Radionuclides In Marine Biota

SUMMARY

The Amchitka *Science Plan* had as its main objectives assuring the safety of foods, and the ecosystem, reducing uncertainty in the groundwater and risk assessment models, and providing information useful for establishing a long-term stewardship plan for Amchitka Island. This chapter provides the quantitative data that will inform these key aspects of the *Science Plan*. In this chapter we examine the following:

1. The levels of radionuclides in a range of marine biota collected near the Amchitka test shots and at Kiska (the reference site).
2. Differences in the concentrations of radionuclides among the target species collected.
3. Differences in the concentrations of radionuclides between biota collected near the test shots at Amchitka, and at Kiska.
4. Differences in the concentrations of radionuclides between biota collected among the Amchitka test shots.
5. A comparison of the levels of radionuclides in biota from Amchitka with those from the general region and other northern Hemisphere places.

The importance, interpretation, and implications of these findings are mentioned briefly at the end of this chapter, but are explored more fully in chapter 12. Values above the Minimum Detection Activity level (MDA) will be referred to as “detects”. Values below the MDA will be called “non-detects”. For Iodine-129 (no detects of 71 samples), cobalt-60 (no detects of 173), europium-152 (no detects of 173), strontium-90 (none out of 85) and technetium-99 (none out of 60), all values were below the minimum detection level, and therefore it was not possible to compare either among species or among locations. For uranium-236 and americium-241 there were a few detectable values (detects). For cesium-137 there were values above the minimum detectable activity (hereafter referred to as detects), but only for 1000 g samples. Uranium -234, 235 and 238 were present in most of the samples analyzed. Time and money constraints limited the number of analyses that could be performed and required optimizing the selection of samples for each analytic stream.

There were significant differences among species in percent of detects for cesium-137. High trophic level organisms (Sea Lion, Octopus, Pacific Cod, Halibut, Black Rockfish) had significantly more detects than all others. Overall, there were no differences in the percent of detects between Amchitka and Kiska for cesium-137, and there were too few detects to compare among Amchitka sites. However, when four, top-level predatory fish were examined, Cs-137 levels were significantly higher at Kiska than at Amchitka

(P<0.056)

There were a number of samples with detectable levels for the actinides, mainly those that are natural in origin (U-238 and U-234). There were species differences, with the primary producers (kelp) and filter feeders (Jingles) having significantly higher percentages of actinide detects than the predators. Of the algae, *Ulva* had the fewest detects. There were no differences in the proportion of actinide detects between Kiska and Amchitka when all analyses are combined. When the algae (the species group with the highest percentage of detects) are considered separately, there are also no significant differences between Amchitka and Kiska, except for Pu-239,240. There was a higher proportion of detectable values for Pu-239,240 at Amchitka than at Kiska. Although there were also significant differences in the mean levels of Pu-239,240 between Amchitka and Kiska, the differences were small, and well below any human health guidance levels.

INTRODUCTION

Understanding the potential risks from possible radionuclide releases following the underground test shots conducted at several sites is a high priority of the Department of Energy. Much of the research examining the risks to the food supply and to ecosystems from radionuclides comes from accidents, such as the Chernobyl disaster in 1986 (Arvela et al. 1990 Svadlenkova et al. 1996, Jagoe et al. 1997), leading to global fallout (AMAP 2003, Dahlgaard et al. 2004), and from studies near existing nuclear power plants or fuel reprocessing facilities (Axelrod 2004). In the case of Chernobyl, radionuclides entered both local and distant food chains as a result of atmospheric deposition (Arvela et al. 1990 Svadlenkova et al. 1996, Jagoe et al. 1997). The DOE has a responsibility to determine whether there is currently, or could be in the future, any risk to humans and the ecosystems surrounding their nuclear tests sites (Crowley and Ahearne 2002).

The *Amchitka Independent Assessment Science Plan* (Appendix 1E) was developed by CRESP (2003) to determine whether there are currently increased radiation health risks related to the underground nuclear test shots to organisms residing around Amchitka Island, and to consumers of these organisms, and to provide a baseline for future monitoring as part of long-term stewardship for the island. The *Science Plan* was developed for Native communities, U.S. Fish & Wildlife Service, the Alaskan Department of Environmental Conservation, U.S. Department of Energy and other stakeholders (Burger et al. 2005). Refinements in the *Science Plan*, suggested by these stakeholders, continued until the expeditions in the summer of 2004.

One of the main purposes of the *Science Plan* was to address public and agency concerns that potential residual radionuclides from the nuclear tests may enter the food chain, causing adverse ecological and human health effects. The *Science Plan* addressed the following questions: 1) Are the foods safe?; 2) Is the biota of Amchitka currently contaminated?; 3) Are the levels of contaminants high enough to pose harm to species or the ecosystem?; 4) What are the current risks?; 5) What species are appropriate for long-term monitoring?; 6. How does the biodiversity at the reference site (Kiska) compare with Amchitka (addressed mainly in chapter 10).

Null Hypotheses

Some of these questions can be answered directly from analysis of the radionuclide data in the biota collected during the two expeditions in the summer of 2004. As scientists, we addressed these questions by examining a number of null hypotheses (the opposite of predicting differences). A null hypothesis is a statement that posits no difference between variables of interest (e.g. kelp vs sea urchins, Amchitka vs Kiska). The main null hypotheses addressed in this chapter are: 1) There are no differences in radionuclide levels among different species, 2) There are no differences in radionuclide levels in biota in the marine environment transects from near the Amchitka Shot cavities compared to the reference site (Kiska), 3) There are no differences in the radionuclide levels in biota among the three test sites on Amchitka, shots, and 4) There are no differences in levels of radionuclides in Amchitka biota and those analyzed elsewhere. Having formulated the null hypotheses, the relevant data are analyzed with statistical tests. If statistically significant differences are found, the null hypothesis is rejected, and the alternative hypothesis ("a difference exists") is accepted. From these data and analyses, we can then address the major objectives listed above (discussed further in chapter 12). Almost all values for some radionuclides were below the minimum detection level, and therefore it was not possible to compare either among species or among locations.

This chapter provides the quantitative data that will inform key aspects of the *Science Plan*. In this chapter we address the following questions:

1. What are the levels of radionuclides in a range of marine biota collected near the Amchitka test shots and at Kiska (the reference site)?
2. Are there significant differences in the concentrations of radionuclides among the target species collected?
3. Are there significant differences in the concentrations of radionuclides between biota collected near the test shots at Amchitka and at Kiska?
4. Are there significant differences in the concentrations of radionuclides between biota collected among the Amchitka test shots?
5. How do the levels of radionuclides in biota from Amchitka compare to those from the general region and other northern Hemisphere locations?

Our overall approach to answering these questions was to 1) Collect marine biota on land, in the intertidal and at benthic dive stations located by GPS coordinates along several transects adjacent to the test shots and from Kiska (reference site, see chapters 4 and 10), 2) Refine our strategy for selecting species for analysis, and to select radionuclides for analysis (see chapter 9), 3) Analyze radionuclides (see chapter 8), and 4) Conduct statistical analyses of the data to answer the above questions. The relationship between the quantitative radionuclide data, and human and ecological risk, model verification, uncertainty reduction, future bioindicator selection, and source of any radionuclides measured, are discussed in chapter 12.

In short, this chapter examines the radionuclide data, makes comparisons among species, between Amchitka and Kiska, and among the test shots, and compares the

radionuclide data from our study to others for the region and the northern hemisphere. Understanding these issues is critical to any consideration of potential risk to humans or other receptors, and to interpreting the data.

Figure. 11.1. Bald Eagle over nest and Common Eider on nest at Amchitka Island (Photos J. Burger).

BACKGROUND ON INTERPRETING RADIONUCLIDE DATA IN BIOTA

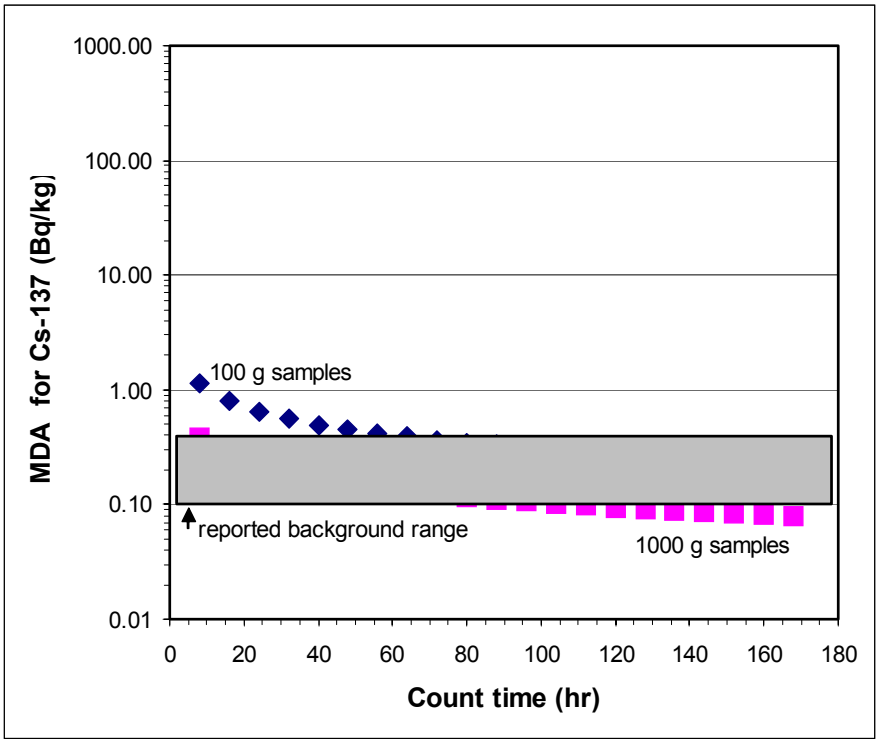
Our analytic approach, discussed more fully in Chapter 9, was 1) to design a human health risk screen for a wide range of species, including a broad gamma analysis of subsistence marine foods and commercial fish, 2) to obtain radionuclide data useful for selecting bioindicators for future biomonitoring at Amchitka, and 3) to obtain isotopic ratio signature informative about the source of radiation. Within this framework, understanding the analytical results from radionuclide analysis requires bearing in mind two factors: 1) how the size of a sample (weight in grams) and the counting time affect the minimum detectable activity (MDA), and 2) because of the low levels of radionuclides, many samples in this (and other studies) will yield results below the detection level (MDA). "Non-detect" refers to a sample value that cannot be statistically distinguished from the background level of radiation in the laboratory counting system (i.e. below the MDA). Time and money constraints required optimizing the selection of samples for analysis and the counting time. The ship's laboratory was able to hold six large chest freezers, which required us to reduce the volume of sample that could be retained.

In the CRESP screening of Amchitka biota, we initially used 100 g samples for gamma analyses, which achieved MDAs that were well below the international food safety standards (FAO 2004, WHO 2004) and the U.S. FDA (2004) derived intervention level (DIL), which is 1200 Bq/Kg for Cesium-134+137. The preliminary results for the gamma screen (including Cs-137) were below MDA (and therefore well below any potential human health risk). Thus, 100 gram samples provide the necessary information to address the question of food safety.

However, samples with concentrations below detectable limits (non-detects) do not help in identifying species for long-term monitoring. That is, if all species have levels below the minimum detectable activity for cesium-137, there is no information on which species

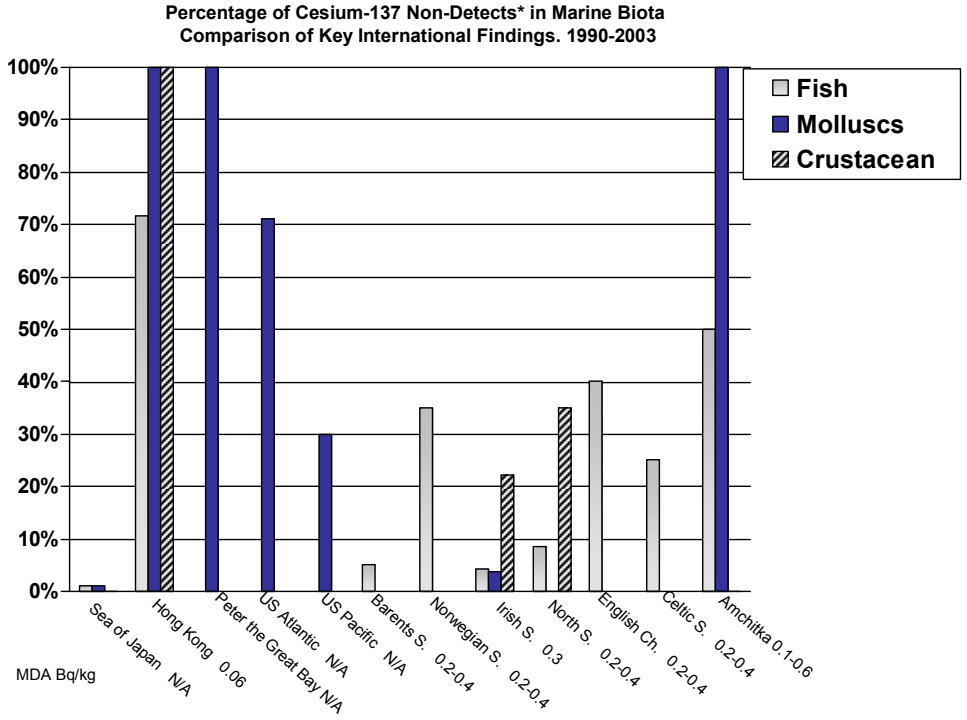
could be used to provide early warning of future contamination or exposure for marine biota and for high trophic level consumers, including humans. Therefore, we began analyzing 1000 g samples for gamma emitters (particularly for cesium-137), to provide detectable levels that would be useful in providing information for the selection of bioindicators. As a general rule, the MDA is improved as a direct function of the mass (weight) of the sample analyzed. For example, a 1000 gram sample provides a 10-fold lower (better) MDA than a 100 g sample. Lengthening the counting time (i.e. from 24 hours to 72 hours) achieves some lowering of the MDA, but it is not proportional. As an example, figure 11.2 provides data from the CRESP Amchitka samples on MDAs as a function of counting time for 100 g and 1000 g samples, as well as indicating the range of published levels in biota. For comparison, the derived intervention level (DIL) is 1200 Bq/kg.

Figure. 11.2. Relationship between sample mass (100 g versus 1000 g), counting time, and typical Minimum Detectable Activity (MDA) for cesium-137. For comparison the U.S. Food and Drug Administration derived intervention level for radioactivity in food is 1200 Bq/kg. The shaded rectangle in the figure gives the range of values reported in the literature for biota from marine environments.



The second important factor in interpreting radionuclide data is the relatively high percentage of non-detects in many different studies. This is particularly true for studies since the late 1990s, when global levels of fallout from above ground nuclear tests had declined due to radioactive decay (refer to Appendix 2.A for a detailed review of radionuclide data). Figure 11.3 shows the percent of samples with concentrations below detection limits for a number of organisms and a number of studies. For comparative purposes, the percent non-detect from the CRESP study falls within the range of these studies for cesium.

Figure 11.3. Relative percent of non-detects for a number of species and studies of radionuclides in marine biota in the northern hemisphere. *Non-detects or less than minimal detectable activity (MDA). The numbers below the location along the bottom are the range of MDAs (Bq/kg).



A third challenge in interpreting radionuclide data is the potential for false positives. The gamma detector scans rapidly along an energy spectrum, recording radioactive emissions of photons with different energies. Each isotope may emit photons with energy at several wave lengths, and there is usually an optimal wave length characteristic of a particular isotope. However, some isotopes emit energy in a range where there are interferences from other isotopes. The counter is “blind”, and the computer analytic software totals the emissions, which may reflect the isotope of interest (for example, Eu-152) as well as contributions from other isotopes with similar

energies. Since Eu-152 emits at several wave lengths, it is therefore necessary to check these other wave lengths to find confirmatory counts. For nuclides with more than one peak, a weighted average value for activity under one or more peaks is calculated. If those confirmatory peaks do not occur, the initial count is concluded to represent interference, rather than Eu-152.

For example, the activity of Eu-152 is calculated by weighting the area under individual peaks at 344, 962, 1112, 779, 1086, 245, 867, and 444 keV. The net activity is proportional to the net area under the peak divided by the peak's photon abundance. The most reliable method for radionuclide identification is to focus on the photon peaks of the highest abundance in the emission spectrum. For the Eu-152 peaks listed above, the absolute abundances for the emissions are 26.5%, 14.6%, 13.6%, 12.9%, 10.2%, 7.6% and 2.5%, respectively. At times, although there is no activity present in the most abundant peaks, a small positive net area from one of the low abundance peaks can suggest a high value for activity in the sample (because some number of positive counts was divided by a very small photon abundance). When a weighted average of all of the results is calculated, it may indicate a high value, perhaps greater than the calculated weighted MDA. When this occurs, the sample spectrum is visually inspected to confirm or negate the reported value. If activity truly above the MDA was present, this will be most evident in the two or three peaks of highest photon abundance. When no activity is seen there, the software result has to be rejected as a false positive, and the sample is treated as if it were below the MDA.

METHODS

The overall protocol described in previous chapters was to collect a range of target species from the marine environment adjacent to the three test shots and from Kiska with appropriate Chain of Custody procedures, reduce mass and prepare some samples on the ships, conduct final preparation of samples at Rutgers (including the reassignment of blind codes), analyze the specimens for radionuclides at INL and Vanderbilt, and analyze the radionuclide data to answer the questions posed above. Detailed methods for each of these phases are described in chapters 8-10, with associated appendices (Figure. 11.4). Some details will be reviewed below.

Figure 11.4. Top: Two teams of UAF divers launched from *Ocean Explorer*; J. Burger and V.M. Vyas receiving samples at Rutgers University. Bottom: T. Stamm fishing on ship, C. Jeitner lab preparation on ship, and S. Burke and M. Donio rad screening at Rutgers (Photos J. Burger, M. Gochfeld) .

The biological sampling plan was a three-pronged approach that was representative of a) Aleut subsistence foods, b) the commercial fishery, and c) different trophic levels on Amchitka food webs. Specimens were collected during two expeditions: *Ocean Explorer* (June 27- July 21), and the *Gladiator* (July 19 - August 4, 2004) by marine ecologists (including divers), Aleut hunters/fishers, and a fisheries biologist. The same species were collected in the marine environments adjacent to each of the three test shots and at Kiska, representing different trophic levels, and ranged from sedentary to mobile. See chapter 8 for methodological details of sample collection and quality control/quality assurance, and chapter 9 for a full description of how the collected species were selected for radionuclide analysis. The specimens collected and analyzed can be characterized as representing:

1. Three groups (Aleut subsistence, commercial fisheries, marine ecosystem).

2. Intertidal, benthic, and sea surface (birds).
3. Several trophic levels from primary producers (kelp) to top level predators (seabirds, halibut, Pacific Cod).
4. A range of mobilities from sedentary to migratory.
5. Spatial proximity to all three test shots, as well as Kiska reference site.
6. Different lifespans, from months to over 50 years of age.

Figure 11.5. Top: Aleut team member, D. Snigaroff, expertly skinning an octopus (left). Commercial trawler unloading Pollock at Dutch Harbor (right) (Photos J. Burger). Bottom: J. Burger in front of the 38 coolers with specimens. (Photo C.W. Powers)

A suite of radionuclides was selected for analysis, based on information obtained from the groundwater models and human health risk assessments (DOE 2003a, 2003b), and our knowledge of radionuclides of interest for human health, ecological health, and source identification. Since the Amchitka radiation information remains classified, this suite of isotopes, identified in the *Amchitka Science Plan*, was reviewed by people with appropriate clearance and access to the classified source term, whom we expected to provide advice if our selection list was missing key isotopes. They indicated that the isotopes we were examining were appropriate.

Isotopes with very long half lives (thousands of years) have relatively few disintegrations per second and therefore low specific activity. Isotopes with relatively short half-lives (such as Iodine 131 at 8 days) have high specific activity and pose acute risks, but also decay rapidly (within weeks). Therefore it is the isotopes with intermediate half-lives such as Cs-137 (30 years), Sr-90 (28.7 years), and Co-60 (5.3 years) which are of the greatest interest.

The main Amchitka isotopes studied for concerns regarding human health and ecological receptors are Cs-137, which is distributed throughout the soft tissue of the body, and Sr-90, which concentrates primarily in bone. Uranium, americium and plutonium isotopes are primarily alpha emitters [their radiation daughter products are not], and accumulate in bone, liver, and kidney. Ratios of plutonium and uranium isotopes, indicative of nuclear detonation and nuclear reactor releases and enrichment processes, can sometimes be used for source identification to distinguish a local source from historic global fallout. If significant contamination had been discovered, presence and ratios of these nuclides could have been used to identify whether the Amchitka test shots were the likely source of measured radionuclides in the biological samples. The source of many radionuclides, such as Cs-137, Sr-90 and Pu-239/240, in terrestrial and freshwater environments, is mainly global fallout from above ground nuclear tests (Dahlgaard et al. 2004). The following analyses were done:

Cs-137, Eu-152, Co-60 (also Am-241)

Pu-238, Pu-239,240, U-234, U-235, U-236, U-238, Am-241

Sr-90, I-129, Tc-99

There were two analytical streams: The analysis at INL focused on testing for a large number of isotopes, but due to time and money constraints, not all samples could be analyzed for all radionuclides. Analyses at Vanderbilt focused on soft tissues for gamma emitters and I-129 for a broader range of species, with more analyses per species (those of primary interest to human health). The MDA (minimum detectable activity) for two of the instruments used at Vanderbilt is shown below. The minimum detectable activity varies slightly for different analytical instruments. Because the detectable activity levels are well below any human health food safety standards, this slight variation does not affect our results. Rather, these data are presented to show the increases in detection activity levels with increases in analytical time (see also Appendix 11.B). We therefore counted 1000 g samples for 72 hours.

Table 11.1. Example of Instrument MDA Variances. MDA values (minimum detectable activity) for two main gamma instruments (Canberra, Ortec) used at Vanderbilt University for analysis of 1000 g samples collected at Amchitka and Kiska. Values are given in Bq/Kg (wet weight).

Isotope	24 hr	48 hr	72 hr
CANBERRA			
I-129	0.645	0.468	0.426
Am-241	0.711	0.493	0.451
Eu-152	0.461	0.323	0.298
Cs-137	0.384	0.335	0.324
Co-60	0.258	0.18	0.168
ORTEC			
I-129	0.882	0.63	0.515
Am-241	0.515	0.362	0.303
Eu-152	0.389	0.279	0.234
Cs-137	0.175	0.124	0.103
Co-60	0.208	0.146	0.133

Quality Control for Radionuclide Analysis

A multi-level quality control program was used for radionuclide analyses that included the following components:

- All sample handling and transfers were under chain of custody documentation.
- Blind analysis: All analytical laboratories were provided with coded samples that did not identify the source location of the sample or the specific species of the sample, although the general type of sample (e.g., soft tissue, bone, algae) was identified to facilitate use of methodology appropriate to the sample type.
- Prior to beginning analysis of actual samples, blind methods validation samples were analyzed by each laboratory. The validation samples were prepared by the RESL laboratory (under the direction of David Sill). The resulting measured value had to be within +/- 30% or 3 standard deviations based on the reported uncertainty of the known value to be considered acceptable.
- Each batch of samples (ca. 20 samples) provided to the analytical laboratories included a blind coded set of two samples that had been prepared by the RESL laboratory. Each of the two samples were spiked with known quantities of some of the radionuclides to be analyzed and blank with respect to other radionuclides to be analyzed. Thus, it was unknown to the analytic laboratory which samples were quality control samples. As with validation samples, the resulting measured value had to be within +/- 30% or 3 standard deviations based on the reported uncertainty of the known value to be considered acceptable.

- For methods requiring preparation through digestion and chemical separations, the analytical laboratory included an additional sample or separate isotopic spike (internal standard) of known concentration to verify recovery. This was in addition to the blind quality control samples. For Tc-99, sample results were adjusted based on individual sample recovery of a rhenium spike (rhenium spike recovery was typically 70-104%). For actinides, chemical yield (recovery) typically ranged between 60-110 %. Recovery of internal standards was reported for each sample.
- For gamma counting and I-129 analysis, instrumentation calibration was checked daily using certified calibration sources. Background counts were carried out weekly. Geometric calibration was carried out using independent certified commercial standards, prepared by the vendor to match the geometry of the specific sample containers (i.e., clean sample containers were supplied to the standards vendor for preparation of calibration standards) and density of the samples.
- Approximately ten percent of the samples were analyzed by both Vanderbilt and INL analytical laboratories for inter-laboratory comparisons. However, because most samples were below method detection limits, this resulted in both laboratories reporting this result and quantitative comparison was limited to a small set of blind quality control spiked samples.
- All analytical results were reported with the uncertainty for each sample analysis.
- All analytical results were independently reviewed for quality assurance.

We also had in place a radiation anti-cross contamination monitoring quality control and assurance plan that included all preparation, whether on the *Ocean Explorer* or in the Rutgers Laboratories (Appendix 11.A). This included screening samples and workplaces with hand-held radiation detectors as well as a program for obtaining wipe samples. There was no evidence of pre-existing sources of radiation on any table surfaces of the boat or laboratories. In-hull radiation activity on the *Ocean Explorer* was appreciably below outdoor and indoor background monitored in Pittsburgh, Pennsylvania and on Adak Island. Scanning of all biological samples brought on board the *Ocean Explorer* detected no values above the background readings of the Ludlum Model 44-9 Alpha Beta and Gamma Detector portable detectors, and all 63 wipe samples taken on board the *Ocean Explorer* demonstrated no detectable activity, when counted in a liquid scintillation detector. In the Rutgers Laboratories wipe samples of all surfaces were taken daily, and all were below background (Appendix 11.A).

Statistical Significance and Power

In this study the emphasis was on detecting a difference in contaminant levels between species and between Amchitka and Kiska. In order to perform a statistical test of a null hypothesis, it is traditional to set a confidence or rejection level. Traditionally scientists have used a 95% level of confidence that means that if a null hypothesis is rejected, there is at least a 95% likelihood that it should have been rejected (i.e. that a difference really does exist). This is accomplished by choosing an alpha level of 0.05 for establishing "statistical significance" which in turn confers 95% confidence that if a difference is detected

it is real (which still carries one chance in 20 of being wrong). The Power is the ability of a study to detect such differences when there really is a difference. Power is largely dependent on the sample size, and when sample size is small a difference may not achieve the level of statistical significance, even if it is real. Readers should be aware that there is no scientific basis for relying on the .05 criterion, which can be traced to R.A. Fisher's study of plant breeding in the 1920's. Any level more or less stringent could be used. We use the .05 "level of significance" in this report because of its general popularity and widespread use. However, it must be emphasized that when the sample size is small (as is the case with many of the sub-analyses reported here), there is inadequate power. Hence, throughout this chapter e also report the absolute probability so that readers can draw their own conclusions, and we refer to P values between 0.05 and 0.10 as "marginally significant". We should note that the power efficiency of the non-parametric tests used throughout this chapter, compared to a parametric one-way ANOVA, is 95%, which makes it a powerful test.

Statistical Tests

Since we did not know in advance what the distribution of radionuclides in organisms would be, all of the statistical tests used in this chapter are non-parametric. This means that no assumptions are made about the underlying distribution of data points (i.e. whether they are normally or otherwise distributed). In comparing the proportion of detects and non-detects, we used contingency table chi square tests. For comparing the actual concentrations we used the non-parametric one way analysis of variance Kruskal-Wallis test based on ranks, which can yield an H-score or a chi square statistic.

Problems arise when many values are below detection levels, hence for most radionuclides, mean values are not reported in the literature, and in this study. In other cases, CRESP has followed the convention of setting non-detectables (all those below the Minimum Detectable Activity) to half the MDA, which despite limitations, provided a reasonable representation of the data (see appendix 11.D).

Methodological Issues

In any large study of contaminants that includes a wide range of species in different trophic levels, with multiple isotopes and multiple study sites, there are methodological issues bearing examination. These include: 1) species and isotope selection, 2) sample and species distribution among study sites, 3) analytical issues. Each will be discussed briefly below.

We selected the species for collection based on trophic level representation, life history information, and cultural factors (are they subsistence or commercial foods? are they endangered? are they of particular interest to resource trustees or others?). Our selection represented an optimization among these factors, on the advice of a wide range of stakeholders, including other scientists, Aleuts, commercial fishermen, and resource trustees. Although all trophic levels were represented, any ecosystem contains so many different organisms at each trophic level that not all ecological equivalents could be represented or captured.

Weather and time constraints while in the Aleutians made it impossible to spend the

same amount of time in each location (*Milrow, Cannikin, Long Shot*, Kiska). Further, inclement weather and surge made it difficult to spend equal diving time at each depth station of every transect. Thus, although our initial design included collection at 3 depths on each transect, this was not always possible. Further, it was too dangerous to land small skiffs at some intertidal coves and beaches. At all times we tried to optimize for collection time at the different locations and dive stations, while always attending to the health and safety of expedition personnel.

Some species were simply not encountered at all locations. For example, although Octopus was one of our target species, we only found them off *Cannikin*. Our Aleut hunters did not find Limpets (Chinese Hats) on all intertidal beaches; some beaches were remarkably devoid of any subsistence foods. "We would starve here", they commented.

There were a number of sample preparation and analyses difficulties that bear mention. It was difficult to obtain sufficient samples of small organisms, such as Limpets and Blue Mussels, or of bones of birds (bird bones are both small and hollow to facilitate flight), particularly for the 1000 g samples. Thus sometimes we had to composite hundreds of individuals to obtain a 1000 g sample. In other cases, it was impossible to composite a 1000 g sample, and smaller amounts were run for longer counting times. However, we felt it was important to examine these species (Blue Mussels, Limpets (Chinese Hats), bird eggs) because of their importance as Aleut subsistence foods). Similarly, where there were limited numbers of samples (Octopus, Halibut), we analyzed individuals or composited fewer than 5 individuals to obtain the maximum amount of information.

Finally, selection of radionuclides in relationship to species and tissues was another difficult process, constrained by available material (some species are too small to obtain large enough samples for analysis), time (Sr-90 analyses takes several weeks depending on the matrix. Sample throughput (some analyses are destructive, making it essential to conduct different isotope analysis in an appropriate sequential order), and money. For example, due to density and the difficulty of separating strontium from calcium, the laboratory could only analyze small quantities of bird bones (about 2 grams), with consequent loss of sensitivity. Therefore, the detection limit for bird bones was much higher.

Any study of this complexity encounters methodological issues dealing with species selection, sample numbers, available tissue (size of individual organisms), sample distribution among habitats and study sites, and isotope selection. Some of these are biological constraints (size of biota, location of biota), others are weather/surge related (when and where we could sample safely) or time/money constraints. Overall these were dealt with by maximizing our use of time and resources while on the expedition and during the analysis phase to achieve a design that balanced subsistence foods/commercial fish, trophic levels, location (among Amchitka test shots and with Kiska), and among different stakeholder concerns.

RESULTS

Number of Analyses

Our overall analysis design was to examine 100 g samples for most radionuclides

(Table 11.2), with 1000 g samples for Cs-137 (Table 11.3). The 100 g samples provided information relevant to assessing the safety of foods, while the 1000 g samples provided information that can be used to select bioindicators for future biomonitoring. To select bioindicators it is necessary to know which species concentrate radionuclides, and thus have higher levels of radionuclides than other species.

Our protocol was to make 100 g composites of five individuals for fish and birds for each location. However, for smaller organisms, such as limpets, mussels, and sea urchins, dozens of individuals were required to achieve 100 g for analysis. In addition, for some species, we analyzed individuals (Sea Lion, Octopus), a small number per composite (Halibut), or combined specimens across Amchitka (gull eggs). Both muscle and liver tissue were analyzed for the one Sea Lion taken in a subsistence hunt by Aleuts. To provide some indication of the number of individuals analyzed, Table 11.2 shows the 100 g samples for each radionuclide and Table 11.3 shows the number of individuals in the 1000 gram samples, by location. The 100 g samples were generally divided evenly between the three test shots and Kiska.

Table 11.2. *Radionuclide Analysis Conducted For Human Health Screen*. Total analyses run by species and radionuclide for 100 g samples (see table 11.3 below for 1000 g samples). This is a complete species list for all analyses. Where there are blanks, all samples were done as 1000 g samples (see following table).

		Number of 100g composite samples for analysis						
SPECIES	**Tissue**	**Cs-137**	**I-129**	**Co-60**	**Eu-152**	**Sr-90**	**Alpha[a] Analyses**	**Tc-99**
PRIMARY PRODUCERS								
Alaria Fistulosa	soft	4	4	4	4	4	10	4
Alaria Nana	soft	4	4	4	4	4	12	4
Fucus	soft	4	4	4	4	4	14	4
Ulva	soft						12	
GRAZERS/FILTER FEEDERS								
Sea Urchin	muscle	4	5	4	4	8		4
Rock Jingle	muscle	4	4	4	4	3	3	3
Limpet (Chinese hats)	muscle							
Gumboot	muscle							
Blue Mussel	muscle							
LOWER PREDATORS								
Dolly Varden	muscle	8		8	8			
Atka Mackerel	muscle	1	1	1	1	2		2
Atka Mackerel	bone	1		1	1	1	1	
Red Irish Lord	muscle	8		8	8			
Rock Greenling	muscle	23	9	23	23	4		4
Rock Greenling	bone	4	4	4	4			
Yellow Irish Lord	muscle	15	4	15	15	4		4
Yellow Irish Lord	bone	3	1	3	3	3	3	
Ocean Perch	muscle	2	2	2	2	2		2
Ocean Perch	bone	1	1	1	1	1	1	
Eider adults (birds)	muscle	4		4	4			
Eider (eggs)	eggs	6	3	6	6	3		3
HIGHER TROPHIC LEVEL								
Black Rockfish	muscle	12	4	12	12	4		4
Black Rockfish	bone	3	2	3	3	1	1	
Walleye Pollock	muscle					2		2
Walleye Pollock	bone					2	2	
Gull (birds)	muscle	18	8	18	18	8		8
Gull (birds)	bone	8		8	8	8	8	
Gull (eggs)	egg							
Pigeon Guillemot	muscle	7	3	7	7	3		3
Pigeon Guillemot	bone	3		3	3	3	3	
Tufted Puffin	muscle	6	3	6	6	3		3
Tufted Puffin	bone	3		3	3	3	3	
TOP TROPHIC LEVEL								
Octopus	muscle							
Halibut	muscle							
Halibut	bone						4	
Pacific Cod	muscle	14	5	14	14	5		6
Pacific Cod	bone	3		3	3		14	
Bald Eagle	muscle							
Sea Lion	muscle							
Sea Lion	Liver							
		173	71	173	173	85	91	60

a. The actinides analyzed were Am-241, Pu-238; Pu-239,240; U-234; U-235; U-236; U-238

Table 11.3. Number of Cs-137 Analyses for Understanding the Food Chain and Bioindicator Selection. Number of analyses of 1000 g samples, with number of organisms in those samples. Given are number of gamma analyses (number of individuals in the analyses). Halibut were very large, and we used fewer than 5 individuals/composite; we analyzed each Octopus separately. Exceptions to 1000 g are indicated in footnotes. Amchitka column is for species that had to be composited from all Amchitka sites.

SPECIES	AMCHITKA	*MILROW*	*LONG SHOT*	*CANNIKIN*	KISKA	NOAA
PRIMARY PRODUCERS						
Alaria fistulosa			1(5)	1(5)	2(10)	
Fucus		1(5)			1(5)	
Alaria nana					1(5)	
Ulva			2[a]		1[a]	
GRAZERS/ FILTER FEEDERS						
Sea Urchin		1(75)	1(52)		1(50)	
Rock Jingle			1(89)	1(142)	1(91)	
Limpets		1(51)[b]	1(99)			
Gumboot	1(109)					
Blue Mussel			1(115)[c]		1(229)	
LOWER PREDATORS						
Dolly Varden	2 (46)					
Rock Greenling		1(8)	2(13)	1(10)	1(6)	
Eiders (adult)			1(6)		1(4)	
Eider (eggs)	1(14)[d]				1(15)	
HIGH TROPHIC LEVEL						
Black Rockfish		1(10)		1(10)	1(11)	
Walleye Pollock						2(10)[e]
Gull (muscle)	1(11)				1(7)	
Gull (eggs)	1(7)[f]				1(7)	
Tufted Puffin	1(9)				1(6)	
TOP TROPHIC LEVEL						
Octopus				4(4)		
Halibut		1(3)	1(2)		1(4)	1(5)
Pacific Cod		2(10)	6(30)	1(5)	3(16)	2(10)[d]
Eagle	1(1)[g]		1(1)			
Sea Lion	1/1[h]					

a. For *Ulva* it is difficult to distinguish individuals, so samples were taken from areas separated by at least 5 m.
b. 51 limpets = 156 g at *Milrow*; 99 limpets = 411 g at Longshot; 109 gumboots = 875 g.
c. 115 mussels = 411 g at Longshot; 2229 mussels = 716 g at Kiska.
d. 14 eggs= 876 g.
e. One composite each from the NOAA trawl near Amchitka, and near Kiska.
f. Combined from all the test shot areas
g. One dead eagle was found near the airport
h. Only one Sea Lion was hunted by the Aleuts, and they requested that the UAF Museum analyze both muscle and liver (the museum subsequently asked CRESP to perform the analysis).

Comparisons among Species

In the human health screen (100g samples) there were no samples with concentrations above minimum detectable activity levels in the radionuclide analyses, except for the alpha analysis. Alpha analysis revealed many samples with concentrations above detection limits mainly for the naturally occurring uranium-238 and U-234. Thus for iodine-129 (no detects of 71 samples), cobalt-60 (no detects of 173), strontium-90 (no detects of 85) and europium-152 (no detects of 173) it was not possible to compare either among species or among locations. However, there were detectable values for cesium-137 in some 1000 g samples.

For Cs-137 it is only the 1000 g samples where a comparison among species can be made (Table 11.4). Even in the 1000 g samples, there were no values above the minimum detectable activity (MDA) levels for primary producers and grazer/filter feeders. High trophic level organisms (Sea Lion, Octopus, Pacific Cod, Black Rockfish, Halibut, Walleye Pollock, Glaucous-winged Gull) had significantly more levels above the MDAs, and significantly higher levels than those species that are lower on the food chain (X^2= 25.4, P<0.0001, 2X2 Contingency Table, see below and Table 11.4). This analysis excludes Dolly Varden, some of which were from *Cannikin* Lake.

	Primary Producers, Filter Feeders, Lower Predators	Higher Trophic Level, Top Trophic Level Predators
Number/Detect	0	21
Number/Non-detect	31	16
Percent Detect	0%	57%

Table 11.4. Percent of Cs-137 Detects by Species. Comparison among species in detectable cesium-137 levels from the 1000 g samples. Detect" = refers to values above the minimum detectable activity (MDA), and "non-detect" refers to those below the MDA.

Species	Number of detects	Number of non-detects	Percent of detects
PRIMARY PRODUCERS			
Alaria fistulosa	0	4	0
Alaria nana	0	1	0
Fucus	0	2	0
Ulva	0	3	0
GRAZERS/FILTER FEEDERS			
Sea Urchin	0	3	0
Rock Jingle	0	3	0
Limpet	0	2	0
Blue Mussel	0	2	0
LOWER PREDATORS			
Dolly Varden	1[a]	0	100
Rock Greenling	0	5	0
Eider adults	0	2	0
Eider eggs	0	2	0
HIGHER TROPHIC LEVEL (predators)			
Black Rockfish	3	0	100
Walleye Pollock	1	1	50
Gull adults	1	1	50
Gull eggs	0	2	0
Tufted Puffin	0	2	0
TOP TROPHIC LEVEL (predators)			
Octopus	4	0	100
Halibut	3	1	75
Pacific Cod	7	7	50
Bald Eagle	0	2	0
Steller Sea Lion	2	0	100

a. We include only the sample from the lake near the Amchitka air strip.

In the Table 11.5 we provide the mean levels of cesium-137 for the 1000 g samples. The number of cesium-137 analyses on 100 g samples (with mean MDA) are shown for comparison. We also provide all of the values above the MDAs, giving an indication of the range of values for samples collected at Amchitka and Kiska. It should be noted that the MDAs for the 1000 g samples lie well below the background level (see discussion), and for both the 100g and 1000g samples, the MDAs lie below the food safety levels for people (see chapter 12).

Table 11.5. Cs-137 Levels. Number of analyses for 1000 g samples (with number of 100 g analyses in parentheses), average radionuclide concentration (1000 g samples only) for muscle or soft tissue, ± standard deviation. All Values Above MDAs and the average MDA for each species for both 1000 g and 100 g samples. MDA = Minimum detectable activity level.

SPECIES	Number of Analyses for 1000 g samples (and for 100 g samples)	Mean ± SD Bq/kg for 1000 g samples[a] (wet weight)	Values above MDA[b]	Mean MDA for 1000 g samples	Mean MDA for 100 g samples
PRIMARY PRODUCERS					
Alaria fistulosa	4 (4)			0.18	5.57
Alaria nana	1(4)			0.36	6.25
Fucus	2(4)			0.34	6.08
Ulva	3			0.25	c
GRAZERS/FILTER FEEDERS					
Sea Urchin	3(4)			0.09	7.82
Rock Jingle	3(4)			0.32	5.96
Limpets	2			0.36	c
Gumboot	1			0.34	c
Blue Mussel	2			0.58	c
LOWER PREDATORS					
Dolly Varden	2(8)		0.70	0.132	3.97
Rock Greenling	5(23)			0.29	2.81
Eider (adults)	2(4)			0.23	4.39
Eider (eggs)	2(6)			0.10	3.12
HIGHER TROPHIC LEVEL					
Black Rockfish	3(12)	0.143 ± 0.040	0.189 0.130 0.111	0.10	3.32
Walleye Pollock	2	0.311 ± 0.311	0.461	0.32	c
Gulls (adults)	2(18)		0.094	0.26	2.78
Gull (eggs)	2			0.24	c
Tufted Puffin	2(6)			0.19	2.74
TOP TROPHIC LEVEL					
Octopus	4	0.262 ± 0.029	0.236 0.249 0.260 0.302	0.09	c
Halibut	7	0.24 ± 0.14	0.190 0.315 0.446	0.15	c
Pacific Cod	14 (14)	0.29 ± 0.20	0.176 0.188 0.209 0.315 0.400 0.472 0.602	0.28	5.70
Bald Eagle	2			0.66	c
Sea Lion	2[d]	0.40[d]	0.554 0.405	0.085	c

a. For sample values below the MDA, one-half the MDA was substituted in calculating the mean and standard deviation. When no value was above the MDA[d], space is blank. Given is mean ± SD.
b. Given for those with few values above MDAs.
c. No 100 g samples analyzed.
d. Liver and muscle were both analyzed from the same individual sampled from an Aleut subsistence hunt. Liver = .55 and muscle = .405 Bq/kg (ww)

Although 85 analyses were run for Sr-90 (both bone and soft tissue); refer to Table 11.2), there were no samples with levels above the MDA. Similarly, for Tc-99, I-129 and Eu-152, there were no samples with concentrations above detection limits. Thus, it was not possible to study differences between species for these isotopes.

For actinides, there were differences among species in the relative percentage of values above the minimum detectable activity levels (Table 11.6). Because Kelp are known to concentrate certain elements and since Kelp figured importantly in the Amchitka *Screening Risk Assessment,* we analyzed more Kelp than other species. Kelp and Rock Jingles (low trophic level organisms) had significantly more detectable levels than did predators (higher trophic level) for Pu-239,240, U-234, U-235, and U-238 (statistical tests for each radionuclide shown on the bottom of Table 11.6).

Table 11.6. Percent of Actinides Detects by Species. Percent of samples with concentrations above minimum detectable activity limits for soft tissue and bone samples for actinides by species. Under source, A = anthropogenic from many human-derived sources, N = naturally occurring (capital letter = the major source, lower case = less of a source), n=secondarily naturally occurring.

SPECIES	Number of analyses	Am-241	Pu-238	Pu-239 -240	U-234	U-235	U-236	U-238
SOURCE Anthropogenic or Natural		A	A	A	N	A,n	A	N,a
PRIMARY PRODUCERS								
Alaria fistulosa	10	0	10	30	100	40	10	100
Alaria nana	12	17	0	25	100	33	0	100
Fucus distichus	14	14	0	29	100	100	8	100
Ulva latuca	12	8	17	0	100	0	0	100
FILTER FEEDERS								
Rock Jingle	3	67	0	33	100	33	33	100
LOWER PREDATORS								
Atka Mackerel	1	0	0	0	100	100	0	100
Yellow Irish Lord	3	0	0	0	100	0	0	100
Ocean Perch	1	0	0	0	100	0	0	100
HIGHER TROPHIC LEVEL								
Black Rockfish	1	100	0	0	100	100	0	100
Walleye Pollock	2	50	0	50	100	50	0	100
Gulls (birds)[a]	8	0	0	0	0	0	0	25
Pigeon Guillemot	3	0	0	33	0	0	0	0
Tufted Puffin	3	0	0	0	0	0	0	33
TOP TROPHIC LEVEL								
Halibut	4	0	0	25	75	25	0	100
Pacific Cod	14	7	0	0	79	0	0	71
Chi square (probability) comparing kelp, Ulva and Rock Jingles versus predators[b]		0.88 (0.34)	2.43 (0.12)	5.01 (0.025)	28.7 (0.0001)	13.2 (0.0003)	2.43 (0.12)	22.9 (0.0001)

a. Adult and young of the year Glaucous-winged Gulls.

b. A probability level of <0.05 means there is a significant difference between Kelp, Ulva and Rock Jingles compared to the predators.

Kelp had both the highest percentage of samples with concentrations above detection limits, and the largest number of analyses, making it possible to look for differences among the species for the different isotopes. Below we examine the mean values (both detect and half the MDA for the non-detects) for the kelp and *Ulva* (Table 11.7). There are clearly species differences for the uranium isotopes, with *Fucus* having significantly higher levels for U-234, U-235 and U-238 than the other species. In general, conducting the analysis on all values (detect + non-detects) yielded similar results as conducting the analysis only with samples above detection limits.

Table 11.7. Mean Actinide Differences Among Algae Species. Mean (± standard deviation) actinide values (Bq/Kg, wet weight) for kelp from both Amchitka and Kiska. Number of analyses was as follows: Alaria fistulosa = 10, A. nana = 12, Fucus = 14, and Ulva = 12. The mean values include both samples with and without measured values above minimum detectable activity level (entered as half the MDA). P values <0.05 indicate a significant difference among algae species using Kruskal-Wallis test.

Isotope	***Alaria fistulosa***	***Alaria nana***	***Fucus***	***Ulva***	**Chi square (p value)**
Am-241	0.013 ± 0.006	0.018 ± 0.01	0.015 ± 0.014	0.017 ± 0.019	1.64 (0.65)
Pu-238	0.014 ± 0.005	0.018 ± 0.01	0.014 ± 0.005	0.021± 0.033	1.59 (0.66)
Pu-239,240	0.057 ± 0.065	0.029 ± 0.016	0.036 ± 0.031	0.014 ± 0.006	11.9 (0.008)
U-234	1.001 ± 0.64	0.77 ± 0.31	3.12 ± 1.087	0.317± 0.121	35.1 (<0.0001)
U-235	0.050 ± 0.035	0.039 ± 0.024	0.15 ± 0.052	0.025 ± 0.004	30.9 (<0.0001)
U-236	0.020 ± 0.013	0.019 ± 0.011	0.018 ± 0.008	0.018 ± 0.006	2.37 (0.50)
U-238	0.856 ± 0.48	0.68 ± 0.30	2.74 ± 0.95	0.246 ± 0.137	37.3 (<0.0001)

Locational Differences: Amchitka vs Kiska and Among Amchitka sites

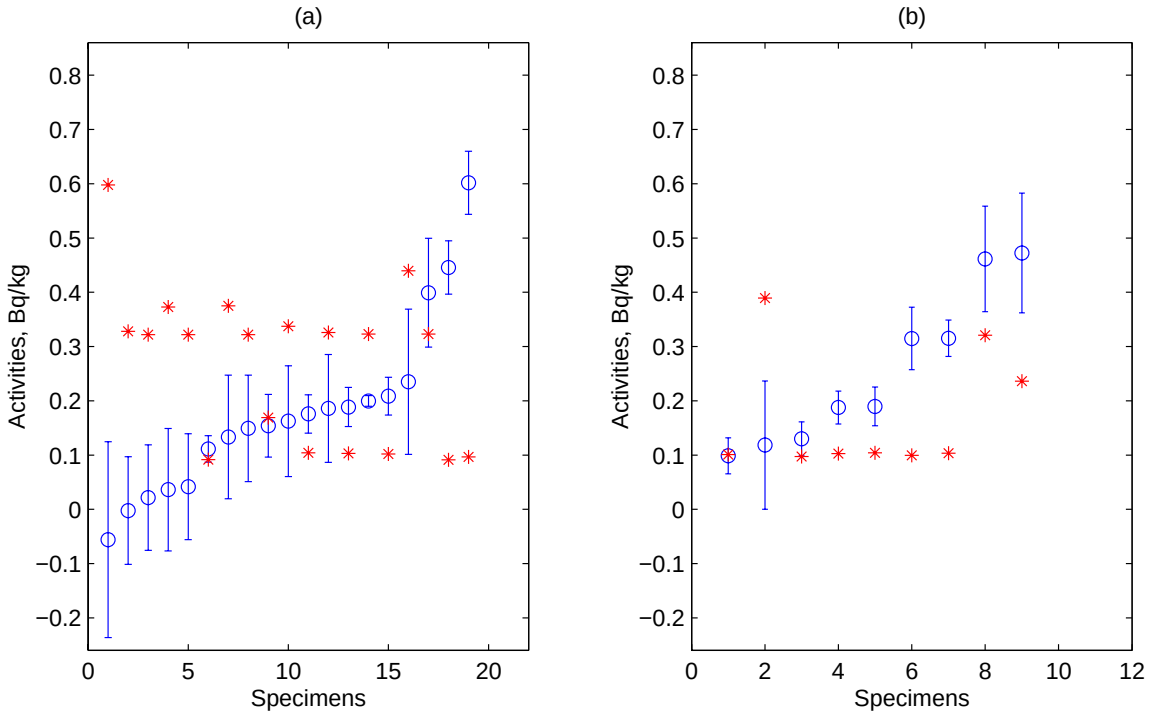
One of our objectives was to compare radionuclide levels in organisms collected around Amchitka and Kiska (our reference site). Again, for strontium, technetium, europium, iodine and cobalt, it was not possible to compare among locations (either within Amchitka test areas, or between Amchitka and Kiska).

It is not possible to make a meaningful comparison among the three test shot regions for Amchitka for cesium-137 because there are too few detects, and many of the

detects were from species that only occurred in one site (i.e. Octopus were only collected at *Cannikin*). It should be noted that the MDAs for the 1000 g samples lie well below the environmental level (background) reported in the literature (see discussion), and for both 100 and 1000 g samples, the MDAs lie below the risk level for people (see chapter 12).

The combined samples for cesium-137 (all analyses) can be used to examine differences between Amchitka and Kiska, and there were no statistically significant differences in the 1000 g samples for the mean Cs-137 levels overall ($X^2 = 0.002$, $P>0.95$). The analysis included all species, however, it is apparent that cesium levels are higher in the high trophic levels birds, fish and mammals than in the lower trophic level filter feeders, grazers, and producers (see above section). Figure 11.6 shows the relationship of MDAs to reported values for Cs-137 for all fish.

Figure 11.6. Comparison of Cs-137 between Amchitka and Kiska. This figure plots the reported concentrations of Cs-137 (in Bq/kg wet weight) for 1000g fish specimens collected at (a) Amchitka and (b) Kiska. The reported values are in open circles with error bars – the error bars represent reported value plus one standard deviation uncertainty on the top, and reported value minus one standard deviation at the bottom. The corresponding method detection activities are shown as stars, and are also in Bq/kg.



To examine species collected at both islands, we confined the analysis to four species of high trophic level fish: Halibut, Pacific Cod, Walleye Pollock, and Black Rockfish. There were fifteen 1000 g samples for Amchitka and eight for Kiska (Table 11.8). For each location there were seven values above the MDA. Although the mean values are similar for the two islands, the proportion of cesium "detects" at Kiska (87%) is marginally significantly higher than at Amchitka (47%) (P=0.056). However, the mean Cs-137 values were not

significantly different between Amchitka and Kiska.

Table 11.8. Comparison of Cs-137 Levels in Fish between Amchitka and Kiska. Comparison of Cs-137 levels in high trophic level fish species (1000 g samples only) for fish collected at both Amchitka and Kiska. This includes Black Rockfish, Halibut, Pacific Cod and Walleye Pollock.

	Amchitka	Kiska	Statistical test
Number of composites	15	8	
Number positive (%)	7 (47%)	7 (87%)	X^2=3.65 (P=.056)[a]
Mean ± SD (using ½ MDA for non detects)	.245 ± .135	.283 ± .130	X^2=1.5 (P=.22)[b]
Mean ± SD for detects only	.304 ± .180	.296 ± .135	X^2=.04 (P=.84)[b]

a. Analysis by 2x2 contingency table and chi square.
b. Analysis by non-parametric Kruskal-Wallis analysis of variance yielding a chi square statistic.

The actinide data for all species combined (all tissues combined) can also be used to examine for differences between Amchitka and Kiska (Table 11.9). Overall, there were no statistically significant inter-island differences in the proportion of detectable levels for most of the actinides, although the proportion of detects was higher at Amchitka for Pu-239,240 (15%) than at Kiska (2%) (P=0.015).

Table 11.9. Comparison of each actinide analysis between Amchitka and Kiska for all species (tissues) combined. Total analyses for Amchitka was 55, and for Kiska was 36. There were no significant differences in the MDAs between Amchitka and Kiska. MDAs are given as mean ± standard deviation (Bq/Kg, wet weight). Last column gives Kruskal-Wallis, non-parametric Chi square value.

Isotope	MDA Amchitka (n=55)	MDA Kiska (n=36)	Percent above MDA Amchitka	Percent above MDA Kiska	Chi square (P) Comparing Number of detects
Am-241	0.052 ± 0.07	0.048 ± 0.07	9%	14%	0.51 (>0.47)
Pu-238	0.059 ± 0.07	0.049 ± 0.06	5%	0%	2.03 (0.15)
Pu-239,240	0.078 ± 0.09	0.069 ± 0.11	15%	2%	5.94 (0.015)
U-234	0.086 ± 0.11	0.070 ± 0.10	82%	78%	0.22 (0.64)
U-235	0.102 ± 0.12	0.089 ± 0.13	31%	28%	0.10 (0.75)
U-236	0.072 ± 0.08	0.062 ± 0.10	4%	3%	0.05 (0.82)
U-238	0.087 ± 0.10	0.067 ± 0.10	84%	83%	0.01 (>0.9)

It should be noted that these data for all actinide analyses can be used to understand something about the possible source of the radionuclides. There were no significant locational differences in the proportion of levels above the MDA (=detects) for the naturally-occurring actinides (U-234, U-235, U-238), nor for some of the anthropogenic

ones (Am-241, Pu-238, and U-236, Table 11.9). However, there was a significant difference for Pu-239,240 (X^2 = 5.94, P = 0.015), which bears further examination.

In the above data analysis, all species were combined. To ensure that differences among species are not masking significant differences within species groups, we compared the percent of values that were above the MDA for algae (including kelp and *Ulva*), the species group with the greatest number of analyses. While there were no significant differences for most actinides (Table 11.10), there was a significant difference both for the proportion of levels above MDAs, and for the mean values for Pu-239,240. Kiska had fewer algae values above the MDA (detect) than did Amchitka. Overall, algae had significantly higher proportions of levels above the MDAs than did predators (fish and birds) for Pu 239,240, U-234, U-235, and U-238 (refer back to Table 11.6), indicating their relative importance as bioindicators for the actinides (see discussion).

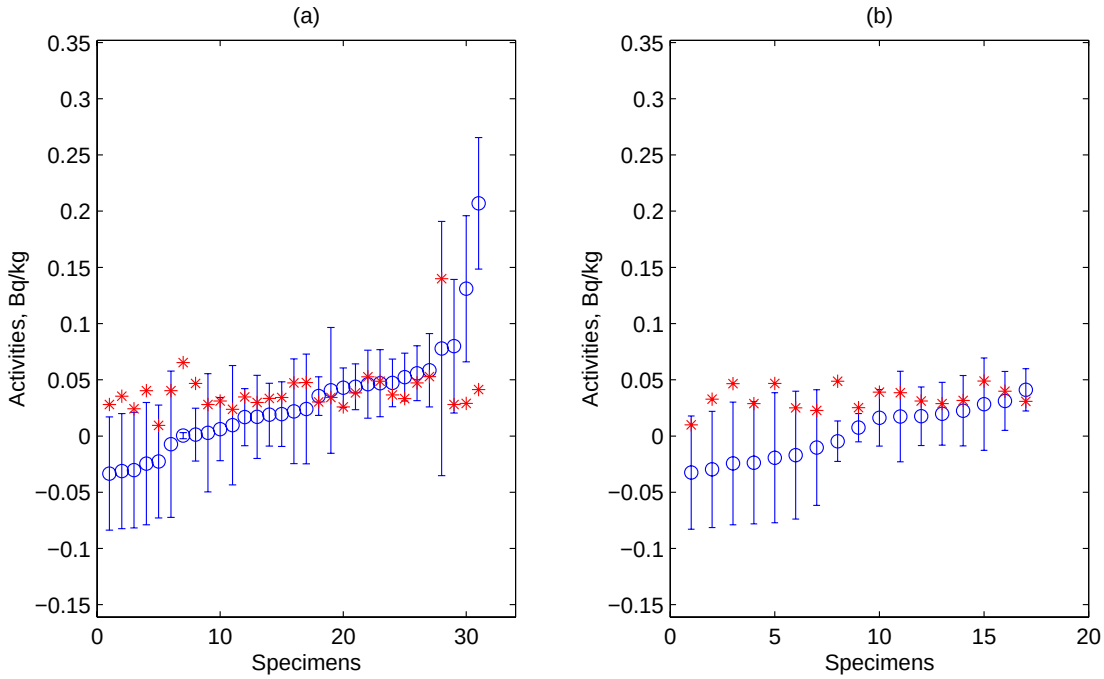
Table 11.10. Comparison of Actinide Levels Between Amchitka and Kiska for Algae. Comparison of radionuclide values in Algae[a] for Amchitka and Kiska islands including the ranges of concentrations reported, the means calculated (using half the MDA for values below the MDA[b]), and the proportion of detects (values > MDA) for each of the actinides. 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 x 2 contingency table. Both tests yield a chi square value. There were 31 algae analyses from Amchitka and 17 from Kiska.

	Range and Means of Isotope Values			**Proportion of Detects**	
ISOTOPE	**Range of reported values**	**Mean + SD**	**Kruskal-Wallis Chi Square (p=)**	**Number of detects (%)**	**Contingency Chi Square (p=)**
Am-241					
Amchitka	<0 - 0.035	0.015 ± 0.008	0.04 (p=.84)	3 of 31 (9%)	0.05 (p=.82)
Kiska	<0 - 0.075	0.018 ± 0.016		2 of 17 (11%)	
Pu-238					
Amchitka	<0 - 0.123	0.019 ± 0.021	0.69 (P=.41)	3 of 31 (9%)	1.75 (p=.18)
Kiska	<0 - 0.006	0.013 ± 0.005		0 of 17 (0%)	
Pu-239,240					
Amchitka	<0 - 0.207	0.039 ± 0.040	5.68 (P=.017	11 of 31 (32%)	4.32 (p=.04)
Kiska	<0 - 0.041	0.018 ± 0.008		1 of 17 (6%)	
U-234					
Amchitka	0.195 - 4.820	1.447 ± 1.221	0.92 (P=.34)	31 of 31 (100%)	0 (p=.99)
Kiska	0.157 - 5.100	1.291 ± 1.526		17 of 17 (100%)	
U-235					
Amchitka	<0 - 0.198	0.071 ± 0.055	1.04 (p=.31)	16 of 31 (52%)	1.18 (p=.28)
Kiska	<0 - 0.254	0.066 ± 0.072		6 of 17 (35%)	
U-236					
Amchitka	<0 - 0.044	0.020 ± 0.011	2.34 (p=.13)	2 of 31 (6%)	1.14 (p=.27)
Kiska	<0 - 0.019	0.016 ± 0.005		0 of 17 (0%)	
U-238					
Amchitka	0.077 - 4.370	1.279 ± 1.100	1.14 (p=.28)	31 of 31 (100%)	0 (p=.99)
Kiska	0.058 - 4.470	1.080 ± 1.291		17 of 17 (100%)	

a=Algae include *Alaria fistulosa, Alaria nana*, *Fucus*, and *Ulva*.
b=There were no significant differences in MDA's for Amchitka and Kiska, including for Pu239,240.

Because of the differences in Pu-239,240 between Amchika and Kiska, we present graphically the data for all algae samples from Amchitka and Kiska. On this graph the MDA, mean, and one standard deviation are presented.

Figure 11.7 Comparison of Reported values and MDAs for Pu-239,240 for algae. This figure plots the reported concentrations of Pu-239,240 (in Bq/kg wet weight) for primary producers (ie all algae species; *Alaria nana, A. Fistulosa, Ulva, Fucus*), for (a) Amchitka and (b) Kiska. The reported values are in open circles with error bars – the error bars represent reported value plus one standard deviation uncertainty on the top, and reported value minus one standard deviation at the bottom. The corresponding method detection activities are shown as stars, and are also in Bq/kg.



Because of the significant difference in both the percentage of values above the MDAs, and the mean levels of Pu-239,240 (see Table 11.10 above), we present more detailed data for kelp (Table 11.11). This table presents all the values above the MDAs for the three kelp species. There was a significant difference in the proportion of values above the MDA among the 4 study sites, but no significant differences in the mean MDAs among *Long Shot, Milrow, Cannikin* and Kiska. That is, the differences in the percent of detects was not due to a difference in analytic sensitivity (MDAs) Three additional points, discussed more fully in the discussion below, should be noted: 1) *Ulva latuca*, the green Sea Lettuce species that is consumed by people, had no levels above the MDAs, 2) the levels found in kelp in this study are well below any human consumption guideline, and 3) the levels are generally within the ranges reported from uncontaminated areas in the Northern Hemisphere (Appendix 2.A, discussion below).

Table 11.11. Plutonium values for Kelp at the Three Test Shots and Kiska. Distribution of Plutonium (Pu-239,240) in kelp species from Amchitka (*Long Shot, Milrow, Cannikin*) and Kiska, including number of values greater than detection level (MDA). The difference in the proportion of values above the MDA between Amchitka and Kiska is small but statistically significant (X^2 = 4.19, P < 0.04). The MDA's did not differ significantly by location. The difference among all four sites is also statistically significant for the proportion above the MDA (X^2=10.1; P=0.018).

	Long Shot	***Milrow***	***Cannikin***	**Kiska**	**Statistic**
Total analyzed[a]	11	9	5	11	X^2=10.1
Number > MDA	2	6	3	1	P=.018. df=3 [c]
MDA for Kelp Bq/kg	0.051 ± .032	0.037 ± .007	0.033 ± .008	0.037 ± .008	X^2 = 3.87 P =.27 (df=3)[d]
Actual Values greater than MDA (Bq/kg)[b] *and number of samples analyzed*					
Alaria fistulosa	0.130 (3 analyzed)	0.207 0.080 (3 analyzed)	0.041 (2 analyzed)	(2 analyzed)	
Alaria nana	None (3 analyzed)	None (1 analyzed)	0.043 0.036 (3 analyzed)	0.041 (5 analyzed)	
Fucus distichus	0.059 (5 analyzed)	0.056 0.052 0.047 0.044 (5 analyzed)	(0 collected)	(4 analyzed)	

a. Each analysis was a composite of fronds from five individual plants.
b. All values are shown as Bq/kg on wet weight basis.
c. Analyzed in 2 x 4 contingency table with chi square (df=3), comparing proportion of detects.
d. Analyzed with Kruskal Wallis non-parametric ANOVA.

DISCUSSION AND IMPLICATIONS

Comparisons among Species

Our approach was to directly analyze radionuclides in the organisms of interest, and examine levels in biota from Amchitka and Kiska, rather than to undertake modeling efforts to estimate radionuclide concentrations in biota based on either soil or water concentrations (Higley et al. 2003a). Partly this approach was taken because it allows an immediate and direct analysis of whether the foods are safe, and whether there is food chain accumulation that might pose a health risk to top-level predators. Dasher et al. (2004) examined radionuclides in the freshwater and terrestrial environments of Amchitka, and did not find evidence of contamination. They further suggested that a radiological assessment of the marine environment around Amchitka was needed.

In general, there are differences among species in contaminant levels because of differences in mobility and habitat, trophic level and diet, and age, gender, and size (Peakall and Burger 2003, Burger et al. 2003b). There are some generalizations about

contaminant distribution that are important. In fish, some contaminants bioaccumulate with size and age (Braune, 1987; Lange et al., 1994; Lacerda et al., 1994; Bidone et al., 1997; Burger et al. 2001b,c, 2002). This is not always the case and the relationship may not exist where food is limiting and fish stop growing, but continue to accumulate contaminants (Downs et al., 1998, MacFarlane et al. 2000). Similarly, in birds, older individuals usually have higher levels of contaminants than younger ones (Thompson et al. 1993; Gochfeld et al. 1996; Burger 1996; Stewart et al. 1997; Burger and Gochfeld 1997b,c; Burger et al. 2002a). These differences are a function of having longer to bioaccumulate contaminants than much younger individuals, but also to age differences in trophic level (Fairey et al., 1997, Monteiro and Furness 1995). Larger individuals can eat bigger prey.

Species and individuals may vary in their susceptibility to contaminants, both because of individual traits (i.e. birth order, age, size, gender, genetics) and because of population traits (i.e. migration patterns, geography, food chain complexity, food chain length, Steinberg et al. 1995). For some contaminants, faster-growing fish have lower concentrations than slower-growing individuals at a given length (Simoneau et al. 2005), further complicating predictions of levels. Susceptibility is of course modified by bioavailability - how available the specific contaminant is to the host organism, including whether it is in a form that can be absorbed.

Further, a range of population and community dynamics can influence the foods available or suitable for a given predator. One key question is whether bottom trophic levels are controlling top-level predator numbers, or whether predators are controlling numbers of prey and producers (e.g. kelp). It is a question of whether there is top-down or bottom-up control of marine food webs (BEST 2004). In times of increasing food supply, top-down control may dominate, but in times of decreasing food supply, bottom-up control may dominate (Hunt et al. 2002). This is important because the movement of contaminants through a food web partly depends upon the number and abundance of different species at every level. While there are data on uptake of contaminants by terrestrial plants and grazing food chains (Pinder et al. 1984), there are few on aquatic grazing food chains. A grazing food web based on kelp dominates the eastern Bering Sea ecosystem, which is associated with high groundfish production (Mito et al. 1999).

In addition, a wide range of physical oceanographic and climate factors affect exposure, such as vertical mixing, wave intervals, eddies and fronts, wind forcing, seasonal and El Niño variations, and other climatic variations (BEST 2004). These factors not only would affect radionuclide movement, but location and movement of biota within benthic environments (Brodeur et al. 1999), although sometimes the differences do not relate to either size or age. Thus oceanographic factors affect both the location of biota and the movement of contaminants.

Differences among species occur for radionuclides as well, even within the same taxon. For example, Burger et al. (2001b) found that cesium-137 varied nearly by an order of magnitude among fish species inhabiting the same environment, and fish from a known-contaminated site had cesium-137 levels an order of magnitude above the reference site.

The data from the CRESP Amchitka/Kiska study indicate differences among species in cesium-137 and some actinides. There were inadequate detectable values for comparing the other radionuclides. For cesium-137, the highest trophic level predators had the

highest number of samples with concentrations above detection limits, compared to all other. That is, over 50 % of the levels were above the MDA for Sea Lion, Octopus, Black Rockfish, Halibut, Pacific Cod, and Walleye Pollock, and none were above the MDAs for lower trophic level organisms.

For the actinides, however, the differences were reversed. That is, primary producers (kelp, *Ulva*) and filter-feeders (Rock Jingles) had significantly more detectable levels of Pu-239,240, U-234, U-235, and U-238 than predators. Thus, the actinides do not appear to bioconcentrate with increasing position on the food web, or with age consistent with other studies. Older organisms, such as the higher trophic-level predators, did not bioaccumulate the actinides. This difference between species has important ramifications for selection of bioindicators for any future biomonitoring plan (see below).

Differences are to be expected among species, given the differences in life style, trophic level and other host-related differences among the species selected. Indeed, our target species were selected to represent different trophic levels, from primary producers (kelp and other algae), through filter feeders (Mussels, Rock Jingles), through grazers or herbivores (limpets, sea urchins, eiders), to different levels of predators (most birds, fish). Even within the predator group, there are wide differences in trophic level. For example, Sea Lion, Octopus, Halibut, Black Rockfish and Pacific Cod all had 50 % or more detect levels for cesium-137, while Bald Eagle and predatory fish had 0 % detect levels. Thus we found a clear trophic level difference in cesium-137 levels, even within the top level predators (Figure 11.8).

Among the fish we sampled, there are differences in size/age, trophic level, and vertical distribution in the water column (Mito et al. 1999) that would lead to different exposure rates. For example, Pacific Halibut are highest on the trophic scale of the fish we examined, but are bottom dwellers, while Pacific Cod are relatively high, but are middle-water column feeders (Mito et al. 1999). On the other hand, Pacific Ocean Perch are bottom dwellers and low on the trophic scale (Mito et al. 1999). Even within fish, age and size differences result in different potential for exposure. That is, larger predatory fish can eat larger prey fish, which themselves may have accumulated higher levels of radionuclides. Further, young Pacific Cod are themselves forage fish for other predatory fish (Brodeur et al. 1999), and dominate in some nearshore communities (Dean et al. 2000). 30-40 cm long Pacific Cod prey mainly on 1-year old Pollock, and Cod over 75 cm long prey on 2-5-year old Pollock (Mito et al. 1999). Tokranov (1992) similarly found that Cod fed mainly on fish, but diet varied seasonally. Pollock play an interesting role in that they are both prey to larger fish, and predators of smaller fish; young Pollock transmit energy from zooplankton to large-sized fishes.

Figure 11.8. Tim Stamm and Sean Burke dissecting Pacific Cod on deck of the *Ocean Explorer* (photo J. Burger).

Birds provided less information than expected, largely because of detection limits. The CRESP MDAs for the 1000 g samples were sufficient to detect concentrations at these levels if they had occurred for most species, and were sufficiently low to be below any human health risk levels. However, in general MDAs for birds, both in the literature and in the CRESP data set, are higher than for other species groups due to instrument difficulty with high density bird bones.

That high trophic level predators accumulate cesium-137 was expected, based on many other studies where predators bioaccumulate contaminants. However, the higher rate of detectable levels for actinides in kelp and Rock Jingles compared to predators was of interest, mainly because it indicates which species might be useful as a bioindicator. When actual levels are examined, the levels in the CRESP study were similar or within the range reported for other studies (see below, Tables 11.13 to 11.18).

Partly, the differences found in this study are consistent with reported bioconcentration factors for marine systems in general. Bioconcentration factors are the ratios between the concentration in an organism, and the concentration in its environment (or food). For Cs-137, the bioconcentration factor for macroalgae, such as the kelp species examined at Amchitka, range from only 50-75, whereas for fish it is 100, and for wading birds it is 400 (IAEA 1985, in press, Fisher et al. 1999, Appendix 2.A). The relative bioconcentration factor for birds should be viewed within the context of the difficulties of analysis (MDAs for birds are often an order of magnitude higher than for fish, partly due to tissue density differences). The bioconcentration factors predict that fish should have higher Cs-137 levels than algae.

In contrast, the bioconcentration factors for Pu-239,240 are reversed: algae have higher values than fish (Figure 11.9). In general, the bioconcentration factors for

macroalgae are up to 4,000 fold, while they are 100 for fish (IAEA in press). Thus, our findings are consistent with previous studies of the bioconcentration factors for these radionuclides, although we did not compute bioconcentration factors because we did not sample water or the food of specific animals.

Figure 11.9. *Fucus* (left) and *Alaria nana* (right) growing in the Amchitka intertidal zone (photo J. Burger).

Study Location Differences

Our sampling was designed to test whether there were differences in radionuclides between Amchitka and Kiska, and whether there were differences among the Amchitka test site marine ecosystems. Overall, we found no differences in the number of detectable and non-detectable values for cesium-137 between Amchitka and Kiska, but there were too few detectable levels for organisms from Amchitka to do an analysis of differences. Among higher trophic level fish, there was a higher proportion of Cs-137 detects at Kiska, but there were no differences in the levels between Amchitka and Kiska for cesium-137, and all other gamma radionuclides were not detected. These data indicate a lack of difference in radionuclide levels between Amchitka and Kiska, suggesting no current significant leakage of radioactive material from Amchitka. We had selected our reference site to be comparable biologically, with the same general benthic physiognomy and biota. Our analyses (see chapter 10) indicates that this is the case.

Overall, there were more detectable levels in the actinide than the gamma analyses, allowing us to compare Amchitka with Kiska. We found no significant differences ($P<0.05$) in the overall percent of detects between Kiska and Amchitka for Am-241, Pu-238, U-234, U-235, U-236, and U-238; with a small difference in Pu-239,240. We also found no statistically significant differences in the percent of samples with concentrations above detection limits in kelp, the species group with the largest number of analyses, and the lowest MDAs, except for Pu239, 240 and Cs-137. Thus, the data indicate few differences in actinides between Amchitka and Kiska, although the uncertainities in Pu-239,240 and CS-137deserve further study. Further, our biological diversity data (see chapter 10) also

indicated no differences, hence the lack of radionuclide differences are not due to differences in species presence or location (depth).

The finding of statistical differences in the proportion of values above MDAs between Amchitka and Kiska in Pu-239,240 and Cs-137, bears further comment. Firstly, it must be stressed that although the differences are significant, they are relatively small, and not likely to be meaningful biologically. That is, the differences are not great enough to suggest differences in biological or ecological functioning (e.g. behavior, growth, survival, reproductive rate). Thus, a difference can be statistically significant, without having biological significance, either because all the values are very low, or because the differences are small. For example, there could be a statistical difference in the mean length of fish, even if the difference might be less than a quarter inch. The difference could derive from there being almost no variance around the mean (all fish in each group were nearly the same size, as occurs in same-age fish). Yet, the quarter of an inch difference would not affect their ability to find food, eat prey fish of a particular size, or avoid predators.

There are two inter-island differences in radionuclide concentrations that bear discussion: 1) higher levels of Pu-239,240 in Amchitka Kelp, and 2) higher levels of Cs-137 in Kiska fish. Each of these will be discussed in terms of six possible explanations:

1. Analytic differences
2. Sampling differences
3. Statistical artifact including chance
4. Regional differences
5. Differences related to the island's locale.
6. Point source differences on Amchitka.

Possible explanations for inter-island plutonium difference in kelp

Although the plutonium values in Amchitka kelp are 200 times below the contamination level reported for the Irish Sea in the vicinity of the Sellafield nuclear reprocessing plant (CEFAS 2003,2004) and about 20 times below food safety standards, they require clarification. The finding of a higher detection rate for Pu-239,240 in kelp species at Amchitka compared with Kiska, prompted extensive discussion within CRESP. Since neither Pu-239 nor Pu-240 occur naturally beyond trace amounts (Myers and Lindner, 1971; Taylor, 2001), CRESP paid particular attention to this finding. There are six possible explanations: 1) analytic differences, 2) sampling differences, 3) statistical artifact including chance, 4) regional differences, 5) differences unique to the Amchitka locale, and 6) point source differences on Amchitka. The latter could indicate one of the three test sites as a local source.

It is also necessary to consider potential sources of the Pu-239 and Pu-240: Amchitka nuclear testing, global fallout from above ground nuclear tests or from accidents such as Chernobyl, other nuclear facility emissions including from production of nuclear materials reprocessing plants(including foreign facilities), and improper nuclear waste disposal including the ocean dumping of reactors and submarines.

Releases from Amchitka could have occurred at the time of the test shots when

there was some release of radionuclides to the surface, or through surface contamination which was considered minimal by Seymour and Nelson (1977) but not by Greenpeace (1996), followed by runoff into the sea, or through underground transport with groundwater (DOE 2002b).

Analytic Differences: If the samples from the two islands had been handled differently, analyzed at different times, or on different instruments, or by different methods, differences could arise as analytic error. The laboratory data were examined; the methods did not differ among islands. The detectable levels were well-distributed among the batches analyzed over a period of months. The method detection levels were not different. If anything the MDAs for the Kiska samples were slightly lower than those for Amchitka, which would have biased the results in the opposite direction. CRESP concluded that the plutonium results did not reflect analytic differences.

Sampling Differences: Attempts were made throughout the study to balance the number of samples from the three Amchitka sites and Kiska, within the analytic time and money constraints. Thus, most species were over-represented at Amchitka, because of the three test sites. A perfectly balanced design was not possible, because of the accessibility differences in the field. But if there were a bias, this could effect the mean values (species high in plutonium would raise the Amchitka average), but would have no effect on the proportion of detections. Thus the difference did not appear due to sampling.

Statistical Artifact: The comparison of proportions of detects and non-detects is the simplest statistical test which lends itself to 2 x 2 contingency table analysis. Since the data do not follow a normal distribution, we also used the non-parametric Kruskal-Wallis one way analysis of variance. There was extensive discussion over how to best represent data where a high proportion of values were below detection level. Table 11.11 illustrates the relationship among the values. The difference between islands is not a statistical artifact. CRESP also considered and rejected the role of multiple comparisons, since the possibility of a difference in Pu-239,240 was a primary hypothesis. However, in any analysis there is always the possibility (at least 1 in 20) that even a statistically significant finding can arise by chance alone. Chance can only be represented, never excluded or affirmed.

Regional Differences: Although Amchitka and Kiska are both in the western Aleutians, west of the International Date Line, they are nonetheless about 80 miles apart and historic global fallout was not perfectly homogeneous. Although CRESP's proposed seismic study was not conducted, there are differences in seismic and volcanic activity across the region. In addition, differences in ocean currents and local ocean floor topography can result in significant differences in sediment deposition, or transport and hence contaminant distribution, over very short distances (i.e., less than 1 km). While Pu-239,240 are almost exclusively anthropogenic (Taylor 2001), these factors can affect their distribution in the marine environment without linking them to a particular origin. Testing this possibility was beyond the scope of the CRESP Amchitka project.

Differences Unique to the Amchitka or Kiska Locale: These differences could include Amchitka as a source, but also could reflect oceanographic features (currents, sediment transport, ocean floor topography) which could result in different fate of radionuclides, even those deposited decades earlier as part of the global fallout from nuclear tests. Wave action, dynamics of the biotic community, sediment burial rates, could all differ between islands. The CRESP *Science Plan* originally included some components to address these issues, but they were not funded. A possibility is that contamination of the island surface occurring at the time of testing in 1965-1971, could have washed off into the sea, affecting several areas of the Amchitka littoral zone. However, the levels of surface contamination documented in the aftermath of the *Cannikin* test were concluded to mostly represent global fallout rather than local release (Seymour and Nelson 1977).

Point Source Differences on Amchitka: The three test shots: *Long Shot, Milrow,* and *Cannikin* are each potential sources, and the CRESP sampling regime was designed to detect seepage from each. The fact that detectable values occurred in kelp from all three sites, argues against point source seepage as an explanation of the differences. The CRESP study did not address the issue of possible runoff into streams (Greenpeae 1996).

In the end no clear explanation exists for the plutonium difference in the CRESP data set. Some local oceanographic features are yet to be characterized but differences between the islands is a real possibility. Further exploration will be required to determine whether the island difference is stable over time, and if so whether there is a distribution around Amchitka unrelated to the three test sites. The addition of another reference site to the east of Amchitka would also clarify the spatial relationship. The separate analysis of Pu-239 and Pu-240 may provide some clarification, since the ratio among these has been used to identify source. Declassification of information on the radionuclides and cavity features of the Amchitka test sites would clarify the source term and would also facilitate both interpretation of data, selection of radionuclides for future study, and communication to the public.

Possible explanation for inter-island cesium difference in fish

Similar explanations were considered for the inter-island cesium difference. The levels in these fish are well below any food safety guidelines, and they are mainly of interest in clarifying baseline and future monitoring issues. The main contribution of Cs-137 was global fallout from above ground nuclear tests, which contributed a more or less uniform “blanket” of fallout. We would not have expected this fallout load to have differed much between Amchitka and Kiska, but the post-fallout fate and transport could have differed as discussed for plutonium.

Analytic differences: The same argument applies to cesium analyses as to plutonium analyses. The laboratory quality control procedures precluded or would have detected differences arising from measurement error.

Sampling differences: Differential representation of species high in Cs-137 would have

biased the results. For example, Octopus and Sea Lion are predators with relatively high Cs-137 levels, but were only collected at Amchitka. Thus we focused on four species of high trophic level fish which were represented at both islands. The difference is, therefore, not due to sampling.

Statistical artifact including chance: In dealing with small samples, a change in just one or two data points can change the significance of the results. And one can neither affirm nor reject chance as the explanation for the higher proportion of detectable levels in Kiska fish. The fact that the mean values (Table 11.8) did not differ between islands, suggests that chance may account for the difference.

Regional differences and Differences Unique to the Amchitka or Kiska Locale: These two points are considered together. Oceanographic currents and sediment transport in the western Aleutians are highly complex, and could account for regional differences in Cs-137. Over the ensuing decades, the fate and transport of cesium could have differed. Coupled with the variable mobility of fish, this possibility would be difficult to assess.

Point source differences on Amchitka: Since the levels were higher in the Kiska fish, this explanation is not relevant to the Cs-137 levels.

In conclusion there may be regional or local geophysical, oceanographic, or climatic factors, as well as differences in fish populations, behavior, or prey availability, that favored the availability, update, and storage of Cs-137 in the Kiska fish. The levels are low and the differences between actual levels in the fish are negligible, but future study could clarify whether the difference in the proportion of detectable values persists. The role of chance cannot be excluded.

Temporal Patterns on Amchitka

Finally, it is useful to compare the CRESP data for fish with those from previous studies on Amchitka prior to 1975 (Isakson and Seymour 1968, Seymour and Nelson 1977). There are five fish for which data are available from both time periods (Table 11.12) and from the samples CRESP collected. The five species include Rock Greenling and Dolly Varden, relatively low trophic level species, as well as species that are top-level predators within the Amchitka marine ecosystem. For all species of fish, the mean Cs-137 levels in 2004 were below those reported prior to 1975 (which was after the nuclear test shots on Amchitka). Further, for three of the four species, the proportion of values above the MDAs was lower in 2004 compared to the pre-1975 levels (Table 11.12). Also Cs-137 has an approximately 30 year half life, so levels are expected to be about one half what would have been measured in 1975. Seymour and Nelson, based on their data from prior to 1975, conclude that there "has been essentially no escape of radionuclides from the sites of the *Long Shot*, *Milrow* and *Cannikin* underground nuclear detonations" (Seymour and Nelson 1977, p 595). The 2004 CRESP data from the same fish species were all below the levels they reported, which provides added assurance of environmental health with respect to fish, and consumers of these fish.

Similarly in 2004, all of our samples for algae were below the MDAs for Cs-137, while Isakson and Seymour (1968) had detectable values for all algae except *Ulva*. For example, their mean for *Fucus* was 0.56 to 0.78 Bq/kg (ww). Thus Cs-137 levels in algae are clearly lower in 2004 compared to before 1975.

Table 11.12. Temporal Patterns for Cs-137 for Fish at Amchitka. Comparison of Cs-137 levels in fish from Amchitka prior to 1975 (after Isakson and Seymour 1968, Seymour and Nelson 1977), and in 2004 (CRESP study). Given are means (with percent above the MDAs) in Bq/kg wet weight.

Species	1967-1968[a]	1965-1975[b]	2004 (CRESP)
Dolly Varden	not given	7.2 (2.4)[c]	0.70[d]
Rock Greenling	0.89 (100)	0.523 [e]	< MDA[f](0)
Walleye Pollock	0.96 (100)	not given	0.32 (50)
Halibut	1.24 (50)	0.58 [e]	0.14 (75)
Pacific Cod	1.14 (100)	not given	0.20 (50)

a. From Isakson and Seymour (1968)
b. From Seymour and Nelson (1977)
c. Average with/without the post *Long Shot* and post *Cannikin* high values.
d. Only the Dolly Varden sample from near the Amchitka air strip is included.
e. Percent above MDA not given
f. For Rock Greenling, the mean MDA for the CRESP study was 0.29, which is below the mean value reported prior to 1975.

The mean Pu-239,240 obtained at Kiska (0.018 Bq/kg) and Amchitka (0.039) were comparable to the only two *Fucus* values reported by Seymour and Nelson (1977) of 0.01 (Bq/kg) in Constantine Harbor and 0.04 Bq/kg near *Cannikin* in 1975.

Geographical Comparisons

One of the important methods of interpreting any contaminants data, including radionuclides, is to compare them with data from elsewhere in the region and elsewhere in the world. Such comparisons require separating the data from known contaminated sites from those that reflect background levels for a given region. Friedlander and other CRESP researchers conducted a detailed review of selected sources to provide the background for understanding radionuclide levels at Amchitka and Kiska (see Appendix 2.A). A second method of interpreting radionuclide data is to compare levels with those known to cause adverse effects on human health or the environment, and this aspect will be discussed further below and in chapter 12.

In the literature on radioactivity in biota there are more data for cesium-137 than for other radionuclides, which provides a comparative base for the CRESP Amchitka data. The source of most of the Cs-137 is global fallout from the era of nuclear testing which deposited Cs-137 and Sr-90 over much of the northern hemisphere (references in Appendix 2.A). Table 11.12 gives representative concentrations of Cs-137 of representative marine biota from the Northern Hemisphere for species groups analyzed in the CRESP study. Samples from the Irish Sea reflect the local contamination from the Sellafield nuclear

fuel reprocessing plant. The table compares the Amchitka data to previously published data. In general, the MDAs in the Amchitka study were similar to those in the literature, although we did not have values above the MDA for algae, mollusks and birds. For example, the average concentration for algae was 1.97 Bq/kg for the Irish Sea. Further, the levels of Cs-137 in fish and the Sea Lion were similar to other Northern Hemisphere values, and well below those of the contaminated Irish Sea. Thus the levels at Amchitka are within the general environmental levels in the Northern Hemisphere (Table 11.13).

Examining the Northern Hemisphere Cs-137 data in more detail illustrates the difference in values within groups, such as algae (Table 11.14), molluscs (Table 11.15), and birds (Table 11.16). In general, the values we found for Amchitka/Kiska were similar or below those reported for other sites, and below the MDAs for Amchitka/Kiska. The CRESP MDAs for algae (1000 g samples, 0.10-0.40 Bq/kg, Cs-137) were within or below the values from the other sites. The key finding is thus, that the Cs-137 levels CRESP found for algae are all below the MDAs, which were below those found in other, previous studies from elsewhere. This was also the case with mussels and jingles (see also Appendix 11.C).

Table 11.13. Geographical Comparison for Cs-137 by Species Group. Cesium-137: Concentration in representative marine biota of the northern hemisphere[a], comparing concentrations in the contaminated Irish Sea versus all other sites from which data are available. Data are primarily from 1999-2003 samples. All concentrations converted to Bq/kg (wet weight). For fish, values below their MDA were converted to half the MDA for calculation of the average. MDA= Minimum detectable activity level

	IRISH SEA	OTHER SITES	CRESP Amchitka Study
ALGAE			
Average Concentration	1.97	0.2	<mda
Range	<mda-7.7	<mda-1.14	0.10-0.40[b]
Number of Analyses	308	135	8/12[c]
MOLLUSKS			
Average Concentration	3.98	0.03	<mda
Range	<mda-16	<mda-0.41	0.09-0.43[b]
Number of Analyses	323	112	12/8
FISH			
Average Concentration	4.64	0.22	0.04-0.31
Range	0.31-13	0.04-0.33	0.09-0.60
Number of Analyses	203	718	34/98
BIRDS			
Average Concentration	124.8	1.62	<mda
Range	9-613	<mda-5.6	0.08-0.75[b]
Number of Analyses	15	15	12/55
MARINE MAMMALS			
Average Concentration	NA	0.31	0.41
Range	NA	<0.2-1.2	0.09[b]
Number of Analyses	NA	19	2/0

a. The Northern Hemispheric data has been narrowed, for the purposes of this report, to Information from CEFAS 2003 and 2004 reports), RPII (2003 and 2004 reports), RAME (2003 and 2004 reports), JCAC (2003 and 2004 reports), Hong Kong Observatory (1999-2004 reports), and selected Russian data (Matishov and Matishov, 2004). The Irish Sea data extracted from database reports from RPII (2003-2004), CEFAS (2003-004 RIFE-8 and 9), and BNFL (2002-2004). Supplementary data were obtained from published studies to address key data gaps.

b. Range of MDAs given when all levels were below MDA for that group. MDA's for 1000 g samples.

c. Given are numbers of 1000 g/number of 100 g analyses.

Table 11.14 . Geographical comparison of Cs-137 levels in marine algae. Cesium-137 in marine algae from the Irish Sea, other northern hemisphere sites and Amchitka-Kiska. All values are converted to Bq/kg on wet weight basis. Values below the MDA were replaced by one half the MDA for computation of the average. Sources given at the bottom.

	Fucus	*Alaria*	*Porphyra*	*Ascophyllum*	*Ulva*	*Undaria*	Source
Irish Sea							
Average (range) Bq/kg	2.17 (nd-8.1)		1.76 (1.7-1.9)	0.54 (0.4-0.70			1,2.3,4
number (% non-detects)	169 (<1%)		121 (0%)	7 (0%)			
North Sea							1
Average (range) Bq/kg	0.23 (0.11-0.30)						
number (% non-detects)	10 (0%)						
Norwegian Sea							1,2,3,4
Average (range) Bq/kg	0.15 (0.03-0.53)						
number (% non-detects)	17 (0%)						
North Atlantic							4,5,6,7
Average (range) Bq/kg	0.19 (0.05-0.73)		0.3 (0.28-0.37)	0.08 (0.08)			
number (% non-detects)	14 (28.6%		16 (0%)	1 (0%)			
Japanese Coast							8,9
Average (range) Bq/kg						0.02 (0.002-0.04)	
number (% non-detects)						18 (0%)	
Hong Kong Coasts							10,11,12,13,
Average (range) Bq/kg			nd (<mda)		nd (<mda)		14
number (% non-detects)			4 (100%)		8 (100%)		
Celtic Seas							5,6,7,8
Average (range) Bq/kg	0.18 (nd-0.41)						
number (% non-detects)	12 (41%)						
Barents Sea							1,2,3
Average (range) Bq/kg	0.12 (0.02-0.29)						
number (% non-detects)	7 (28.5%)						
Greenland Sea							4
Average (range) Bq/kg	0.39 (0.27-0.52)			0.06 (0.06)			
number (% non-detects)	3 (0%)			1 (0%)			
Baffin Bay							4
Average (range) Bq/kg	0.18 (0.06-0.58)						
number (% non-detects)	6 (0%)						
Skagerrak							1,2
Average (range) Bq/kg	0.70 (0.27-1.14						
Number (% non-detects)	8 (0%)						
Amchitka-Kiska							
Average (range) Bq/kg	<mda		<mda		<mda		
Number (% non-detects)	4 (100)		13 (100)		3 (100)		

Weighted mean of all sites (Bq/kg ww) 1.57; Unweighted Mean of averages of all sites (Bq/kg ww) 0.48

Unweighted mean average of all sites excluding Irish Sea (Bq/kg ww) 0.27

1. Gafvert, T et. al.2003.; **2** Norwegian Radiation Protection Authority. 2004.; 3. Gwynn JP, et.al., 2004.; 4. Dahlgaard H, et. al. (2004) 53-67.; 5-6.CEFAS,Radioactivity in Food and the Environment, 2003 and 2004.; 7.Radiological Protection Institute of Ireland (RPII). 2002.; 8-9.Japan Chemical Analysis Center. Number 138, October, 2003, Number 139, August, 2004.; 10-13.Hong Kong Observatory. Environmental Radiation Monitoring in Hong Kong. Technical Report(s) No 19-22, 1999-2002.; 14.Li SW and Yeung KC. Hong Kong Observatory Summary of Environmental Radiation Monitoring in Hong Kong 2003. Technical Report No.23, 2004.

Table 11.15. Geographical Comparison for Cs-137 for Molluscs. Cesium-137 levels in mollusks from the Irish Sea and other areas of the Northern Hemisphere[a] including CRESP's Amchitka data. Shown are mean values (ranges) and number of samples analyzed including the percent of non-detects for each. All values have been converted to Bq/kg on a wet weight basis and values below the MDA have been converted to half the MDA for computation of the mean. If (mda) is shown for range, then all values were below the MDA. References given in footnote a.

	Mussel	Oyster	Scallop	Winkle	Cockle	Whelks	Limpets	Jingles
Irish Sea								
average (range) Bq/kg	2.4(nd-4.6)	0.18(0.1-.3)	.16(<.17-.3)	6.69(<.2-16)	3.7(1.5-5.2)	1.1(0.6-1.9)	7.2(5.7-10)	
Number (% non-detects)	91(1.1%)	12 (0%)	27 (55.6%)	111 (7.2%)	50 (0%)	16 (0%)	16 (0%)	
North Sea								
average (range) Bq/kg	.1(<.1-0.17)			.15(<.1-.41)				
Number (% non-detects)	5(20%)			6(50%)				
Norwegian Sea								
average (range) Bq/kg	.16(.07-.34)							
Number (% non-detects)	4 (0%)							
North Atlantic								
average (range) Bq/kg	0.03(nd-.06)							
Number (% non-detects)	17(29.4%)							
Japanese Coast								
average (range) Bq/kg	0.01(0.01)	0.01(.01)	0.02(0.02)					
Number (% non-detects)	2 (0%)	2 (0%)	4 (0%)					
Hong Kong Coasts								
average (range) Bq/kg	<.02(mda)	<.02(mda)		<.02(mda)	<.02(mda)			
Number (% non-detects)	40 (100%)	2 (100%)		11 (100%)	19 (100%)			
Amchitka-Kiska								
average (range) Bq/kg	<mda						<mda	<mda
Number (% non-detects)	2 (100)[b]						2 (100)[c]	3 (100)[d]

a. REFERENCES: Japan Chemical Analysis Center. Radioactivity Survey Data in Japan: Environmental and Dietary materials. Number 138, Oct.2003. Japan Chemical Analysis Center: Radioactivity Survey Data in Japan: Environmental and Dietary Materials. Number 139, Aug. 2004. Norwegian Radiation Protection Authority. Radioactivity in the Marine Environment 2002: Results from the Norwegian Marine Monitoring Programme (RAME), 2004. Gafvert T, Foyn L, Brungot AL et al. Radioactivity in the Marine Environment 2000 and 2001. Results from the Norwegian National Monitoring Radiation Programme (RAME), 2003. Ryan TP, McMahon CA, Dowdall A, et al. Radioactivity monitoring of the Irish marine environment 2000 and 2001 (The Radiological Protection Institute of Ireland), 2003. Radiological Protection Institute of Ireland. Environmental Radiation Marine Monitoring Programme. 2002 Report (2004). Hong Kong Observatory. Environmental Radiation Monitoring in Hong Kong. Technical Report(s) No 19-23, 1999-2003.

b. 344 individuals in the 2 samples.

c. 150 individuals in the 2 samples.

d. 322 individuals in the 3 samples.

Table 11.16. Geographical Comparison for Cs-137 for Birds. Cesium-137 concentrations in marine birds from the Irish Sea and the Barents Sea. Reported are average concentrations and ranges converted to wet weight basis. For values below the MDA a value of half the MDA was used to compute the average. The time period for the two samples is separated by a decade.

EAST IRISH SEA REGION	**1980-1984**	**Concentration Bq/kg (w/w)**	**Range**	**Number of Analyses**
Grey Lag Goose	*Anser anser*	57.7	57.7	1
Black-headed gull	*Larus ridibundus*	13.8	<mda-27.9	8
Great Black-backed Gull	*Larus marinus*	158	158	1
Lesser Black-backed Gull	*Larus fuscus*	9	9	1
Herring Gull	*Larus argentatus*	155.8	9.7-301.9	2
Oystercatcher	*Haematopus. ostralegus*	612.8	578.8-647	2
BARENTS SEA REGION	**1995-1996**	**Concentration**	**Range**	**Number**
Great Black-backed Gull	*Larus marinus*	4	2.4-5.6	2
Common Gull	*Larus canus*	1	1	1
Great Skua	*Stercorarius skua*	3.5	3.0-4.0	2
Spotted Redshank	*Tringa erythropus*	4.3	4.3	1
Little Sandpiper	*Calidris minuta*	1.5	1.5	1
Common Eider	*Somateria mollissima*	0.17	nd-0.30	4
Black Guillemot	*Cepphus grylle*	0.43	0.43	1
European [Great] Cormorant	*Phalacrocorax carbo*	0.45	<0.2-0.64	3
AMCHITKA-KISKA 2004				
Common Eider	*Somateria mollissima*	Non-detect		12
Tufted Puffin	*Fratercula cirrhata*	Non-detect		13
Glaucous-winged gull	*Larus glaucesceus*	<0.09[c]	<mda – 0.09	32
Pigeon Guillemot	*Cepphus columba*	Non-detect		13
Bald Eagle	*Haliaeetus leucocephalus*	Non-detect		1

a=Samples of 1980-1984 reported by Lowe 1991.
b=Samples of 1995-1996 reported by Matishov and Matishov 2004
c= One gull had a value <mda (=0.09 Bq/kg).

Fish provide the best biota group for further analysis of Cesium-137. For the gamma analysis, there were detectable levels for cesium-137. Below we present data on cesium-137 from a range of studies with fish that are closely-related to those we examined at Amchitka (Table 11.17). Mean values for CRESP data (for 1000 g samples), and those presented in this table, were computed substituting half the MDA for each sample below the MDA. If we add our 100 g samples, the number of pooled analyses would be greater for each species, and the mean values would be lower (since all of the 100 g samples were non-detect for cesium-137).

Algae provide the best biota group for comparison of other radionuclide (Table 11.18). The values from Amchitka/Kiska are generally similar (or below) those reported from elsewhere in the Northern Hemisphere.

Table 11.17. Geographical Comparison for Cs-137 for Fish. Cesium-137 concentrations in representative marine fish from the Irish Sea and other northern hemisphere sites, with Amchitka data shown for comparison. All values have been converted where necessary to Bq/kg wet weight. Values below the MDA were converted to half the MDA for computing the mean value. Data are primarily from 1999 through 2003.

Location/Sea	Species	Concentration	# (pooled)	Reference
Japan	Tilefish	0.12	2	Japan Chemical Analysis
	Greenling	0.12	2	Center, 2003
	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&Matishov, 2004
	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
	Bartail Flathead	0.06	11	1999-2003
Barents Sea	Cod	0.29	53	Gafvert et al, 2003
	Haddock	0.2	10	CEFAS, 2003&2004
				Ryan et al, 2003
North Sea	Cod	0.38	21	CEFAS, 2003 & 2004
	Haddock	0.2	10	Gafvert et al, 2003
	Plaice	0.21	19	
Norwegian	Cod	0.32	20	Gafvert et al, 2003
	Saithe	0.27 to 0.64	4	CEFAS, 2003& 2004
	Mackerel	0.14	4	Ryan et al, 2003
N. Atlantic	Cod	0.28	3	CEFAS, 2003&2004
	Plaice	0.36	3	Gafvert et al, 2003
	Haddock	0.47	3	
	Mackerel	0.09	5	
Channel	Cod	0.2	8	CEFAS, 2003&2004
	Plaice	0.06	16	
	Mackerel	0.19	8	
Irish	Cod	6.44	75	Ryan et al, 2003
	Plaice	3.77	60	CEFAS, 2003&2004
	Mackerel	0.31	39	
	Flounder	11.0	19	
	Haddock	1.1	10	
Baltic	Cod	8.86	7	CEFAS, 2003&2004
Amchitka	Rock Greenling	0.04	5	CRESP expedition 2004
	Pacific Cod	0.20	14	(1000 g samples)
	Halibut	0.14	4	
	Black Rockfish	0.14	3	

Table 11.18. Geographical Comparison for Actinides for Algae. Comparison of levels in Kelp from elsewhere and Amchitka

	Waterways with Reprocessing Plants		**Selected Regions of the Northern Hemisphere**					
Seaweed	Irish Sea *Fucus*	Channel France *Fucus*	Greenland *Fucus*	Baffin Bay *Fucus*	North Sea *Fucus*	Norwegian & Skagerrak *Fucus*	Japan *Undaria*	Amchitka algae[a]
Sr-90								
Average	2.5						0.02	<mda
Range	1.7-3.1						0.009-.056	-[b]
Number	16						18	12
Tc-99								
Average	319.3		6.71	0.97	52.3	41.9		<mda
Range	17.5-742		4.04-7.25	0.72-1.20	19.2-80.2	5.05-69		
Number	24		11	6	9	16		12
I-129*								
Average		5.38						<mda
Range		0.82-12.16						
Number		17						12
U-234**								
Average	0.32							0.311-1.00
Range	0.23-0.86							
Number	28							48
U-235**								
Average	0.02							0.025-0.15
Range	0.007-0.28							
Number	28							48
U-238**								
Average	0.78							0.246-2.74
Range	0.42-3.53							
Number	28							48
Pu-239,40								
Average	8.4			0.05		0.01		0.014-0.057
Range	6.0-11			<.01-.09		0.004-0.02		
Number	20			6		21		48
Am-241								
Average	4.03							0.013-0.018
Range	3.2-4.8							
Number	16							60
Cm-242								
Average	0.005							
Range	0.003-.006							
Number	8							

a. *Alaria, Fucus and Ulva*; b. Range of MDAs when no values were above the MDAs.; ** MARINA II, 1989-1990 samples; *Frechov and Calmet, 2003 in samples near La Hague reprocessing plant region of Channel. CEFAS (RIFE-8), 2003; CEFAS (RIFE-9), 2004.

Implications of Radionuclide Data

The radionuclide data generated by this study have implications for evaluating whether there is currently a risk to human and environmental health, which species might be useful for designing a future biomonitoring plan, and whether they reduce any uncertainties in the groundwater or human health risk assessments. The presence in the marine environment, including the sea surface, intertidal, and benthic zones, of a wide range of biota at different trophic levels clearly indicates that there are organisms that would be a risk should seepage occur in the future. The degree of exposure of biota to radionuclides depends on the location and levels of the seepage of radionuclides into the marine interface, uptake rates, and to the complexity of food webs, among other factors, as well as to depth, since most organisms live in a particular zone (refer back to Figure. 10.4 in chapter 10). The similarity in the organisms found and collected at Amchitka and Kiska suggests that there are flourishing marine communities adjacent to the nuclear test shots. An earlier study (Baskaran et al. 2003) compared levels of radionuclides in sea otter (*Enhydra lutris*) skulls from near Amchitka with those collected near Adak, and did not find significant differences attributable to the Amchitka nuclear blasts (Baskaran et al. 2003).

As mentioned above, the initial field radiologic scanning of specimens did not reveal any elevations above background. Although relatively crude, the detection of elevated levels would have triggered a safety protocol described in the HASP (Appendix 4.E) as well as notification of the DOE and the Advisory board members. The laboratory analysis was much more sensitive than the shipboard screening values. Nonetheless, the levels of radionuclides relevant to human and ecological health were well below human health risk levels (see chapter 12 for a full discussion). Further, the differences among species in levels of different radionuclide isotopes can provide insights into which species may be most useful as bioindicators. While there are many different factors that influence the selection of bioindicators, from a biological perspective, the most useful bioindicators are those species that first accumulate radionuclides because they can be used as an early warning of any potential future problems. One difficulty, of course, is that no single species has the highest accumulation of all the radiological isotopes of interest. In the present study, there were few detects from the gamma analyses, except for cesium-137. Thus, for iodine (no samples with a concentration above detection limits of 71 samples), cobalt-60 (no detect of 173), americium-141 (no detects of 91), and europium (no detects of 173) the data do not provide any useful information for selection of biota for bioindicators. For cesium-137, Sea Lion, Glaucous-winged Gull, Octopus, Black Rockfish, Halibut, Walleye Pollock and Pacific Cod, had the highest percent of detectable levels. They are, interestingly enough, some of the species of greatest interest to Aleut subsistence fishers (all seven), commercial fisheries (the latter three), and resource trustees (all nodes on the food web).

In contrast, for the actinides, low trophic-level organisms had the most samples with detectable levels. That is, kelp and Rock Jingles had a higher percentage of detects than did predators. Further, the intertidal kelp (*Alaria nana* and *Fucus*) had higher percentages of detects than the subtidal kelp (*Alaria fistulosa*). This was true for both the naturally-occurring and the anthropogenic actinides (except for U-236, where *A. fistulosa* had more

detects). These data suggest that a suite of bioindicator species must be selected to adequately address the issues of isotope presence and potential health risks to biota and humans.

APPENDICES FOR CHAPTER 11 (See attached CD-ROM)

11.A. Preparation and Analysis Phase: Anti-cross Contamination Quality Control and Assurance Report by C. Volz
11.B. Relationship between Reported Values and Minimum Detectable Activities by V. Vyas
11.C. Additional Comparative Levels of Radionuclides in Marine Biota by B. Friedlander, M. Gochfeld, J. Burger, V. Vyas, C.W. Powers
11.D. Statistical Analysis of Data Sets with Values below Detection Limits by M. Gochfeld, J. Burger, and V.M. Vyas