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An assessment of the reported leakage of anthropogenic radionuclides from the underground nuclear test sites at Amchitka Island, Alaska, USA to the surface environment

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Abstract

Three underground nuclear tests representing approximately 15–16% of the total effective energy released during the United States underground nuclear testing program from 1951 to 1992 were conducted at Amchitka Island, Alaska. In 1996, Greenpeace reported that leakage of radionuclides, ^{241}Am and $^{239+240}$Pu, from these underground tests to the terrestrial and freshwater environments had been detected. In response to this report, a federal, state, tribal and non-governmental team conducted a terrestrial and freshwater radiological sampling program in 1997. Additional radiological sampling was conducted in 1998. An assessment of the reported leakage to the freshwater environment was evaluated by assessing ^{3}H values in surface waters and ^{240}Pu/^{239}Pu ratios in various sample media. Tritium values ranged from 0.41 Bq/l$\pm$0.11 two sigma to 0.74 Bq/l$\pm$0.126 two sigma at the surface water sites sampled, including the reported leakage sites. Only at the Long Shot test site, where leakage of radioactive gases to the near-surface occurred in 1965, were higher ^{3}H levels of 5.8 Bq/l$\pm$0.19 two sigma still observed in 1997, in mud pit #3. The mean ^{240}Pu/^{239}Pu for all of the Amchitka

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samples was 0.1991 ± 0.0149 one standard deviation, with values ranging from $0.1824 \pm 1.43\%$ one sigma to $0.2431 \pm 6.56\%$ one sigma.

The measured ^{3}H levels and $^{240}Pu/^{239}Pu$ ratios in freshwater moss and sediments at Amchitka provide no evidence of leakage occurring at the sites reported by Buske and Miller (1998 Nuclear-Weapons-Free America and Alaska Community Action on Toxics, Anchorage, Ak, p. 38) and Miller and Buske (1996 Nuclear Flashback: The Return to Anchitka, p. 35). It was noted that the marine sample; $^{240}Pu/^{239}Pu$ ratios are statistically different than the global fallout ratios presented by Krey et al. (1976) and Kelley, Bond, and Beasley (1999). The additional non-fallout component $^{240}Pu/^{239}Pu$ ratio, assuming a single unique source, necessary to modify the global fallout $^{240}Pu/^{239}Pu$ ratio to that measured in the marine samples is on the order of 0.65 (Hameedi, Efurd, Harmon, Valette-Silver, & Robertson, 1999; Kelley et al., 1999). While this potentially suggests another plutonium source, such as high burn-up nuclear reactor fuel, rather than underground nuclear tests, the uncertainties in analyses and environmental processes need to be fully assessed before any conclusion can be reached.

Further work is needed to evaluate these findings and to support any radiological assessment of the marine environment surrounding Amchitka. Based on geohydrological testing and modeling, leakage from the Amchitka Underground Nuclear Tests is projected to occur to the marine environment (Claassen, 1978; Fenske, 1972; Wheatcraft, 1995).

Keywords: Amchitka Island; Alaska; Underground nuclear testing; Cannikin; Long shot; Milrow; Tritium; Plutonium

1. Introduction

Amchitka Island, Alaska (Fig. 1) was the site of three United States underground nuclear tests carried out from 1965 through 1971. Long Shot was detonated at a depth of 716 m on October 29, 1965, and had an explosive yield of approximately 80 kt. Milrow was detonated at a depth of 1220 m on October 2, 1969, and had an explosive yield of approximately 1 Mt. Cannikin, the largest American underground nuclear test, was detonated at a depth of 1790 m on November 6, 1971, and had an explosive yield of approximately 5 Mt (Merritt & Fuller, 1977). These three tests on Amchitka represent approximately 15–16% of the total effective energy released during the United States underground nuclear testing program from 1951 to 1992 (Robbins, Makhijani, & Yih, 1991; US Department of Energy, 1994).

The Alaska Department of Environmental Conservation (ADEC) is conducting an ongoing independent assessment of the terrestrial and freshwater radioecology of Amchitka Island as part of its oversight work under an Agreement in Principle with the US Department of Energy (DOE). This work is being conducted in close coordination with the University of Alaska (UA) and with the Aleutian/Pribilof Island Association (A/PIA).

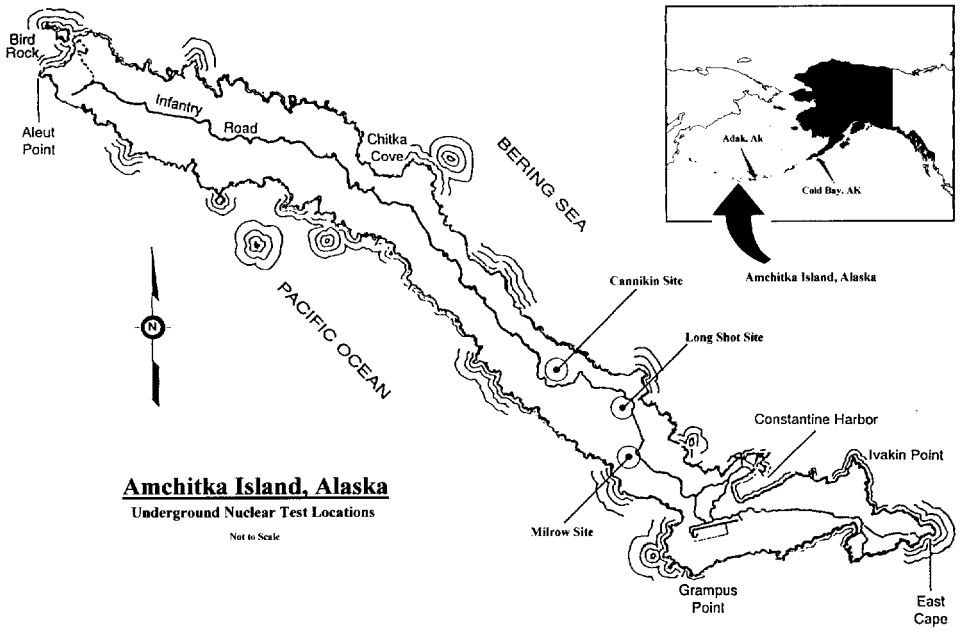

Fig. 1. Amchitka Island, Alaska, location map.

In 1997, it was stated in *Arctic Pollution Issues: A State of the Arctic Environment Report* that surface radioactive contamination was detected at the Amchitka underground test sites (Nilsson, 1997). This statement was based on a 1996 report, *Nuclear Flashback: Return to Amchitka* which concluded that leakage of ^{241}Am and $^{239+240}$Pu was occurring into the freshwater environment (Miller & Buske, 1996). The authors hypothesized that global fallout transuranics would be associated primarily with the freshwater sediments and that underground test transuranics would be soluble in water thus, incorporated in biota. This was based on their hypothesis that in underground nuclear tests ^{241}Am formed dramatically different compounds or complexes than in an above ground test. They reasoned ^{241}Am detected in the sediments would be of global fallout origin and ^{241}Am incorporated in the biological tissue would originate from the underground tests. Thus, by washing the sediment from the freshwater biota samples only radionuclides originating from the underground tests would be present. Since ^{241}Am was determined to be present they also indicated that $^{239+240}$Pu was leaking from the underground tests. A later report by the same authors in 1998 revised this statement, indicating that only ^{241}Am was observed to be leaking from the underground test areas into freshwater environments (Buske & Miller, 1998). The hypothesis was that ^{241}Am was much more mobile than $^{239+240}$Pu and would significantly move ahead of it in groundwater.

Leakage of anthropogenic radionuclides from the Amchitka underground test areas into the Bering Sea and North Pacific Ocean is a serious concern, as these are two of the most productive commercial fishing grounds in the world. Amchitka is within the migratory pathways of many subsistence food species that are used by the Aleut Natives living in the Aleutian and Pribilof Islands.

This study evaluates the reported (Miller & Buske, 1996; Buske & Miller, 1998) leakage from the nuclear tests into the environment by evaluating recent ^{3}H levels in Amchitka freshwater samples and $^{240}Pu/^{239}Pu$ ratios in aquatic vegetation and sediments. In addition, the results of the observed $^{240}Pu/^{239}Pu$ ratios are compared with global fallout values. First, though, a discussion of the island environment and testing history is warranted.

2. Geographic setting

Amchitka Island is about 56 km long in a northwesterly direction and 5–8 km wide and lies between longitudes 178°37′E. and 179°29′E. and between latitudes 51°21′N. and 51°39′N. Landforms vary from a mountainous area in the islands northwestern section, dropping to a high plateau forming the central section and then dropping to a flat swampy tableland in the southeast (Powers, Coats, & Nelson, 1960). The three underground nuclear tests were carried out in the central section.

As part of the Aleutian arc defining the boundaries between the North Pacific Ocean and the Bering Sea, Amchitka Island exhibits a maritime climate with moderate temperatures, but with extremes in fog and wind. The mean temperature is 3.9°C, with extremes recorded of minus 10°C and 18.3°C. Annual wind speeds average 9 m/s. Summer winds seldom exceed 31 m/s, but winter wind speeds frequently exceed 45 m/s. Precipitation for the period from 1943 to 1948 averaged 828 mm a year, including snowfall of approximately 1778 mm (Armstrong, 1977).

Vegetation in the low plateaus of Amchitka, where the underground nuclear testing was carried out, is composed of heath blankets comprising low shrubs of *Empetrum*, *Vaccinium*, *Loiseleuria*, and varying amounts of many species of sedges, grasses, forbs, lichens and bryophytes (Shacklette et al., 1969).

Geologically, the island is made up of volcanic ash, tuff, breccias, and lava flows, interbedded with marine conglomerate that has experienced deformation, uplifting, and glacial erosion and deposition (Powers et al., 1960). The underground nuclear test working points were generally within volcanic pillow lava, basalt, andesite or breccias.

3. Amchitka underground nuclear test site history and environmental issues

There have been significant environmental impacts on the terrestrial, freshwater and marine ecosystems, occurring not just from the physical effects of the nuclear tests, but also from construction activities (Merritt & Fuller, 1977). There were documented releases of radionuclides from two of the underground nuclear tests, Long Shot and Cannikin.

4. Long shot underground nuclear test

Long Shot was part of the US Department of Defense (DoD) Vela Uniform program, which was designed to determine whether seismic signals from an underground nuclear test could be differentiated from those of earthquakes. As the focus was on distant measurements of seismic signals, there were minimal on-island measurements of ground shock or environmental effects (US Department of Energy, 1995).

A rubble chimney formed above the Long Shot point of detonation, but it did not reach the surface, and no surface depression occurred. Surface uplift and cracking did occur, resulting in the disruption of some surface pond and stream features. Rockfalls along the island ocean cliffs did occur, though to a smaller extent than in the subsequent Milrow and Cannikin tests (US Department of Energy, 1995).

Documented leakage of ^{3}H, ^{131}I and ^{85}Kr into the surface environment occurred after the Long Shot test. The leakage was first detected in late November and December of 1965 as traces of ^{131}I in a pond 100 m north-northwest of ground zero (GZ)(US Atomic Energy Commission, 1974). The general explanation for the leakage was a combination of migration of the gaseous radionuclides to the top of the rubble chimney and then through inadequately sealed pre-shot drill holes or failed emplacement hole blocks, resulting in accumulation in the shallow sub-surface around GZ. It is also thought that infilling of the rubble chimney with groundwater forced the gaseous radionuclides upward (Castagnola, 1969; Merritt, 1970). Long-term ^{3}H monitoring by the US Environmental Protection Agency (EPA), started in 1977 and continued periodically through 1997, recorded a maximum surface water value of 125.9 Bq/l $\pm$ 13 two sigma in mud pit #3 near GZ in 1977. Since then levels have generally declined, with a 1997 level of 5.80 Bq/l $\pm$ 0.19 two sigma. Fig. 2 documents the observed decline in tritium levels in the surface environment in Long Shot Mud Pit #3 (Faller & Farmer, 1998). Increases observed in ^{3}H levels in September 1980 and July 1986 are likely related to China's last atmospheric nuclear test (Natural Resource Defense Council, 2000) and the 1986 Chernobyl accident.

5. Milrow underground nuclear test

Milrow was part of the large-yield weapons testing involving the Spartan missile warhead program. It was also considered a "calibration" test to establish if large-yield tests could be safely conducted at Amchitka. This test resulted in substantial surface effects within the nearby GZ area as well as several miles away. Initial doming was estimated to be 2 m above grade at GZ. About 37 hr. after the test the collapse of the cavity resulted in the formation of a surface crater about 4.5 m deeper than the pre-shot grade and 915 m in diameter (U.S. Department of Energy, 1995).

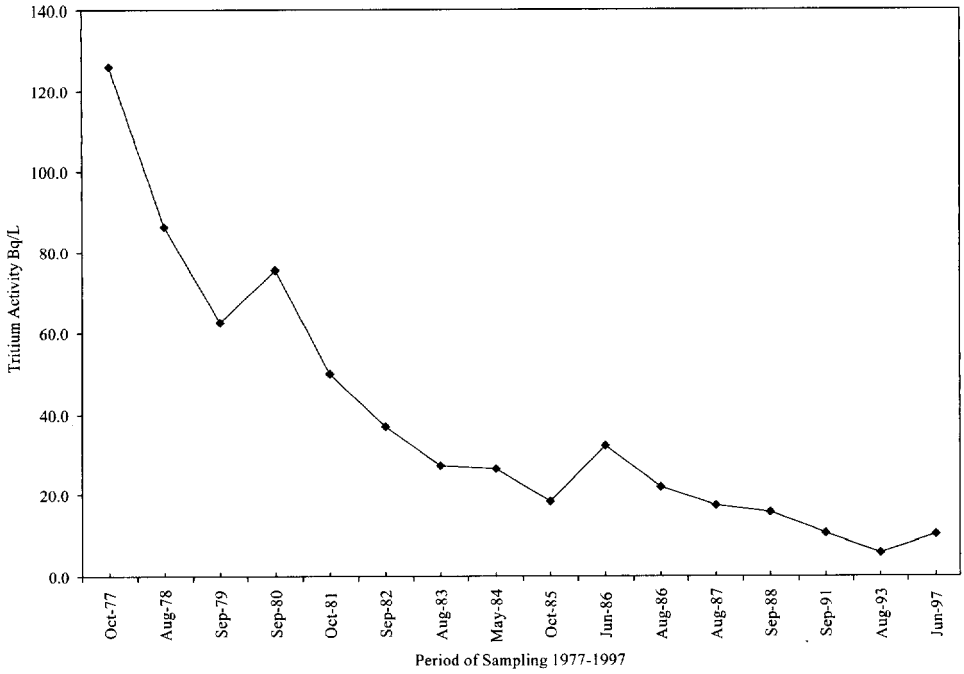

Fig. 2. Tritium activity Bq/l Long Shot Mud Pit #3, Amchitka Island, Alaska 1977–1997 (Faller & Farmer, 1998).

6. Cannikin underground nuclear test

The Cannikin underground nuclear test vaporized and melted a large amount of rock, forming an underground cavity that collapsed about 38 h after the test. A rubble chimney formed between the cavity and the land surface, creating a large irregular collapse crater. This depression intercepted both the North and South Forks of White Alice Creek allowing inflow of surface water into the rubble chimney. The depression in the North Fork of White Alice Creek resulted in the formation of a small pond. The depression on the South Fork took 13 months to fill before it began to discharge again to White Alice Creek. This formation, now called Cannikin Lake, is the largest lake on the island, covering approximately 12 ha, with a maximum depth of 10 m (Fuller & Kirkwood, 1977).

During Cannikin post-shot drilling of well UA-1-P1 in 1972 approximately 4000 m^3 of radioactive gases, principally ^{85}Kr, ^{3}H, and ^{14}C, were vented from the drill pipe to the atmosphere (Douthett, 1972; Littlejohn, 1972; Miller, 1972; Smith, 1972). There is no documented report of any surface deposition or leakage of these radioactive gases within the Cannikin watershed other than during this drilling

operation (Seymour & Nelson, 1977). After completion of the post-shot drilling work, items containing possible internal ^{3}H contamination were cut into small pieces, placed at the bottom of the well cellar and covered with more than 1.2 m of concrete when the well was sealed on May 15, 1973 (Eberline Instrument Corporation, 1973).

7. Environmental sampling history summary 1964–1998

During 1965–1976, an extensive effort was undertaken by various contractors for the Atomic Energy Commission to sample the air, soil, water, and biota for radionuclides to determine if the testing had released any radionuclides to the land surface. Sampling during this period included marine waters surrounding the island. Other than the documented leakage at Long Shot and the Cannikin drill-back well, no evidence of leakage from the underground tests to the terrestrial or marine environment was documented. Anthropogenic radionuclides from atmospheric tests conducted by China, though, were detected at various times (Seymour & Nelson, 1977).

From 1977 to 1997, the EPA collected and analyzed water samples for ^{3}H and gamma-ray emitting radionuclides from surface lakes and streams, springs, various groundwater wells, and precipitation. This work was conducted as part of the EPA Long Term Hydrological Monitoring Program. No anthropogenic gamma-ray emitting radionuclides were detected and ^{3}H levels continue to decrease at the Long Shot site due to radioactive decay and dilution (Faller & Farmer, 1998). No marine sampling offshore of the underground test areas occurred during this period.

In 1996, a survey of selected aquatic biota on the island by Greenpeace raised the issue of the migration potential via groundwater for several long-lived, man-made radionuclides from detonation cavities several thousand feet below the surface of the island. The first report stated that leakage of ^{241}Am and $^{239+240}Pu$ were detected at the Cannikin site (Miller & Buske, 1996). Later in a second report (Buske & Miller, 1998) the same authors claimed leakage of ^{241}Am, but not $^{239+240}Pu$, from all three nuclear test sites.

As a result of the 1996 Greenpeace Report, the EPA's routine surveillance of ground and surface waters for ^{3}H was expanded in 1997 to include selected sampling of aquatic biota and sediments (US Environmental Protection Agency, 1977). During this sampling, conducted jointly between EPA, DOE, ADEC, A/PIA and Greenpeace, water, biota and sediment samples were collected for the ^{3}H and $^{240}Pu/^{239}Pu$ ratio analytical work discussed in this paper. In 1998, the ADEC, A/PIA and DOE carried out an expanded sampling of Amchitka that encompassed additional sites and re-sampled some 1997 locations. Terrestrial and freshwater vegetation were targeted, along with lichen, soil and sediment samples. Freshwater fish samples were collected within the underground test area watersheds and in watersheds outside the test areas. The additional ^{3}H samples discussed in this paper were collected

during the 1998 sampling. In 1998 samples of biota, soil and stream sediments were also taken on Adak Island and at Cold Bay, Alaska, as additional control locations.

In this paper we report, for the first time, the 1998 ^{3}H and 1997 $^{240}Pu/^{239}Pu$ ratio results of samples collected at Amchitka. This information is relevant to addressing the issue raised of leakage into the Amchitka freshwater environment from the underground nuclear tests.

8. Rationale For using ^{3}H and $^{240}Pu/^{239}Pu$ atom ratios

Tritium was produced in significant quantities by these three underground tests, especially from the fusion components of the weapons, and is highly mobile in groundwater. For example, at the Nevada Test Site ^{3}H is the most abundant radionuclide in the groundwater and will remain so for 100–200 years (Bryant, 1992; Nimz & Thompson, 1992; Smith & Bradley, 1995). These properties make it one of the first radionuclides likely to be detected above the background if leakage is occurring.

Results of the 1997 and 1998 ^{3}H measurements of freshwater sources can be used to assess whether leakage is occurring from an underground nuclear test versus atmospheric input. Significant quantities of ^{3}H remain in the deep groundwater from the nuclear tests at the Long Shot, Milrow and Cannikin sites. In the Long Shot GZ-1 well, which only represents the early phase surface release that occurred at this site, the 1997 ^{3}H level was close to 37 Bq/l (Faller & Farmer, 1998). This represents only a small fraction of what remains in the deeper groundwater at this site that is being transported away from the test cavity. A maximum ^{3}H level of 33.3 MBq/l was observed in the groundwater during the Cannikin drill-back in 1972 (Claassen, 1978).

The $^{240}Pu/^{239}Pu$ atom ratio can be used to differentiate between global fallout plutonium and other local or regional sources. Global fallout $^{240}Pu/^{239}Pu$ isotopic composition is a complex mixture that is regulated by the design of the nuclear weapon, physical and chemical processes occurring during the explosion, location of the test sites, and the process of atmospheric transport (Eisenbud & Gesell, 1997). The global mean $^{240}Pu/^{239}Pu$ atom ratio for fallout, neglecting local effects near test or accident sites, is 0.176 ± 0.014 one standard deviation, corrected to 1971 (Krey et al., 1976). A recent reassessment by Kelley, Bond, and Beasley (1999) of the work undertaken by Krey et al. (1976) provides a current regional, latitude 71–30 N, $^{240}Pu/^{239}Pu$ atom ratio of 0.180 ± 0.014 two standard deviations. Differences from the global or regional mean ratio can be indicative of other sources of plutonium in addition to global fallout.

Low $^{240}Pu/^{239}Pu$ atom ratios in comparison to the global fallout ratio are characteristic of weapon-grade plutonium, which generally contains less than 5% ^{240}Pu (Oughton et al., 1999). Early tests (1951–1955) at the Nevada Test Site produced a mean ratio of 0.028 compared to later tests (1957–1961) that produced a mean ratio of 0.054 (Hicks & Barr, 1984).

Local or regional inputs of weapon grade plutonium from production facilities or that remaining and produced in a low yield nuclear test will result in lowering the mean global fallout $^{240}Pu/^{239}Pu$ atom ratios. A high yield thermonuclear test can, however, produce $^{240}Pu/^{239}Pu$ atom ratio higher than global fallout. This is dependent in part upon the device design and thermal neutron flux (Beasley et al., 1998; Kelley et al., 1999). For example, the 1952 10.4-Mt Ivy/Mike test and the 1954 Castle/Bravo test at Enewetak Lagoon in the South Pacific produced $^{240}Pu/^{239}Pu$ atom ratios of 0.363 ± 0.004 and 0.32 ± 0.03, respectively (Kelley et al., 1999).

Higher $^{240}Pu/^{239}Pu$ atom ratios are produced when nuclear fuel is irradiated under controlled conditions, such as in a nuclear power reactor. The more highly irradiated the nuclear fuel the greater the production of the plutonium isotopes ^{238}Pu, ^{240}Pu, and ^{241}Pu compared to ^{239}Pu.

Production of plutonium under low versus high irradiation conditions will produce $^{240}Pu/^{239}Pu$ isotopic atom ratios, respectively, of 0.063 and 0.65 (Sanders & Boni, 1980).

Americium exists in global fallout and in the underground test debris. Its existence in trace levels in environmental samples cannot be used to uniquely identify leakage from specific Amchitka tests, as can the $^{240}Pu/^{239}Pu$ ratio.

In the event that transuranics are transported from the test cavities to the Amchitka freshwater environment it is likely that plutonium would be a component in this transport and it would alter the $^{240}Pu/^{239}Pu$ ratios from that of global fallout.

9. ^{241}Am transport vs $^{239+240}Pu$ transport in groundwater

Is it possible for ^{241}Am to be transported significantly ahead of the $^{239+240}Pu$ isotopes in groundwater as hypothesized by Buske and Miller (1998)? The physical and chemical properties of Am and Pu result in very high distribution coefficients for adsorption to particles, greatly retarding their movement in groundwater. Properties of Am make its movement slightly less retarded in groundwater than Pu, but typically, transport is still only on the order of tens of meters (Penrose, Polzer, Essington, Nelson, & Orlandini, 1990; Penneman & Keenan, 1960; Penneman, 1968; Nitsche, 1991; Silva & Nitsche, 1995). Inorganic and organic complexes can be formed with Am or Pu in aqueous solutions, increasing their migration rates. Americium typically exists in the groundwater in a $+3$ oxidation state, while Pu typically exists in a $+5$ state, though it may also be found in the $+4$ and $+6$ state (Triay et al., 1997; Silva & Nitsche, 1995). It is also likely that the formation of both intrinsic and pseudo-colloids of Am and Pu would occur in the groundwater at underground nuclear test sites (Honeyman, 1999; Nitsche, 1991). Colloids of Am and Pu have been reported to have moved over 1 km in the groundwater at sites at Los Alamos, New Mexico, and the Nevada Test site (Kersting et al., 1999; Penrose et al., 1990). It remains to be determined what masses of the actinides present in an underground nuclear test are actually transported as colloids. The foregoing

referenced studies on complexation and colloid formation do not lend support to the theory that Am would move significantly ahead of Pu if colloidal transport proves the primary transport mechanism for long distance travel in groundwater. If ^{241}Am was transported to the surface environment at Amchitka, it is expected that ^{239,240}Pu would also be present, because contaminant transport would likely be as colloids, especially over distances of 0.5–1.0 km or more.

For these reasons the results of the ^{240}Pu/^{239}Pu atom ratios were utilized in lieu of ^{241}Am values to determine if transuranics were leaking from the reported sites at Cannikin and Long Shot.

10. Sampling and analytical procedures

Sampling and analysis methods are described in the *Amchitka, Alaska Special Sampling Project 1997 Sampling and Analysis Plan* (US Environmental Protection Agency, 1977). The primary targeted sample species for 1997 and 1998 were the freshwater moss *Fontinalis neomexicanus*, and the marine alga *Fucus distichus*. These species were selected as indicator species during the 1960s environmental work at Amchitka and have been utilized in Europe for radionuclide biomonitoring in freshwater or marine environments (Coquery, Carvallo, Azemard, & Horvat, 1999; Hongve, Brittain, & Bjørnstad, 1999; Kershaw, McCubbin, & Leonard, 1999; Seymour & Nelson, 1977). Emphasis was placed on collecting as few species of freshwater moss or marine alga as possible to reduce variability due to inter-specific differences in radionuclide concentration capability.

Sample transects of varying lengths were established on a stream in each of the test watersheds and background areas. Each stream transect included three to four inter-transect sample locations. About 3 kg wet weight of aquatic moss or marine alga was required for radionuclide analyses. Sediment samples were collected at each sample location. In addition, the fine sediment and organic debris removed while field washing the aquatic moss samples were recovered by filtering the wash water through sieves.

Surface waters and shallow groundwaters were sampled in 1997 by the EPA for ^{3}H as part of the Long Term Hydrological Monitoring program. However, no samples for ^{3}H were specifically taken at the Long Shot or Cannikin sites reported to be leaking underground test radionuclides. In 1997, no measurable rainfall occurred during the sampling period, precluding the collection of an ^{3}H precipitation sample. As part of the 1998 sampling we collected water from the reported Long Shot and Cannikin leakage sites and obtained precipitation samples for ^{3}H analysis.

11. Sample preparation

The 1997 vegetation samples prepared for Los Alamos National Laboratories (LANL) plutonium analyses were dried at 100°C for 24 h and then were Wiley-milled with a 2 mm screen to reduce volume for ashing. The samples were shipped under

chain of custody to LANL where they were further processed by first placing them in a muffle furnace and heating to 300°C then raising the temperature to 550°C and ashing the samples to a constant weight (Efurd, 1998). The sediment samples were collected for LANL from the stream sites, within the vegetation transects, not dried in the field lab, and shipped under chain of custody to LANL for analysis.

Tritium samples collected in 1997 and in 1998 followed sampling procedures described in the *Standard Operating Procedure CER-203* and the *Amchitka, Alaska Long Term Hydrologic Monitoring Plan* (Faller & Farmer, 1998). Water samples were collected in 500-ml glass bottles prepared for the sampling program by EPA and were not chemically preserved.

12. Instrumentation

The thermal ionization mass spectrometer (TIMS) was utilized at LANL to determine the $^{240}Pu/^{239}Pu$ ratios of selected vegetation samples and sediment samples. The TIMS instrument is a modified version of one developed at the US National Bureau of Standards (now called the National Institute of Standards and Technology) (Efurd, Rokop, & Roensch, 1994). This instrument is capable of measuring the $^{240}Pu/^{239}Pu$ atom ratio to 0.07% precision and accuracy at the 95% confidence level. Additional documentation on the LANL TIMS instrumentation and QA/QC can be found in Perrin, Knobeloch, Armijo, and Efurd (1985).

The EPA Radiation and Indoor Environments National Laboratory in Las Vegas, Nevada conducted tritium analyses for the 1997 and 1998 samples. Conventional analysis was performed to screen the water samples for gross contamination to detect concentrations above 15 Bq/l. This screening was done to prevent contamination of the equipment used in the more sensitive tritium enrichment method. The conventional analysis was done by distillation and liquid scintillation analysis. The only sample found to exceed this level was from the Long Shot GZ-1 well. The remaining samples were analyzed by the tritium enrichment method utilizing slow electrolysis to preferentially concentrate tritiated water (Faller & Farmer, 1998).

13. Statistical analysis

For statistical analysis the program PROPHET was used. Reported sigma values represent the counting error for a single sample. The reported standard deviation represents the error for the mean of multiple sample values. Due to limited sample size the $^{240}Pu/^{239}Pu$ atom ratios were pooled for comparison with global fallout data. Statistical comparison of the mean $^{240}Pu/^{239}Pu$ ratios between Amchitka and observed worldwide ratios reported by Krey et al. (1976) and Kelley et al. (1999) were carried out by utilizing a two-sample equal-variance t test. The pooled Amchitka and global fallout data both failed the Shapiro-Wilk test for normality. Hence the non-parametric two-sample Mann-Whitney rank sum test (2 sided) was also utilized to compare medians.

14. Results

14.1. $^{240}Pu/^{239}Pu$ ratios

For the 8 *Fontinalis neomexicanus* samples analyzed the $^{240}Pu/^{239}Pu$ the ratios ranged from $0.1824 \pm 1.43\%$ one sigma to $0.1976 \pm 1.42\%$ one sigma. The mean for these samples is 0.1893 ± 0.0060 one standard deviation.

The 10 freshwater sediment samples had a mean $^{240}Pu/^{239}Pu$ ratio of 0.1984 ± 0.010 one standard deviation with a range from $0.1837 \pm 3.35\%$ one sigma to $0.2179 \pm 8.64\%$ one sigma. This sediment analysis set included duplicate analyses of two of the samples to assess sample inhomogeneity. Fig. 3 compares the mean $^{240}Pu/^{239}Pu$ ratios for the various sampled Amchitka media and the reference $^{240}Pu/^{239}Pu$ ratios used in our assessment.

Three samples of *Fucus distichus* from the Cannikin, Milrow and reference site marine littoral zones were analyzed. The mean $^{240}Pu/^{239}Pu$ ratio was 0.2173 ± 0.016 one standard deviation with a range from $0.2046 \pm 6.18\%$ one sigma to $0.2354 \pm 10.25\%$ one sigma.

The three marine sediment samples had a mean of 0.2161 ± 0.023 one standard deviation with a range from $0.1828 \pm 1.86\%$ one sigma to $0.2421 \pm 6.56\%$ one sigma. This sediment analysis set included duplicate analyses of two of the samples to assess sample inhomogeneity.

14.2. Tritium analysis

Results of the two 1998 samples taken from the 1996 site of reported leakage (XCN-5) at Cannikin were $0.737 \text{ Bq/l} \pm 0.126$ two sigma and $0.533 \text{ Bq/l} \pm 0.115$ two sigma. This sample site is within a wetland adjacent to White Alice Creek, which drains Cannikin Lake and the watershed boundaries around Cannikin. The mean ($n = 4$) for the 1997 ^{3}H samples from White Alice Creek locations upstream of XCN-5 was $0.53 \text{ Bq/l} \pm 0.06$ one standard deviation. The 1998 ^{3}H samples from the site of reported leakage (XLS-7) at Long Shot, which is characteristic of a soligenous bog, contained $0.448 \text{ Bq/l} \pm 0.204$ two sigma and $0.404 \text{ Bq/l} \pm 0.122$ two sigma. For the two precipitation samples obtained in 1998 the results were $0.24 \text{ Bq/l} \pm 0.11$ two sigma and $0.39 \text{ Bq/l} \pm 0.11$ two sigma. Two 1998 samples, one from Duck Cove Creek and the other from an adjacent wetland draining into Duck Cove Creek, resulted in levels of $0.41 \text{ Bq/l} \pm 0.11$ two sigma and $0.47 \text{ Bq/l} \pm 0.12$ two sigma. A comparison of the mean ^{3}H values for the various Amchitka sampling locations are represented in Fig. 4.

15. Discussion

15.1. $^{240}Pu/^{239}Pu$ ratios

Mean and individual $^{240}Pu/^{239}Pu$ ratios for *Fontinalis neomexicanus* samples from the three underground nuclear test watersheds and the reference stream

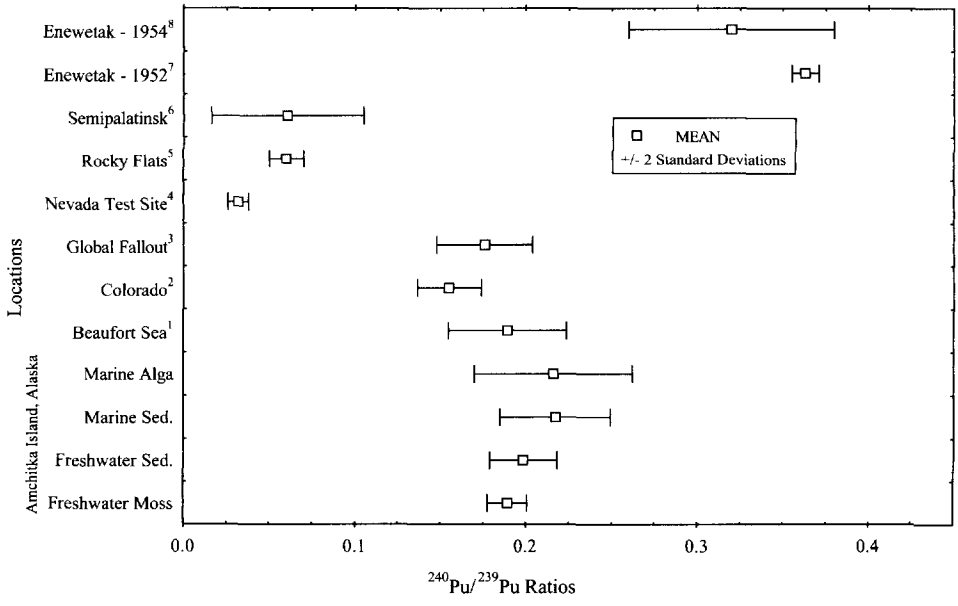

Fig. 3. $^{240}Pu/^{239}Pu$ atom ratios for Amchitka Island, Alaska sample sites and comparison with reference sites.

(1) Beaufort Sea sediments near Alaska, USA.
(Efurd et al., 1997)
(2) Background levels Colorado, USA.
(Efurd et al., 1994)
(3) Global fallout background.
(Krey et al., 1976)
(4) Nevada Test Site, Nevada, USA.
(Hicks et al., 1984)
(5) Rocky Flats Facility, Colorado USA.
(Efurd et al., 1993)
(6) Semipalatinsk Test Site, Kazakhstan.
(Beasley et al., 1998; Yamamoto et al., 1996)
(7) Enewetak Lagoon, Marshall Islands
Ivy/Mike (Thermonuclear Test) – 1952
(8) (Kelley et al., 1999)
Enewetak Lagoon, Marshall Islands
Castle/Bravo (Thermonuclear Test) — 1954
(Kelley et al., 1999)

are in agreement with the average 0.176 ± 0.014 one standard deviation ratio observed for global fallout by Krey et al., (1976) and the latitudinal regional average of 0.180 ± 0.014 two standard deviations observed by Kelley et al. (1999).

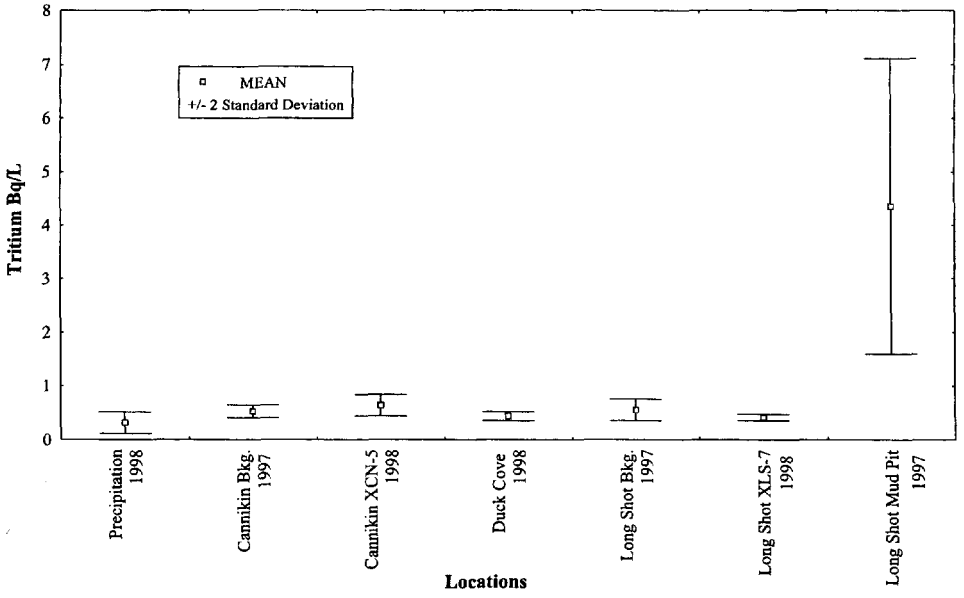

Fig. 4. Comparison of tritium activity in Bq/l for Amchitka Island, Alaska, sample sites.

Only one of the freshwater sediment sample $^{240}Pu/^{239}Pu$ atom ratios was slightly outside the 95% confidence interval for the global fallout ratio of 0.176 ± 0.014 one standard deviation (Krey et al., 1976).

For *Fucus distichus* the resulting mean $^{240}Pu/^{239}Pu$ atom ratio of 0.2173 ± 0.016 one standard deviation is slightly outside the 95% confidence interval (± 2 standard deviations) of the reported global $^{240}Pu/^{239}Pu$ atom ratio of 0.176 ± 0.014 one standard deviation (Krey et al., 1976).

The littoral zone marine sediment samples mean $^{240}Pu/^{239}Pu$ atom ratio of 0.2161 ± 0.023 one standard deviation was consistent with the higher ratio seen for the *Fucus distichus*. Duplicate analyses of several of the marine sediment samples raised some concern for inhomogeneity in the samples. These samples were not milled and had different matrix compositions, which are likely to account for the inhomogeneity observed at the 95% confidence level, based on counting error. This inhomogeneity, however, does not impact our observations.

In Fig. 3, these $^{240}Pu/^{239}Pu$ atom ratios for the freshwater samples do not suggest influence from the Amchitka underground nuclear tests when compared to the ratios observed at the nuclear test sites of Semipalatinsk, Nevada Test Site, the US Rocky Flats facility, or the Enewetak Lagoon Ivy/Mike or Castle/Bravo tests.

Pooled Amchitka $^{240}Pu/^{239}Pu$ atom ratios ($n = 26$) were compared with global fallout values ($n = 58$). The two-sample equal-variance t-test with a significance level

of 0.05 resulted in a t statistic of 6.624 and a corresponding P value (82 degrees of freedom) of $P = 0.0001$. The inference is that the two means are significantly different. The non-parametric two-sample Mann–Whitney rank sum test (2-sided) with a significance level of 0.05, a calculated U statistic of 1397.5, a normal Z approximation 6.222 and a $P = 0.001$ infer that two medians are significantly different. While recognizing that the sample sizes were substantially unbalanced and exhibited differences in skewness, these tests infer significant differences between the two sample sets.

The $^{240}Pu/^{239}Pu$ atom ratios for the *Fucus distichus*, marine sediments and pooled Amchitka samples differ from the global fallout (Krey et al., 1976) and latitudinal regional (Kelley et al., 1999) ratio values. The marine sample values in fact are comparable to the mean $^{240}Pu/^{239}Pu$ atom ratio values recently reported for the Eastern Bering Sea ($n = 36$) of 0.200 ± 0.012 one standard deviation (Hameedi et al., 1999). For the mean Eastern Bering Sea ratio it was estimated that potentially 10% of the observed plutonium input could be attributed to a highly irradiated fuel source having a $^{240}Pu/^{239}Pu$ atom ratio of 0.65, but only if sampling, analytical and environmental processes effects can be ruled out. This $^{240}Pu/^{239}Pu$ atom ratio of 0.65 is greater than that produced in the high yield thermonuclear tests at Enewetak Lagoon in the South Pacific (Kelley et al., 1999) and more characteristic of a highly irradiated nuclear reactor fuel (Sanders & Boni, 1980). In comparison, local or near-field Pu contributions from low yield weapon testing lower the mean global fallout $^{240}Pu/^{239}Pu$ atom ratio (Beasley et al., 1998; Efurd et al., 1997, 1994; Efurd, Rokop, & Perrin, 1993; Hicks & Barr, 1984; Kelley et al., 1999; Krey et al., 1976; Oughton et al., 1999; Yamamoto, Tsumura, Katayama, & Tsukatani, 1996). However, we have not identified a known source of highly irradiated nuclear fuel in this region that would contribute to an increase in the global fallout $^{240}Pu/^{239}Pu$ atom ratio observed in these samples.

Our interpretation of the $^{240}Pu/^{239}Pu$ atom ratios must be evaluated with caution in light of the very low levels of plutonium observed in these samples. There are inherent statistical errors when measurements approach the detection levels. In addition, uncertainties may be introduced in the analytical process at these low levels and potentially by physical and chemical processes occurring in the environment. These results bear further investigation; additional work still needs to be done to establish if there is another source of plutonium isotopes to this region or whether the higher observed $^{240}Pu/^{239}Pu$ ratios are the result of sampling and analysis uncertainty or environmental fate and transport processes.

15.2. Surface water tritium values

Measurements of ^{3}H at the sites reported to be leaking at Cannikin and Long Shot do not support the conclusion that leakage is occurring at these locations from the underground test cavities. Observations of ^{3}H at sites within the Long Shot watershed continue to provide evidence of an early escape of radioactive gases to the near-surface shortly after the October 1965 test (Faller & Farmer, 1998). These

measurements do not appear to reflect long-term movement from the contaminated groundwater to the Long Shot GZ surface environment.

15.3. ^{241}Am behavior

A review of the literature was made to evaluate the hypothesis that ^{241}Am has special properties in an underground nuclear test that made its fate and transport significantly different from the other transuranics. One key to the Miller and Buske (1996) hypothesis was that ^{241}Am would form +1 ions and behave as a cesium ion in the groundwater environment.

Our review of the literature, even that referenced (Penneman, 1968) by Miller and Buske (1996), did not support the original hypothesis. Americium typically exists as a +3 ion in groundwater, and while this is dependent upon groundwater chemistry, pH and E_h, no evidence suggests that it exists as a +1 ion (Cotton & Wilkinson, 1988; Silva & Nitsche, 1995). Transuranics, including americium, will tend to form slightly soluble complex ions in the groundwater (Penneman, 1968; Silva & Nitsche, 1995; Triay et al., 1997). In addition, as previously discussed, the transuranics may form or be incorporated into colloid fractions and be readily transported by groundwater (Honeyman, 1999; Kersting et al., 1999; Kim, 1991).

15.4. Hydrogeological considerations

Another important aspect in evaluating leakage from the underground test sites at Amchitka is the physical hydrogeological regime. Hydrogeological tests at Amchitka found decreasing head with depth, indicating a downward flow of recharge water to a freshwater/saline water diffusion zone where movement occurs laterally, providing for eventual exit into the marine environment (Claassen, 1978; Dudley, Ballance, & Glanzman, 1977). Results of the hydrogeological tests and a conceptual model for groundwater movement on Amchitka Island do not indicate flow paths from the shot cavities to the surface environment at the Cannikin and Long Shot sites reported to be leaking radionuclides from the underground tests (Claassen, 1978; Fenske, 1972; Wheatcraft, 1995).

16. Conclusion

The measured $^{240}Pu/^{239}Pu$ ratios and ^{3}H levels do not provide any evidence for leakage of ^{241}Am or other radionuclides from the underground test shot cavities into the terrestrial or freshwater environments on Amchitka. The only documented exception is the early phase release of gaseous radionuclides at Long Shot. In addition the hydrogeological regime as understood for Amchitka does not provide the physical means to transport transuranics from the test cavities to the reported

surface locations. Finally, a review of the literature on the physical and chemical properties of americium in the environment did not support the unique hypothesis proposed by Miller and Buske (1996) for the movement of ^{241}Am movement in groundwater.

Perturbations from the global fallout mean ^{240}Pu/^{239}Pu atom ratio that were observed in marine alga, sediment and pooled Amchitka samples may suggest another plutonium source to the marine environment. However, uncertainties in analyses and environmental processes need to be fully assessed before drawing any conclusions. Further work is needed to determine if there are any other sources for plutonium besides global fallout to the Bering Sea and North Pacific regions.

17. Future marine environmental issues

These results do not mean that leakage from the Amchitka underground nuclear tests is not occurring or will not occur into the North Pacific Ocean or the Bering Sea. Hydrogeological modeling predicts that leakage can begin initially for ^{3}H from the test sites into the marine water over ranges from 20 to 3000 years (Claassen, 1978; Dudley et al., 1977; Fenske, 1972; Wheatcraft, 1995). The ranges bridge the various hydrogeological parameter assumptions that can be made. No sampling has been conducted in the marine environment surrounding Amchitka in the past 25 years and, thus, it remains an important area to be addressed.

A current modeling effort is being done by DOE to improve the ability to further evaluate the movement of radionuclides from the shot cavities to the marine waters and the near-shore environmental pathways and risks posed to the biota and human health. This effort will provide a better conceptual knowledge of the fate and transport of radionuclides from the Amchitka underground nuclear test area.

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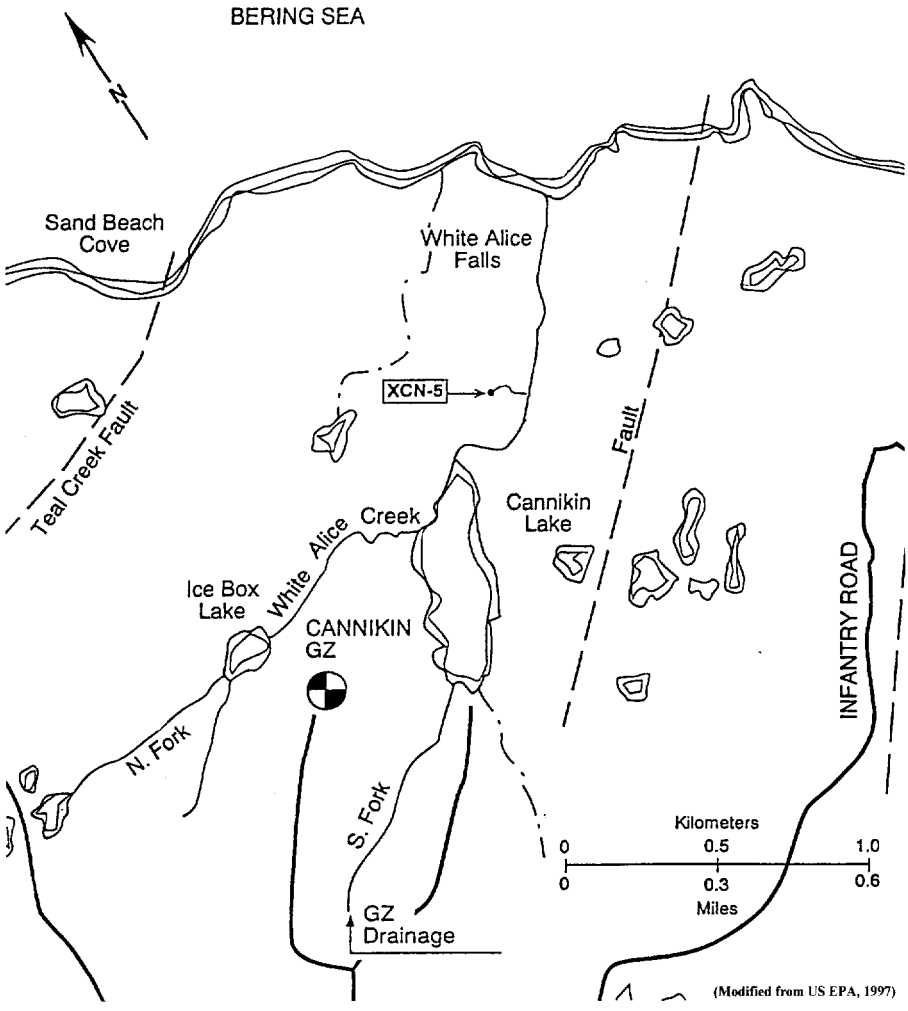
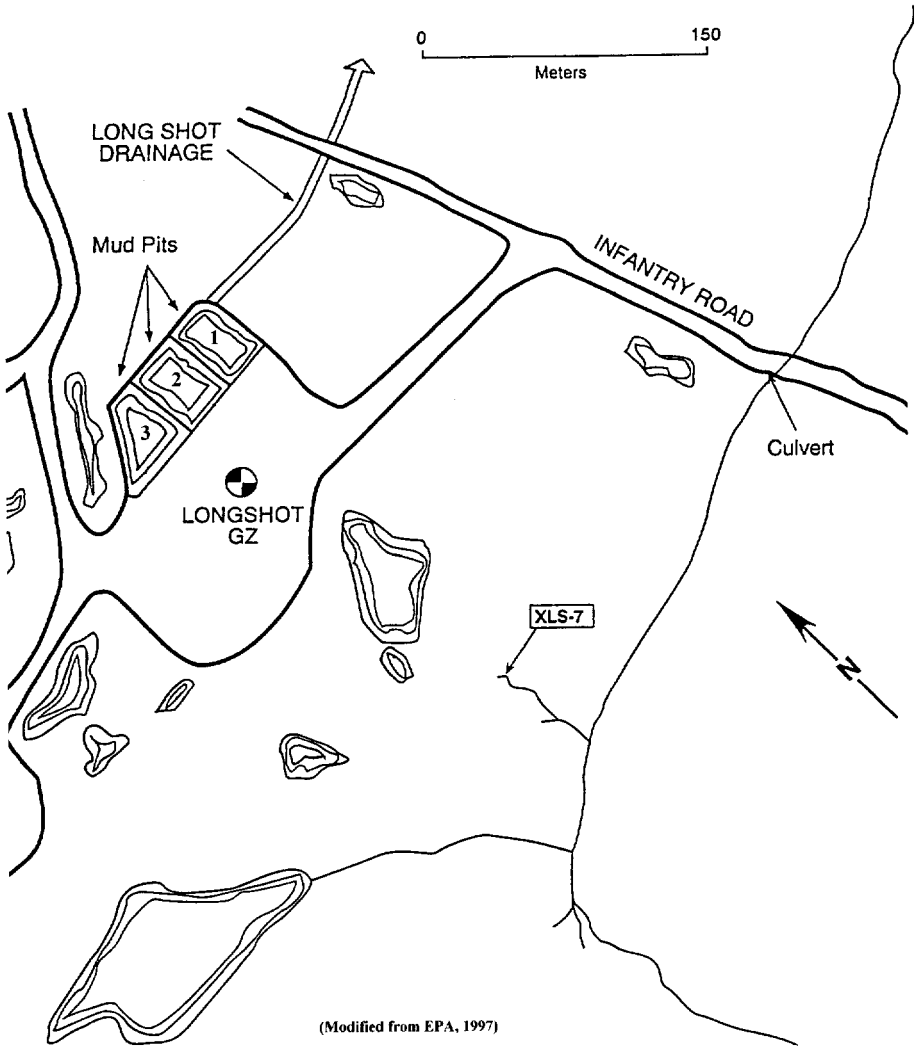
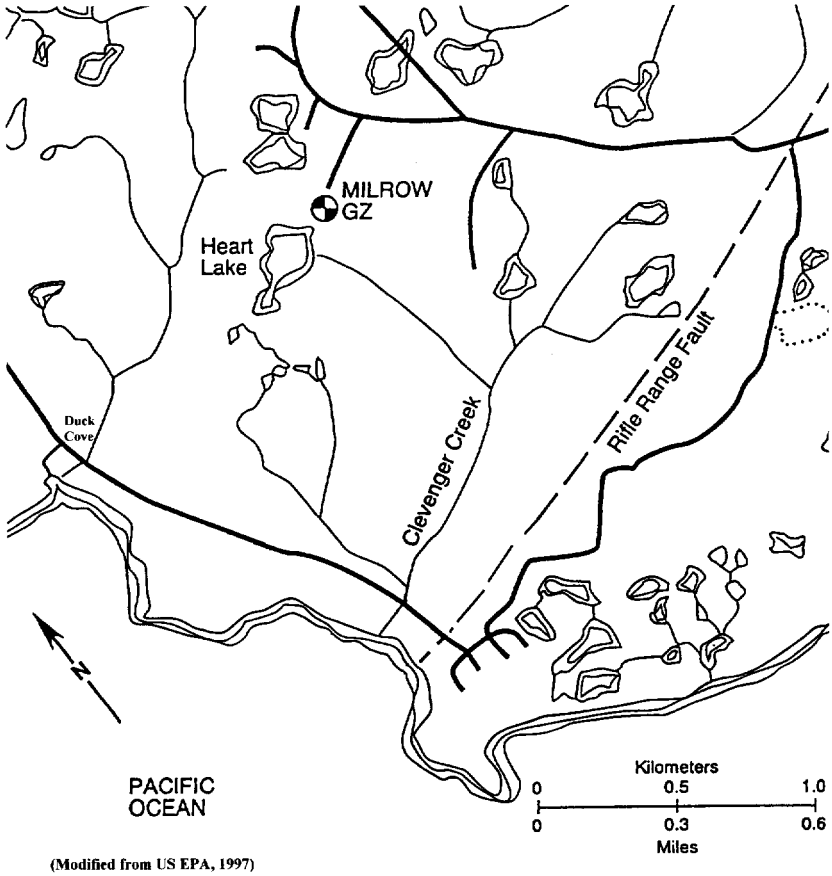
Appendix

Site maps for Cannikin, Long Shot and Milrow


BERING SEA
N
Sand Beach Cove
White Alice Falls
XCN-5
Teal Creek Fault
Fault
Cannikin Lake
White Alice Creek
Ice Box Lake
CANNIKIN GZ
INFANTRY ROAD
N. Fork
S. Fork
Kilometers
0
0.5
1.0
0
0.3
0.6
Miles
GZ Drainage
(Modified from US EPA, 1997)


0
150
Meters
LONG SHOT
DRAINAGE
Mud Pits
1
2
3
INFANTRY ROAD
Culvert
LONGSHOT
GZ
XLS-7
N
(Modified from EPA, 1997)



(Modified from US EPA, 1997)

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