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Concentrations of ^{207}Bi and ^{210}Pb-^{210}Bi-^{210}Po Disequilibrium in Fish[1]

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ABSTRACT: Radioactive ^{207}Bi, produced during nuclear testing at the Pacific Proving Grounds, concentrates in the muscle tissue and organs of goatfish and certain pelagic lagoon fish from Bikini and Enewetak Atolls. It is reasonable to expect that fish capable of accumulating ^{207}Bi could also be efficient accumulators of other bismuth isotopes—namely ^{210}Bi, the daughter of naturally occurring ^{210}Pb. Therefore, ^{210}Bi and consequently ^{210}Po, the decay product of ^{210}Bi, would be expected in notable excess over the precursor ^{210}Pb in specific tissues. To test this assumption, we compared concentrations of ^{210}Pb, ^{210}Bi, and ^{210}Po in muscle, liver, and bone separated from some reef species from the Marshall Islands. Concentrations of ^{210}Bi in muscle and liver were found to exceed those of its precursor by factors of 2 to 15. The excess ^{210}Bi in some species, however, is not from the environmental sources (either food or water) from which ^{207}Bi is derived. The data suggest that the excess ^{210}Bi may be translocated to muscle and liver tissue following the decay of ^{210}Pb in bone.

OUR PURPOSE HERE IS TO focus attention on the presence of excess ^{210}Bi (unsupported by its precursor ^{210}Pb) in fish. Part of the justification for initiating this study, however, was developed from an assessment of ^{207}Bi concentrations in fish.

The presence of ^{207}Bi ($t_{1/2} = 33.4$ y) was first reported in marine samples obtained from the Pacific Proving Grounds during 1961 (Lowman and Palumbo 1962). It was formed during a series of nuclear tests conducted by the United States at Bikini and Enewetak Atolls between 1946 and 1958, possibly by reactions such as ^{207}Pb(p, n) or ^{206}Pb(p, γ), assuming stable lead was present during testing as shielding material near the nuclear devices (Beasley 1969).

In addition to its presence in the marine sediments at these atolls and in the lagoon water mass, ^{207}Bi is accumulated by a variety of marine organisms. Table 1 shows concentrations of ^{207}Bi determined by gamma spectrometry in tissues and organs of several species of fish collected from Bikini and Enewetak Atolls during different years. Most striking is the range of concentrations in tissues and organs among different species of fish collected simultaneously from the same location. Highest concentrations of ^{207}Bi were consistently detected in the muscle and other tissues of goatfish and some of the pelagic lagoon fish compared to those of other reef fish such as mullet, surgeonfish, and parrotfish. Over 80 percent of the whole-body activity of ^{207}Bi in goatfish is associated with the muscle tissue, whereas less than 1 percent is found in the muscle of surgeonfish and mullet.

Concentrations of ^{210}Po were determined in the muscle of a few goatfish, mullet, and

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TABLE 1

CONCENTRATIONS OF ^{207}Bi IN FISH FROM ENEWETAK AND BIKINI ATOLLS

ATOLL AND ISLAND†	YEAR	COMMON NAME	^{207}Bi (pCi/kg wet)† MUSCLE	BONE	VISCERA	STOMACH CONTENT	SKIN	LIVER
E-24	80	Mullet (*Crenimugil*)	1.1 (27)	<2	578 (1)	1490 (2)	<1	30 (20)
		Surgeonfish	1.8 (29)	5 (30)	91 (2)		4.5 (20)	391 (2)
		Goatfish	656 (1)	283 (2)	661 (1)		465 (2)	1190 (1)
		Snapper (*Lethrinus*)	263 (2)	102 (4)	713 (2)	92 (4)	266 (3)	
		Snapper (*Lutjanus*)	191 (2)	198 (2)	1130 (2)		150 (2)	
		Bonito	186 (3)	60 (24)	421 (3)	124 (12)	90 (18)	662 (7)
E-10	78	Mullet (*Crenimugil*)	<2	<5	1150 (1)	1420 (2)	<2	<11
		Surgeonfish	<1	<6	20 (18)	65 (16)	<8	89 (10)
		Goatfish	6530 (2)	1770 (11)	9560 (1)	1220 (7)	4670 (1)	7460 (2)
		Parrotfish	<2	<10	<12		<4	
B-5	81	Mullet (*Crenimugil*)	<2	<14	44 (9)		<10	<40
		Surgeonfish	<4	<11	21 (35)	<40	<14	36 (24)
		Goatfish	56 (4)	22 (3)	145 (12)	242 (18)	37 (25)	50 (10)
		Parrotfish	<4	<14	<49	51 (23)	<23	<50
B-1	78	Mullet (*Crenimugil*)	4 (20)	<4	54 (11)	124 (11)	<3	116 (15)
		Mullet (*Neomyxus*)	<1	<8	68 (6)	100 (28)	<4	48 (25)
		Surgeonfish	<2	<25	49 (20)	<190	<30	<100
		Goatfish	1360 (2)	400 (4)	206 (2)	1070 (11)	960 (2)	3020 (2)
		Jack	121 (2)	19 (35)	120 (11)	<60		190 (20)

*E = Enewetak Atoll and B = Bikini Atoll. Numbers designate islands.
†The 1 σ counting error expressed as the percentage of the listed value appears in parenthesis; 1 pCi = 37 mBq.

surgeonfish from Bikini in 1982. The average concentration in goatfish flesh (0.67 pCi/wet g) exceeded the level in surgeonfish (0.065 pCi/wet g) and mullet (0.38 pCi/wet g). These data, coupled with the observation of higher levels of ^{207}Bi in goatfish, led us to speculate that similar enrichment might be expected for ^{210}Bi ($t_{1/2}$ = 5.01 *d*), the precursor of ^{210}Po and direct daughter product from decay of ^{210}Pb (^{210}Pb → ^{210}Bi → ^{210}Po).

Numerous data exist documenting that ^{210}Po ($t_{1/2}$ = 138.4 *d*) is accumulated in different tissues of marine organisms to levels greater than those resulting from the decay of its long-lived precursor ^{210}Pb (Folsom and Beasley 1973, Beasley et al. 1973, Cherry and Shannon 1974). Although according to Cherry and Shannon (1974) the "(^{210}Bi – ^{210}Pb) disequilibrium situation is relatively uninteresting," our results may generate some interest because any unsupported ^{210}Bi detected could be a potential source for some fraction of the excess ^{210}Po in the organism.

At Bikini and Enewetak, we expected high concentrations of ^{207}Bi that would interfere with the beta-counting detection of ^{210}Bi. Therefore, we also collected fish for analysis from the relatively uncontaminated environment of Kwajalein Atoll. Some additional fish were collected later from Rongelap Atoll for analysis of ^{210}Pb and ^{210}Po in bone. Mullet (*Crenimugil crenilabis* and *Neomyxus chaptalii*, second trophic level), surgeonfish (*Acanthurus triostegus*, second trophic level), goatfish (*Mulloidichthys samoensis*, third trophic level), flagtail (*Kuhlia taeniura*, third trophic level), and parrotfish (*Scarus sordidus*, fourth trophic level) were segregated from throw-net collections at several islands of Bikini, Enewetak, and Kwajalein Atolls in 1983. The relative ability of some of these fish to accumulate bismuth is shown in Table 1.

MATERIALS AND METHODS

The fish were dissected within hours of collection and pooled samples of tissues were

prepared from four to twenty individual fish of similar size. Wet weights were determined and the samples were decomposed by wet digestion with one part of concentrated nitric (15.5*N*) and three parts of concentrated hydrochloric acid (12.5*N*) in the presence of standardized lead and bismuth carriers and ^{209}Po tracer as yield monitors. After dissolving the sample concentrated hydrochloric acid was added to volatilize the nitric acid. This step is necessary to optimize the chemical recovery. Lead, bismuth, and polonium fractions were sequentially separated by an anion exchange column using 3, 8, and 12*N* nitric acid. The separations were made within 24 h of collection to minimize the unavoidable growth–decay corrections. The samples were prepared as precipitates of lead chromate and bismuth oxychloride and counted on low-background beta detectors for approximately 20 to 30 d to follow the decay of ^{210}Bi in the bismuth fraction and its growth in the lead fraction. The polonium radionuclides were deposited on silver disks and measured on low-background alpha spectrometers. Reagent blanks and standards for each radionuclide were processed along with the samples.

RESULTS AND DISCUSSION

The concentrations of ^{210}Pb, ^{210}Bi, and ^{210}Po determined in the tissues (corrected to the date of collection) and computed activity ratios are given in Table 2. Concentrations of ^{210}Po in muscle are variable in fish from different trophic levels, and significant differences are noted in the ^{210}Pb : ^{210}Po ratio in tissues among species and even between two species from the same family (*Neomyxus* and *Crenimugil*). Concentrations of ^{210}Po are greater than ^{210}Pb in muscle and liver of these atoll species. This enrichment has been emphasized in previous studies with fish collected from other global locations where it has been generally acknowledged that the main source of unsupported ^{210}Po (and ^{210}Pb) in fish is the food chain (Folsom and Beasley 1973, Beasley et al. 1973, Cherry and Shannon 1974).

Although ^{210}Po is enriched relative to ^{210}Pb in the muscle and liver of surgeonfish, there is no significant magnification of ^{210}Po relative to ^{210}Pb between the items ingested by the fish and the tissues. A clear distinction is seen between ^{210}Pb and ^{210}Po in the bone of fish collected at Rongelap—^{210}Pb levels exceed the concentrations of ^{210}Po by 25 to 35 percent and are also much higher than in muscle measured in species from the other atolls. Whereas ^{210}Po concentrations are, in general, higher in goatfish muscle than in other species, the reverse trend is noted for the concentrations in bone. Further, ^{210} Po concentrations are higher in the muscle of smaller sized goatfish from island B-6, indicating there may be variations in concentrations related to size among some reef species. Concentration factors for ^{210}Po in muscle to that in filtered seawater have been calculated using a mean value of 31 ± 3 fCi/L for ^{210}Po measured in 12 lagoon and ocean surface water samples. Values range from 8×10^2 for surgeonfish muscle to 2.3×10^4 for goatfish.

The ^{210}Bi concentrations in liver and flesh from all species exceed those of its precursor ^{210}Pb measured in these tissues. Therefore some account of excess ^{210}Bi in edible portions of freshly caught and rapidly consumed fish should be made in radiological dose estimates from natural radionuclides in marine food pathways.

Based on the ^{207}Bi results and on the assumption that any unsupported ^{210}Bi accumulated by fish from food or water would relate to ^{207}Bi concentrations, we anticipated levels of ^{210}Bi in the muscle of goatfish and flagtail several orders of magnitude larger than the unsupported concentrations in surgeonfish, mullet, and parrotfish. Rather, we found no significant differences in concentrations of ^{210}Bi in the muscle among all the species. It is clear, therefore, that the excess ^{210}Bi in the muscle and liver of surgeonfish, mullet, and parrotfish is not from its precursor nor from the environmental sources from which ^{207}Bi is derived.

From the results in Table 1 and the percent wet weight of the different fish tissues (Noshkin et al. 1981), it can be determined that fish bones are poor reservoirs for ^{207}Bi. The contribution of ^{207}Bi in bone to the total activity ranges from less than 1 percent in sur-

TABLE 2

CONCENTRATIONS AND CONCENTRATION RATIOS OF ^{210}Pb, ^{210}Bi, AND ^{210}Po IN MARSHALL ISLANDS FISH

ATOLL AND ISLAND*	COMMON NAME	ORGAN†	MEAN STANDARD LENGTH OF FISH (mm)‡	pCi/kg WET WEIGHT§				
				^{210}Pb	^{210}Bi	^{210}Po	^{219}Bi:^{210}Pb	^{210}Pb:^{210}Po
Kwaj.	Surgeonfish	M	121 (6)	128 (3)	230 (9)	280 (3)	1.8	0.46
B-5	Surgeonfish	M	123 (6)	42 (8)	120 (20)	208 (2)	2.9	0.20
B-5	Surgeonfish	L	123 (6)	3,550 (3)	16,000 (7)	23,800 (2)	4.5	0.15
B-1	Surgeonfish	M	143 (6)	7 (20)	70 (30)	26 (3)	10.0	0.27
B-1	Surgeonfish	L	143 (6)	1,220 (6)	4,800 (9)	2,150 (2)	3.9	0.57
B-12	Surgeonfish	M	142 (5)	10 (5)	50 (15)	34 (3)	5.0	0.29
B-12	Surgeonfish	S	142 (5)	170 (20)		4,600 (1)		0.037
B-1	Mullet (*Neomyxus*)	M	225 (5)	36 (6)	200 (8)	280	5.6	0.13
B-1	Mullet (*Neomyxus*)	L	225 (5)	1,330 (2)	7,500 (4)	10,400 (1)	5.6	0.13
B-12	Mullet (*Neomyxus*)	M	198 (6)	56 (5)	140 (8)	147 (5)	2.5	0.38
E-38	Mullet (*Neomyxus*)	M	211 (5)		55 (50)	146 (3)		
Kwaj.	Mullet (*Crenimugil*)	M	221 (5)	30 (11)	60 (19)	330 (4)	2.0	0.09
B-1	Mullet (*Crenimugil*)	M	203 (5)	<3	lost	160 (5)		<0.02
B-1	Mullet (*Crenimugil*)	L	203 (5)	150 (13)	950 (7)	3,840 (1)	6.3	0.04
B-5	Mullet (*Crenimugil*)	M	216 (5)	<3		156 (2)		<0.02
B-5	Mullet (*Crenimugil*)	L	216 (5)	140 (20)		5,200 (2)		0.03
Kwaj.	Goatfish	M	180 (5)	17 (10)	50 (60)	450 (3)	2.9	0.04
B-12	Goatfish	M	173 (6)	9 (30)	140 (7)	600 (5)	15.5	0.02
B-6	Goatfish	M	149 (7)	4 (45)		720 (8)		0.006
B-6	Goatfish	M	181(5)	8 (30)		570 (3)		0.014
B-6	Goatfish	M	203 (5)	6 (30)		290 (3)		0.021
Kwaj.	Flagtail	M	179 (5)	6 (30)	40 (30)	560 (5)	6.7	0.01
B-5	Parrotfish	M	223 (4)	3 (60)	40 (30)	122 (4)	13	0.02
R	Surgeonfish	B	80 (12)	13,100 (2)		9,600 (3)		1.36
R	Mullet (*Neomyxus*)	B	215 (10)	6,500 (2)		5,200 (3)		1.25
R	Mullet (*Crenimugil*)	B	168 (8)	2,700 (2)		2,100 (3)		1.29
R	Goatfish	B	175 (20)	1,700 (5)		1,230 (3)		1.38

*Kwaj. = Kwajalein Atoll, B = Bikini Atoll, E = Enewetak Atoll, and R = Rongelap Atoll. Numbers designate islands. Fish collected 22–28 August 1983 except at Rongelap where collection was on 17 October 1983.

†M = muscle, L = liver, S = stomach contents, and B = bone.

‡Number of fish pooled per sample appears in parenthesis.

§The 1 σ counting error expressed as the percentage of the listed value appears in parenthesis; 1 pCi = 37 mBq. All data corrected to date of collection.

TABLE 3

CONTRIBUTION TO TOTAL FISH ACTIVITY OF ^{210}Pb, ^{210}Bi, AND ^{210}Po FOR ORGANS ANALYZED

FISH AND TISSUE	MEAN % OF WHOLE BODY WET WEIGHT	CONTRIBUTION TO TOTAL ACTIVITY IN FISH (pCi)		
		^{210}Pb	^{210}Bi	^{210}Po
Surgeonfish (*Acanthurus*)				
Muscle	66.3	0.005–0.09	0.033–0.15	0.022–0.19
Liver	0.7	0.009–0.025	0.034–0.11	0.015–0.17
Bone	8.0	1.05		0.77
Total in above tissues		1.06–1.17		0.81–1.13
Mullet (*Neomyxus*)				
Muscle	55.3	0.02–0.03	0.03–0.11	0.08–0.15
Liver	0.7	0.023	0.052	0.18
Bone	5.5	0.36		0.29
Total in above tissues		0.40–0.41		0.37–0.62
Mullet (*Crenimugil*)				
Muscle	58.9	0.002–0.02	0.035	0.091–0.19
Liver	0.9	0.001	0.009	0.034–0.047
Bone	6.9	0.19		0.14
Total in above tissues		0.19–0.21		0.27–0.38
Goatfish (*Mulloidichthys*)				
Muscle	66.3	0.004–0.011	0.033–0.093	0.19–0.48
Liver	0.41			
Bone	8.0	0.14		0.098
Total in above tissues		0.15		0.29–0.58

geonfish to a maximum of 3 percent in goatfish. Shown in Table 3 are the estimated contributions of ^{210}Pb, ^{210}Bi, and ^{210}Po activities in muscle, liver, and bone to the total activity in the fish. Fractions of the whole-body concentration cannot be computed because we lack data for remaining organs such as skin, viscera, and so on. The original bone samples from the Bikini–Enewetak collections were discarded. We have to assume that the bone concentrations from the Rongelap fish are representative for the species from any location. Display of the results in this form reveals several features not evident from the concentration data in Table 2.

In surgeonfish and mullet (*Neomyxus*), the total concentration of ^{210}Pb in muscle, liver, and bone is nearly equivalent to the total ^{210}Po concentration in these tissues. Had the concentrations been determined in these bulked tissues, one would have concluded that there is little or no unsupported ^{210}Po. This conclusion differs from the assessment suggested by the data in Table 2 and general conclusions from the literature. The excess concentration of ^{210}Po in muscle and liver could be explained as redistributed amounts generated in the bone from the decay of accumulated ^{210}Pb. This mechanism was proposed to account for ^{210}Po in humans (Holtzman 1964). Since we have ruled out environmental sources, a source must also be found, however, for the excess ^{210}Bi in muscle and liver. Because bismuth is poorly retained by bone, as shown by the ^{207}Bi data, it is conceivable that the ^{210}Bi, rather than ^{210}Po, is translocated rapidly from bone surface following its production by decay of accumulated ^{210}Pb.

In goatfish and mullet (*Crenimugil*), the range of the cumulative ^{210}Pb activities is less than that of ^{210}Po. There is, in addition, unsupported ^{210}Bi in the muscle and liver of these fish to be accounted for as well. A fraction of the ^{210}Po accumulated by these fish probably originates directly from the food

chain, but the remainder could also be produced by decay of a mobile source of ^{210}Bi relocated to the tissues from the bone.

This assessment shows that all unsupported ^{210}Po measured in fish, and possibly in other organisms, does not necessarily have to result directly from the food chain. Some fraction, which may vary with the species, of unsupported ^{210}Po in specific tissues such as muscle and liver may result from redistribution and decay of ^{210}Bi generated from ^{210}Pb accumulated in bone.

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