^{210}Po AND ^{239}Pu, ^{240}Pu IN
BIOLOGICAL AND WATER SAMPLES FROM
THE BIKINI AND ENIWETOK ATOLLS

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MEASUREMENTS of the radioactivity in water and biological samples from Bikini and Eniwetok lagoons indicate that although the samples were collected from the most plutonium-polluted waters of the world, the values of the naturally produced radionuclide, polonium-210 were usually greater than the values of plutonium-239 and 240, which are produced by nuclear detonations by factors as great as 100.

With the advent of nuclear testing at the Bikini and Eniwetok atolls in the central Pacific, an environmental laboratory became available for investigating the concentrations of many radionuclides in the marine environment and their uptake by biota. Over the 16 years since the conclusion of nuclear testing, the long lived radionuclides created during the tests and deposited in sediments have had time to approach equilibrium with biota and with natural radionuclides present.

It is of particular interest to compare the concentrations of natural radionuclides, such as ^{210}Po, with concentration of ^{239}Pu and ^{240}Pu produced by nuclear bombs, in that part of the world which has been subject to most radioactive fallout. Both radionuclides decay by α-particle emission of approximately equal energy and both become concentrated in biota. The amount of plutonium to be used in nuclear power reactors is expected to increase by a factor of 1,000 in the next 30 years[1] and with this increased use comes the possibility of accidental or planned releases of plutonium into the food chain of man. The uptake of plutonium by biota in the marine environment is not well understood and a full evaluation of their potential hazard cannot yet be made.

Natural waters contain ^{210}Po as a member of the ^{238}U decay chain. Atmospheric aerosols also contain ^{210}Po which are scavenged[2] by precipitation and enter the hydrosphere. Once in the aquatic environment, polonium and plutonium undergo processes such as precipitation, redistribution, and changes in physical–chemical and oxidation states which may not be similar for the two elements. Both, however, are apparently available biologically as they are accumulated by algae, phytoplankton, zooplankton, vertebrates and invertebrates[3–6].

The samples discussed here were collected by the Laboratory of Radiation Ecology (LRE) during the 1972 joint expedition with the Puerto Rico Nuclear Center and the Lawrence Livermore Laboratory to Bikini and Eniwetok atolls. The locations of the collecting stations are shown in Fig. 1. The analytical results and collection locations of both biological and water samples, and the number of individual species of biological samples measured, are given in Table 1. Samples labelled as 'eviscerated whole' are whole fish which have had the viscera (liver, kidney, gonads, gut contents, and gastrointestinal tract) removed.

As several months elapsed between the collection and analysis of samples, the disintegration rate of ^{210}Po was corrected to allow for decay between the date of counting and the date of collection. The lower and upper limits given in Table 1 indicate the possible range of values if, first, ^{210}Po was derived entirely from the decay of ^{210}Pb in the sample (the lower limit) or, second, no ^{210}Pb was present

A single juvenile collected from a non-detonation area

Table 1 ^{210}Po, ^{239}Pu and ^{240}Pu values in biological and water samples, and the corresponding concentration factors for samples collected in October, 1972 at Bikini and Eniwetok atolls

	Biological samples			Water samples			Concentration factor	
Species and tissue	Location	^{210}Po range* (d.p.m. g^{-1}, wet)	^{239,240}Pu* (d.p.m. g^{-1}, wet)	Location	^{210}Po (d.p.m. m^{-3})	^{239,240}Pu (d.p.m. m^{-3})	^{210}Po range	^{239,240}Pu
Turtle liver (1)†	Eniwetok rubbish dump dock	0.20–0.69	0.200	Runit dock	188 ± 40	190 ± 11	(1.1–3.7) × 10^3	1.1 × 10^3
Goatfish viscera (6)	Eniwetok, Runit seaward reef	2.60–9.27	0.650	200 yards off Runit	146 ± 20	190 ± 11	(1.8–6.3) × 10^4	3.4 × 10^3
Wavyback skipjack (1), dark muscle	Eniwetok Lagoon off Runit	2.50–8.94	0.280	" "	146 ± 20	190 ± 11	(1.7–6.1) × 10^4	1.5 × 10^3
Mullet (2) eviscerated whole	Eniwetok, Runit seaward reef	0.50–1.90	0.003	" "	146 ± 20	190 ± 11	(3.4–13) × 10^3	16
Surgeon fish (3), bone	Bikini *a*–6	8.15–49.70	<0.001	*b*–15	107 ± 10	73 ± 9	(7.6–46.0) × 10^4	<20
Surgeon fish (3), muscle	Bikini *a*–6	0.02–0.15	<0.001	*b*–15	107 ± 10	73 ± 9	(1.9–14.0) × 10^2	<20
Surgeon fish (1), eviscerated whole	Bikini *a*–2	0.40–2.50	0.018	*b*–2	124 ± 13	138 ± 9	(3.2–20.0) × 10^3	130
Surgeon fish (3), viscera	Bikini *a*–6	0.51–3.09	1.330	*b*–15	107 ± 10	73 ± 9	(4.7–29.0) × 10^3	1.8 × 10^4
Convict surgeon (3), muscle	Bikini *a*–3	0.10–0.59	<0.001	*b*–6	125 ± 13	51 ± 7	(8.0–47.0) × 10^2	<20
Convict surgeon (17), muscle	Bikini *a*–7	0.06–0.34	<0.001	*b*–18	145 ± 14	102 ± 9	(4.1–23.5) × 10^2	<20
Convict surgeon (13), muscle	Bikini *a*–4	0.13–0.77	<0.001	*c*–12	135 ± 14	45 ± 5	(9.6–57.0) × 10^2	<20
Convict surgeon (6), muscle	Bikini *a*–5	0.09–0.56	<0.001	*b*–16	153 ± 15	7 ± 2	(5.9–36.5) × 10^2	<20
Convict surgeon (13), eviscerated whole	Eniwetok, Runit seaward reef	0.80–2.75	0.005	200 yards off Runit	146 ± 20	190 ± 11	(5.5–19) × 10^3	26
Convict surgeon (21), eviscerated whole	Eniwetok, Runit	0.20–3.31	0.025	" "	146 ± 20	190 ± 11	(1.4–22.6) × 10^3	130
Convict surgeon (4), eviscerated whole	Bikini *a*–2	1.40–8.28	0.028	*b*–2	124 ± 13	138 ± 9	(1.1–6.7) × 10^4	200
Convict surgeon (1), eviscerated whole	Bikini *a*–1	0.46–2.82	0.010	*c*–3	141 ± 15	138 ± 9	(3.3–21.7) × 10^3	72
Convict surgeon (4), eviscerated whole	Bikini *a*–1	0.34–2.06	0.017	*c*–3	141 ± 15	138 ± 9	(2.4–14.6) × 10^3	120
Convict surgeon (17), liver	Bikini *a*–7	2.04–12.50	0.913	*b*–18	145 ± 14	102 ± 9	(1.4–8.6) × 10^4	9 × 10^3
Convict surgeon (21), iscera	Eniwetok, Runit seaward reef	1.20–4.20	0.140	200 yards off Runit	146 ± 20	190 ± 11	(8.2–28.7) × 10^3	7.4 × 10^2
Convict surgeon (13), viscera minus liver	Bikini *a*–4	1.67–10.21	0.350	*c*–12	135 ± 14	45 ± 5	(1.2–7.6) × 10^4	7.8 × 10^3

*Analytical error is ± 10% for polonium and ± 20% for plutonium.

†Numbers in parentheses indicate the number of fish or organs combined in a given sample. Concentration factors are calculated as a ratio of activity per gram of wet tissue to activity per gram of water.

and the ^{210}Po measured was the true concentration present at the sampling time (the upper limit). The real value is expected to be near the upper limit for most samples since, in pelagic fish, the ^{210}Po is obtained by direct uptake and not as much by ingrowth from ^{210}Pb; measurements showed 1.5 times more ^{210}Po than ^{210}Pb (refs 8–10).

Table 1 shows that ^{210}Po activity is highest in bone, decreasing in order in liver, viscera and muscle. Although bone and liver have been reported as the major repositories for plutonium[10], the plutonium content of the single bone samples reported was at the detection limit of 0.001 d.p.m. per g. wet weight. The high concentrations of both ^{210}Po and ^{239}Pu and ^{240}Pu in viscera and viscera minus liver (convict surgeon), together with the fact that algae in the diet of these species concentrates those radionuclides from seawater, seems to support the idea that both radionuclides enter the fish during the ingestion of food.

Values of plutonium radioactivity have been reported in a large number of biological samples from Eniwetok Atoll[11]. These data also show that the highest plutonium concentrations occur in the viscera of fish. The food contents of the digestive organs were not separated, however, so it is not possible to estimate what fraction of the activity is associated with food and how much occurs in the tissues or organs of the fish.

The convict surgeon and surgeon fish were selected for ^{239}Pu, ^{240}Pu and ^{210}Po analysis because they are local feeders and would concentrate radionuclides from nearby waters. The concentrations of ^{239}Pu, ^{240}Pu and ^{210}Po measured in water from near the sites of the fish collection were used to determine the concentration factors shown in Table 1. The ^{210}Po concentration in the water was calculated from the ^{210}Pb concentration measured in the samples after most of the ^{210}Po had decayed. The error on the ^{210}Po calculation is expected to be small, as the ratio of ^{210}Po/^{210}Pb is approximately 1 : 2 in seawater[6,7]; thus, the ^{210}Pb and ^{210}Po would be near secular equilibrium in the marine environment at Bikini and Eniwetok atolls. The highest ^{239}Pu and ^{240}Pu contents in the water column were found in areas where the highest activity of those radionuclides occurred in sediments, as reported already for Bikini[12]. The highest ^{210}Po concentration was found outside the lagoon in the deep ocean waters, and one possible explanation could be that biological uptake results in a shorter resident time of ^{210}Po–^{210}Pb in the lagoon. The concentration of ^{210}Pb in the particulate phase, (>0.3 μm in size) was less than 10% of the total; the concentrations of ^{239}Pu and ^{240}Pu in the particulate phase varied from 2 to 60% of the total depending on where it was collected from[12]. The overall result indicates that inside the lagoon the radioactivity values of plutonium were more variable than those of ^{210}Po.

Concentration factors of ^{210}Po, ^{239}Pu and ^{240}Pu in organisms (Table 1) are calculated using the total concentrations (particulate plus soluble) measured in the water. All specimens measured have concentration factors for ^{210}Po that are greater (as much as 100 times greater) than those for ^{239}Pu and ^{240}Pu with the exception of samples of turtle liver and surgeon fish viscera. Although these samples were collected in the most plutonium-polluted waters of the world we conclude that the plutonium radioactivity is only a fraction of the natural polonium radioactivity, and that a greater radiation dose to the marine organisms of Bikini and Eniwetok lagoons would be received from ^{210}Po than from ^{239}Pu and ^{240}Pu. This conclusion should not, however, lead to neglect in studies of plutonium concentrations in the higher trophic levels and of its transport through the food web to man.

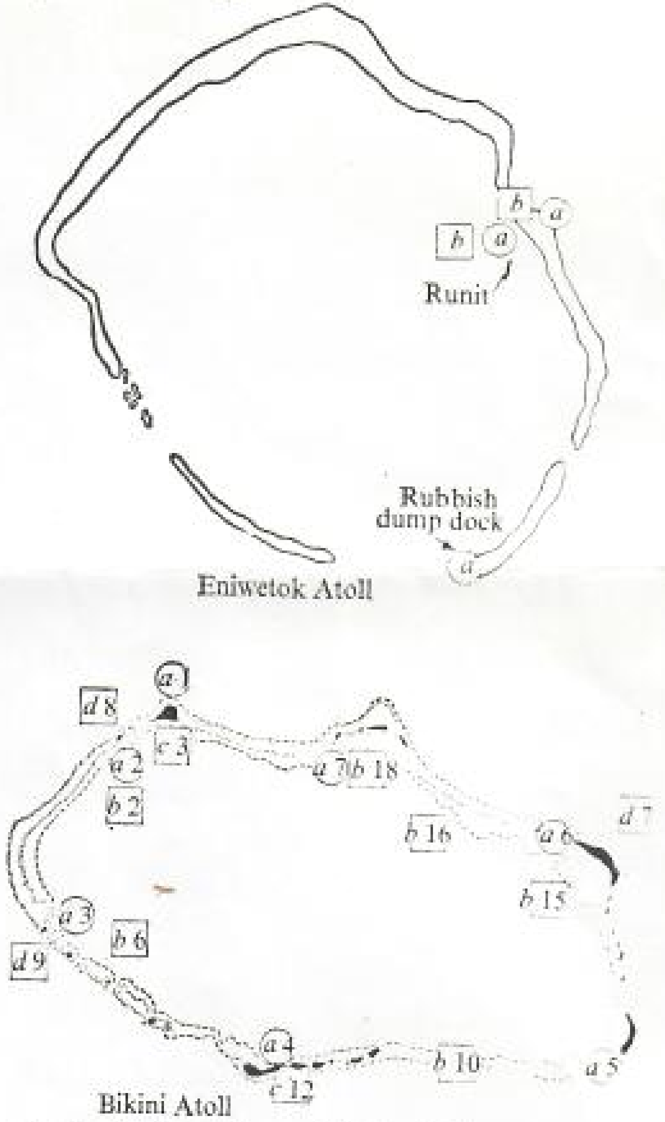

Fig. 1 Sampling stations at Bikini and Eniwetok atolls. *a*, Intertidal zone, biological stations; *b*, lagoon stations; *c*, crater stations; *d*, ocean stations.

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