Model

Table 1. List of species, with scientific name of organism collected at Amchitka and Kiska.

Common name	Scientific name	Eaten by Aleuts
Lower predators		
Dolly Varden	*Salvelinus malma*	Yes
Atka Mackerel	*Pleurogrammus monopterygius*	Yes
Red Irish Lord	*Hemilepidotus hemilepidotus*	Yes
Yellow Irish Lord	*Hemilepidotus jordani*	Yes
Northern Rock Sole	*Lepidopsetta polyxystra*	Yes
Rock Greenling	*Hexagrammos lagocephalus*	Yes
Pacific Ocean Perch	*Sebastes alutus*	Yes
Black Rockfish	*Sebastes melanops*	Yes
Common Eider adults (birds)	*Somateria mollissima*	Yes
Common Eider (eggs)	*Somateria mollissima*	Yes
Higher trophic level		
Walleye Pollock	*Theragra chalcogramma*	Yes
Glaucous-Winged Gull (bird)	*Larus glaucescens*	No
Glaucous-Winged Gull (eggs)	*Larus glaucescens*	Yes
Pigeon Guillemot	*Cepphus columba*	Yes
Tufted Puffin	*Fratercula cirrhata*	Yes
Top trophic level		
Pacific Halibut	*Hippoglossus stenolepis*	Yes
Pacific Cod	*Gadus macrocephalus*	Yes
Bald Eagle	*Haliaeetus leucocephalus*	No

44-9 alpha, beta, gamma pancake type halogen quenched G-M detector, both attached to a Ludlum Model 2241-2 ratemeter (Ludlum Instruments, Inc., 501 Oak Street, P.O. Box 810, Sweetwater, TX 79556). The instrument was factory calibrated on 23 May 2004, before expedition use. Scanning was accomplished to identify any specimens with high radioactivity that might contaminate the ship or personnel. Such specimens would have been stored in lead bags. However, none of the samples scanned during the expedition exceeded background readings, so the lead-bag contingency was not activated. The scanning was for safety only, and the results were not recorded.

Processing the fish and bird samples included filling in the chain of custody forms, and measuring, weighing, dissecting, packaging, labeling and freezing muscle and organs. The 100-g composites contained five similarly sized individual (±15%) fish or birds of one species from one sampling station that were prepared for analysis. To enhance the sensitivity for ^{137}Cs we prepared 1,000-g composites that required up to 24 individuals for small species (Table 2). Samples were prepared for analysis and 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). Since one of the study objectives was to assess whether there was any cause for concern about human health, we analyzed the tissues that would be expected to have the highest concentrations (bioaccumulate the highest levels) as a screen (e.g., muscle for ^{137}Cs and bone for ^{90}Sr).

Wipe samples of laboratory table tops and blenders were taken every day in the laboratory and read in a Ludlum Model 44-9 detector. No wipe samples were above background or blanks. The filter paper discs were then archived in scintillation vials, marked with the date and time taken, and were sent to Vanderbilt University for corroborative analysis by liquid scintillation analysis (LSA). All wipe samples were below the minimum detectable activity (MDA) level (see Powers et al. 2005, appendices).

Our radionuclide analysis design was based on trophic level considerations and sample availability and

Table 2. Radionuclide analysis conducted for human health screen. Total analyses run by species and radionuclide for 100 g samples. For cesium the number of 1,000 g samples is shown in parentheses. This is a complete species list for all analyses.

		Number of 100 g composite samples for analysis						
Species	Tissue	^{137}Cs	^{129}I	^{60}Co	^{152}Eu	^{90}Sr	Alpha[a] analyses	^{99}Tc
Lower predators								
Dolly Varden	muscle	8 (2)		8	8			
Atka Mackerel	muscle	1 (3)	1	1	1	2		2
Atka Mackerel	bone	1 (3)		1	1	1	1	
Red Irish Lord	muscle	8		8	8			
Yellow Irish Lord	muscle	15 (3)	4	15	15	4		4
Yellow Irish Lord	bone	3	1	3	3	3	3	
Northern Rock Sole	muscle	(2)						
Rock Greenling	muscle	23 (5)	9	23	23	4		4
Rock Greenling	bone	4	4	4	4			
Pacific Ocean Perch	muscle	2 (3)	2	2	2	2		2
Pacific Ocean Perch	bone	1	1	1	1	1	1	
Black Rockfish	muscle	12 (3)	4	12	12	4		4
Black Rockfish	bone	3	2	3	3	1	1	
Eider adults (birds)	muscle	4 (2)		4	4			
Eider (eggs)	eggs	6 (2)	3	6	6	3		3
Higher trophic level								
Walleye Pollock	muscle	(2)				2		2
Walleye Pollock	bone					2	2	
Gull (birds)	muscle	18 (2)	8	18	18	8		8
Gull (birds)	bone	8		8	8	8	8	
Gull (eggs)	egg	(2)						
Pigeon Guillemot	muscle	7	3	7	7	3		3
Pigeon Guillemot	bone	3		3	3	3	3	
Tufted Puffin	muscle	6 (2)	3	6	6	3		3
Tufted Puffin	bone	3		3	3	3	3	
Top trophic level								
Pacific Halibut	muscle	(4)						
Pacific Halibut	bone						4	
Pacific Cod	muscle	14 (14)	5	14	14	5		6
Pacific Cod	bone	3		3	3		14	
Bald Eagle	muscle	(2)						

[a] The actinides analyzed were ^{241}Am, ^{238}Pu, ^{239,240}Pu, ^{234}U, ^{235}U, ^{236}U, ^{238}U.

quantity. Desired detection sensitivity was initially set using a lifetime human cancer risk level of 10^{-6}, and sample numbers, counting times, and quantities were selected based on achieving detection levels well below this risk level. We used this process to determine how many samples we needed to run of each species (and thus how many to collect in the field). However, when we found that most samples were below the MDA for ^{137}Cs, we shifted to larger quantities (1,000 g) and longer counting times (72 h) to provide quantitative results useful for bioindicator selection.

Detailed analytic and quality assurance (QA) methods are provided for Vanderbilt (2005) and INL (INEEL 2005). Both laboratories shared and reviewed their proposed analytic methods. The Data Quality Objectives were established in advance of sample collection (CRESP 2003) and informed the establishment of target MDAs and numbers of samples. The MDA was derived following the method originally proposed by Currie (1968). Isotopes analyzed included 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 INL provided inter-laboratory validation (Powers et al. 2005). Samples below the MDA in one laboratory were below the MDA in the other. Samples above the MDA were within 20%, except for one sample that was re-analyzed.

Gamma emitters (^{137}Cs, ^{152}Eu, ^{60}Co) were analyzed using homogenized samples loaded into 0.5 L Marinelli beakers and counted on high purity germanium detectors (HPGe) calibrated to the container geometry for 24–72 h. ^{99}Tc was "trapped" on an Eichrom TEVA resin (Eichrom Technologies, Inc., 8205 S. Cass Ave., Suite 106, Darien, IL 60561) to preconcentrate the analyte and remove potential interferences prior to measurement by inductively coupled plasma-mass spectrometry (ICP-MS). The method uses rhenium spikes as a recovery surrogate. ^{129}I was analyzed with a HPGe detector optimized for low energy photon counting. Solid phase extraction was used for selective separation of strontium, americium, plutonium, and uranium using a serial configuration of Eichrom TRU resin and TEVA columns. The beta emitter, ^{90}Sr, was separated by sulfate precipitation and analyzed at Vanderbilt by LSC of its decay product ^{90}Y. The actinides (uranium, plutonium, and americium) were quantified radiochemically using co-precipitation with neodymium fluoride, followed by alpha spectroscopy (INEEL 2001 Method ACM 3816).

Counts were adjusted for background counts, and evaluated against detection limits based on the method of Currie (1968). All values are presented in Bq kg^{-1}, wet weight. Average MDAs ranged from 0.029 Bq kg^{-1} for ^{241}Am to 0.102 Bq kg^{-1} for ^{235}U. Initially for gamma emitters, 100-g samples were counted for 24 h, but all results were below the MDA; thus, to enhance sensitivity, 1,000-g samples were analyzed for 72 h. MDAs for ^{137}Cs ranged from 5.57–6.25 Bq kg^{-1} for 100-g samples and 0.18–0.36 Bq kg^{-1} for 1,000-g samples (Table 3).

Table 3. Examination of predators for use as bioindicators for ^{137}Cs. Given are the values in Bq kg^{-1} (wet weight) for 1,000 gram samples only. For comparative purposes, all predators are listed. Trophic levels were based on previous information. Low trophic levels eat mainly invertebrates. Medium trophic levels eat small fish. Higher trophic levels eat medium-sized fish. These distinctions are not absolute; for example, smaller or younger individuals feed at a lower trophic level than older or larger individuals of the same species.

Species	Number of 1,000 g analyses	Mean MDA Bq kg^{-1} (wet weight)	Percent above the MDA	All values above the MDA are shown below in Bq kg^{-1} (wet weight)
Low trophic level	(Total number of individuals)			
Dolly Varden	2 (46)	0.12	100	0.697, 0.780
Atka Mackerel	3 (30)	0.10	33	0.102
Rock Greenling	5 (37)	0.25	0	—
Yellow Irish Lord	3 (15)	0.10	33	0.131
Northern Rock Sole	2 (10)	0.10	0	—
Pacific Ocean Perch	3 (15)	0.12	33	0.108
Black Rockfish	3 (31)	0.10	100	0.189, 0.130, 0.111
Eider (birds)	2 (10)	0.23	0	—
Eider (eggs)	2 (29)	0.10	0	—
Medium trophic level				
Walleye Pollock	2 (10)	0.32	50	0.461
Gulls (birds)	2 (18)	0.26	50	0.094
Gulls (eggs)	2 (14)	0.24	0	—
Tufted Puffin	2 (15)	0.12	0	—
Top trophic level				
Pacific Halibut	4 (14)	0.15	75	0.190, 0.315, 0.446
Pacific Cod	14 (71)	0.28	57	0.176, 0.200, 0.209, 0.315, 0.323, 0.399, 0.472, 0.602
Bald Eagle	2 (2)	0.66	0	

Sample results above the MDA are referred to as "detects" and values below the MDA are "non-detects" (Helsel 2005). For calculating the means, "non-detects" were assigned half their respective MDA, a common but not ideal convention in environmental chemistry (Helsel 2005). This is the least biased statistical approach, although if the concentration of an analyte is truly zero, it introduces an upward bias (Gochfeld et al. 2005). Because of the large number of values for composites that fell below the MDA, the actual values for "detects" is given in Table 3.

RESULTS

The raw data tables for this study are available at http://www.cresp.org/Amchitka/RadionuclideData.html. For analytic quality assurance see Vanderbilt (2005) and INEEL (2005). For fishes and birds, there were no values above the MDA for several radionuclides, including ^{129}I, ^{60}Co, ^{152}Eu, ^{90}Sr, and ^{99}Tc. The remainder of the paper focuses on radiocesium (^{137}Cs) and the actinides.

Actinides

There were significant inter-specific differences among the actinide radionuclides, in terms of percent of samples above MDA and activities measured for "detects." For fishes, 73 composites of 234 (31%) were above the MDA (Table 4). For birds, only 3 composites of 98 (3%) had results above the MDA. There were no composites above the MDA for ^{238}Pu or ^{236}U in fishes or birds. For ^{241}Am, there were only three out of 36 composites above the MDA for fishes and none for birds. Only three samples exceeded the MDA for ^{239,240}Pu (one each for Walleye Pollock, Halibut, and Pigeon Guillemot), and only four composites were above the MDA for ^{235}U (one each for Atka Mackerel, Black Rockfish, Walleye Pollock, and Pacific Cod) (Table 4).

^{234}U and ^{238}U, radionuclides that are primarily natural in origin, had the greatest number of samples that were above the MDA. All fish species had some composites above the MDA for both ^{234}U and ^{238}U, but birds only had detectable concentrations of ^{238}U. Most composites of Halibut and Pacific Cod had values above the

Table 4. Number of composites, number above the maximum actinides level (MDA) and highest value above the MDA for radionuclides in biota from Amchitka and Kiska. Top line is the number of composites above the MDA/number of composites. Lower line for each species is the maximum level in Bq kg^{-1} wet weight. For Source, A = anthropogenic and N = naturally occurring. X = no analyses conducted.

Species	^{137}Cs	^{241}Am	^{238}Pu	^{239,240}Pu	^{234}U	^{235}U	^{236}U	^{238}U
Main source	A	A	A	A	N	A	A	N
Lower predators								
Dolly Varden	3/3 0.78	X	X	X	X	X	X	X
Atka Mackerel	1/3 0.102	0/1	0/1	0/1	1/1 0.963	1/1 0.065	0/1	1/1 0.94
Rock Greenling	0/5 0.16	X	X	X	X	X	X	X
Yellow Irish Lord	1/3 0.25	0/3	0/3	0/3	3/3 0.567	0/3	0/3	3/3 0.607
Northern Rock Sole	0/2	X	X	X	X	X	X	X
Pacific Ocean Perch	1/3 0.108	0/1	0/1	0/1	1/1 0.655	0/1	0/1	1/1 0.654
Black Rockfish	3/3 0.189	1/1 0.029	0/1	0/1	1/1 2.18	1/1 0.116	0/1	1/1 1.83
Eider (birds)	0/2	X	X	X	X	X	X	X
Eider (eggs)	0/2							
Higher trophic level								
Walleye Pollock	1/2 0.461	1/2 0.022	0/2	1/2 0.02	2/2 0.857	1/2 0.053	0/2	2/2 0.779
Gulls (birds)	1/2 0.094	0/8	0/8	0/8	0/8	0/8	0/8	1/8 0.449
Gulls (eggs)	0/2	X	X	X	X	X	X	X
Pigeon Guillemot	X	0/3	0/3	1/3 0.312	0/3	0/3	0/3	0/3
Tufted Puffin	0/2	0/3	0/3	0/3	0/3	0/3	0/3	1/3
Top trophic level								
Pacific Halibut	3/4 0.445	0/7	0/7	1/7 0.017	6/7 1.2	1/7 0.048	0/7	7/7 0.179
Pacific Cod	8/14 0.602	1/21 0.015	0/21	0/21	17/21 0.29	0/21	0/21	17/21 0.2575
Bald Eagle	0/2	X	X	X	X	X	X	X

MDA for ^{234}U (means ± standard error of 0.289 ± 0.15 and 0.109 ± 0.01 Bq kg^{-1} wet weight, respectively) and ^{238}U (means ± standard error of 0.21 ± 0.12 and 0.112 ± 0.01 Bq kg^{-1}, respectively).

For most radionuclides the relatively low percentage of composites above the MDA precluded examining inter-island and inter-specific differences. However, it is clear that a higher percent of fishes accumulated actinides than birds, and that all trophic levels of fishes had detectable concentrations of the naturally-occurring ^{234}U and ^{238}U. Halibut and cod combined had sufficient composites above the MDA to examine inter-island differences for the naturally-occurring uranium isotopes. Using Kruskal Wallis non-parametric One-Way Analysis of Variance there were no significant inter-island differences in concentrations for ^{234}U ($X^2 = 1.22, p = 0.27$) or ^{238}U ($X^2 = 2.29, p = 0.138$). Nor were there inter-island differences in the MDAs for ^{234}U ($X^2 = 1.12, p = 0.29$) or ^{238}U ($X^2 = 0.93, p = 0.33$).

There were also no significant differences between cod and halibut for ^{234}U in either of the concentrations ($X^2 = 1.43\ p = 0.23$) or the MDAs ($X^2 = 0.77, p = 0.38$). Also, there were no differences between cod and halibut for ^{238}U in concentrations ($X^2 = 0.44$, $p = 0.51$) or MDAs ($X^2 = 0.31$, $p = 0.58$). Combined, these data indicate that the concentrations of naturally-occurring uranium isotopes were higher than the anthropogenic radionuclides, and that there were no differences among islands or species for cod and halibut, the species with the largest number of composites analyzed and with detectable concentrations.

Radiocesium

None of the ^{137}Cs values for fishes and birds were above the MDA for the 100-g samples counted for 24 h ($N = 173$ composites), so 1,000-g samples were then counted for 72 h (Table 3). Most of these samples were either below or close to the MDA. Except for one gull sample, all 1,000-g bird samples were below the MDA ($N = 12$ samples representing 88 birds or eggs).

The percent of values above the MDA for fishes ranged from 0% (Rock Greenling and Northern Sole) to 100% (for Dolly Varden, Black Rockfish). The highest values were for Dolly Varden, Walleye Pollock, Pacific Halibut, and Pacific Cod. The mean ± standard error for ^{137}Cs value in the species with the most composites are as follows (values below the MDA were entered as half the MDA): Black Rockfish = 0.143 ± 0.04, Halibut = 0.24 ± 0.14, Pacific Cod = 0.29 ± 0.20, Walleye Pollock = 0.311 ± 0.311, and Dolly Varden = 0.74 ± 0.04 Bq kg^{-1}. Pacific Cod, the species with the most composites analyzed, did not show any difference in ^{137}Cs as a function of the size of the individual fish sampled, although the mean total fish lengths in composites varied from 42 to 92 cm.

When fish were compared, there was not a significant difference in either the percent of values above the MDA or in the mean value of ^{137}Cs for fish collected at Amchitka and Kiska (Table 5). Dolly Varden was excluded from this analysis because it was collected only from Amchitka.

DISCUSSION

Establishing baselines

Radionuclides in the environment come from natural sources, from fallout from historic nuclear weapons testing (Duran et al. 2004), from nuclear facility accidents (Baeza et al. 1994; Cooper et al. 1998; Livingston and Povinec 2000), and from discarded nuclear wastes (Fisher et al. 1999; IAEA 1999), as well as other nuclear accidents (UNSCEAR 2000; Sanchez-Cabeza and Molero 2000; Amundsen et al. 2002; Aumento et al. 2005). A total of 543 atmospheric nuclear weapons tests were conducted from 1945–1980, primarily in the Northern Hemisphere (UNSCEAR 2000), although radionuclides have been found in fishes from as far away as the Antarctic (Marzano et al. 2000). The disposal of large quantities of radioactive wastes in the Arctic Seas by the former Soviet Union prompted interest in radionuclides in the Bering Sea ecosystem as well (Fisher et al. 1999). Early on, Moscati and Erdmann (1974) recognized the implications of accidental releases of radioactivity into the marine environment, often noting the importance of

Table 5. Comparison of ^{137}Cs levels in fishes between Amchitka and Kiska. Comparison for Black Rockfish, Pacific Halibut, Pacific Cod, Walleye Pollock, Pacific Ocean Perch, Atka Mackerel and Yellow Irish Lord (1,000 gram samples only, fish species where cesium was detected in at least one composite sample). Dolly Varden is not included because samples came only from Amchitka.

	Amchitka	Kiska	Statistical test
Number of composites	20	12	
Number positive (%)	10	8	0.84, $p < 0.36$
Mean ± SD (using 1/2 MDA for non detects)	0.152 ± 0.160	0.184 ± 0.139	0.61, $p < 0.43$
(range)	(<0.05–0.602)	(0.069–0461)	
Mean ± SD for detects only	0.257 ± 0.167	0.252 ± 0.120	0.08, $p < 0.93$

^{137}Cs, ^{239}Pu, and other radionuclides in producing adverse health impacts.

Because of the importance of fish and shellfish consumption throughout the world, and the occurrence of atmospheric deposition into marine environments, a number of monitoring programs have been established in Asia (Duran et al. 2004), in the Sea of Japan (Togawa et al. 1999; JCAC 2003, 2004), in the Irish Sea (RPII 2003, 2004), and in the Black Sea (Bologa 2000). Other biomonitoring programs have been established to evaluate possible exposure from nuclear facility operations (Poon and Au 2002; Shinohara 2004), as well as exposure due to dumping by the former Soviet Union (Togawa et al. 1999; Yamada et al. 1999; Matishov et al. 2001). Some of this biomonitoring is ongoing near decommissioned nuclear power plants and reprocessing plants (e.g., Sellafield in the UK, Sanchez-Cabeza and Molero 2000; and in Taiwan, Hung et al. 1998). The present data set can serve as a baseline for the Northern Pacific/Bering Sea ecosystem for possible future seepage from the Amchitka underground nuclear test shots and for nuclear waste dumping or accidents from the former Soviet Union. While fishes and birds often have relatively low concentrations of radionuclides (Skwarzec 1997), they are key species both for the ecosystem and (except for the eagle) for both subsistence and commercial human consumption. All of the fishes and most birds we studied are regularly eaten by native people of the Aleutians (Hamrick and Smith 2003).

Inter-specific and inter-island differences

There were inter-specific differences in concentrations of radionuclides for fishes and birds: 1) for actinides, 31% of the fish composites exceeded the MDA, while only 3% of the bird composites had concentrations above the MDA; 2) for ^{234}U and ^{235}U, some fish composites were above the MDA, but no bird composites were above the MDA; 3) for ^{238}U, a higher percent of composites for fishes were above the MDA than for birds; 4) for ^{137}Cs, 46% of the composites were above the MDA for fishes, compared to 8% for birds, and 5) some species of fishes had no composites above the MDA, while others had up to 100% above the MDA. Thus, there were inter-specific differences in the radionuclide concentrations both among fishes and birds, and within fish species. For birds, ^{234}U was below MDA even when ^{238}U was detectable. This unexpected result is most likely related to the generally low levels of ^{238}U in the bird tissues, not to some fundamental toxicokinetic difference between fish and birds.

Inter-specific differences in concentrations of contaminants, including radionuclides, are usually due to differences in trophic level (Denton and Burdon-Jones 1986; Jackson 1991; Kasamatsu and Tshikawa 1997; Watras et al. 1998; Wiener and Spry 1996; Wiener et al. 2003; Burger et al. 2001), size and age (Lange et al. 1994; Bidone et al. 1997; Burger et al. 2001; Pinho et al. 2002; Green and Knutzen 2003), and habitat (Burger et al. 2002). In general, concentrations are higher in species that are larger, older, and at a higher trophic level. Sanchez-Cabeza and Molero (2000) found higher ^{137}Cs concentrations in pelagic fishes, compared to more sedentary demersal fishes. These differences may be partly due to some of the larger pelagic fishes being higher on the food chain. Trophic level relationships have been previously reported for ^{137}Cs (Pentreath 1973).

In this study, species that were higher on the trophic scale and were large, such as pollock, cod, and halibut, generally had higher concentrations than the smaller fishes and than birds, several of which consume small fish (Puffins, Guillemots) or invertebrates (Eiders). However, ^{137}Cs concentrations were highest in Dolly Varden, which is a smaller fish than most others examined in this study, and is short-lived (up to 8–16 y; ADFG 1994), although the differences were not great. Halibut range in age up to 55 y, and Pacific Cod up to 25 y (Merrell 1977; Munk 2001). As large predators within the marine ecosystem, both cod and halibut eat other medium-sized fishes. There was no relationship between size or weight of cod and ^{137}Cs concentrations, however, unlike the fishes examined by Kasamatsu and Tshikawa (1997). This lack of difference may be due either to small sample sizes, or to lower concentrations of ^{137}Cs in the Amchitka/Kiska fish. However, Kasamatsu and Tshikawa (1997) also found no relationship of ^{137}Cs with size for 12 of the 16 species of fish they examined. The percent of composites above the MDA for ^{239,240}Pu was similar for birds and fish, and they were very low.

There were no inter-island differences in this study for fishes, and the concentrations were relatively low for all radionuclides. One potential source of radionuclides could have been seepage from the underground nuclear test cavities at Amchitka, but we found no evidence that this has occurred.

Temporal patterns for ^{137}Cs

Radiological analyses immediately after the *Cannikin* detonation (5 megatons in 1971) found no contamination in marine fishes (Held et al. 1973). The concentrations of ^{137}Cs in fishes in 2004 could be compared to those found immediately after the nuclear tests (Table 6). For all fishes there was a decline in the concentrations of

Table 6. Temporal patterns of ^{137}Cs for fish from Amchitka, compared to 1967–1968 (Isakson and Seymour 1968), and 1965–1974 (Seymour and Nelson 1977). Given are the means in Bq kg^{-1} (wet weight). Some have been calculated from dry weight data in the literature using the average percent moisture content for that species. The percent of samples above the MDA is given in parentheses.

Species	1967–1968	1965–1975	This study
Dolly Varden	not given	7.2 (2.4%)	0.74 (100%)
Rock Greenling	0.89 (100%)	0.523 (not given)	< MDA of 0.29 (0%)
Walleye Pollock	0.96 (100%)	not given	0.31 (50%)
Pacific Halibut	1.24 (50%)	0.58 (not given)	0.24 (75%)
Pacific Cod	1.14 (100%)	not given	0.29 (57%)

^{137}Cs from the time of the tests until the present. The decline ranged from 60% in pollock, to over a 75% decline for halibut and cod. The individual fishes examined in this study from 2004 were not present at the time of the last underground nuclear test (1971), since no very large halibut were collected, and the oldest cod (aged by otoliths) was less than 10 y old. Mainly, radioactive decay of ^{137}Cs from earlier atmospheric testing (radiologic half-life of 30 y; biologic half-life about 110 d) can account for the lower concentrations.

Worldwide, the concentrations of radionuclides in seawater and biota have been declining since the period of intense above-ground nuclear testing in the 1950's and 1960's; this includes fishes (Waldichuk 1989; Noshkin et al. 1997; Duran et al. 2004). In the Marshall Islands, several radionuclides decreased significantly in fishes between 1958 (the end of above-ground nuclear testing there) and 1994, due to both radioactive decay and other natural processes (recycling or environmental decay; Noshkin et al. 1997). Similar declines have occurred in the Irish Sea (Kanisch et al. 2000) where there are multiple sources.

Geographical comparisons

One of the objectives of this study was to compare the concentrations of radionuclides found in the fishes and birds at Amchitka and Kiska with those from other regions in the Northern Hemisphere, both uncontaminated sites and contaminated sites such as the Irish Sea. Part of the difficulty with comparisons of results from this study with other studies reported in the literature is that the suite of radionuclides and associated detection limits examined varies by site and, of course, the objectives of the study reported.

Actinides. There are fewer data for actinides in fishes and birds from the Northern Hemisphere than there are for ^{137}Cs. However, Hunt and Smith (1999) noted that actinides released from Sellafield constitute a major pathway to seafoods consumed by humans. Information on actinides may be less available in the literature because of inconsistency in the suite of radionuclides examined. This inconsistency is partly due to the great expense of radionuclide analysis, an emphasis on the radionuclides of high health concern, such as ^{137}Cs and ^{90}Sr, and because of local efforts to analyze only for the most likely radionuclides released from local nuclear activities, dumping, or accidents.

The concentrations of actinides from Amchitka and Kiska for fishes were an order of magnitude lower for ^{241}Am, higher for ^{238}U, similar for ^{238}Pu, and higher for ^{239,240}Pu, compared to other Northern Hemisphere sites (Table 7). From the Japanese coast, Yamada et al. (1999) reported ^{239,240}Pu concentrations of 0.0004 Bq kg^{-1} in fishes, indicating that the fishes were not currently affected by nuclear waste dumping by the Former Soviet Union. Some studies report ^{239}Pu alone (Marzano et al. 2000), making comparison difficult.

Rollo et al. (1992) examined mainly naturally-occurring radionuclides in the marine environment around the United Kingdom (UK), and found the following maximum concentration for Atlantic Cod: ^{234}U = 0.011 Bq kg^{-1} (compared to 0.29 Bq kg^{-1} for Pacific Cod at Amchitka) and ^{238}U = 0.009 Bq kg^{-1} (compared to 0.258 Bq kg^{-1} at Amchitka). Thus the concentrations of the naturally-occurring radionuclides were higher at Amchitka/Kiska than in the marine environment around the UK. In some places, such as the UK, radioactivity from natural sources is by far the most significant source of exposure to communities remote from nuclear sites (CEFAS 2004). Similarly, the anthropogenic ^{235}U of 0.0007 Bq kg^{-1} in cod around the United Kingdom (Rollo et al. 1992) was lower than the values for Amchitka. Around Amchitka all values of ^{235}U

Table 7. Geographical comparisons of some actinides (Bq kg^{-1}, wet weight) for birds and fish from the Irish Sea and other Northern Hemisphere locations.[a] Given are means or ranges of means when multiple data are available. Some results were originally reported in pCi or in dry weight and have been converted.

Group	Irish Sea	Other sites	Amchitka/Kiska (2004)[b]
Birds			
^{241}Am			< MDA
^{238}U			< 0.45
^{238}Pu			< MDA
^{239,240}Pu			0.31
Fish			
^{241}Am	0.0001–0.23	0.0012	0.029
^{238}U	0.003–.005	0.008–0.015	0.94
^{238}Pu	< MDA–0.02	< MDA	< MDA
^{239,240}Pu	< MDA–0.08	< MDA–0.07	0.02

[a] Sources: Ryan et al. (2003), RPII (2003, 2004), CEFAS (2003, 2004).
[b] The maximum value is given because the concentrations were so low and close to the MDA.

for cod were below the MDA, but for Walleye Pollock the high value was 0.053 Bq kg^{-1}.

There are even fewer data on actinides in birds from the Northern Hemisphere.

Radiocesium. Table 8 lists the mean concentrations for ^{137}Cs for some representative fishes and birds from the Northern Hemisphere. Several conclusions can be drawn from these data for ^{137}Cs. The ^{137}Cs mean for fishes from Amchitka (mean of 0.04 to 0.74 Bq kg^{-1}, depending upon species) was similar to concentrations for the Northern Hemisphere generally (mean of 0.22 Bq kg^{-1}, range up to 0.33 Bq kg^{-1}), but lower than the concentrations reported in fishes from the Irish Sea (mean of 4.64 Bq kg^{-1}, range up to 13 Bq kg^{-1}). More recently, Duran et al. (2004) reported median ^{137}Cs values of 0.20 Bq kg^{-1} in fish from the Asian-Pacific and Yamada et al. (1999) reported ^{137}Cs concentrations of 0.30 Bq kg^{-1} from the Japanese coast. Concentrations are similar from the Southern Hemisphere: Godoy et al. (2003) found ^{137}Cs concentrations to be 0.19 Bq kg^{-1} in Brazil. Concentrations can be much higher in fishes living near nuclear waste dump sites; fishes from the Farallon Islands nuclear waste dump had mean muscle concentrations of 1.1 Bq kg^{-1} (Suchanek et al. 1996).

There are a number of studies that report values for cod (Table 9), and the means range from 0.2 Bq kg^{-1} for the Arctic and the English Channel, to 0.3 Bq kg^{-1} for the Barents Sea, Norway, and North Atlantic, to 6.44 Bq kg^{-1} in the Irish Sea and 8.86 Bq kg^{-1} for the Baltic Sea. Concentrations in cod in the Barents and Kara Seas can also range as high as 4 Bq kg^{-1} ^{137}Cs (Matishov et al. 1995, 2001). At Amchitka, Pacific Cod were at the low end of this range (mean of 0.29 Bq kg^{-1}).

The only value above the MDA for birds from Amchitka (0.094 Bq kg^{-1}) was below the average concentration for other Northern Hemisphere sites (mean of 1.62 Bq kg^{-1}, range up to 5.6 Bq kg^{-1}), as well as being considerably below the mean for birds in the Irish Sea (mean of 124.8 Bq kg^{-1}, range up to 613 Bq kg^{-1}). Dietz et al. (2000) reported lower concentrations of ^{137}Cs in birds from Greenland (means of 0.2 and 0.5 Bq kg^{-1}).

Overall, the concentrations of cesium in fishes and birds from Amchitka were similar to those of other uncontaminated sites in the Northern Hemisphere and were well below those from the contaminated Irish Sea. In any case, all of the concentrations of ^{137}Cs in all the organisms from the Amchitka study were well below the allowable concentrations in foods as set by the European Economic Community (600 Bq kg^{-1}; EEC 1986).

Table 8. Geographical comparisons of ^{137}Cs (Bq kg^{-1}, wet weight) for birds and fish. Concentrations or ranges of mean concentrations are compared from the contaminated Irish Sea with marine birds and fish from other Northern Hemisphere[a] locations. Data from other locations are primarily from 1999 to the present. Some results were originally reported in pCi or in dry weight and have been converted.

Group	Irish Sea	Other sites	Amchitka/Kiska (2004)
Birds			
Mean level	124.8	1.62	< MDA
Range	9–613	< MDA–5.6	0.08–0.94
Number of analyses	15	15	12
Fish			
Mean level	4.64	0.22	0.04–0.74[b]
Range	0.31–13	0.04–0.33	< MDA–0.78
Number of analyses	15	718	44

[a] The Northern Hemisphere data comes from CEFAS (2003, 2004), RPII (2003, 2004), NRPA (2003, 2004), JCAC (2003, 2004), Hong Kong Observatory (2000, 2003, 2004), and Matishov and Matishov (2004). The Irish Sea data were extracted from RPII (2003, 2004), CEFAS (2003, 2004), and BNG (2004).

[b] For different fish species.

CONCLUSION

The only radionuclides above the minimum detectable activity (MDA) were ^{137}Cs, ^{241}Am, ^{239,240}Pu, ^{234}U, ^{235}U, and ^{238}U for a wide range of fishes and birds. There were significant differences in ^{137}Cs as a function of species, but not location for top predatory fish. Most of the fishes (8 of 10 species) had ^{137}Cs concentrations above the MDA. However, Glaucous-winged Gull was the only bird species that had a value above the MDA. The highest concentrations of ^{137}Cs were in Dolly Varden and Pacific Cod. ^{137}Cs concentrations for all fishes and birds were low and close to the MDA, providing no evidence of any seepage from the Amchitka test shots. In aggregate, fish had 73 of 234 (31%) actinide analyses above the MDA, while birds had only 3 of 98 (3%) actinide composite analyses above the MDA, suggesting that fishes are better accumulators, and are thus more useful bioindicators to provide any early warning.

^{234}U and ^{238}U, nuclides that are primarily natural in origin, had the highest detection rates, and there were no significant differences in mean concentrations between Amchitka and Kiska. Thus we did not document evidence of higher levels or leakage at Amchitka. The concentrations of all radionuclides examined at Amchitka are similar to those of other uncontaminated Northern Hemisphere sites, and are lower than those reported for fishes and birds from the Irish Sea. The values for all samples in this study fell below the European Economic Community allowable health guidance concentrations of 600 Bq ^{137}Cs kg^{-1} fresh weight (EEC 1986). The data from this study provide reassurance that the concentrations of radionuclides in fishes

Table 9. Geographical comparison for ^{137}Cs for representative marine fishes in the Irish Sea and other Northern Hemisphere sites, with Amchitka data shown for comparison. All values have been converted where necessary to Bq kg^{-1} wet weight. Values below the MDA were converted to half the MDA for computing the mean value. Data are primarily from 1999 to the present.

Location/Sea	Species	Concentration	# (pooled)	Reference
Japan	Tilefish	0.12	2	Japan Chemical Analysis Center (2003)
	Greenling	0.12	2	
	Flounder	0.07	12	
	Rockfish	0.09	4	
	Mackerel (various)	0.12	18	
Arctic	Sculpin	0.3	10	Jensson et al. (2004)
	Flounder	0.3	6	Matishov and Matishov (2004)
	Pacific Cod	0.2	394	
	Haddock	0.3	65	
Hong Kong	Melon Coat	0.07	11	Li and Yeung (2004)
	Hair Tail	0.09	19	Hong Kong Observatory (2003)
	Bartail Flathead	0.06	11	
Barents Sea	Atlantic Cod	0.29	53	Gafvert et al. (2003)
	Haddock	0.2	10	CEFAS (2003 and 2004)
				Ryan et al. (2001)
North Sea	Atlantic Cod	0.38	21	CEFAS (2003 and 2004)
	Haddock	0.2	10	Gafvert et al. (2003)
	Plaice	0.21	19	
Norwegian	Atlantic Cod	0.32	20	Gafvert et al. (2003)
	Saithe	0.27 to 0.64	4	CEFAS (2003 and 2004)
	Mackerel	0.14	4	Ryan et al. (2001)
N. Atlantic	Atlantic Cod	0.28	3	CEFAS (2003 and 2004)
	Plaice	0.36	3	Gafvert et al. (2003)
	Haddock	0.47	3	
	Mackerel	0.09	5	
Channel	Atlantic Cod	0.2	8	CEFAS (2003 and 2004)
	Plaice	0.06	16	
	Mackerel	0.19	8	
Irish	Atlantic Cod	6.44	75	Ryan et al. (2001)
	Plaice	3.77	60	CEFAS (2003 and 2004)
	Mackerel	0.31	39	
	Flounder	11	19	
	Haddock	1.1	10	
Baltic	Atlantic Cod	8.86	7	CEFAS (2003 and 2004)
Amchitka	Dolly Varden	0.74	2	Powers et al. (2005, 2006)
	Pacific Cod	0.29	14	(1,000 g samples)
	Pacific Halibut	0.24	4	
	Black Rockfish	0.14	3	

and birds, all of which are important commercial or subsistence species, are currently below levels established as safe for human consumption (see above and Powers et al. 2005). The database can be used for bioindicator selection as part of a long-term biomonitoring plan for the region. These data are fundamental to establishing a baseline for the Bering Sea ecosystem.

Finally, transparency, trust, and sustainability, as well as the public's participation in decision-making, helped shape the Amchitka investigation. These will be needed to shape the public policy agenda with respect to nuclear wastes in the future (Omenn 2001; Florig 2001). Trust is particularly an issue with chemical and nuclear wastes (Slovic 1987), and some people feel that the government and experts downplay issues of nuclear safety and risk (Ahearne 2001; Thomas 2001). The CRESP study benefited from stakeholder input at several levels. Transparency encouraged stakeholders to participate in the planning, participation, interpretation, and acceptance of the results, improving both the quality and usefulness of the study.

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