# Selecting Species and Radionuclides for Assessment

SUMMARY

The biological component of the Amchitka *Science Plan* had two main components: collection of biological specimens according to a plan that reflected subsistence foods, commercial fish, and food chain and ecosystem receptors, and the radiological analyses of these species. Two critical steps that occurred between these two phases were the selection of specimens from our inventory (species, tissues and locations) for radionuclide analysis, and the selection of radionuclides for analysis. While both were discussed in the original Amchitka *Science Plan*, they required refinement. Radionuclide analysis was conducted at the Idaho National Laboratory (INL) and at Vanderbilt University. In this chapter we address the following:

1) Our overall radiological analysis plan.
2) The species (and tissues) to be examined in the human health screen.
3) The species to be selected for additional analyses of alpha emitters to achieve statistical comparisons.
4) The species (and tissues) to be analyzed for gamma emissions at Vanderbilt for the broad food web analysis.
5) The samples to be analyzed in larger quantities for longer count times to achieve more sensitive detection levels in the gamma counters.
6) The spatial distribution across the four sampling locations (three Amchitka test sites and Kiska (the reference site).
7) The radionuclides that should be examined in each species and tissues.

These questions are interrelated, and iteration was required to achieve the optimal balance between: 1) species and tissues, 2) Aleut foods, commercial fish, and species at different trophic levels on the food web, 3) collection locations (three Amchitka test shots, and Kiska), 4) radionuclides of interest for human or ecological health, and for possible identification of a source signature.

In this chapter we report on the rationales for selection of biota for analysis, selection of radionuclides for analysis, and the melding of these two to arrive at an overall analytic sampling plan. The factors that entered into our selection of species were important for human health and the food chain, potential as bioindicators for future biomonitoring, distribution among the test shots and reference sites, mobility, trophic-level, potential age, and availability in our inventory. The factors that entered into our selection of radionuclides were those of primary importance to the health of humans or biota, and those that might contribute to our understanding of the source of any radionuclide levels detected. A five-pronged radionuclide strategy was developed that included:

1) Initial radionuclide analysis at INL, primarily Human Health Screening and Analysis at INL , including gamma spectroscopy, and strontium and actinide analysis,
2) Follow up analysis of kelp and bone to achieve statistical power,

3) Gamma Analysis of a broad range of Aleut foods and food chain organisms at Vanderbilt,
4) Analysis of larger samples of 1000 grams for longer times, at Vanderbilt for gamma analysis to improve quantification of Cs-137, and
5) Multi-faceted quality control program for radionuclide analysis.


INTRODUCTION

Selection of the species for radionuclide analysis, and the selection of radionuclides for analysis were both critical to our ability to achieve the goals of the biological component of the Amchitka *Science Plan*. The goals that are particularly relevant to the selection of species and radionuclides for analysis include:

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 species are appropriate indicators for long-term monitoring?

These questions guided our discussions of the main issues surrounding the selection of species and radionuclides for analysis (see Appendix 8.A). The initial selection of species to be collected, and the radionuclides for analysis, were set forth in the Amchitka *Science Plan*. However, refinement was necessary to reflect changes in target species that resulted from meetings with stakeholders between the completion of the *Science Plan* (summer 2003) and the initiation of the Expeditions (summer 2004) (see chapter 8), changes that resulted because of presence or abundance in the marine environment around Amchitka, or logistical consideration imposed by weather, diving constraints, and available sampling time. Similarly, some refinement in the suite of radionuclides for analysis occurred because of stakeholder discussions, additional information from an extensive literature review, and logistical considerations of timing, specimen flow through laboratories, and available funds for analysis.

The main questions we address in this chapter are:

1) What should be our overall radiological analysis plan?
2) What species (and tissues) should be examined in the initial broad radionuclide screen?
3) What species should be selected for additional analyses to achieve statistical comparisons?
4) What species (and tissues) should be examined for gamma emissions at Vanderbilt in the broad food web analysis?
5) Which samples should be analyzed in larger quantities for longer count times to achieve more sensitive detection levels in the gamma counters.
6) How should the analyses be distributed across the Amchitka test shots and Kiska (the reference site)?

7) Which radionuclides should be examined in what species and tissues?

These questions are interrelated, and iteration was required to achieve the optimal balance between: 1) species and tissues, 2. Aleut foods, commercial fish, and species at different trophic levels on the food web, 3) collection locations (three Amchitka test shots, Kiska), 4) radionuclides of interest for human or ecological health, and for possible identification of the source of the radionuclides if found at significant levels.

Within the framework of examining radionuclides in Aleut/Pribilof Islanders' foods, commercial fish, and ecoreceptors, we aimed to integrate considerations of trophic level, mobility, species type (kelp, invertebrates, fish, birds), and collection location. Our overall plan was to be able to assess, where possible, whether there were differences in radionuclide levels among the three Amchitka test shots, and between Amchitka and the reference site (Kiska). Because different organisms have different mobilities, we also wanted to ensure that our radionuclide analysis plan reflected these differences.

Our ability to have a meaningful comparison between Amchitka and Kiska depended upon Kiska being an appropriate reference site for Amchitka. This required an ability to find and collect the same species at both places in sufficient numbers to allow for radionuclide analysis. For the most part we were able to collect the same target species at both Amchitka and Kiska. See chapter 10 for an analysis of the species found and collected at both places. Our ability to extrapolate the radionuclide results from the fish caught by scientists to subsistence Aleut/Pribilof Islanders was also an important aspect of this study. This issue is also addressed in chapter 10.

In this chapter we discuss our overall strategy for selecting species and radionuclides for analysis to meet our goals listed above (fig 8.1), and the specific considerations that were used to select species and radionuclides for analysis. Thus, this chapter first discusses our overall radionuclide analysis approach, then discusses in detail the selection of species for analysis and then the selection of radionuclides for analysis within the over all approach.



Fig. 9.1. Commercial fisheries interests in the Aleutians. Left: Unloading a haul of Pollock at Dutch Harbor. Right: Aleut fishing co-op at Atka, preparing bait for Halibut long-lining. (Photos J. Burger)

METHODS AND APPROACHES

Selecting the species (and tissues) to examine, and the radionuclides to analyze was a multi-step process that involved: 1) Discussions among the CRESP team about the major issues involved, 2) Proposals by the Biological Team Leader and the Radiological Team Leader about which species, tissues and radionuclides to examine, 3) Additional discussions by the CRESP team about each plan to optimize use of laboratory and financial resources, 4) Finalization of an initial analytic sampling strategy, 5) Review of preliminary analytic screening data and detection levels, and 6) Revision of analytic strategy to enhance sensitivity and validity.

CRESP accomplished its species selection and radionuclide selection as follows. Our approach was to: 1) develop an overall analytical approach, 2) select species for each phase of the analytical approach, 3) select radionuclides for analysis. These three approaches are related and continued iteration and refinements were necessary throughout the analysis phase (August 2004 to July 2005). Our overall approach was set forth in the initial Amchitka *Science Plan*, and refined according to the principles described in this chapter.

Our selection and collection of organisms benefited greatly from the detailed studies in The Environment of Amchitka Island, Alaska (Merritt and Fuller 1977); particularly chapters on marine algae (Lebeduik and Palmisano 1977); invertebrates(O'Clair 1977); fish (Simenstad et al 1977); birds (white et al 1977); and mammals (Abeqqlen et al 1977).

RESULTS

Overall Radionuclide Approach

An overall radionuclide approach was developed, and modified as a result of specimen availability, analytical constraints, time and money constraints, and preliminary data. Our overall protocol for the biological component was to select species as indicators of the safety of the subsistence marine foods and commercial fish and the health of the food chain, and to provide useful information for the future biomonitoring plan. We developed a five pronged approach, including multi-faceted quality control for radionuclide analyses (see Table 9.1, Fig. 9.2).

Table 9.1 Overall radionuclide analytical approach
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1. Human Health Screening and Analysis (INL and Vanderbilt)

      Representative Aleut Foods
      Represent study sites (*Milrow, Long Shot, Cannikin*, Kiska)
      Represent all levels of food chain (kelp to top predators)
      Human health and Signature radionuclide analysis

2. Additional actinide analysis for bone and kelp (INL)
      Additional analyses of species considered in the human health screen for statistical analysis of alpha emitters in bone and kelp.
      Analysis of species of special concern available in low numbers, along with the health screen, to provide information on source.

3.  Larger samples for enhanced detection levels (Vanderbilt)
      Analysis of larger samples (1000 g) for longer count times to achieve still lower detection limits for Cs-137, I-129, Eu-152, and Co-60.

4. Gamma Analysis of a broad range of Aleut foods and food chain organisms (Vanderbilt)
      Specific isotopes of biological interest
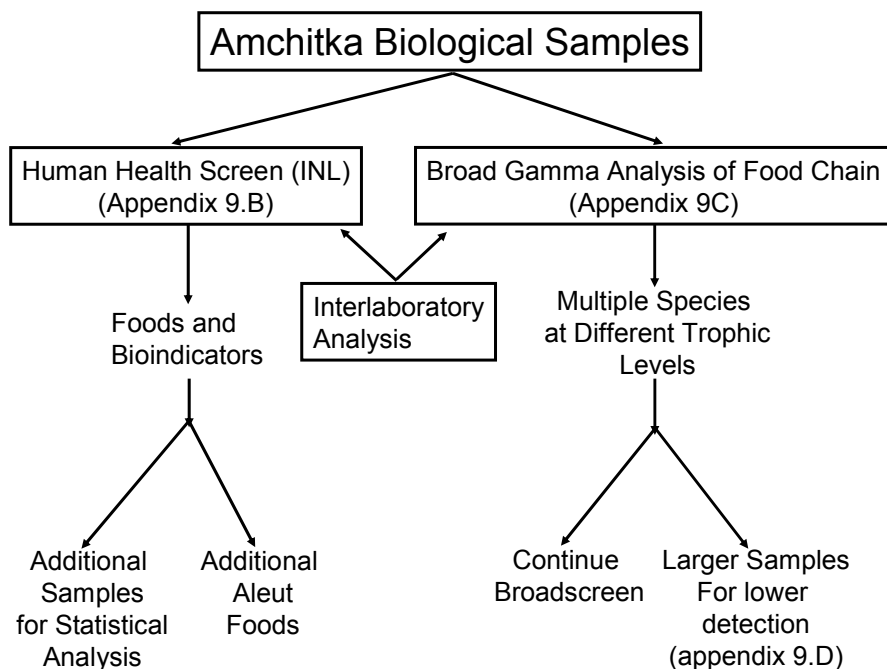      Wider spectrum of species and locations
      More samples of each species to achieve statistical power

5. Interlaboratory Analyses for quality control
      Analyze subset of sample at both INL and Vanderbilt Laboratories for quality control
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Fig. 9.2. Overall plan for analysis of biological samples from the Amchitka Expedition.

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                    ┌─────────────────────────────────┐
                    │  Amchitka Biological Samples     │
                    └─────────────────────────────────┘
                       ╱                        ╲
        ┌──────────────────────────┐   ┌──────────────────────────────┐
        │ Human Health Screen (INL)│   │ Broad Gamma Analysis of Food  │
        │     (Appendix 9.B)       │   │   Chain  (Appendix 9C)        │
        └──────────────────────────┘   └──────────────────────────────┘
                │          ╲         ╱           │
                │       ┌──────────────────┐     │
                │       │ Interlaboratory  │  Multiple Species
           Foods and    │ Analysis         │  at Different Trophic
          Bioindicators └──────────────────┘  Levels
                │                                  │
           ╱         ╲                        ╱          ╲
     Additional    Additional           Continue      Larger Samples
      Samples        Aleut             Broadscreen      For lower
    for Statistical  Foods                              detection
      Analysis                                        (appendix 9.D)
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Thus, there were four primary sets of radionuclide analyses (refer to Table 9.1 above):

1.  Human health screening of a wide range of biota for radionuclides that have the potential to accumulate in, soft tissues and algae that are typically consumed or are important for the ecological food chain. The primary radionuclides of interest for this set of analyses were Cs-137, I-129, Sr-90, Tc-99. Sr-90 also preferentially accumulates in bone based on its chemical behavior, so bone samples from biota were also analyzed for Sr-90. While these radionuclides are predominantly man-made, their presence may have originated from a variety of sources, including world-wide historic atomic testing, production of nuclear materials, or nuclear reactor accidents. For these measurements, detection limits were based on being substantially below food safety levels.

2.  Screening a wide range of biota for radionuclides that have the potential to provide insights into the origin of any detected radionuclide contamination. The primary radionuclides of interest for this set were U-234, U-235, U-236, U-238, Pu-238, Pu-239,240, and Co-60. On an activity basis, uranium isotopes U-234 and U-238 occur in nature in equal proportion (U-234/U-238 between 0.914 and 1.14 for naturally occurrence) while U-235 is present in nature at levels 1/10 of U-234 and U-238 (activity basis; Fetter, 1993). When uranium isotopes are present from man-made activities (e.g., nuclear reactors or atomic blasts), they have an increased proportion of U-235. Pu-238, Pu-239,240 are only present from man-made sources. When present at sufficient levels to accurately determine the isotopic ratios, these ratios are indicative of their source. Co-60 is not a signature isotope for atomic blasts, but does originate from other types of nuclear activities

(e.g., nuclear materials production, medical devices, etc.). Based on the chemical nature of these radionuclides, if present, they were expected to be found in the bone of fish and birds and in kelp. Eu-152 also was of interest in the bone because of its potential origin from the Amchitka test shots. For these measurements, detection limits were based on being as low as practical, considering available sample size and analytical methods. These detection limits also were below food safety levels.

3. Analysis of a selected set of soft tissue and kelp (as in the first set above) for Cs-137 and I-129 with larger sample quantities to achieve lower detection limits comparable to those reported from other studies for background levels from historic nuclear activities.

4. Additional sample analyses of selected species and sample types for statistical purposes or because of specific local interest in particular species of biota but where insufficient samples were collected for full comparative analysis.

SELECTION OF BIOTA SPECIES FOR RADIONUCLIDE ANALYSIS

1.  Human Health Screen

The most important criteria for selection was whether the species was eaten by people, or was an important link in the food chain leading to people. The factors that influenced our selection of species (and tissues) related to trophic level, mobility, and suitability. We wanted to select species that were both consumed by people and represented different levels on the food chain. Understanding the mobility of species allowed us to examine species that represented local exposure (sedentary and locally mobile) as well as regional exposure (highly mobile or migratory). Mobility is a reflection of individual range, and how far an organism moves during its life. The characteristics used to select species are shown in Tables 9.2, 9.3 and figure 9.3. Thus, two of the features that needed to be considered are mobility and location. Location was critical because we wanted to have organisms at different depths and distances from the Amchitka test shots to insure that, if there were seepage, we would have selected appropriate indicators (Fig. 9.3 and 9.4).

In addition, species selected for the initial screen were available from all four study sites (the three Amchitka test shots and Kiska). Using these criteria, we selected the species for the initial human health screen (see Tables 9.5 and 9.6).

Table 9.2. Bioindicator qualities for human exposure, top level predators, and self-exposure (after Burger and Gochfeld 2001,2004).

| FEATURE | IMPORTANCE | SPECIES | |
| --- | --- | --- | --- |
| Human Exposure | Can it directly affect people because they are eaten by people? | All species, Except: Kelp (brown algae), and eagle | |
| Food-chain Exposure | Is it at the base of the food chain? | All algae | |
| Receptor Exposure | Can it directly impact the health of top-level predators (predatory fish, marine birds, and marine mammals? | Blue Mussel Limpets Chiton Sea Urchin Atka Mackerel Black Rockfish Yellow Irish Lord Red Irish Lord Dolly Varden Rock Greenling | |
| Top-level Predators | Bioindicators of exposure of humans and of other top-level predators | Eagle Glaucous-winged Tufted Puffin Pigeon Guillemot Octopus Halibut Pacific Cod | Gull |
| Self-exposure | Bioindicator of effects of exposure on the organisms themselves | All species | |

Table 9.3. Life history traits influencing selection of species for human health. Species with * were used for the initial human health screen, all others were used for the 2$^{nd}$ tier gamma analyses discussed in the next section

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| MOBILITY | IMPORTANCE | SPECIES |
|---|---|---|
| Sedentary<br>*Or Sessile* | Provides an indication<br>*of point exposure* | *Fucus*<br>*Alaria nana*<br>*Alaria fistulosa*<br>Ulva latuca<br>Blue Mussels<br>Limpets<br>Chiton |
| Locally mobile | Integrates exposure<br>over a small area | *Sea Urchin<br>*Rock Jingle<br>*Black Rockfish<br>*Rock Greenling<br>*Glaucous-winged Gull |
| Mobile | Provides an indication<br>local movement within<br>a few km of designated<br>site | *Yellow Irish Lord<br>*Ocean Perch<br>*Walleye Pollock<br>*Tufted Puffin<br>*Pigeon Guillemot<br>*Common Eider<br>Brown King Crab<br>Red-Irish Lord<br>Dolly Varden<br>Bald Eagle |
| Migratory | Provides an indication<br>of regional exposure | *Atka Mackerel<br>*Pacific Cod<br>Halibut<br>Steller Sea Lion |

Figure 9.3. Major Ecoreceptors Potentially at Risk in the Amchitka Marine Ecosystem.

|  | Intertidal | Subtidal | Deepwater | Surface |
|---|---|---|---|---|
| **Sessile/ benthic** | Kelp Sea Lettuce (*Ulva*) Chiton Blue Mussels Limpets | Tunicate*, Sponges* Sea Cucumber*, Kelp, Giant Chiton*, Blue Mussel, Rock Jingle |  |  |
| **Mobile** | Gulls Common Eider Puffins Eagle | Sculpin, Rock Greenling, Octopus, Sea Urchin, Basket Star*, Rock Fish | King Crab Irish Lord Pollock Rock Fish Ocean Perch Atka Mackerel | Sea Otter* Harbor Seal* Sea Lion* |
| **Migratory** | Oystercatcher* |  | Halibut Pacific Salmon* Dolly Varden Pacific Cod | Harbor Seal* Sea Lion* Eagle Puffins & other seabirds |

*Not part of CRESP study

Figure 9.4. Rock greenling (left), and Sean Burke and Tim Stamm with Pacific Cod. (Photos S.Jewett, J. Burger).

Table 9.4. Rationale for Species Selection for Human Health Screening Analysis (Scientific names are provided in Table 9.1)

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PRIMARY PRODUCERS:  The following species are all primary producers in the marine ecosystem, are sedentary (and thus represent local exposure), and are the base of food chains.  There is good representation of the sedentary species from the four study sites (*Milrow, Long Shot, Cannikin*, Kiska), and for the mobile species from Amchitka and Kiska (Lebednik and Palmissano 1977).

Alaria fistulosusa - This kelp occurs at several depths, representing the subtidal environment. It grows upward from holdfasts in the sediment.

Alaria nana - This kelp occurs mainly in the intertidal water column.

Fucus - This brown algae occurs on rocks in the intertidal, and there is extensive reference data from other places.


INVERTEBRATES: Invertebrates are often the primary consumers in marine ecosystems, are eaten by organisms higher on the food chain, and are fairly sedentary representing local exposure. They are also eaten by the Aleut people.

Green Sea Urchin - Urchins were abundant in most of the diving transects at 15, 30 and 60 feet and thus represent good coverage of the marine floor environment.  They are a primary food of Sea Otters, a species of concern. They are also eaten by Eiders and Gulls (based on the literature and on stomach contents we examined). And they are considered a delicacy by Aleuts. They feed on algae and move over short distances.

Rock Jingle - They are less abundant, but are sedentary.

FISH: Serve multiple roles in the food web, and all species selected are eaten by people.

Rock Greenling - This is a sedentary species, each male maintaining a small territory in the kelp zone, hence representing local exposure.  It is eaten by Aleuts (as are its eggs), and is eaten by fish higher on the trophic chain, such as Cod and also by Gulls. Young fish are eaten by Puffins and Guillemots.

Black Rockfish - This is a relatively sedentary species (representing local exposure) that lives in the kelp zone and just outside the kelp zone.  It is eaten by Aleuts and is a little higher on the food chain than the Rock Greenling.

Yellow Irish Lord - This is a less sedentary (but not migratory) species that is larger than Black Rockfish, eats invertebrates, and is eaten by Aleuts, although it is not a preferred food.

Atka Mackerel - This is a deep water, bottom fish that is relatively low on the food chain, but is of commercial value and is migratory. It was of particular interest to the NOAA Fisheries.

Pacific Cod - This fish can reach 50-60 pounds, and eats smaller fish, such as Rock

Greenling and Atka Mackerel, as well as Octopus, squid, fish eggs, and crabs (all found in our specimens' stomachs). It is both a preferred fish for the Aleut people and a major commercial species.  It is mobile to migratory.

Ocean Perch -  Top level predator of commercial interest that is mobile.

Walleye Pollock - This predatory fish is currently the leading commercial species in the area.  It is a mobile species and a major food of Steller's Sea Lion.  Juvenile Pollock are a major food of Cod and Halibut

Halibut - This fish is a top-level predator, can reach large sizes (up to 500 pounds) and advanced ages, and is highly prized both by Aleuts and commercial fisheries. It is bottom dwelling, but is migratory.

BIRDS: Birds were selected because they are at different trophic levels, they (or their eggs) are eaten by subsistence hunters, and they are excellent bioindicators of exposure and effects for other top-level predators, including humans. There are contaminant data from other locations in the Pacific. All species are resident.

Eiders - Common Eiders are hunted extensively by Aleuts and their eggs are also eaten. They nest mainly in tall grass around lakes and along coastlines. Our Aleut team showed us how they locate the nests. The eggs are prized for food and the adults are eaten.  It represents a low trophic level for birds, eating mussels, snails, and urchins.

Gulls - Glaucous-winged Gull eggs are considered a delicacy by Aleuts, and gulls represent an omnivorous species.  We found urchins, starfish, and fish (including Dolly Varden and Greenlings) in their stomachs.  Since there are nesting colonies at each of the Amchitka test sites, and they normally feed within 5 miles of their colony, they represent local exposure.  They do not migrate and so represent longer term exposure in the vicinity of Amchitka.  They also can live to be 30 + years old.  Gulls nest in vegetation on small knolls in the uplands of Amchitka and Kiska.

Young Gulls - There were nesting colonies adjacent to each of the 3 test shot areas, and on Kiska.  Since parents feed their young entirely from local foods (usually within 5 miles of nesting colonies), they represent local exposure.

Tufted Puffin – Puffins nest in cliffs and crevices on the coasts of the islands. They eat entirely fish of small to intermediate sizes.  They are less localized to test shots, and represent local exposure within a regional area. Birds were moving back and forth from the *Long Shot* to the *Cannikin* shoreline. We thus combined the Bering coastal birds and had three groups (*Milrow*, *Long Shot-Cannikin*, Kiska).

Pigeon Guillemot – They nest in coastal rocks and under the docks at both islands. They eat mainly small fish, and are localized to the sides of islands during the breeding season. Birds were moving back and forth from the Long Shot to the Cannikin shoreline. Thus there were also three groups for analysis.

Table 9.5.  Rationale for Selections for Human Health Screen
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1.  One sample each from *Milrow, Long Shot, Cannikin*, Kiska for sedentary species or
        Locally mobile ones (N=4).

|  |  |
|---|---|
| Fucus | Sea Urchin |
| Alaria nana | Rock Jingle |
| Alaria fistulosa | Black Rockfish |
| Glaucous-winged Gull | Rock Greenling |
| (adults, chicks) | Yellow-Irish Lord |

2.  Locally, mobile examined from both sides of Amchitka, and from Kiska (N=3).

        Common Eider (eggs)
        Pigeon Guillemot

        Tufted Pigeon
3.  Highly mobile or migratory, examined from Amchitka and from Kiska (N=2).

        Ocean Perch
        Atka Mackerel
        Walleye Pollock

4.  Pacific Cod were available for both the inshore sampling and the NOAA trawl.  We thus
        analyzed for both sides of Amchitka and Kiska from Amchitka and Kiska for the
        NOAA trawl (N=5).

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        For all species, including those that were included in the human health screen, the
optimal pattern for the broad gamma analysis was to examine 4 composites (typically five
individuals per composite) per site per species where we had sufficient samples and
sufficient analytical time.  Samples sizes by species are given in chapter 11.

2.  Follow up Analysis of Actinides in Kelp and Bone (INL)

        The second prong of our radionuclide analysis strategy had the following
objectives: 1) to select one or more species from the list used in the Human Health
Screen, with the objective of allowing for a balanced statistical analysis, 2) to examine
species of special concern to stakeholders where few individuals were obtained,
        The first objective was met by examining the data obtained from the Human Health
Screen, and selecting species for additional analysis at INL.  When the results of the
health screen were almost all below the detection levels, we determined that kelp and fish
bone would be most productive in terms of achieving levels of some radionuclides that
were above detection levels in the first prong, and would allow statistical comparison
among test shots, and between Amchitka and Kiska (our reference site).  Kelp were

selected because there were detectable levels of some radionuclides, they form the base of the food chain, and they are sedentary, representing very local exposure around the three test shots and at Kiska.  Fish bone was selected because there were detectable levels of some radionuclides, these species are eaten by Aleuts and used generally by commercial fisheries, and they represent different levels of the food chain.

The second objective was met by selecting species of special concern to the Aleuts and resource trustees.  The list included Octopus, Halibut, Bald Eagle, and Steller Sea Lion.  While we had sufficient Halibut to make composites, the other species were in sufficiently low number to preclude compositing, and individuals were prepared as the analysis unit.  These included Halibut, Octopus, Eagle and Sea Lion, and all individuals were used.

3.  Large Sample for Enhanced Detection Levels

The third prong  was to analyze larger samples (1000 g compared to 100 g) for longer periods (48 to 72 hours compared to 24 hours) to achieve very low detection levels for Cs-137.  Within this objective we had two approaches: select some species where a large enough number of samples would be available for statistical analysis (kelp, Pacific Cod), and select additional species of special concern.  Our selection of samples was constrained by the sizes of the organisms involved.  That is, it was not possible to produce several 1000 g samples of species such as Sea Urchin because of the small amount of soft tissue in each Sea Urchin.  Thus, we had an adaptive strategy for the broad gamma screen (Fig. 9.5), which allowed us to maximize the information we could obtain from our samples by continuing with the 100 gram samples for a broad range of species, but analyzing 1000 samples for key species.

While being able to detect such low levels is not essential for assuring human health safety or for ecological risk assessments, it is essential for providing information to aid in selection of bioindicators for future biomonitoring at Amchitka.  That is, it is important to know in which species radionuclides of health interest would first be detectable.

Fig. 9.5. Overall Analysis Strategy for Cesium-137

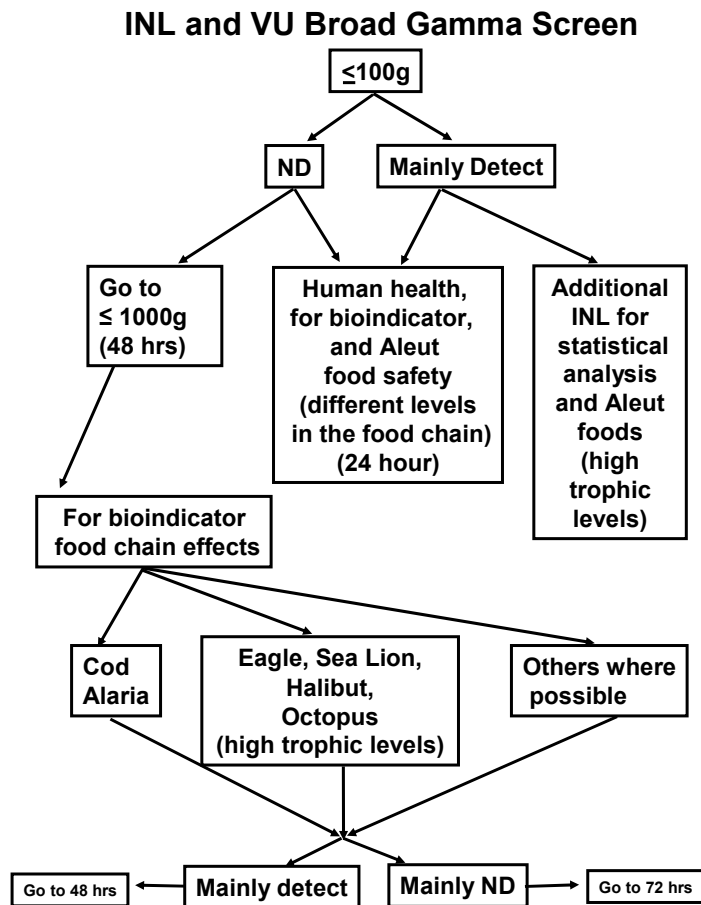**INL and VU Broad Gamma Screen**



Figure 9.5

4.   Gamma Analysis of a broad range of Aleut foods and food chain organisms (Vanderbilt)

The fourth prong of our radionuclide analysis strategy was to analyze for gamma radionuclides in two ways: 1) to increase the sample size by location, of species examined in the first prong (human health screen), and 2) to increase the number of species to include other Aleut foods and key nodes on the food chain.  The rationale for the additional species is shown in Table 9.6.

Table 9.6. Rationale for the additional species selected for the broad gamma screen at Vanderbilt .

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PRIMARY PRODUCERS:  The following species are all primary producers in the marine ecosystem, are sedentary (and thus represent local exposure), and are the base of food chains.

*Ulva latuca*- This green algae is eaten by people, and radiological data are available from other sites.

INVERTEBRATES: Invertebrates are the primary consumers in marine ecosystems, are eaten by organisms higher on the food chain, and are fairly sedentary representing local exposure. They are also eaten by the Aleut people.

Blue Mussel - They are not very abundant, but are eaten by people and marine birds, and are used extensively in other regions as a bioindicator.

Limpets - Known as Chinese Hats by Aleuts, they are prized as food, and are eaten by marine birds.  They were collected by our Aleut collectors in their traditional manner

Octopus - Top level predators of extreme interest to Aleuts.  This was a species added after meetings with Aleuts in Nikolski, Atka, and Unalaska.

VERTEBRATES: vertebrates are often secondary or tertiary consumers, and have different degrees of mobility.  The species selected, at some stage in their life cycle, are all eaten by Aleuts and some are part of commercial fisheries.

Red Irish Lord - This is a less sedentary (but not migratory) species that is larger than Black Rockfish, eats invertebrates, and is an Aleut food.

Atka Mackerel - This is a deep water, bottom fish that is relatively low on the food chain, but is of commercial value and is migratory.

Ocean Perch -  Top level predator of commercial interest that is mobile.

Walleye Pollock - This predatory fish is a major commercial species that is mobile. It comprises about 70% of the Bering Sea fishery.

Halibut - This fish is a top-level predator, can reach large sizes (up to 500 pounds) and advanced ages, and is highly prized both by Aleuts and commercial fisheries, and is migratory. The Aleuts preferred fish under 50 pounds.

Dolly Varden - Of interest to the Aleuts, this species is unusual in having a fresh/saltwater phases.  The Aleuts on our collecting trip were particularly interested in this species.

Eiders - Common Eiders are hunted extensively by Aleuts and their eggs are also eaten.  It represents a low trophic level for birds, eating mussels, snails, and urchins.  Only eggs were analyzed for the health screen.

Eagle – Bald Eagle is a top level predator of particular interest to a wide range of stakeholders, particularly the U.S. Fish & Wildlife Service

Sea Lion - The Aleut hunters/fishermen on our expedition conducted a subsistence hunt and obtained one Steller Sea Lion at Amchitka.  They requested that we analyze radionuclides in this animal that they were eating.

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SELECTION OF RADIONUCLIDES FOR ANALYSIS

Our selection of radionuclides for analysis was based on the following questions:

1. What are the radionuclides of concern to human health and biota?
2. What are the radionuclides that will provide information on the source of the radionuclides.
3. Which radionuclides formed in the nuclear tests are likely to be measurable in the marine environment (excludes short-lived radionuclides such as I-131).

The first question directly relates to the concerns of people who consume food items from the marine environment around Amchitka, and to food chain bioaccumulation that might lead to adverse effects in the organisms themselves, and to predators that eat them. The second question addresses whether it is possible to determine the source of the radionuclides in organisms in the marine environment, by examining ratios among isotopes.

Several types of analytical methods were required to measure the radionuclides indicated (see Appendix 8.E and 8.F for detailed methodology):

1. Gamma spectroscopy was used to measure Cs-137, Co-60 and Eu-152. High purity germanium (HPGe) detectors in conjunction with appropriate shielding to limit background interference were used for gamma spectroscopy. All of these isotopes (Cs-137, Co-60, Eu-152) are measured simultaneously. Am-241 is also measured in this manner, but at much higher detection limits than are achievable by alpha spectroscopy (see below).
2. Low energy photon spectroscopy (LEPSI)or gamma spectroscopy with specialized low energy HPGe detectors were used to measure I-129.
3. Chemical digestion followed by inductively coupled mass spectroscopy (ICP-MS) was used to measure Tc-99.
4. Chemical digestion followed by chemical separations were used provide one sample fraction with Sr-90 and a separate chemical fraction with Pu-238, Pu-239, 240, U-234, U-235, U-236,U-238, and Am-241. Sr-90 then was quantified based on it's daughter decay product, Y-90, using beta detection. Alpha spectroscopy was used to measure plutonium, uranium and americium isotopes. Pu-239 and Pu-240 were measured in combination through this method. If sufficient levels of Pu-239/240 were detected to warrant further speciation, ICP-MS would have been used to achieve separate isotopic quantification.

To achieve the sets of measurements described above, radionuclide measurement laboratories at Idaho National Laboratory (INL) and Vanderbilt University were employed. As a practical matter, analyses were according to the following groups:

1. I-129
2. Cs-137, Co-60, Eu-152, Am-241
3. Sr-90
4. Tc-99
5. Pu-238, Pu-239/240, U-234, U-235, U-236, U-238, and Am-241

INL served as the primary laboratory for Sr-90, Tc-99, Pu-238, Pu-239/240, U-234, U-235, U-236, U-238 and Am-241 with Vanderbilt analyzing a limited number of samples for inter-laboratory comparison. Vanderbilt served as the primary laboratory for I-129 and gamma analysis (Cs-137, Co-60, Eu-152, Am-241), with additional samples and inter-laboratory comparison samples analyzed by INL.

Quality control was maintained for radionuclide analyses through the following approaches (see Appendix 9E for a full description of procedures and quality control):

1. An independent national radionuclide certification laboratory (RESL), also located at INL (but separate from the laboratory used for the radionuclide analyses in this study) prepared double-blind quality control samples for initial methods validation and for inclusion with batches of samples sent to each laboratory for analyses. Rutgers sent RESL prepared tissues of representative biota, which were spiked and returned to Rutgers for coding and blind submission to the analytic laboratories as part of a batch. Typically, two quality control samples (one blank with respect to some isotopes and spiked with respect to other isotopes, and a second sample with a different mix of blank and spikes) were included with each batch of biota samples (typically 20 samples per batch) sent to the laboratory for analyses. The analytical laboratory did not know which samples were quality control samples or the identity and amount of each isotope in each sample. After completion of analyses, the laboratory results were compared to the known quantities of each spiked isotopes to verify correct isotope identification and quantification.
2. Approximately 10 percent of the samples, or similar samples when replicates were not possible, were analyzed by both INL and Vanderbilt to provide a basis for inter-laboratory comparison. This approach was included in the analytical methodology on the anticipation of measuring a large number of detected values for the variety of radionuclides measured. However, as will be seen, only a small number of samples and isotopes were present above detection limits. Thus, inter-laboratory comparison primarily confirmed less than detection limit measurements.
3. INL and Vanderbilt laboratories were blind with respect to sample identification. All biota samples were encoded at Rutgers prior to shipment to either INL or Vanderbilt for analysis. A full chain of custody protocol was followed.
4. INL and Vanderbilt both followed standard laboratory procedures for internal quality control, including instrument calibration, background measurements, and sample chain of custody.

The details on the "Source term", the radionuclides produced in the three tests, remains classified. Based on information obtained from the groundwater models and human health risk assessments conducted (DOE 2003a, 2003b), and our knowledge of radionuclides of interest for human health, ecological health, and source identification, we selected a suite of radionuclides for analysis. This suite of isotopes was delineated in the Amchitka *Science Plan*, and thus was reviewed by people with appropriate clearance who we expected to provide advice if our selection list was missing key isotopes. Isotopes of interest for analysis for this study were:

Cs-137, Eu-152, Co-60 (by gamma spectroscopy)

Pu-238,239,240,241, U-234,235,236,238, Am-241 (by alpha spectroscopy)
Sr-90 (by beta analysis)
I-129 (by low energy gamma spectroscopy)
Tc-99 (by ICP-MS)

The main isotopes of interest for human health and ecological receptors are Cs-137, and I-129, which accumulate in soft tissue, and Sr-90 which mainly accumulates in bone. Uranium (U) and plutonium (Pu) isotopes, which provide information on the potential source of the isotopes, mainly accumulate in skeletal material. Ratios of isotopes of Pu (indicative of nuclear detonation) and U (indicative of nuclear reactor releases and enrichment processes) will be used to the extent possible to identify whether Amchitka test shots are the likely source of measured radionuclides in the biological samples.

Combining Species Selection and Radionuclide Selection

Determining the analysis plan for the Human Health Screen involved two steps, described above: 1) selecting species for analysis, and 2) selecting radionuclides for analysis as a function of both species and tissue type. The numbers in each column indicate the number of samples, reflecting the locations represented (4 sites for sedentary species, 3 for mobile species, 2 for migratory species). This resulted in the following Human Health Screen at INL (Fig. 9.4 and Table 9.7)

Table 9.7. Human Health Screen of Samples Analyzed at INL

| Species | Tissue | I-129 | Gamma Spectroscopy | Sr-90 | Tc-99 | Actinides |
|---|---|---|---|---|---|---|
| Alaria fistulosa | soft | 4 | 4 | 4 | 4 | 10 |
| Alaria nana | soft | 4 | 4 | 4 | 4 | 12 |
| Fucus | soft | 4 | 4 | 4 | 4 | 14 |
| Ulva | soft | | | | | 12 |
| Sea Urchin | soft | 4 | 4 | 4 | 4 | |
| Rock Jingle | soft | 4 | 4 | 4 | 4 | |
| Pacific Halibut | bone | | | | | 4 |
| Pacific Cod | muscle | 5 | 5 | 5 | 5 | |
| | bone | | | | | 14 |
| Black Rockfish | muscle | 4 | 4 | 4 | 4 | |
| | bone | | 4 | 4 | | 4 |
| Rock Greenling | muscle | 4 | 4 | 4 | 4 | |
| | bone | | 4 | 4 | | 4 |
| Yellow Irish Lord | muscle | 4 | 4 | 4 | 4 | |
| | bone | | 4 | 4 | | 4 |
| Ocean Perch | muscle | 2 | 2 | 2 | 2 | |
| | bone | | 2 | 2 | | 2 |
| Atka Mackerel | muscle | 2 | 2 | 2 | 2 | |
| | bone | | 2 | 2 | | 2 |
| Walleye Pollock | muscle | 2 | 2 | 2 | 2 | |
| | bone | | 2 | 2 | | 2 |
| Common Eider eggs | egg | 3 | 3 | 3 | 3 | |
| Tufted Puffin | muscle | 3 | 3 | 3 | 3 | |
| | bone | | 3 | 3 | | 3 |
| Pigeon Guillemot | muscle | 3 | 3 | 3 | 3 | |
| | bone | | 3 | 3 | | 3 |
| Glaucous wing gull | eggs | | | | | |
| adult | muscle | 4 | 4 | 4 | 4 | |
| | bone | | 3 | 3 | | 3 |
| young | muscle | 4 | 4 | 4 | 4 | |
| | bone | | 3 | 3 | | 3 |

## DISCUSSION AND IMPLICATIONS

In this chapter we describe on the rationales for selection of biota for analysis, selection of radionuclides for analysis, and the melding of these two to arrive at a sampling plan. The factors that entered into our selection of species were importance for human health and the food chain, potential as bioindicators for future biomonitoring, distribution among the test shots and reference sites, mobility, trophic-level, potential age, and availability in our inventory. The factors that entered into our selection of radionuclides were those of primary importance to the health of humans or biota, and those that might contribute to our understanding of the source of any radionuclide levels found. With this overall plan we were able to analyze a wide range of Aleut foods and commerical fish that had diverse life styles, mobilities, and trophic levels representing the complex marine ecosystems around Amchitka and Kiska.

The selection of both species for analysis, and radionuclides for analysis resulted in an analysis strategy which provided data that will be useful for understanding current risks to humans and biota, providing some information on possible source of the radionuclides, and providing information which will be useful in selecting biota for biomonitoring, a critical element of any long term stewardship plan for Amchitka. The human health screen at INL provided the base data from assessing any potential risk to human from radionuclides in biota at Amchitka, while the in-depth gamma screen at Vanderbilt provided sufficient sample numbers to back up the human health screen, added additional species of interest to subsistence hunters/fishers and to understanding the food web, and provided sufficient numbers for statistical comparisons. The 1000 gram samples used for gamma analysis at Vanderbilt allowed us to reach very low detection levels, well below any human health or ecological risk concerns, but that will be useful in further selecting bioindicators for long-term stewardship plans.

A study of this scope can detect broad contamination, and was designed to optimize the use of resources. It does not eliminate the possibility of local seepage that was missed by the specimens we obtained, and it does inform the approach for detection of future seepage.

APPENDICES FOR CHAPTER 9 (See attached CD-ROM)

9.A. Amchitka Expedition Update: Initial Specimen Screening and Future Laboratory Testing C.W. Powers, Report Coordinated and Edited by H. Mayer
9.B. Selection of Samples for Human Health Screen by J. Burger
9.C. Broad Gamma Analysis of Food Chain Organisms by J. Burger
9.D. Adaptive Analysis Strategy for Radionuclide Analysis by J. Burger

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