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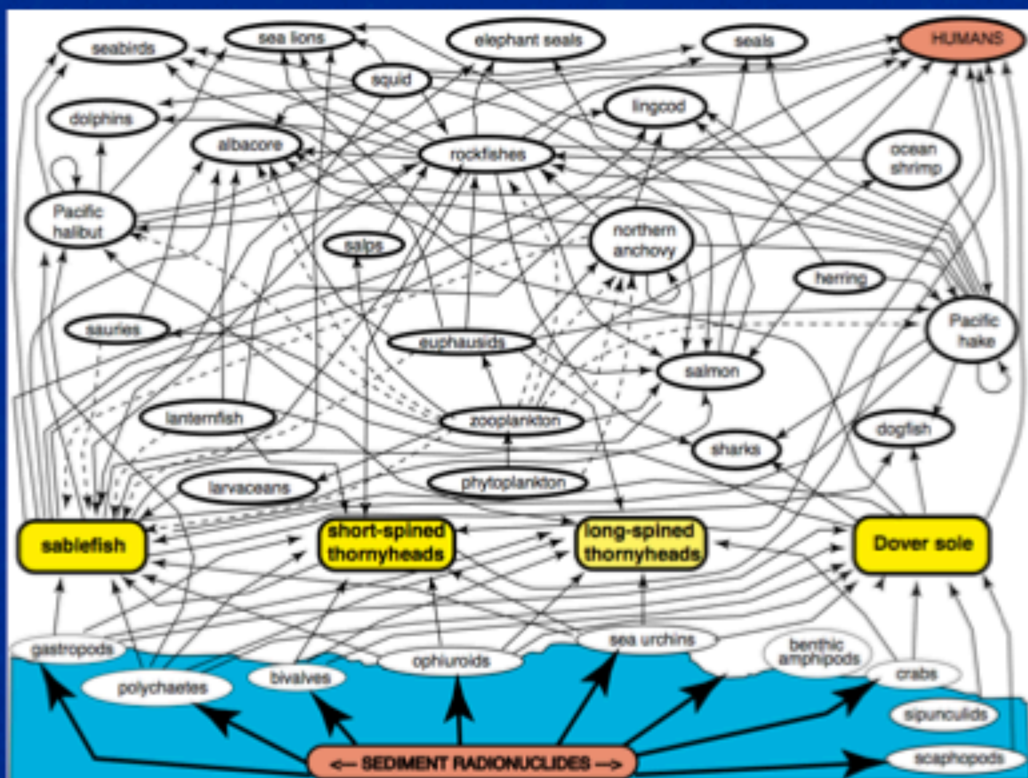

On the Cover: Simplified food web for species analyzed for radionuclides from the Farallon Islands Nuclear Waste Dump Site. See article by Suchanek et al. on page 167 for more information.

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RADIONUCLIDES IN FISHES AND MUSSELS FROM THE FARALLON ISLANDS NUCLEAR WASTE DUMP SITE, CALIFORNIA

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Abstract—The Farallon Islands Nuclear Waste Dump Site (FINWDS), approximately 30 miles west of San Francisco, California, received at least 500 TBq encapsulated in more than 47,500 containers from approximately 1945 to 1970. During several seasons in 1986/87 deep-sea bottom feeding fishes (Dover sole = *Microstomus pacificus*; sablefish = *Anoplopoma fimbria*; thornyheads = *Sebastolobus* spp.) and intertidal mussels (*Mytilus californianus*) were collected from the vicinity of the FINWDS and from comparable depths at a reference site near Point Arena, CA. Tissues were analyzed for several radionuclides (^{137}Cs, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am). Radionuclide concentrations for fish mussel tissue ranged from non-detectable to 4,340 mBq kg^{-1} wet weight, with the following means for Farallon fishes: ^{137}Cs = 1,110 mBq kg^{-1}; ^{238}Pu = 390 mBq kg^{-1}; $^{239+240}$Pu = 130 mBq kg^{-1}; and ^{241}Am = 1,350 mBq kg^{-1}. There were no statistically significant differences in the radionuclide concentrations observed in samples from the Farallon Islands compared to reference samples from Point Arena, CA. Concentrations of both ^{238}Pu and ^{241}Am in fish tissues (from both sites) were notably higher than those reported in literature from any other sites worldwide, including potentially contaminated sites. Concentrations of $^{239+240}$Pu from both sites were typical of low values found at some contaminated sites worldwide. These results show ~10 times higher concentrations of $^{239+240}$Pu and ~40-50 times higher concentrations of ^{238}Pu than those values reported for identical fish species from 1977 collections at the FINWDS.

Radionuclide concentrations were converted to a hypothetical per capita annual radionuclide intake for adults, yielding the following values of annual Committed Effective Dose Equivalent (CEDE) from ionizing radiation emitted from these radionuclides: 0.000 mSv y^{-1} for ^{137}Cs, 0.009 mSv y^{-1} for ^{238}Pu, and 0.003 mSv y^{-1} for $^{239+240}$Pu. For ^{241}Am, projected CEDE for Dover sole, sablefish, and thornyheads were higher, averaging 0.03 mSv y^{-1}. The observed isotopic ratio of ^{238}Pu/$^{239+240}$Pu was about 4 (which is two orders of magnitude higher than the ratio of 0.03 associated with fallout from weapons tests and accidental releases in the north temperate zone of the earth), indicating a considerably higher environmental mobilization for ^{238}Pu compared to $^{239+240}$Pu. Likewise, the observed ratio of ^{241}Am/$^{239+240}$Pu of about 30 was nearly two orders of magnitude higher than the fallout ratio of 0.43 in the north temperate zone of the earth. The projected ionizing radiation CEDE to people from the ingestion of fish with fallout radionuclides was three times higher for ^{241}Am than from the plutonium isotopes.
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Key words: ^{137}Cs; ^{238}Pu; ^{239}Pu; fallout

INTRODUCTION

Background

Marine communities worldwide are subjected to a wide range of anthropogenic pollutants, including radionuclides (Suchanek 1994). Between 1946 and 1970 the Farallon Islands Nuclear Waste Dump Site (FINWDS), ~40 km SW of San Francisco, received at least 47,500 barrels of radioactive waste [reportedly low-level radioactive waste, but see Davis (1980a, b) for an alternative opinion], with a radionuclide inventory of at least 500 TBq, excluding tritium (Noshkin et al. 1978). Most of the radioactive wastes deposited at the FINWDS are believed to be contained in recycled 55-gallon 16-gauge steel drums, deposited either individually or in clusters of barrels at three primary sites with the following approximate depths (A = 100 m, B = 900 m, and C = 1,800 m; Fig. 1), although the present day location of the barrels is more widely distributed (Karl et al. 1991). Historical records indicate that approximately 150 barrels were deposited at the 100-m site (A); 3,600 barrels at the 900-m site (B); and 44,000 barrels at the 1,800-m site (C). The polygon shown in Fig. 1 probably more accurately describes the actual distribution of most of the waste containers.

The life expectancy of these metal waste containers (to effectively contain wastes) has been calculated to be ~10 y in seawater, whereas the life expectancy of the concrete enclosing the radioactive wastes has been calculated at ~30 y (Waldichuk 1960); but the important

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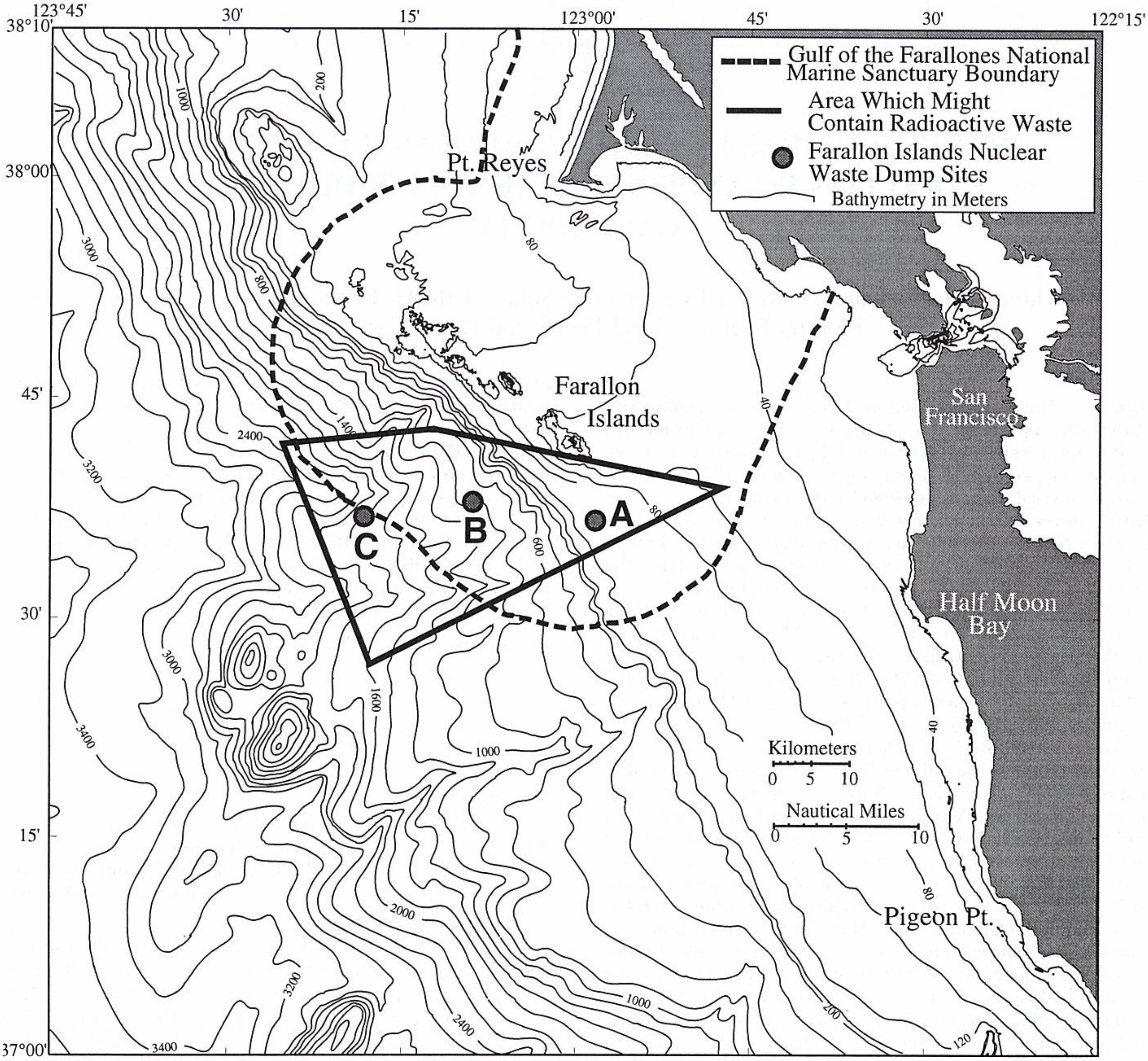

Fig. 1. Transverse Mercator Projection of the Farallon Islands Nuclear Waste Dump Site. A = 300 m dump site, B = 900 m dump site, C = 1,800 m dump site. Collections for this study were taken from the 900 m dump site.

radionuclides contained within the barrels have half-lives ranging from 30 to 24,065 y. However, no *in-situ* testing of these barrels has been conducted under deep ocean water pressures, which might further reduce barrel integrity. During the 1970's, an extremely small proportion (0.34%) of the waste barrels was examined by deep-sea photography, indicating that a substantial number of barrels (25-30%) had been breached, presumably from implosion (Dyer 1976). Because these barrels have been in the deep sea for 25-50 y, even intact barrels are now likely reaching their functional life span. Therefore, radionuclides originally contained in these barrels may be deposited in or on nearby sediments or they may already have been widely dispersed by ocean currents.

The primary focus of the study was to determine whether radionuclides concentrated in tissues typically consumed by the public posed a potential threat to human health. The organisms chosen for study included Dover sole (*Microstomus pacificus*), sablefish (*Anoplopoma fimbria*), thornyheads (*Sebastolobus* spp.), and the California mussel (*Mytilus californianus*). Field collections were conducted in 1986 and 1987 and subsequent radionuclide analyses continued through 1991 (Suchanek and Lagunas-Solar 1991, 1993).

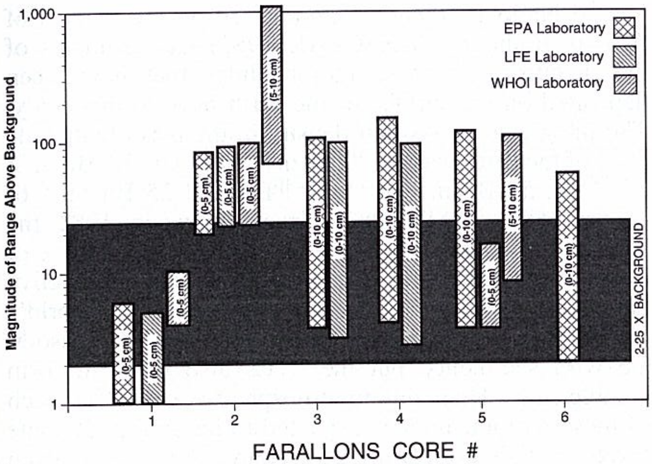

Fig. 2. "Magnitude Above Background" concentrations of radionuclides from FINWDS core samples analyzed by three different analytical laboratories (EPA, LFE, and WHOI) as reported by Dyer (1976). Vertical bars represent a range of magnitude for core sediment data above estimated global fallout levels. Shaded region (2-25 X background) represents magnitude above background reported by Dyer (1976). Values in parentheses indicate depth of core section analyzed.

Previous data on radionuclides in sediments and biota at the FINWDS

From a 1974 study at the 900 m site, Dyer (1976) reported that sediment radionuclide concentrations ranged from 2-25 times expected background levels. However, a recalculation of Dyer's data shows that for $^{239+240}$Pu, the actual concentrations ranged up to 1,064 times background (see Suchanek 1987 and Fig. 2). Another report on sediments from near the waste barrels estimated the range of $^{239+240}$Pu concentrations at 8 to 2,208 times expected background (LFE 1979; and further interpretation by Davis 1980b). From a human health perspective, there was concern that commercially exploited deep-sea fishes (e.g., channel rockfish, Dover sole, sablefish) may be contaminated with short- and/or long-lived radionuclides (e.g., ^{137}Cs, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am) with half-lives ranging from 30 y (for ^{137}Cs) to 24,065 y (for ^{239}Pu).

Several species of marine organisms have been observed and/or collected from the immediate vicinity of the waste containers. These include invertebrates such as vasiform sponges, brittle stars, polychaetes, sea pens, squid, sea cucumbers, anemones, snails, shrimp, and crabs as well as fishes including sablefish, thornyheads, Dover sole, deep-sea sole, Pacific flatnose, Pacific rattail, Pacific sanddab, lanternfish, loosejaw, eared blacksmelt, midshipman, rock sole, hake, deep-sea smelt, and two-line eelpout (Dyer 1976; Schell and Sugai 1980). Fig. 3 provides a considerably simplified food web diagram constructed from available literature and data (Allen 1982, 1990; MMS 1987; Cailliet et al. 1988; Haugen 1990[#] that indicates the connectedness between various benthic invertebrate species near the FINWDS, some of their predators and the trophic linkage of these species to commercially important fishes and humans.

Some fishes and invertebrates from this region were found to contain elevated levels of radioactivity, and some fishes were believed to have radionuclide concentrations of 10 to 8,500 times expected background levels (Schell and Sugai 1980; Davis 1980b). These studies showed that the organisms with the highest radionuclide concentrations (i.e., >100 pCi kg^{-1} = 3,700 mBq kg^{-1} dry wt) were invertebrates (= polychaete worms, sea cucumbers, sponges) and fishes (= deep-sea smelt, Dover sole, hake, lantern fish, midshipman, Pacific flatnose, rattail fish, and Pacific flatnose).

Because mussels are efficient filter feeders, they have been used effectively in statewide, nationwide, and worldwide programs to monitor pollutant levels in the natural environment. Mussels have been found to concentrate radionuclides about 200 to 300 times the level found in surrounding seawater. The Farallon Islands is one of many California sites where national pollutant levels in intertidal mussels have been compared through the Mussel Watch Program (Goldberg et al. 1978). The U.S. EPA reported that some radionuclides in mussel tissues (visceral mass only) from Southeast Farallon Island were roughly 3.3 times higher than the mean concentration found at all other California sites. Mussel samples from this site had mean dry weight radionuclide concentration, for $^{239+240}$Pu of 130 $\pm$ 5 mBq kg^{-1} compared with a mean of 37 $\pm$ 25 mBq kg^{-1} for all other California sites (n = 19, range = 5-77 mBq kg^{-1}). Mean concentrations for ^{241}Am were 330 $\pm$ 25 mBq kg^{-1} compared with 99 $\pm$ 109 mBq kg^{-1} for all other California sites (n = 17, range = 1.5-290 mBq kg^{-1}).

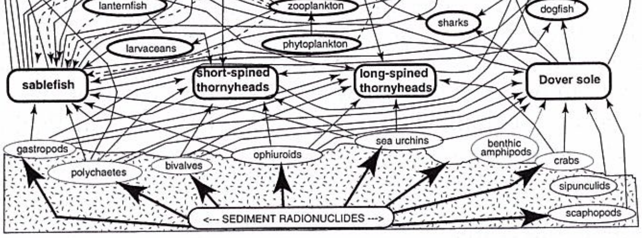

Fig. 3. Simplified food web for fish species analyzed for radionuclides from the Farallon Islands Nuclear Waste Dump Site, emphasizing the connectedness to other trophic levels. Dashed lines represent connections for larvae or juveniles and solid lines represent connections for adults.

[#] P. Adams. Personal Communication. NationalOceanic and Atmospheric Administration, 1995.

Ecology of bottom-feeding fishes sampled

Three taxa of fishes (Dover sole, sablefish, and thornyheads) were collected in this study because of their direct applicability to potential human health risk through consumption. The following information provides natural history data that are relevant to each of these taxa.

Dover sole (*Microstomus pacificus*) is a bottom-feeding species that generally occurs on muddy substrata and ranges from about 55-1,100 m depth. Its larvae have been collected to ~450 km offshore, yet it is not known as a widely migrating species. The Dover sole does, however, undergo extensive seasonal onshore/offshore movements related to its spawning cycle. Adults exhibit limited coastwise movement and several isolated sub-populations are believed to exist (Frey 1971). During spring through summer it typically feeds extensively in inshore waters; during November to March it moves offshore for spawning, where it produces buoyant pelagic eggs. This species feeds exclusively on benthic invertebrates including bivalves, scaphopods, sipunculids, polychaetes, echinoids, ophiuroids, gastropods, and crustaceans (Frey 1971).

Sablefish (*Anoplopoma fimbria*) prefer soft bottom habitats like the Dover sole and are found at depths greater than about 100 m. They are not known to migrate for spawning purposes, but migration is likely important in this species, as one individual tagged in Japan was later found in the United States (Frey 1971). It is believed that sablefish from central to southern California represent a single stock population. Spawning generally occurs from December to April with a peak in January/February. Juveniles are known to feed on the following benthic invertebrates: copepods, amphipods, euphausids, fish eggs, fish larvae and the larvacean *Oikopleura*. Subadults and adults are generally known to feed on euphausids, tunicates and fish, especially anchovy (Frey 1971).

Thornyheads (also called idiot-fish or channel rock-fish) (shortspine = *Sebastolobus alascanus*; longspine = *S. altivelis*) are non-migratory deep-water species that are generally known to range from 550-1,600 m depth although they likely occur deeper (Phillips 1957; Frey 1971; Wakefield and Smith 1990). These species spawn gelatinous egg masses that float to the surface; this process (Wakefield and Smith 1990) could represent a significant conduit for potentially contaminated deep-sea particulate organic material to reach upper regions of the water column and higher trophic levels. Both species are known to feed on numerous benthic invertebrates and the shortspine has been shown to prey on the longspine (see Fig. 3).

Background occurrence of ^{137}Cs, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am

The world's oceans contain background levels of ^{137}Cs, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am as a result of worldwide fallout from nuclear weapons tests and accidental releases of these radionuclides to the atmosphere. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 1982) has estimates of the quantities of these radionuclides that have been deposited on the surface of the earth prior to this study. The integrated deposition density in the north temperate zone of the earth was 5,170 Bq m^{-2} of ^{137}Cs, 1.5 Bq m^{-2} of ^{238}Pu, 58 Bq m^{-2} of $^{239+240}$Pu, and 25 Bq m^{-2} of ^{241}Am (UNSCEAR 1982). In addition, in 1982 the effective deposition of ^{241}Am was growing by the rate of about 1 Bq m^{-2} per year because of the radioactive decay of 730 Bq m^{-2} of deposited ^{241}Pu. In the world's oceans these radionuclides might be expected to associate with sediments, but the ^{137}Cs also tends to form soluble salts. Thus, due to atmospheric deposition, each of these radionuclides is expected to be present at some levels in fish even without a source originating from nuclear dump leakage. However, the intention was to compare the levels of such radionuclides observed in fish obtained from the FINWDS with a reference site (see below) in order to evaluate the significance of the measured levels.

MATERIALS AND METHODS

Selection of a reference site

The location of the reference site was chosen on the basis of the best available data on water movement in the FINWDS region in order to provide the least potential for reference site contamination. Although the directionality and speed of currents in the Gulf of the Farallons are seasonally variable, the largest annual mass movement of water from the vicinity of the FINWDS is southward (Conomos et al. 1971; Conomos 1975; Conomos and Peterson 1977). Surface currents from FINWDS generally show significant northward and/or southward movement along the coast, whereas bottom currents are more complex (Conomos et al. 1971; Conomos 1975; D. Lindberg;** Dyer 1976; Conomos and Peterson 1977). One current meter placed on the bottom at the 1,829 m site during 1975 showed essentially northward bottom current movement at speeds of approximately 1.17 km d^{-1} (Dyer 1976; Crabbs 1983). Bottom currents moving in this direction would likely transport particles toward the vicinity of Cordell Bank, a region used extensively for commercial and sport fisheries. However, another bottom current study (using seabed drifters released in the Gulf of the Farallons) indicated consistent eastward movement of bottom currents at speeds of at least 0.5 km d^{-1} (Conomos et al. 1971; McCulloch et al. 1970; Conomos and Peterson 1977). Significantly, these currents move particles eastward along the sea floor, with final destinations in San Francisco Bay and San Pablo Bay. Therefore, a reference location was chosen at Point Arena, ~100 km north of the FINWDS.

Field collections

Fish and mussel collections were made during three different seasons: (1) winter = December 1986/January

**Personal communication, University of California, Berkeley, CA, 1994.

1987, (2) summer = May/June 1987, and (3) fall = August/September 1987. Deep-sea fishes were collected using commercial fishing trawlers (typical trawling period = 1-4 h) in the vicinity of the 900 m depth FINWDS dump site and from a comparable depth off the Point Arena coast. Individuals of each target species were selected haphazardly (but included a range of sizes) by hand from hauls that typically contained several tons of material. After collection, fish were immediately iced and placed in polyethylene plastic bags on board the vessel (1-2 d holding time). After return to port they were preserved by freezing (−20°C) until dissection. California mussels were collected intertidally by hand from Southeast Farallon Island and from a reference intertidal site at the base of the Point Arena Lighthouse. Mussels were placed in polyethylene bags and held on ice during transit; they were subsequently frozen whole at −20°C until dissection.

Sample preparation: Dissection, homogenization, lyophilization, and preservation

Fishes were sexed, weighed, and measured for total length, fork length, and standard length. The following organs/tissues were dissected and weighed for each fish: fillets (muscle tissue only), skin, liver, gonads, kidney, otoliths, and gastrointestinal tract. Only fillets were analyzed for radionuclides. Mussels were dissected using methods from Goldberg et al. (1983). Mussel shells were measured for total length, width, and height. Wet weight of the shell, byssal threads (if still attached), and composite viscera were recorded. Remaining organs/tissues for mussels and fishes (as well as the remainder of the fish fillets and carcasses, including the bones) remain archived in a frozen state (−20°C).

Each fish or mussel tissue sample (~750-1,200 g wet wt) was placed in a stainless steel Waring™†† blender and homogenized. Double-distilled deionized water was added to some samples in order to achieve a smooth consistency in the homogenate. This homogenate slurry was then poured into aluminum foil "pans," flash frozen (at −50°C) and lyophilized for about 4-5 d in a Virtis™‡‡ freeze-drier to form a thick wafer of tissue. Water, which originated from the tissues and had accumulated in the freeze-drier during the lyophilization process, was analyzed and no above-background alpha- or gamma-ray activity was detected in these samples. Wet to freeze-dried weight ratio approximated 10:1 but varied among samples. Samples (30-40 g) of lyophilized fish or mussel tissues were compressed into a geometry suitable for gamma-ray spectrometry. In order to prevent decomposition of samples during a long storage period, the samples were sterilized by gamma radiation (35-40 kGy).

^{137}Cs gamma spectrometry

Quantification of ^{137}Cs in fish samples was performed by measurement of gamma emissions (for 20 h each) from the lyophilized samples using scintillation pulse-height analysis of the 0.662 MeV gamma photon emitted by the short-lived ^{137m}Ba progeny, with two opposing 10.2 cm × 20.3 cm NaI(Tl) crystals§§ with near 4π geometry. The gamma detectors were located within a shielded enclosure whose outer shell was a 12.7-cm-thick steel cylinder made of pre-atomic-era armor plating, with one end closed by a 15-cm-thick steel plate and the other by two 23-cm-thick steel doors. A lead-cadmium-copper inner lining served as an additional shield to further reduce background radiation levels.

In addition to ^{137}Cs, natural ^{40}K was measured with its 1.461 MeV gamma photon. The net count rate associated with ^{137}Cs+Ba was calculated by subtracting background counts in both the ^{137}Cs-Ba channels and the ^{40}K channels and subtracting the crossover count rate from the ^{137}Cs-Ba count rate. The minimum detectable activity for this system was 20 mBq per sample with about 30% counting efficiency. The normal approximation for Poisson statistics and cross-over correction factors were used to calculate the variance associated with each observed count rate, the variance associated with the calculated net ^{137}Cs count rate, and the variance associated with the measured ^{137}Cs concentration in each sample. Calibrations were done with standards traceable to the National Institute of Standards and Technology (NIST).

^{238}Pu, $^{239+240}$Pu, and ^{241}Am alpha spectrometry

Approximately 40 g of each lyophilized tissue sample was first reconstituted with doubly distilled water to a manageable consistency and spiked with radiotracers (approximately 5 dpm of ^{243}Am and 5 dpm of ^{242}Pu from NIST referenced solutions)‖‖. Samples were wet ashed using concentrated HNO_3 at 70°C. Ashing was continued in a muffle furnace at increasing temperatures up to 500°C for up to ~24 h. The reduction of fresh (lyophilized) sample to ash weight was approximately 10:1.

The radiochemical methods used to separate americium and plutonium from fish and mussel samples were adapted from Singh et al. (1984) and Boyd et al. (1979). Americium, thorium, plutonium, and uranium were coprecipitated with $Fe(OH)_3$. Plutonium, uranium, and americium were separated by solvent extraction using tri-lauryl amine (TLA) (also known as tridodecylamine). Plutonium and uranium were extracted using 1.0 M $NaNO_2$ and the plutonium was further separated using 0.05 M NH_4I. Plutonium was further purified by solvent extraction using ether. Americium was separated using ether solvent extraction and anion exchange procedures using AG/MP-1 anion exchange resin. Plutonium and americium from each sample were then separately electrodeposited onto a 2.54 cm OD stainless steel planchet using methodologies adapted from Singh et al. (1984).

Radioassays of ^{238}Pu (87.74 y), $^{239+240}$Pu (24,000 y; 6,600 y), and ^{241}Am (432 y) were performed for 60 to 800 min in a vacuum using alpha spectrometry with

†† Waring, Model CB-4, Winsted, CT.

‡‡ Virtis, Model 50-SRC, Gardiner, NY.

§§ Harshaw Chemical Co., Cleveland, OH.

‖‖ ^{243}Am [7.4×10^3 y; alpha's 5.28 MeV (88%), 5.23 MeV (11%); measured as approximately 5.27 MeV (100%)] and ^{242}Pu [3.8×10^5 y; alpha's 4.90 MeV (74%), 4.86 MeV (26%); measured as approximately 4.89 MeV (100%)].

Table 1. Measured mean radionuclide concentrations (mBq kg^{-1} ± SE) in fish and mussel tissues. Sample sizes in parentheses.

	Farallon Islands	Pt. Arena	Pooled means
^{137}Cs			
mussels	1,010 ± 321 (8)	817 ± 167 (6)	926 ± 275 (14)
Dover sole	742 ± 221 (9)	711 ± 242 (8)	727 ± 224 (17)
sablefish	1,050 ± 260 (9)	1,530 ± 751 (9)	1,290 ± 599 (18)
thornyheads	1,220 ± 158 (8)	1,290 ± 266 (8)	1,260 ± 214 (16)
^{238}Pu			
mussels	16 ± na (1)	nd	16 ± na (1)
Dover sole	572 ± 945 (3)	141 ± 105 (6)	284 ± 526 (9)
sablefish	204 ± 20 (2)	186 ± 293 (4)	192 ± 227 (6)
thornyheads	330 ± 529 (3)	49 ± 44 (3)	190 ± 370 (6)
$^{239+240}Pu$			
mussels	nd	nd	nd
Dover sole	228 ± 199 (3)	232 ± 488 (6)	231 ± 398 (9)
sablefish	45 ± 26 (2)	116 ± na (1)	69 ± 45 (3)
thornyheads	44 ± 59 (3)	20 ± 14 (3)	32 ± 41 (6)
^{241}Am			
mussels	6 ± na (1)	126 ± 88 (2)	86 ± 93 (3)
Dover sole	1,630 ± 2,360 (3)	1,320 ± 1,560 (6)	1,420 ± 1,710 (9)
sablefish	2,850 ± na (1)	1,000 ± 1,400 (5)	1,310 ± 1,470 (6)
thornyheads	186 ± 35 (2)	299 ± 157 (3)	254 ± 128 (5)

surface barrier detectors coupled to a 4,096-multichannel analyzer.¶¶ The surface barrier detectors were calibrated using alpha standards referenced to the NIST. The detector efficiencies ranged from 37.2% (detector A) to 35.6% (detector B). The uncertainties for the alpha radioassays were estimated from the maximum individual uncertainties (errors) using the root-mean-square method. The individual uncertainties included: *Fixed Sources* = sample dry weight (± 1%); timing errors (± 1%); detector efficiencies (± 3%); source-detector geometry variations (± 3%); sample self-absorption errors (estimated) (± 15%); peak fitting errors (± 8%); secondary references (± 5%); natural background subtractions (± 5%); and *Variable (Sample Dependent) Sources* = radiotracer count rate uncertainties; unknown count rate uncertainties; and radiochemistry yield (spike recovery) errors. The alpha counting protocol included periodic background counting, alpha energy resolution tests, and detector efficiency measurements.

In order to determine the specific radionuclide concentration for each treatment group of fishes and mussels, the total activity (mBq kg^{-1} in freeze-dried wt) for each radionuclide was divided by the wet:dry ratio (a function of the amount of water removed during lyophilization) to obtain the extrapolated radionuclide concentration for wet weight tissues (mBq kg^{-1} wet wt). All results and calculations are reported as wet weight values.

¶¶ ^{241}Am [5.49 MeV (86%), 5.44 MeV (13%); measured as approximately 5.48 MeV (100%)]; ^{238}Pu [5.50 MeV (71.1%), 5.46 MeV (27.8%); measured as approximately 5.49 MeV (100%)]; and $^{239,240}Pu$ [5.16 MeV (73.3%), 5.15 MeV (15.1%), 5.10 MeV (11.5%); and 5.16 MeV (75.5%) and 5.12 MeV (24.4%)]; measured combined as approximately 5.15 MeV (100%)].

RESULTS

Radiochemical and electroplating yields

Radiochemical yields were based on the recovery of radiotracer spikes (^{243}Am and ^{242}Pu) which were added after reconstituting the sample water, but prior to the radiochemical process. The plutonium radiochemical yields averaged 19% for Dover sole samples (range 2-58%, n = 10); 14% for sablefish samples (range 3-28%, n = 9); 27% for thornyhead samples (range 11-65%, n = 6); and 17% for mussel samples (range 16.6-17.4%, n = 2). For americium, the radiochemical yields averaged 13% for Dover sole samples (range 1.4-40%; n = 10), 9% for sablefish samples (range 1-35%, n = 6), 9% for thornyhead samples (range 2-17%, n = 5) and 21% for mussels (range 6-51%, n = 3). Average electroplating yields for plutonium were measured at 61% while americium yields were 40%. The average overall radiochemical yields for plutonium and americium in this study were 20% and 14%, respectively.

Radionuclide concentrations

The radionuclide concentrations (mBq kg^{-1} wet wt) in fish and mussel tissues from both the FINWDS and the Point Arena region are presented in Table 1 and plotted in Fig. 4. In general, most of the variances of sample group means were unequal (Bartlett's test). Therefore, non-parametric statistics (Wilcoxon Test) and Welch's ANOVA (allowing unequal variances) were used to analyze differences between group means. However,

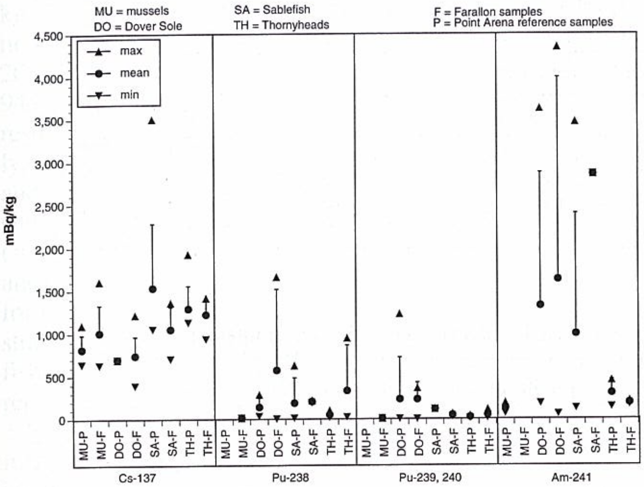

Fig. 4. Total radioactivity (mBq kg^{-1} wet wt) measured in mussel and fish tissues from the Farallon Islands Nuclear Waste Dump Site and from Point Arena. Vertical bars represent +1 standard deviation.

without exception, results obtained using both standard parametric and non-parametric tests were consistent.##

On a gross level (all samples pooled for each radionuclide), there were significant differences between the concentrations of the four radionuclides analyzed in fishes and mussels (Welch's ANOVA). Variances were unequal between the distributions of the four radionuclides. Further analysis using a Tukey-Kramer Multiple Comparisons Test showed that for comparisons between ^{137}Cs and ^{241}Am and between ^{238}Pu and $^{239+240}$Pu there were no significant differences between radionuclides within each of those comparisons, but there was a highly significant difference between the first and second pair. Thus, for the pooled tissue data, there were significantly higher concentrations of ^{137}Cs and ^{241}Am compared to ^{238}Pu and $^{239+240}$Pu.

Gamma ray results. Results of ^{137}Cs radioassays are shown in Fig. 4 and summarized in Table 1. For all three fish species and the mussel species, no statistically significant differences were found for ^{137}Cs concentrations between the Farallons dump site and the Point Arena comparison site ($p > 0.05$). In addition, each species was tested for potential seasonal differences, but none was found. For data pooled by species, mean values for ^{137}Cs in muscle tissue of Dover sole (727 mBq kg^{-1}) were statistically different ($p < 0.001$) and were nearly half the concentrations found in sablefish (1,290 mBq kg^{-1}) or thornyheads (1,260 mBq kg^{-1}).

Alpha particle results. The results of the alpha radioassays (for ^{238}Pu, $^{239+240}$Pu, and ^{241}Am) showed a high degree of variability in all sample groups (see Table 1 and Fig. 4). This variability reflects the various procedural uncertainties and sample variability such as the high fat content exhibited in most of the fish tissues, since fat probably has lower concentrations of radionuclides. Using both parametric and non-parametric tests (for individual species and for pooled fish data), there were no statistically significant differences ($p > 0.05$) in the level of alpha emitting radionuclides between samples originating from the Farallon Islands Nuclear Waste Dump Site and those from the Point Arena reference site. There were also no significant differences for any species between the three different seasons (winter, summer, and fall) during which samples were collected.

Radiation doses from eating fish containing ^{137}Cs, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am

The International Commission on Radiological Protection (ICRP) provides estimates of (1) committed equivalent dose conversion factors (Sv per Bq to age 70 y) for all major organ systems and (2) the whole-body Committed Effective Dose Equivalent (CEDE) for ingestion of ^{137}Cs, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am by children of various ages or adults as summarized in Table 2 (ICRP 1990). From these dose conversion factors it is possible to estimate the annual dose commitment to age 70 y (or to 50 y after ingestion for adults) from the regular consumption of fish from the Farallon Islands using the average concentrations of the radionuclides observed in the samples from this study and assuming the ingestion of 0.36 of fish per kg body weight per year equivalent to about 25 kg fish per adult as estimated by the World Health Organization for a Mediterranean diet (WHO 1988).

Mean annual CEDE values are given in Table 3 and individual values are shown in Fig. 5. These calculated doses are relatively small, with the highest total dose being about 0.05 mSv, an added amount of approximately 1.5% of typical annual doses (about 3.6 mSv). The primary calculated exposure dose results from ingestion of the three alpha radionuclides, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am, estimated here as about 0.01-0.05 mSv per year of exposure. For comparison, a round trip transcontinental commercial jet airline flight from San Francisco to New York City results in an elevated exposure from cosmic radiation of about 0.05 mSv.

The NCRP (1987) also provides recommendation limits for exposure to non-medical man-made sources of radioactivity. Non-occupational exposure limits (for members of the general public) are categorized into three ranges: (1) Infrequent Exposure: Annual effective dose equivalent not to exceed 500 mrem (= 5 mSv); (2) Continuous (= Frequent) Exposure: Annual effective dose equivalent not to exceed 100 mrem (= 1 mSv); (3) Negligible Individual Risk Level (NIRL): Annual effective dose equivalent below 1 mrem (= 0.01 mSv). According to the NCRP (1987) the NIRL is defined as: "a level of average annual excess risk of fatal health effects attributable to irradiation, below which further effort to reduce radiation exposure to the individual is unwarranted."

Parametric and non-parametric statistical analyses were performed using JMP (SAS Institute, Inc.) for Macintosh(TM).

Table 2. Dose conversion factors by exposure age for whole body effective dose equivalent (Sv/Bq) committed to age 70 for ingestion of ^{137}Cs, ^{238}Pu, $^{239+240}Pu$, and ^{241}Am from ICRP (1990).

Radionuclide	Sv/Bq Given By Age At Intake					
	3 mo	1 y	5 y	10 y	15 y	adult
^{137}Cs	2.0×10^{-8}	1.1×10^{-8}	9.0×10^{-9}	9.8×10^{-9}	1.4×10^{-8}	1.3×10^{-8}
^{238}Pu	1.3×10^{-5}	1.2×10^{-6}	1.0×10^{-6}	8.8×10^{-7}	8.7×10^{-7}	8.8×10^{-7}
$^{239+240}Pu$	1.4×10^{-5}	1.4×10^{-6}	1.1×10^{-6}	1.0×10^{-6}	9.8×10^{-7}	9.7×10^{-7}
^{241}Am	1.2×10^{-5}	1.2×10^{-6}	1.0×10^{-6}	9.0×10^{-7}	9.1×10^{-7}	8.9×10^{-7}

Table 3. Calculated whole body committed effective dose equivalent (CEDE, mSv) to age 70 by age at intake for annual ingestion of 0.36 kg of fish per kg of human body weight with mean concentrations of ^{137}Cs, ^{238}Pu, $^{239+240}Pu$, and ^{241}Am assuming consumption of fish from the Farallon Islands (using data in Table 1 and Table 2).

Radionuclide	Annual CEDE (mSv) Given By Age At Intake					
	3 mo	1 y	5 y	10 y	15 y	adult
^{137}Cs	0.000	0.000	0.000	0.000	0.000	0.000
^{238}Pu	0.012	0.002	0.003	0.005	0.008	0.010
$^{239+240}Pu$	0.003	0.001	0.001	0.001	0.002	0.003
^{241}Am	0.038	0.006	0.009	0.014	0.024	0.030
Total	0.053	0.009	0.013	0.020	0.034	0.043

It is apparent that most of the projected CEDE values for ^{137}Cs, ^{238}Pu, and $^{239+240}Pu$ in Fig. 5 fall below the NIRL. For ^{241}Am the projections are more variable. Based on pooled fish data, predicted mean CEDE values exceed the NIRL for a number of cases, most notably for ^{238}Pu in Farallons samples, and for ^{241}Am for both Farallons and Point Arena samples. For individual species, Dover sole registered both the highest total radioactivity (4,340 mBq kg^{-1} for ^{241}Am) in muscle tissue as well as the highest predicted mean CEDE values calculated at 0.122 mSv for exposure to a 3-mo-old child (on a fish diet).

DISCUSSION

A selected subset of collected fish and mussel samples was analyzed to draw conclusions regarding (1) potential significant differences between the FINWDS and the comparison site at Point Arena, and (2) the projected radiation dosage that would be expected from human consumption of these species. Low sample numbers lowered the statistical power of the tests. Also, when evaluating these results, consideration should be given to the type of tissues being analyzed. For fish, analyses were restricted to muscle tissue (fillets) only. Other work, however, has shown that skin tissue often contains considerably higher radionuclide concentrations than does muscle tissue (see Schell and Sugai 1980). Because many ethnic groups consume other fish tissues besides muscle (including skin and bone), the use of only fish fillet tissues for assessing human health risks may be misleading in that it provides an incomplete picture of total potential exposure.

Radionuclide concentrations

Concentrations of ^{137}Cs in fish muscle tissues (~500 to 1,500 mBq kg^{-1}) are generally within the range of values found in most other non-contaminated regions (e.g., Suzuki et al. 1973), yet considerably lower than values found at contaminated sites such as Enewetak and Bikini Atolls in the Pacific Testing Grounds (e.g., Noshkin et al. 1980, 1981) or near the Windscale Nuclear Plant (e.g., Pentreath and Jefferies 1971; Pentreath 1973).

Considerable data from the literature exist on fish muscle tissue concentrations of $^{239+240}Pu$, but little was found for ^{238}Pu. These results indicate that concentrations of ^{238}Pu and $^{239+240}Pu$ in Farallons and Point Arena fish were very similar, with means ranging from ~20-230 mBq

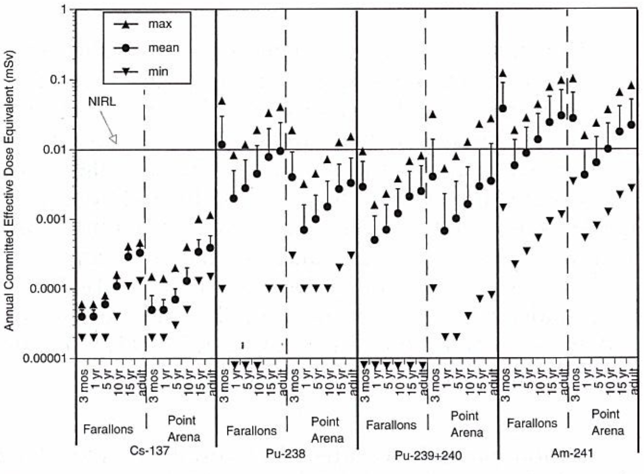

Fig. 5. Whole body committed effective dose equivalents for pooled fish values (pooled values for Dover sole, sablefish and thornyheads from Table 1) committed to age 70 by age at ingestion. The Negligible Individual Risk Level (NIRL) is shown at 0.01 mSv y^{-1} (NCRP 1987). Vertical bars represent +1 standard deviation. Note log scale.

kg^{-1} (see Fig. 4). Results for ^{238}Pu (from 1986/87 collections), however, yield mean values of approximately 572, 204, and 330 mBq kg^{-1} (with maxima of 1,660, 218, and 944 mBq kg^{-1}) for Dover sole, sablefish, and thornyheads, respectively. Interestingly, Schell and Sugai (1980) analyzed some of the same species (i.e., Dover sole, sablefish, and thornyheads) from a 1977 collection near the Farallons site for ^{238}Pu and found values of < 7.4 mBq kg^{-1} dry wt (= < 0.74 mBq kg^{-1} wet wt) for Dover sole and sablefish and < 11.1 mBq kg^{-1} dry wt (= < 1.1 mBq kg^{-1} wet wt) for thornyheads. Could the 40-50-fold higher values in this study indicate a real increase in the levels of ^{238}Pu in these fishes during the period from 1977 to 1986? Curtis (1988) reported wet weight values for ^{238}Pu in fish muscle tissue of ~63 mBq kg^{-1} from a Boston seafood market and ~303 mBq kg^{-1} from a San Francisco seafood market (possibly including some fish from near the Farallons). The only other value found for ^{238}Pu in fishes was from Aarkrog (1971; Noshkin 1985), who reported values of ~259 mBq kg^{-1} for ^{238}Pu from a single Greenland halibut near the site of a nuclear weapons incident resulting from a B-52 crash at Thule, Greenland. Therefore, these results yield the highest concentrations of ^{238}Pu reported in fish tissues for any sites worldwide, including some potentially contaminated sites.

Concentrations of $^{239+240}Pu$ are much more commonly reported, with the results of many studies summarized in Noshkin (1985). Those studies yielded widely varying results, with concentrations of $^{239+240}Pu$ in fish muscle tissues from generally non-contaminated sites ranging from ca. 0.03 to 8.25 mBq kg^{-1}. Curtis (1988) reported on the results of a 1981-1982 monitoring program of seafood markets in Boston, Atlantic City, and San Francisco for $^{239+240}Pu$ in fish muscle tissues and found no values over the minimum detectable limit of 3.7 mBq kg^{-1}. On the other hand, many other studies from various radioactively contaminated sites yielded typical concentrations of $^{239+240}Pu$ in fish muscle tissues of ~30-3000 mBq kg^{-1}, up to a maximum of ~10,000 mBq kg^{-1} from Enewetak Atoll in Micronesia, the site of post World War II atom and hydrogen bomb testing (Noshkin et al. 1976). From their 1977 fish collections at the FINWDS, Schell and Sugai (1980) found concentrations of $^{239+240}Pu$ ranging from about 3.7 to 22.2 mBq kg^{-1}. Therefore, these results for $^{239+240}Pu$ from 1986 collections, with mean concentrations of ~32-231 mBq kg^{-1}, are approximately 10 times higher than Schell and Sugai's results and are representative of typical low end values for radioactively contaminated sites.

The radionuclide with the highest concentration in fish tissue in this study was ^{241}Am. The limited literature on ^{241}Am in marine fishes indicates that concentrations found in this study (muscle tissue with mean values of 1,000 to 1,500 mBq kg^{-1}) are higher than those values for comparable tissue reported from the Pacific Testing Grounds at Enewetak, Bikini, and Kwajalein Atolls in Micronesia (Schell and Watters 1975; Noshkin et al. 1980) and from the Irish Sea near the Windscale Nuclear Plant (Pentreath and Lovett 1976, 1978), none of which exceed approximately 740 mBq kg^{-1}.

The ^{238}Pu:$^{239+240}Pu$ ratio

The cumulative fallout of plutonium isotopes from weapons tests and accidental releases was inventoried in 1973 at 9,472±1,221 TBq (256±33 kCi) of ^{239}Pu and 340±41 TBq (9.2±1.1 kCi) of ^{238}Pu in the Northern Hemisphere (Hardy et al. 1973). Assuming no major additional inputs since 1986, radioactive decay would have reduced the ^{238}Pu in the Northern Hemisphere to 307±37 TBq (8.3±1.0 kCi). In studies using alpha spectroscopy ^{239}Pu values also include ^{240}Pu. From these values the average ratio of ^{238}Pu:$^{239+240}Pu$ is 307:9,470 = 0.03. The ratio of values given by UNSCEAR (1982) for the north temperate zone of the earth is 1.5:58 = 0.03. Therefore, the average ratio of ^{238}Pu:$^{239+240}Pu$ would be expected to be about 0.03. In this study the observed ratio of ^{238}Pu:$^{239+240}Pu$ was 3.5±0.7 in samples from Pt. Arena reference fishes and 4.2±1.2 in fish from the Farallon Islands. The obvious enrichment of ^{238}Pu in these samples (a factor of 120 times higher than expected from gross fallout) can be explained by the higher environmental mobility of ^{238}Pu over ^{239}Pu.

About one-third of environmental ^{238}Pu was released to the atmosphere as small particles of ^{238}Pu oxide from a satellite power source that burned re-entering the atmosphere after an aborted launch (Hardy et al. 1973). Raabe et al. (1973) have shown that fine particles of ^{238}Pu oxide exhibit about a 200 times faster dissolution rate at neutral pH than does ^{239}Pu oxide because of radiolytic fragmentation effects. The specific activity of ^{238}Pu oxide is about 200 times the specific activity of ^{239}Pu oxide. Also, Hakonson and Johnson (1973) found that there was a greater biological uptake of ^{238}Pu over ^{239}Pu in the fallout region of the Trinity atomic bomb test site in New Mexico 27 years after the test (since they used alpha spectroscopy their ^{239}Pu values included ^{240}Pu). They found ^{238}Pu:$^{239+240}Pu$ ratios of 0.05±0.01 in surface soil but a ratio of 2.3±0.5 in the livers of native rodents. This suggests that the vaporization/condensation of plutonium in an atomic bomb fireball may result in some separation of the plutonium isotopes facilitating the radiolytic fragmentation of ^{238}Pu. Also, these rodent liver plutonium ratios from the desert of New Mexico are remarkably similar to the isotopic ratios observed in fish in this study. Near the site of the Trinity test, Hakonson and Johnson (1973) found an average of about 400 mBq kg^{-1} of ^{239}Pu+^{238}Pu in rodent livers, a value that is less than that found for some fish samples in this study.

The ^{241}Am:$^{239+240}Pu$ ratio and the ^{137}Cs:$^{239+240}Pu$ ratio

Using the worldwide fallout values in the north temperate zone given by UNSCEAR (1982) of 58 Bq m^{-2} for $^{239+240}Pu$ and 25 Bq m^{-2} for ^{241}Am, the expected ratio of ^{241}Am:$^{239+240}Pu$ in environmental samples is 25:58 = 0.43. The observed mean ratio values were 55±24 for Pt. Arena fish samples and 13±4 for Farallon Islands fish samples. These results can be explained by the greater solubility of americium compounds compared to plutonium compounds in biological systems (Boyd et al. 1974; Raabe 1980), which would result in higher concentrations of americium than plutonium in biological tissues. Using the worldwide fallout values in the north temperate zone given by UNSCEAR (1982) of 5,170 Bq m^{-2} for ^{137}Cs, the expected ratio of ^{137}Cs:$^{239+240}Pu$ in environmental samples is 5,170:58 = 90. The observed mean ratio values were 62±21 in Pt. Arena fish samples and 47±21 in Farallon Islands fish samples, which are not too different from the expected ratio. Since cesium compounds tend to be water soluble and since cesium clears readily from living tissues, the observed comparability of ratios is probably fortuitous.

Other recent and potential future studies near the FINWDS

Other recent studies have also shown an abnormally high incidence (up to 38.6%) of pigmented skin tumors (chromatophoromas) and related hyperplastic lesions in five species of Pacific rockfish (*Sebastes* spp.) from Cordell Bank (Okihiro et al. 1993). The prevalence of pigmented lesions (neoplasia and hyperplasia), for at least one species (*S. flavidus*), had increased over a 5-y period from 1985-1990. In addition, a 1992 collection of sablefish from one of the three major dump sites at the FINWDS (Site B on Fig. 1) has revealed a 37% prevalence of similar chromatophore lesions (Okihiro[##]). Although lesions in sablefish were, on average, much smaller than those in rockfish, 7% were histologically classified as neoplasms (amelanotic melanophoromas). Although there is no direct evidence linking these skin lesions with materials deposited within the FINWDS, the high prevalence in both groups of fishes is certainly suggestive of exposure to xenobiotic or radioactive carcinogens.

With renewed interest in the potential that radionuclide contamination from the FINWDS may be affecting human health, congressional funds were appropriated in 1991 from both the National Oceanic and Atmospheric Administration (NOAA) and U.S. EPA to reevaluate the status of the dump site. The first stage in this reevaluation is an inventory search of records to determine the following as accurately as possible: (1) the contents of the waste barrels; (2) the radionuclide inventory, originally estimated to be ~500 TBq (exclusive of tritium, ^{3}H); and (3) the location of sites where barrels were believed to be deposited. While this is clearly a useful exercise, the essential elements needed to determine the current level of radionuclide contamination at the FINWDS and any subsequent potential human health risk are a complete survey of the physical and biological components at the site today.

Results indicate that for the fish species studied, no significantly elevated radionuclides were found at the Farallon Islands dump site in comparison to the Pt. Arena comparison site. This result could indicate a lack of contamination at the Farallons site. However, all the fish species tested are quite mobile, with seasonal or annual migration patterns moving on/off shore and/or along the coast with some species potentially migrating as far as Japan (see earlier discussion under natural history of fish species). This may provide an alternative explanation why they do not show significantly elevated radionuclide concentrations in the region of the FINWDS. Species, however, that spend a majority of their life cycles at or very near the dump site (e.g., benthic invertebrates), might be expected to show a clearer indication of site-specific radionuclide burdens. Many species of benthic invertebrates colonize sediments and remain within a relatively restricted zone throughout their life cycle (see Fig. 3). These organisms would likely represent a better choice to evaluate whether the local marine ecosystem in the proximity of the FINWDS is experiencing significant exposures from radionuclide contamination, and it is recommended that a more careful examination of the benthic invertebrate community be undertaken.

CONCLUSIONS

No statistically significant differences were observed for radionuclide concentrations in fish muscle tissue between the Farallon Islands Nuclear Waste Dump Site and the reference site at Point Arena. The lack of statistical significance may be due to (1) a lack of contamination at the FINWDS, (2) lowered statistical power due to low sample sizes, (3) high mobility of the fish species studied, and/or (4) movement of contaminated fishes from the FINWDS to the Pt. Arena region.

Fish samples collected in 1986/87 from the FINWDS exhibit ~10 times higher concentrations of $^{239+240}Pu$ and ~40-50 times higher concentrations of ^{238}Pu than those reported from a 1977 collection of identical species at the FINWDS reported by Schell and Sugai (1980). This could have been due to differences in analytical procedures, but could also have been due to increased contamination of the FINWDS deep-sea environment from the radionuclide waste containers during the period from 1977 to 1986/87.

Concentrations of ^{241}Am were also higher than comparable fish muscle tissue values reported at relatively contaminated sites from the Pacific Testing Grounds in Micronesia (Enewetak and Bikini Atolls) and from a potentially contaminated site in the Irish Sea in the vicinity of the Windscale Nuclear Plant. It is unclear why these values for ^{241}Am from the Farallons site and the Pt. Arena site are elevated in comparison with other sites worldwide.

A whole body effective dose equivalent value for pooled radionuclide concentrations committed to age 70 y by age at first intake yielded an added amount of ~1.5% of typical annual doses, with the primary calculated exposure dose being derived from ingestion of ^{241}Am.

The observed isotopic ratio of ^{238}Pu:$^{239+240}Pu$ was about 4, which is two orders of magnitude higher than the ratio 0.03 associated with fallout from weapons tests and accidental releases in the north temperate zone of the earth, which indicates a considerably higher environmen-

[##] Okihiro, University of California, Davis, CA., 1996.

tal mobilization for ^{238}Pu compared to $^{239+240}Pu$. Likewise, the observed ratio of ^{241}Am:$^{239+240}Pu$ of about 30 was nearly two orders of magnitude higher than the fallout ratio of 0.43 in the north temperate zone of the earth, which is likely explained by the greater solubility of americium compounds compared to plutonium compounds in biological systems (as compared with soil or sediment matrices).

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