Temporal variations of natural and anthropogenic radionuclides in sea otter skull tissue in the North Pacific Ocean

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Abstract

Marine mammals being among the top predators in the food web tend to accumulate organic and inorganic contaminants from the environment. The body burden of contaminants in these species could reflect their foods and thus contaminant levels could serve as proxies on the changes of ecosystem. A pilot study was carried out to investigate the possibility of radio-nuclide leakage at Amchitka using a suite of sea otter (*Enhydra lutris*) skulls collected near Amchitka nuclear test-sites before (1950s) and after the testing (1990s), and at Adak, another Aleutian Island, about 300 km from Amchitka, where the potential impact of radionuclide leakage from Amchitka is expected to be negligible. In addition, the naturally occurring and anthropogenic radionuclide content on the sea otter skull was also utilized to investigate if there was any significant ecosystem changes in the environment.

Concentration of ^{210}Pb in sea otter bones collected during the 1950s was significantly higher than those collected in the 1990s. We propose that among the various factors that could cause this higher enrichment in ^{210}Pb, changes in the sea otter prey is the most likely one. Comparison of the ^{137}Cs, ^{90}Sr, $^{239,240}Pu$ concentrations appear not to be significantly higher in sea otter

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skulls collected in 1990s from Amchitka where the underground tests in 1965–71 than those from Adak, although significant differences were detected among different groups collected at various times.

1. Introduction

The US Atomic Energy Commission jointly with the Department of Defense detonated three underground nuclear tests at Amchitka Island in the central Aleutian Archipelago between 1965 and 1971. The total yield of these detonations (6.08 million tons) is estimated to be about 15% of the total yield of all US underground nuclear explosions [Long Shot: 80 kton at 690 m below the island's surface (mbs); Milrow: 1 million ton, 1969 at 1200 mbs; and Cannikin: 5 million-ton, 1971 at 1763 mbs (the world's largest underground test)]. The Department of Energy has conducted monitoring of this site for a number of years and has found little evidence of radionuclide leakage (Sibley & Tornberg, 1982). However, there has been growing public concern on the possible leakage from the test sites. In 1997 we initiated a pilot study to investigate the possibility of radionuclide leakage at Amchitka using a suite of sea otter (*Enhydra lutris*) skulls collected near Amchitka nuclear test-sites before and after the testing, and at Adak, another Aleutian Island, about 300 km from Amchitka, where the potential impact of radionuclide leakage from Amchitka is expected to be negligible. Any potential leakage of radionuclides from the three underground nuclear tests detonated at the Amchitka Island could lead to increased concentrations of Pu and other anthropogenic radionuclides in top-level predators (such as skulls of sea otters) in the coastal areas of the Amchitka Island.

Uranium (^{238}U)-series isotopes have been utilized to date bones of marine and terrestrial mammals, fish, bivalves, etc. (Bennett, Boehlert, & Turekian, 1982; Kimura & Kastelle, 1995; Burton, Andrews, Coale, & Cailliet, 1999). The isotopic pairs that have been commonly utilized include ^{230}Th–^{234}U and ^{210}Pb–^{226}Ra. These pairs are utilized because of biodepletion of thorium and lead with increased trophic status in marine food webs while uranium and radium get incorporated into the bone (radium is a bone-seeking element while some amount of uranium is also incorporated). The concentrations of ^{210}Pb and ^{226}Ra in sea otter skull will provide information on the feasibility of dating sea otter skulls using the disequilibrium between ^{210}Pb and ^{226}Ra. Considerable deposition of plutonium (as much as 50% of the intake) on human bone surface has been reported (ICRP, 1991; Kathren, McInroy, & Swint, 1987). Variations in the concentrations of these nuclides could provide insight into the variations of the sea otter prey and other ecological changes in the coastal areas.

In order to establish the base line data for the natural and anthropogenic radionuclides, we analyzed 18 sea otter skull tissues collected from Amchitka, and Adak
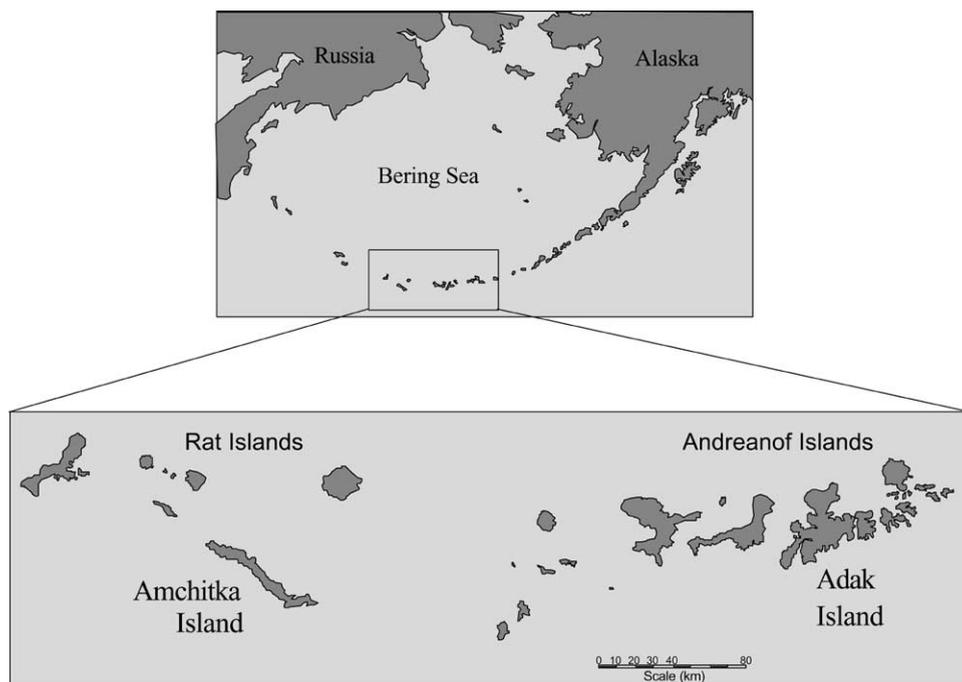
Fig. 1. Map of the sampling area.

during the time periods between 1950s and 1995 (Fig. 1). Our investigation was designed to shed light on the following questions:

1. What are the concentrations of natural and anthropogenic radionuclides in sea otter skull tissue? Does it vary with the age, and spatial and temporal locations of the sea otters? Is there any disequilibrium between ^{210}Pb and ^{226}Ra that can be employed to obtain the age of the skull since the death of the otters?
2. Are there any increased levels of anthropogenic radionuclides in sea otter skulls that could potentially be attributed to the radionuclide source adjoining Amchitka island (such as nuclear material leakage from Amchitka Island test site)?
3. Is there any particular natural radionuclide that is elevated which could provide information on the sources of that nuclide in the food chain? Can we utilize the concentration(s) of natural and/or anthropogenic radionuclides as a proxy to determine the ecological changes in the coastal marine environments of the Aleutian Island chains?

2. Rationale for the selection of sea otters

It has been widely reported that some of the environmental contaminants (including heavy metals and some radionuclides) exhibit biomagnifications as they progress through the food chain (e.g. Connel, 1990; Becker, 1993; Thomas, Sheard, & Swanson, 1994; Thomas & Gates, 1999), reaching greatest levels in apex predators. Sea otters as top-level predators in coastal marine communities could potentially accumulate high and potentially toxic residue levels of contaminants. Energetic requirements of sea otters are high, about 22% of their body mass per day (Costa, 1982) in food. The diet of the sea otter at Amchitka and elsewhere in the Aleutian Islands consists largely of the green sea urchin (*Strongylocentrotus drobachiensis*), various mollusks (mussels and clams) and some fishes (Hexagrammidae, Cottidae, Cyclopteridae) (Lensink, 1962; Kenyon, 1969; Riedman & Estes, 1990; Watt, Siniff, & Estes, 2000). Consumption of large amounts of food, required by a metabolic rate of 2.4 times that of a similar terrestrial mammal (Costa & Kooyman, 1984) could lead to elevated accumulation of contaminants that are present in the prey. In addition, sea otters provide a unique opportunity for eco-toxicological and environmental contaminant studies because both the otters and many of their prey are relatively sedentary, home ranges for sea otters most commonly include a few to tens of km of coastline and seldom range more than 1–2 km from shore (Riedman & Estes, 1990; Jameson, 1989; Ralls, Eagle, & Siniff, 1996; Bodkin & Udevitz, 1998). Most invertebrate sea otter prey have limited mobility (e.g. urchins, clams and mussels) and although fishes may be capable of long movements those species consumed by sea otters do not (except the cyclopterids) move along Island groups within the Aleutians, thus, contaminant burdens on sea otters should reflect conditions of local habitats (Estes et al., 1996; Bacon, Jarman, Estes, Simon, & Norstrom, 1999). If there were any leakage of radionuclides from any of the Amchitka test sites, then, it is likely that the leakage led to increased concentration of radionuclides close to the shoreline. Since female otters are reported to be more sedentary, we restricted our tests to this gender.

3. Materials and methods

3.1. Materials

The skull specimens were donated to the project by the USGS Biological Resources Division, and US Fish and Wildlife. These skulls were collected from various beach locations in the 1950s and 1990s and were archived immediately after collection. Individual sea otter skull samples without tooth were pulverized using a powder mill. Precleaned plastic balls were utilized in the mill. In one skull, the tooth was pulled out and analyzed separately for radionuclides.

3.2. Gamma spectrometric analysis

About 10–15 g of the powdered sample were packed into a 10-ml counting vial and assayed for gamma-emitting radionuclides. Typically, the samples were counted for 24–36 h, depending on the activities of various gamma-emitting radionuclides. There was no peak background in any of the radionuclides analyzed, except [40]K for which the background corrections were made.

The concentrations of ^{137}Cs, [226]Ra, [228]Ra, [40]K, [238]U, and ^{210}Pb were determined using a high-purity, germanium Well-Detector coupled to a Canberra InSpector multichannel analyzer. The gamma ray detector was calibrated with sediment standards for those gamma energies that were used for determining the concentrations of radionuclides. The standards utilized were obtained from the International Atomic Energy Agency (IAEA). These standards include IAEA-300 for [137]Cs (661.6 keV), RGU-1 for [210]Pb (46.5 keV) and [226]Ra (via [214]Pb, 352 keV), RGTh-1 for [228]Ra (via [228]Ac, 338 and 911 keV), and RGK-1 for [40]K (1460.8 keV). The standards were calibrated for various geometries. In the case of [226]Ra, the powdered sample was packed in the counting vial and stored for about 4 weeks for the growth of [222]Rn and its daughter products to reach secular equilibrium with its parent, [226]Ra. Typical resolution (full-width at half-maximum) was about 1.3 keV at 46 keV and 2.2 keV at 1.33 MeV. The peak/Compton ratio for [60]Co (1332 keV) was 45.0:1. Peak analysis was performed with the software peak analysis that came with the InSpector (Canberra Company). [210]Pb activities were not specifically corrected for self-absorption. The matrix densities of the standard and sample are quite comparable, and the self-absorption corrections were not applied for both the standard and the samples. In the past, we had measured ^{210}Pb concentrations in selected sediment samples by alpha counting of its daughter, [210]Po, and compared the activities obtained by alpha counting with the gamma counting. It was found that no self-absorption corrections were necessary when the standard used for calibration (dpm/cpm ratios for standards for various geometries), and the sample has comparable densities. However, the propagated errors on ^{210}Pb values are relatively high and thus we also carried out ^{210}Pb analysis via [210]Po plating followed by alpha spectrometry (e.g. Baskaran & Naidu, 1995).

3.3. Radiochemical procedures for plutonium

About 10–15 g of the pulverized sea otter skull sample was suspended in 100 ml of distilled water. To this solution, 1.00 dpm of [242]Pu spike (obtained from National Institute of Standards and Technology) followed by 20 ml of H_2O_2 (30% solution) were added and the solution was gently boiled. 25 ml of concentrated HNO_3 was added to this solution and boiled for another 30 min to remove all traces of organic matter. The solution was centrifuged and the supernatant was utilized for subsequent analysis. To this solution, 0.5 ml of purified $FeCl_3$ (equivalent to 25 mg Fe) was added. The solution was allowed to equilibrate for 12 h. Ammonium hydroxide was added drop wise while stirring to form a precipitate in the pH range of 3.2–3.4. The

first precipitate was usually white (~60% of Ca-phosphate is precipitated at this pH, while 95% of $Fe(OH)_2$ is precipitated. Actinides are associated with the Fe phase). The slurry was transferred to a 50 ml centrifuge and centrifuged at 3000 rpm for 15 min. The supernatant was discarded. The precipitate was redissolved in conc. HNO_3 and the precipitate was repeated until the precipitate was less than 5 ml in volume and had an obvious red color. Usually, three to five fractional precipitations were necessary. The final precipitate was dried at 60°C. The precipitate was dissolved in 8 ml of 8 M HNO_3 and centrifuged. The supernatant was used for radiochemical separation and purification as described in Baskaran et al. (1996). The pure Pu was electroplated onto stainless steel planchets as described in Baskaran et al. (1996). The electroplated Pu source was assayed for ^{238}Pu and ^{239,240}Pu by using 8-input alpha spectrometer with surface barrier detectors coupled to Octete-PC (Ortec Company). The background of each of the eight detectors was carefully monitored over a period of a week to 10 days time period and the background count rates were utilized to subtract the background counts in each of the detector.

Since the concentrations of ^{238}Pu and ^{239,240}Pu were very low, it is critical to use alpha detectors with very low backgrounds. Eight alpha detectors were used for Pu. Background of the detectors was determined after leaving the detector counting for 7–12 days. The backgrounds in the ^{238}Pu and ^{239,240}Pu regions varied between 0.006 and 0.073 counts per hour (cph) for different detectors but remained constant for any given detector. The errors on the activities of ^{238}Pu and ^{239,240}Pu were propagated for the errors arising due to background, counting statistics, and the spike activity.

3.4. Radiochemical analysis for ^{90}Sr

The analytical procedure was modified from the one published by Wong, Jokela and Noshkin (1994). About 4 g of pulverized sample was taken for analysis. About 100 Bq of ^{85}Sr was added to each sample for Sr yield determination and about 1 g stable Sr was added as a chemical carrier. The oxalate was ashed at 550°C for one day and the ashes were dissolved in concentrated HNO_3, which precipitates Sr, Ca, and Ba in nitrate form. After purification, the chemical recovery of Sr was determined by gamma ray spectrometric analysis of ^{85}Sr. The sample solution was set aside for at least 14 days to enable the full ingrowth of ^{90}Y. Thereafter, stable yttrium oxalate was added to the solution and Yr was precipitated as yttrium oxalate and filtered onto a pre-weighed membrane filter. The chemical recovery of Yr was determined gravimetrically from the dried filter and precipitate. The ^{90}Y samples were counted in a low background beta counter (Risø National Laboratory) for 300–600 min continuously over a period of 2 weeks. The accuracy and precision is more than 90% and less than 10%, respectively. The detection limit for ^{90}Sr was 0.14 Bq kg^{-1}. The chemical procedure was periodically checked with standard reference materials for ^{90}Sr analysis.

3.5. Statistical analysis

The 18 skull samples were divided into 3 groups for comparison. They were: Adak Control (6 samples) collected from Adak in 1992 and 1995 served as control; Amchitka Post-testing (6 samples of skull and 1 tooth that came out of one of these skulls) from Amchitka area collected in 1992 (Amchitka Island was used as underground nuclear tests in the period of 1969–1971); and Amchitka Pre-testing from Amchitka area collected in 1954 (5 samples) and 1957 (1 sample in 1957; these were collected prior to nuclear testing in Amchitka). The concentrations of radionuclides in all three groups were analyzed using a Kruskal–Wallis test to determine if there were significant differences among these three groups. Non-parametric Tukey test (Zar, 1984) was conducted to determine which groups differed from one another.

4. Results and discussion

Of all the radionuclides measured in the otter skulls, five (^{137}Cs, ^{90}Sr, ^{238}Pu, ^{239,240}Pu) are anthropogenic and are largely derived from stratospheric fallout (global fallout), releases from production and reprocessing plants, and possible leakage from test sites. Three (^{238}U, ^{226}Ra and ^{210}Pb) of the remaining four is daughter products in the U-series, and ^{40}K is a naturally occurring primordial radionuclide. Concentrations of ^{137}Cs and ^{90}Sr were corrected to January 1, 2000. Lead-210 values were corrected for the year of collection (discussed below).

4.1. Concentrations of anthropogenic radionuclides ^{90}Sr, ^{137}Cs, ^{238}Pu, ^{239,240}Pu and activity ratios of ^{137}Cs/^{90}Sr and ^{238}Pu/^{239,240}Pu

The ^{90}Sr ($t_{1/2}$=28.5 yr), ^{137}Cs ($t_{1/2}$=30.0 yr), ^{238}Pu ($t_{1/2}$=87.8 yr), and ^{239,240}Pu (^{239}Pu ($t_{1/2}$=2.41×10^4 yr), ^{240}Pu ($t_{1/2}$=6538 yr)), concentrations in eighteen sea otter skulls and one tooth sample are given in Table 1. Strontium-90 concentrations in all the Amchitka pre testing group samples were below detection limit. Since more than 50% of the ^{90}Sr present in the samples collected in 1950s would have undergone radioactive decay, and the depositional input during 1950 was relatively low compared to global fallout maximum period of 1962–1964 (HASL, 1977), the concentration in Amchitka pre testing group is likely below detection limit. The mean concentration in Amchitka post testing group, 5.6±2.4 Bq kg^{-1}, is about 30% higher than the mean concentration in Adak control group, 4.3±1.4 Bq kg^{-1}. However, a Kruskal–Wallis test indicates that Amchitka post testing and Adak control groups are not significantly different. The mean concentration of ^{90}Sr for Adak control and Amchitka post testing samples is 4.97 Bq kg^{-1}.

In most of the samples, the ^{137}Cs concentrations were low and in 6 samples, they were below detection limit. In the remaining 12 samples, the concentrations varied between 0.7 and 1.3 Bq kg^{-1}, and were barely above the detection limit (0.5 Bq kg^{-1}). Of the three groups, the average value for Amchitka post testing group was slightly higher (1.1±0.5 Bq kg^{-1}) as compared to Adak Control group (0.81±0.25).

Table 1
Concentrations of ^{137}Cs, ^{234}Th, ^{210}Pb, ^{40}K, ^{226}Ra, ^{238}Pu, $^{239,240}Pu$ and ^{90}Sr in sea otter skull and tooth samples

Sample	Year of collection	Age (yr)	Area collected	^{137}Cs (Bq kg^{-1})	^{234}Th (Bq kg^{-1})	^{210}Pb (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{238}Pu (mBq kg^{-1})	$^{239,240}Pu$ (mBq kg^{-1})	^{90}Sr (Bq kg^{-1})	$^{238}Pu/^{239,240}Pu$ (AR)
SO A01	1992	10	Adak	0.64±0.52	BD	12.1±3.8	12.5±4.8	3.04±0.77	2.3±1.6	0.9±1.8	3.03±0.84	
SO A08	1992	14	Adak	0.66±0.43	6.1±4.7	19.4±4.7	12.4±4.8	2.00±0.85	−5.3±5.7	−3.2±5.3	6.42±1.32	
SO A09	1992	15	Adak	1.25±0.71	10.8±5.8	135.3±7.3	25.8±3.8	3.66±0.93	5.8±11.7	−9.2±18.3	4.72±1.35	
ADC 9501	1995	16	Adak	0.74±0.64	16.1±4.8	14.2±4.7	7.04±2.01	1.77±0.75	15.3±19.6	15.3±19.6	2.40±0.78	
ADC 9502	1995	9	Adak	0.77±0.49	14.2±3.8	13.8±3.7	10.1±4.6	2.19±0.73	39.7±28.4	13.5±12.5	4.72±0.90	
ADC 9505-A	1995	10	Adak	BD	9.32±3.5	5.61±3.45	48.5±8.2	0.81±0.59	28.6±29.3	71.5±26.2	4.52±1.13	
9209	1992	13	Amchitka	BD	11.2±4.6	165.8±6.3	8.7±4.0	2.70±0.85	−1.8±3.9	62.0±11.6	3.58±0.69	
9210-A	1992	6	Amchitka	1.9±1.5	2.23±0.54	49.0±4.5	2.8±1.8	0.82±0.24	0.37±1.4	62.3±8.5	3.04±1.29	0.006±0.023
9210-B*	1992		Amchitka	BD	72.1±7.2	38.9±5.5	195.0±11.2	4.34±0.14	NM	NM		
9214	1992	16	Amchitka	0.90±0.53	5.2±4.5	89.7±5.8	7.5±4.8	3.38±0.81	−33.3±22.1	73.3±31.5	9.16±1.86	
9215	1992	15	Amchitka	1.03±0.53	BD	11.9±4.1	BD	2.43±0.81	0.60±3.4	23.5±6.3	4.84±0.58	
9220	1992	9	Amchitka	0.67±0.49	7.47±4.47	25.3±4.9	8.83±5.76	1.97±0.83	7.10±5.0	22.0±11.3	5.37±1.41	0.32±0.28
9224	1992	11	Amchitka	1.08±0.54	8.8±4.2	188.4±6.9	11.7±5.1	3.35±0.69	6.5±2.7	15.3±4.3	7.87±1.48	0.42±0.21
D 157	1957	8	Amchitka	1.24±0.65	12.9±5.4	109.3±6.3	16.8±5.1	3.72±0.96	BD	4.5±3.5	BD	
D 7-57	1957	15	Amchitka	1.03±0.56	19.4±5.9	187.8±8.0	22.3±5.6	4.93±2.90	15.7±8.8	8.0±7.5	BD	
D 1057	1957	15	Amchitka	BD	BD	16.5±4.6	18.1±5.3	2.43±0.70	9.9±14.6	34.5±18.0	BD	0.29±0.45
D 1157	1957	3	Amchitka	0.70±0.63	10.2±5.2	207.5±7.3	32.6±5.8	2.40±0.79	−6.8±8.2	−5.5±6.9	BD	
D 1457	1957	10	Amchitka	BD	BD	78.9±5.5	16.7±5.2	4.08±0.91	BD	−1.9±12.3	BD	
AIWR 1154	1954	20	Amchitka	BD	21.1±4.2	144.3±5.5	12.0±4.3	2.06±0.67	−14.2±4.9	21.0±7.5	BD	

NM: Not measured; BD: Below detection limit: *sea otter tooth taken out of skull of sample 9210-A. Age of sea otter skull was taken from Bodkin 2000, (unpublished data) Adak Control group: SO A01, SO A08, SO A09, ADC 9501, ADC 9502, ADC 9505-A (n=6) Amchitka post-test group: 9209, 9210, 9214, 9215, 9220, 9224 (n=6) Amchitka pre-test group: D 157, D 7-57, D 1057, D 1157, D 1457, AIWR 1154 (n=6)

Statistical analyses conducted on the ^{137}Cs concentrations indicate that there was no significant difference among groups.

Plutonium-239,240 concentrations varied between below detection limit to 73.3 mBq kg^{-1}, with a mean value of 30.5 mBq kg^{-1} (n=14, assuming that all the below detection limit values are zero values). The mean concentration in Amchitka post testing group (43.1±25.4 mBq kg^{-1}) was significantly higher than those in Amchitka pre-testing (16.9±27.6 mBq kg^{-1}, assuming that the negative values are zero) control group. It must be pointed out that the errors associated with the measurements are high due to extremely low concentrations and it is difficult to conclude unequivocally if the sea otters collected from Amchitka area after the nuclear tests (collected in 1992) were elevated in Pu concentrations. However, three samples (9209, 9210 and 9214) collected in 1992 from Amchitka area had the highest values, yet, they are significant. Statistical analyses of Pu data indicate that ^{239,240}Pu was significantly lower at the Amchitka pre-testing group than at Amchitka post-testing or Adak control groups in 1990s ($P < 0.05$), which is expected due to very low global fallout in the 1950s. There is no significant difference between Adak control and Amchitka post-testing groups.

^{238}Pu concentrations were very low with high errors and it is difficult to interpret the data. However, in 5 of the 18 samples, the one sigma propagated error is less than the value and in only one sample (sample number 9224, collected near Amchitka in 1992), the ^{238}Pu concentration is significant within 2 sigma errors. The ^{238}Pu/^{239,240}Pu activity ratio in that sample is 0.42±0.21. This is significantly different than the global value of 0.03 in the Northern hemisphere (Baskaran et al., 1995), and if this value is confirmed, there is additional ^{238}Pu source in this region. With one data point we are unable to firmly discuss further on the validity of this ratio. Thus, it is not feasible to determine the amount of Pu, if any, derived from the nuclear tests in Amchitka. The atomic ratios (such as ^{239}Pu/^{240}Pu) and activity ratios (^{238}Pu/^{239,240}Pu) provide information on the sources of Pu (e.g. Baskaran et al., 1995; Buesseler, 1997; Cooper et al., 1999; Cooper, Kelley, Bond, Orlandini, & Grebmeier, 2000). Future studies on the atomic ratios of ^{239}Pu/^{240}Pu and activity ratios of ^{238}Pu/^{239,240}Pu in these and other natural samples (sediments and biota) could potentially provide information on the sources of Pu to this site.

The activity ratios of ^{137}Cs/^{90}Sr varied between 0.10 and 0.63, with a mean value of 0.22. Since the errors associated with the ^{137}Cs measurements are high (due to low levels of ^{137}Cs), we do not interpret this data further (Table 2). However, it is pertinent to point out the ^{137}Cs/^{90}Sr ratios in global fallout had a constant value of 0.67 (HASL, 1977; Baskaran & Naidu, 1995).

4.2. Concentrations of naturally occurring radionuclides ^{40}K, ^{210}Pb, ^{226}Ra and ^{238}U and activity ratios of ^{210}Pb/^{226}Ra and ^{226}Ra/^{238}U

The concentrations of ^{40}K in all the 18 samples varied between below detection limit to 48.5 Bq kg^{-1}, with a mean of 16.1 Bq kg^{-1} (except one tooth sample with a value of 195.0 Bq kg^{-1}). The samples from the Amchitka post-test group contain the lowest ^{40}K concentrations (average value of 7.9 Bq kg^{-1}, as compared to the

Table 2
Activity ratios of $^{210}Pb/^{226}Ra$, $^{226}Ra/^{238}U$, $^{137}Cs/^{90}Sr$, and $^{238}Pu/^{239,240}Pu$ in sea otter samples

Sample	Year of collection	Area collected	$^{210}Pb/^{226}Ra$ Activity ratio	$^{226}Ra/^{238}U$ Activity ratio	$^{137}Cs/^{90}Sr$ Activity ratio	$^{238}Pu/^{239,240}Pu$ Activity ratio
SO A01	1992	Adak	4.0±1.5	–	0.21±0.20	NC
SO A08	1992	Adak	9.7±4.1	0.33±0.33	0.10±0.06	NC
SO A09	1992	Adak	37.0±4.4	0.34±0.18	0.26±0.16	NC
ADC 9501	1995	Adak	8.0±4.1	0.11±0.06	0.31±0.33	NC
ADC 9502	1995	Adak	6.3±2.4	0.15±0.06	0.16±0.10	NC
ADC 9505-A	1995	Adak	6.9±7.9	0.09±0.08	0	0.40±0.46
9209	1992	Amchitka	61.4±8.4	0.24±0.12	0	NC
9210-A	1992	Amchitka	59.8±10.6	0.37±0.13	0.63±0.65	0.006±0.022
9210-B			9.0±1.3	0.06±0.01	ND	ND
9214	1992	Amchitka	26.5±3.2	0.65±0.64	0.10±0.05	NC
9215	1992	Amchitka	4.9±2.2	–	0.21±0.08	0.03±0.15
9220	1992	Amchitka	12.8±4.8	0.26±0.21	0.12±0.10	0.32±0.31
9224	1992	Amchitka	56.2±4.4	0.38±0.17	0.14±0.06	0.42±0.21
D 157	1957	Amchitka	29.4±3.7	0.29±0.12	–	NC
D 7-57	1957	Amchitka	38.1±14.6	0.25±0.17	–	NC
D 1057	1957	Amchitka	6.8±2.5	–	–	0.29±0.50
D 1157	1957	Amchitka	86.5±12.4	0.14±	–	NC
D 1457	1957	Amchitka	19.3±2.3	–	–	NC
AIWR 1154	1954	Amchitka	70±10	0.10±0.04	–	NC

NC: Not calculated, as the ratios are either negative or >1. ND: No data Adak Control group:SO A01, SO A08, SO A09, ADC 9501, ADC 9502, ADC 9505-A (n=6) Amchitka post-test group:9209, 9210, 9214, 9215, 9220, 9224 (n=6) Amchitka pre-test group:D 157, D 7-57, D 1057, D 1157, D 1457, AIWR 1154 (n=6)

values of 19.4 and 19.8 Bq kg^{-1} for Adak control and Amchitka post-test groups, respectively). Statistical analysis indicates that ^{40}K were significantly lower in Amchitka post-test group than in Amchitka pre-test group ($P < 0.05$) and Adak control group ($P < 0.10$). This significantly lower ^{40}K value in the Amchitka post-testing group could be due to significant changes in the K-bearing dietary food intake by sea otters. No significant difference between Adak control group and Amchitka pre-test group was found.

The concentrations of ^{238}U in all the samples varied between below detection limit to 21.1 Bq kg^{-1}. Except one tooth sample (9210-B), most of the concentrations fall within a narrow range of 5.2–21.1 Bq kg^{-1}, with a mean value of 11.1±5.3 Bq kg^{-1} (excluding 4 samples where the concentrations were below detection limit). The tooth sample was enriched with ^{238}U (as well as ^{40}K) as much as one or two orders of magnitude higher than skull samples. The samples in the Amchitka post-test group contain the lowest ^{238}U concentrations, with an average value of 7.0±3.4 Bq kg^{-1}, as compared to 11.3±4.0 Bq kg^{-1} in Adak control group and 15.9±5.2 Bq kg^{-1} in Amchitka pre-test group. Statistical analyses indicate that ^{238}U was significantly higher in Amchitka pre-test group than in Amchitka post-group and Adak control group ($P < 0.05$). There is no significant difference between Adak control group and Amchitka post-test group.

Lead-210 concentrations were initially obtained by gamma ray spectrometry. Since the propagated errors associated with the data were relatively high, alpha spectrometry (via ^{210}Po) was employed. Both values are given in Table 3. In most of the samples, the agreement between the two methods is excellent. The data obtained by alpha spectrometry are only discussed below.

Lead-210 concentration in all the samples varied between 10.2 and 248.5 Bq kg^{-1}, with a mean value of 75.7 Bq kg^{-1}. Highest ^{210}Pb concentrations were found in samples in the Amchitka pre-test group (124±71 Bq kg^{-1}) compared to Adak control group samples (33±50 Bq kg^{-1}) and Amchitka post-testing group (88±74 Bq kg^{-1}). The ^{226}Ra concentrations in the whole set of samples ranged between 0.81 and 4.93 Bq kg^{-1}, with a mean value of 2.65 Bq kg^{-1}. The mean value of ^{226}Ra concentration in Adak control group (2.2±1.0 Bq kg^{-1}) is comparable to Amchitka post-test group (2.4±1.0 Bq kg^{-1}) and the mean value in Adak control samples (3.3±1.1 Bq kg^{-1}) is slightly higher than Amchitka post-test group and Adak control groups. In all samples, the ^{210}Pb concentration is significantly higher than ^{226}Ra. A similar enrichment of ^{210}Pb over ^{226}Ra was observed in the bones of caribou (Thomas & Gates, 1999). Statistical analyses indicate that ^{210}Pb was significantly higher in Amchitka pre-test group than in Amchitka post-test group or Adak control group ($P < 0.06$). There is no significant difference between Adak control group and Amchitka post-test group.

There are four major potential sources of ^{210}Pb to sea otter bones: (i) from the decay of ^{226}Ra that incorporated prior to the death of the sea otter (assuming that the bone remained a closed system); (ii) direct intake from the water column by the animal which incorporated into the bone; (iii) production from the decay of ^{222}Rn that is taken-up by the animals; and (iv) intake of radioactive lead from the food. Of these four sources, contributions from (ii) are likely negligible, as Pb is discrimi-

Table 3
Comparison of ^{210}Pb activities obtained by alpha spectrometry and gamma spectrometry, initial activities of ^{210}Pb in the skull at the time of sample collection, and activity ratios of ^{210}Pb/^{226}Ra

Sample	Year of collection	^{210}Pb activity (alpha) – Bq kg^{-1}	^{210}Pb activity (gamma) – Bq kg^{-1}	^{210}Pb initial activity (Bq kg^{-1})	^{210}Pb/^{226}Ra initial activity ratio
SO A01	1992	11.6±1.0	12.1±3.8	13.9	4.6±1.2
SO A08	1992	24.1±0.8	19.4±4.7	30.3	15.2±6.5
SO A09	1992	134.9±4.0	135.3±7.3	171.8	46.9±12.0
ADC 9501	1995	11.0±0.6	14.2±4.7	12.5	7.1±3.0
ADC 9502	1995	12.0±0.7	13.8±3.7	13.7	6.2±2.1
ADC 9505-A	1995	10.2±0.4	5.61±3.45	11.8	14.5±10.6
9209	1992	159.9±3.4	165.8±6.3	204.2	75.6±23.9
9210-A	1992	52.0±2.1	49.0±4.5	66.4	81.0±23.9
9214	1992	86.0±2.0	89.7±5.8	109.3	32.3±7.8
9215	1992	15.8±0.6	11.9±4.1	19.6	8.1±2.7
9220	1992	20.7±0.7	25.3±4.9	25.9	13.2±5.6
9224	1992	101.1±1.6	188.4±6.9	128.7	38.4±7.9
D 157	1957	112.9±4.2	109.3±6.3	418.2	112.4±29.3
D 7-57	1957	196.2±5.6	187.8±8.0	731.1	148.3±87.3
D 1057	1957	20.4±0.7	16.5±4.6	70.5	29.0±8.4
D 1157	1957	248.5±13.6	207.5±7.3	936.9	390.4±130.3
D 1457	1957	69.8±2.2	78.9±5.5	253.7	62.2±14.0
AIWR 1154	1954	NM	144.3±5.5	595.1	288.8±94.6

NM: Not measured Adak Control group: SO A01, SO A08, SO A09, ADC 9501, ADC 9502, ADC 9505-A (n=6) Amchitka post-test group: 9209, 9210, 9214, 9215, 9220, 9224 (n=6) Amchitka pre-test group: D 157, D 7-57, D 1057, D 1157, D 1457, AIWR 1154 (n=6)

nated by most organisms and ^{210}Pb is removed very fast from the water column by suspended particles (Rama, Koide, & Goldberg, 1961; Baskaran & Santschi, 1993; Brenner, Peplow, & Schelske, 1994). Being a noble gas, radon is not retained in the body of any animals. In addition, concentration of ^{222}Rn in surface water is significantly lower than ^{226}Ra and this was caused by loss to the atmosphere (Broecker & Peng, 1974) and hence most likely the contribution from (iii) is negligible. Since ^{210}Pb concentrations at the time of collection are excessively higher than those of ^{226}Ra, it is likely that most of the radioactive lead is primarily derived from the intake of lead from the foods. Of the daughter products of ^{226}Ra, ^{210}Po has been reported to accumulate in biological organisms (Heyraud & Cherry, 1979; Cherry & Heyraud, 1981; Kim et al., 2002). Since the samples were collected many years ago, all the ^{210}Po would have decayed away.

Because exchange of ^{226}Ra and ^{210}Pb with the surrounding since the death of otters presumably has not occurred, the measured ^{210}Pb concentration in the sea otter samples is due to the following factors: (a) decay of initial ^{210}Pb from the death of organism to the time of counting; and (b) production of ^{210}Pb from the parent, ^{226}Ra. From the measured activity and time elapsed since collection, the ^{210}Pb activity present at the time of death (called 'initial' activity, ^{210}Pb$_i$, from now onwards) of the otter can be calculated from the following relationship:

$$[^{210}\text{Pb}]_m = {}^{210}\text{Pb}_i e^{-\lambda t} + {}^{226}\text{Ra}[1 - e^{-\lambda t}] \tag{1}$$

where $[^{210}\text{Pb}]_m$ is the measured activity, $[^{210}\text{Pb}]_i$ is the initial activity (when the otter died), λ is the decay constant of ^{210}Pb (0.03108 yr^{-1}) and 't' is the time elapsed from the death of organisms to counting of the samples. Knowing the date of collection and concentrations of $[^{210}\text{Pb}]_m$ and ^{226}Ra, the initial $[^{210}\text{Pb}]_i$ activities can be calculated and are given in Table 3. The initial ^{210}Pb concentration in all the 18 samples varied between 11.8 and 731 Bq kg^{-1}, with a mean value of 200 Bq kg^{-1}. The mean value for Amchitka pre-test group is significantly higher (501 Bq kg^{-1}) than those in Adak control group (42.3 Bq kg^{-1}) and Amchitka post-test group (92.4 Bq kg^{-1}). Such variation from samples collected in the same region within a time span of 40 years is likely due to differences in the relative proportions of various prey ingested, changes in the ecosystem, and lifespan of the otters. Although data analysis of fecal matter and stomach collections (Bodkin, 2000, unpublished data) indicate there was no large-scale dietary shift in this area, variations on the abundance of individual sea urchins (which is likely enriched in Pb) could lead to variations in the uptake of radioactive lead. Sea otters have highly individualized diets and hence, the 'typical prey' could vary considerably from one another (Riedman & Estes, 1990). Although there is a general trend that the older sea otters contain higher ^{210}Pb concentration, there is no strong indication of systematic change with time or space. There is no correlation between the age of the otters and the initial ^{210}Pb concentration in the otters. Goldberg, Koide, Hodge, Flegal and Martin (1983) reported highly varying concentrations of heavy metals and radionuclides in bivalves that were collected around the US coastal waters over a period of 2 years.

Most of the radioactive lead to the coastal waters is delivered through atmospheric deposition, and that the long-term annual input has remained relatively constant

(Benninger, Aller, Cochran, & Turekian, 1979; Baskaran, Coleman, & Santschi, 1993), the changes in the ^{210}Pb is likely due to variations in the amount of radioactive lead retained in some of the sea otter prey. Being a steady-state tracer, the concentration of ^{210}Pb in the ocean water does not change appreciably with time (e.g. Baskaran & Santschi, 2002), and body burden of ^{210}Pb of marine plants, zooplanktons and fishes also are not expected to change with time, and thus change in ^{210}Pb activity in sea otter skull is not possible without change of major food items for sea otter. Since the sea urchins have remained the dominant prey, the increase in ^{210}Pb levels in otters collected in 1954 could be either due to increased levels of ^{210}Pb in urchins in the 1950s and/or due to some episodic prey that contains high concentrations of ^{210}Pb. Data on the population density of sea otters during the last 90 years in the Amchitka area indicate that there was a population crash of ~60% in early 1950s and the urchins were small with low biomass and the habitat was kelp dominated (Estes, Tinker, Williams, & Doak, 1998). The specific activity of ^{210}Pb per unit biomass would have been significantly higher during that time, which could have resulted in higher intake of radioactive lead by the sea otters. The population is declining at a rate of ~25%/year since 1987 until the present, and the density and sizes of urchins are more abundant and larger and the kelp forests are in decline (Estes et al., 1998; Bodkin, 2000, unpublished data). It is also likely that urchins in the 1950s were more reliant on grazing on kelps, while those in the 1990s, may have been grazing on diatoms as urchin barrens were becoming prevalent due to otters decline in abundance and urchins increase in size and number resulting in urchin barrens. Decadal scale ecosystem changes have been reported in the Bering Sea (Mantua, Hare, Zhang, Wallace, & Francis, 1997; Hare & Mantua, 2000). From the concentrations of initial ^{210}Pb in sea otter skull, one can say the radioactive lead-enriched sea otter prey has changed considerably over the last 50 years. By extension, it appears that there had been a significant ecosystem change since 1950s in those waters under which these otters lived and the nature of prey has significantly altered. Ideally, analyses on sea otter prey samples collected in the 1950s would enable us to test our hypothesis, but unfortunately no such samples were collected in the 1950s and the current food items for sea otter may not reflect the food items for 1950s. As of now, there is no way to confirm the high level of ^{210}Pb accumulation in the 1950s unless we do time-series measurement for the longer period and the similar ecosystem change is repeated.

It has been reported that there are variations in the accumulation patterns of heavy metals and trace elements in the liver, and kidneys that were dependent on the size/age of sea otters (Riedman & Estes, 1990). Certain heavy elements do not get accumulated in the higher trophic levels of the food chain. An earlier study by Smith, Niemeyer, Estes and Flegal (1990) on the stable lead concentration in teeth of sea otters showed that the sea otter lead burdens have not increased significantly in contemporary sea otters (collected in 1986 and 1987) compared to the pre-industrial sea otters (collected in 1937). Although Smith et al. (1990) did not find significant increases in the lead content of the teeth of contemporary sea otters, the isotopic ratios of lead indicated substantial changes in the source of lead. Enrichment of lead from seawater to plants due to strong chelation of lead on plant surfaces has been

observed (Burnett & Patterson, 1980; Smith et al., 1990). In addition, some plants such as algae have large surface area/volume ratio (Smith et al., 1990). In subsequent transfers to consumer organisms, lead gets depleted due to discrimination of lead against calcium during the metabolic assimilation of elements (Smith et al., 1990). In a simple food chain consisting of algae and gastropod, Burnett and Patterson (1980) reported the following: a 5-fold increase in Pb/Ca in algae decreased to 3-fold increase in the Pb/Ca ratio in the shell of a gastropod and only 50% increase in its muscle.

The activity ratios of $^{210}Pb_i/^{226}Ra$ are given in Table 3. If all the ^{210}Pb to the skull is derived from the radioactive decay of ^{226}Ra (as Ra is a bone-seeking element), then, the $^{210}Pb/^{226}Ra$ activity ratio is expected to be less than 1. In all the 18 samples, this ratio varied between 4.6 and 390, with a mean value of 76. The propagated error associated with this ratio is high, due to low concentrations of ^{226}Ra in most of the samples. This ratio is the lowest in Adak group samples (15.8), as compared to Amchitka post-test samples (mean: 41.4) and Amchitka pre-test samples (mean: 172). This excess of ^{210}Pb over its parent in the sea otter skull can be employed to date the bones. Radium-226/Uranium-238 activity ratios varied between 0.09 and 0.65, with a mean value of 0.26. This is likely due to more U deposition on to the bone than ^{226}Ra.

5. Conclusions

We report the first set of radionuclide measurements on sea otter skulls collected from the marine environments surrounding a nuclear underground test site. We have analyzed a suite of radionuclides, ^{40}K, ^{90}Sr, ^{137}Cs, ^{210}Pb, ^{226}Ra, ^{238}Pu, and $^{239,240}Pu$ on 18 sea otter skulls and 1 tooth collected from two areas: Amchitka Island and Adak. Overall, the concentrations of all the radionuclides are very low. In 3 samples collected in 1992 from Amchitka, >20 years following the underground nuclear tests, the $^{239,240}Pu$ concentrations are the highest. On the other hand, in 6 samples collected from Amchitka in 1950s, the $^{239,240}Pu$ concentrations are low. Due to low concentrations, we could not see any distinct $^{238}Pu/^{239,240}Pu$ activity ratios (with propagated error on the activity ratios). It is not clear if some of the Pu could have come from leakage from Amchitka test site. Since we have only two groups of samples (1950s and 1992), we are unable to conclude how the Pu concentration would be on those sea otters that were collected in between these time periods. In addition, it would be highly desirable to collect a few marine sediment samples as well as sea otter prey adjacent to the Amchitka Island (on the Bering Sea side) and investigate the Pu concentrations (inventories for sediments, and concentrations and activity ratios for sediments and biological samples) to determine if there is any input of Pu derived from nuclear test site.

The concentration of ^{210}Pb is significantly higher than ^{226}Ra, possibly suggesting

additional source(s) of ^{210}Pb to the sea otter skulls. We propose that one or more of the prey of sea otters are highly enriched in radioactive lead (and possibly stable also). Further work is needed to track the source of radioactive lead to the sea otters.

Acknowledgements

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