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Concentration of ^{210}Po and ^{210}Pb in the diet at the Marshall Islands

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Abstract

The concentrations of ^{210}Po and ^{210}Pb have been determined in many local foods consumed by societies residing on different atolls in the Marshall Islands. The average daily intake of these two naturally occurring radionuclides from local and imported food is estimated to be 2.18 and 0.36 Bq, respectively. Local foods contribute 87% of the ^{210}Po and 47% of the ^{210}Pb associated with the diet. The items contributing the majority of the activity to the diet are derived from the marine environment and include parts of fish, invertebrates, seabirds and eggs of seabirds. The committed effective dose from ingestion of ^{210}Po and ^{210}Pb is ~ 2 mSv/year (200 mrem/year). This pathway now contributes 83% of the natural background irradiation received by residents in the Marshall Islands. Because the naturally occurring radionuclides are omnipresent in terrestrial and marine foods at all atolls, the annual intake and computed dose can be considered as typical values for individuals with comparable diets and inhabiting other islands in the Pacific.

Keywords: Marshall Islands; Effective dose; Diet; Radioactivity; ^{210}Po; ^{210}Pb

1. Introduction

A large fraction of the radiation exposure experienced by individuals through ingestion of food is from the naturally occurring radionuclides ^{210}Po and ^{210}Pb (UNSCEAR, 1988). Conclusions from previous dietary studies (Holtzman, 1980) indicate that the intake rate of these radionuclides may vary considerably because of differences in concentration among classes of food in the diet. According to UNSCEAR (1988) the average dose rate from ^{210}Pb and ^{210}Po intake with food is ~ 0.12 mSv/year. Greater than average dose is experienced by populations with diets high in seafood (Holtzman, 1980).

Average values for intake of ^{210}Pb and ^{210}Po are available from a number of cities in eighteen countries (Holtzman, 1980) but there exists little information on the levels of these naturally occurring radionuclides in many local foods from the Marshall Islands or from other populated

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small islands in the Pacific Ocean where consumption of local seafood can be significant. Many of the items in the Marshallese diet such as breadfruit, pandanus, coconut crab, seabirds, some species of local fish, and the *Tridacna* clam are unknown as food items to societies outside coral atolls. Refrigeration does not exist on many islands so that locally collected food products are prepared for immediate consumption thereby maximizing the intake of any ^{210}Po.

The best available dietary information for the Marshall Islands (Robison et al., 1980) reveals that, on the average, 60% of the solid and liquid foods prepared for consumption is now imported. Twenty-seven percent of the local solid food consumed is derived from the marine environment; the remainder from domestic animals and terrestrial crops. When imported foods are not available, marine foods are a larger percentage of the solid food intake. For comparison, marine products in the average diet of individuals from the UK represents only 1% of the total solid food intake (Smith-Briggs and Bradley, 1984). A preliminary dose assessment, using the dose coefficients suggested in ICRP 30 (ICRP, 1979), revealed that ingestion of the flesh only from some species of local reef fish at Enewetak Atoll in the Marshall Islands resulted in an annual dose of 0.21 mSv from ^{210}Po alone (Robison et al., 1987).

A number of scientific expeditions have been made to atolls in the Marshall Islands during the last 20 years to collect environmental samples for the measurement of different man-made radionuclides that resulted from the series of USA nuclear tests conducted at Bikini and Enewetak Atolls between 1946 and 1958. Since the calculated annual effective dose from ingestion of ^{210}Po associated with reef fish intake alone was greater than the global average effective dose, a decision was made to broaden the available database for ^{210}Po and ^{210}Pb. Concentrations of ^{210}Po and ^{210}Pb have now been determined in many dietary foods that were, in part, also collected for the man-made radionuclide studies. An assessment is made of the annual intake and the estimated effective dose from these naturally occurring radionuclides in the food ingestion pathway.

2. Diet information

The variety and quantity of food consumed in the Marshall Islands is based on available knowledge of dietary habits for adult Marshallese determined from questionnaires and interviews conducted by the Micronesian Legal Services Corporation (MLSC) and a Marshallese school teacher on Ujelang (Robison et al., 1980). The MLSC data has been used to estimate radiation exposure to members of several populations from the residual man-made radioactivity generated at Bikini and Enewetak during the period of USA nuclear testing (Robison, 1983). Predictions of the ^{137}Cs body burden and dose using this diet model are very close to the ^{137}Cs body burdens determined in the population from whole-body measurements (Robison, 1983). Consequently the MLSC diet is selected, rather than other proposed diets, to assess the intake of ^{210}Po and ^{210}Pb with food and water.

During the last 45–50 years many Marshallese have experienced major changes in their lifestyle. At most atolls there is a preference for imported foods that have been substituted for local traditional foods consumed in the past. Commercial transport to even the most remote atoll is now available and is reasonably reliable so it is unlikely that there will be a total absence of any desired imported food from the diet.

The best estimate of the type and quantity of imported and different local plants, organisms, and water ingested is shown in the first two columns of Table 1. The type and quantity of local and imported food described in Table 1 (IA diet) is considered to represent the current ('normal') adult diet at many Pacific atolls (Robison, 1983).

The diet survey also considered a situation where imported foods are unavailable (IUA) and individuals have to rely only on domestic (local) foods. The average amounts of different local food ingested when imported foods are absent from the diet (IUA) are also listed in Table 1. The total intake of 1.54 kg/day in this diet converts to a caloric intake of 1256 kcal/day (Robison et al., 1987) which is less than an individual's

Table 1
Dietary intake in the Marshall Islands

	Imported food available (IA) (kg/day)	Imported food unavailable (IUA) (kg/day)
Local food		
Reef fish	0.024	0.043
Pelagic fish	0.018	0.047
Marine crab	0.002	0.010
Lobster	0.004	0.018
Clams and trochus	0.006	0.035
Coconut crab	0.003	0.013
Octopus	0.005	0.025
Turtle	0.004	0.006
Turtle eggs	0.009	0.117
Chicken flesh	0.008	0.016
Chicken liver	0.005	0.009
Chicken gizzard	0.002	0.002
Chicken eggs	0.007	0.021
Pork	0.019	0.021
Local bird flesh	0.003	0.013
Bird viscera	0.002	0.005
Bird eggs	0.002	0.011
Terrestrial vegetation	0.259	0.604
Water and water products	0.947	0.530
Total local food and water	1.328	1.543
Imported food		
Bread	0.102	
Pancake-cake	0.062	
Rice	0.234	
Potatoes	0.127	
Sugar	0.065	
Canned meat	0.134	
Canned chicken	0.013	
Canned fish	0.146	
Juice	0.491	
Carbonated drinks	0.338	
Powdered milk	0.073	
Evaporated milk	0.201	
Noodles (pasta)	0.006	
Total imported food	1.992	
Total local and imported food	3.320	1.543

recommended allowance of 1600–4000 kcal/day (Robison et al., 1987). Near famine conditions have occurred at some atolls during the 1970s and domestic foods were used exclusively (Robison et al., 1980). It is probably unrealistic to consider these conditions will recur today. However, in another context, a diet of only local foods could be looked upon as an example of the minimum amount of food available for consumption by individuals prior to the early 1950s when supply of imported food was relatively unreliable at many atolls. This diet and the normal (both imports and indigenous foods available) diet will be used in conjunction with the radiological concentration data to compare past with present intakes of ^{210}Po and ^{210}Pb.

3. Methods

3.1. Collection of local samples

Many of the marine and terrestrial samples for this study were collected from Bikini and Enewetak Atolls, the sites of the USA nuclear testing program in the Pacific from 1946 to 1958. Some years ago Beasley (1969) measured the ^{210}Pb content in 15 samples of local sediment and soil that were contaminated with fission and activation products from the test series. On the basis of the concentration data and other available information, Beasley concluded that the ^{210}Pb levels in the samples did not exceed the levels expected to occur naturally. However, a review of test data and other information shows that some long-lived precursors of ^{210}Pb may possibly have been associated with unburnt weapon fuel or with other components containing uranium having a high percentage of ^{238}U (Lynch and Gudiksen, 1973; Schell et al., 1980). Therefore additional comparative data was collected to affirm that the levels of ^{210}Pb (^{210}Po) found were naturally occurring and not artificially enhanced at Bikini or Enewetak, or at Rongelap Atoll which received some intermediate range fallout from tests conducted at the Pacific Proving Grounds. Samples from control sites at Kwajalein, Majuro, Pohnpei, and in the equatorial Pacific ocean were collected to generate comparative concentration data in fish, terrestrial foods and surface seawater. Data from these comparisons and results from other samples are discussed in another section of this report but all results support the conclusion that ^{210}Pb and/or ^{210}Po levels in different environmental samples from Bikini, Enewetak, or elsewhere in the Marshall Islands, do not exceed the levels expected to occur naturally.

Fig. 1 shows the geographical location of the Marshall Islands in the North Equatorial Pacific Ocean and most of the atolls visited on sampling programs.

Samples collected for analysis of ^{210}Po and, in some cases, ^{210}Pb include species of reef and pelagic fish; *Tridacna* clam; lobster and marine crab; coconut and other land crabs; seabirds and seabird eggs; chicken and chicken eggs; breadfruit; pandanus; coconut; papaya; pumpkin; banana; and limes. Terrestrial vegetation and organisms were collected by hand from locations where samples were abundant and available on different islands of an atoll. Surface seawater samples were collected from lagoons of atolls and the open ocean. The species of reef fish collected include: mullet, *Crenimugil crenilabis* and *Neomyxus chaptalii*; convict surgeonfish, *Acanthurus triostegus*; unicornfish, *Naso lituratus*; rabbitfish, *Siganus rostratus*; bonefish, *Albula vulpes*; flagtail, *Kuhlia taeniura*; goatfish, *Mulloidichthys samoensis*; and parrotfish, *Scarus sordidus*. Throw nets were used exclusively to catch the reef fish at the different atolls. The pelagic species collected include: grouper, *Epinephelus spilotoceps*; ulua, *Caranx melanpygus*; jack, *Caranx* sp.; snapper, *Aprion virescens*; rainbow runner, *Elegatis bipinnulatus*; mackerel, *Grammatorcynus billineatus*; and bonito, *Euthynnus affinis*. All pelagic and benthic fish were collected in the lagoons on sport fishing gear using feathered jigs and baited hooks. Divers collected clams and marine invertebrates and the terrestrial invertebrates were collected by shore parties.

Several local food items identified in the diet survey were not sampled. We were unable to obtain an octopus or turtle from any lagoon. A pig and turtle eggs were the only terrestrial foods not sampled.

3.2. Preparation and analysis

Marine and/or terrestrial biota were segregated by type, transferred to plastic bags, frozen, and shipped by air or sea in a frozen state to Lawrence Livermore National Laboratory (LLNL) for further processing and analysis. Polonium was separated from 60-l sea water samples (and from ^{210}Pb) shortly after collection in the field. Samples were returned to the lab for plating and counting.

On two occasions fish were dissected in the field within hours of collection and pooled samples of tissues from the same species were prepared from four to twenty individual fish of similar size for analysis. These samples were immediately decomposed by wet acid digestion onboard ship to separate and measure the levels of any unsupported ^{210}Bi, as well as ^{210}Po and ^{210}Pb (Noshkin et al., 1984). These separations were

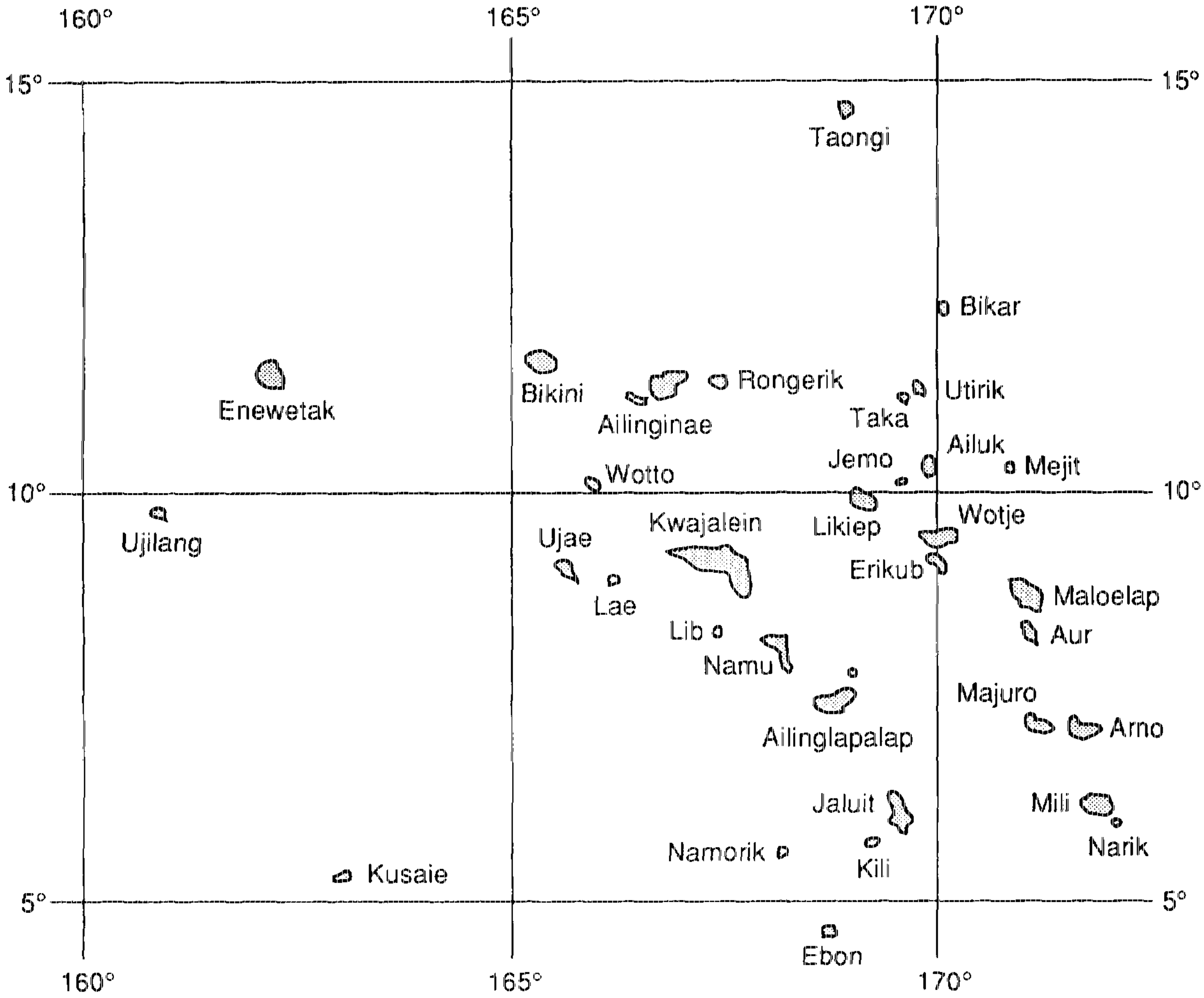

Fig. 1. Location of the Marshall Islands showing some of the atolls where samples were collected.

completed within 24 h of collection to minimize the unavoidable growth-decay corrections.

At the LLNL laboratory the biota is thawed and the sample weighed. Specific parts and tissues are separated from the different plants and animals. Aliquots of the fresh sample are weighed and, together with ^{209}Po as a tracer and stable lead carrier, dissolved in HNO_3 and $HClO_4$ acids. Lead and polonium are precipitated from a basic solution with iron hydroxide. The hydroxide precipitate is dissolved in 0.5 M HCl and the polonium removed by spontaneous deposition onto silver discs in the presence of ascorbic acid at 90°C. The separation time of ^{210}Po from ^{210}Pb is recorded and the ^{210}Po is measured, along with the ^{209}Po yield tracer, by alpha spectrometry.

After the ^{210}Po separation, the ascorbic acid is decomposed with nitric acid and the lead chromate is precipitated. This precipitate is dissolved in HCl and lead, with ^{210}Pb, is separated from interfering cations, including any remaining ^{210}Po and ^{209}Po by anion exchange. After the lead is eluted from the column it is precipitated as the chromate for yield determination. The identification and concentration of ^{210}Pb are determined from the ^{210}Bi daughter by following the growth of this radionuclide on low background beta detectors until equilibrium is established.

3.3. Calculation of ^{210}Po concentrations in the environmental samples

^{210}Po accumulates in all food items with and without immediate support from its long-lived precursor ^{210}Pb. When months elapse between collecting and processing the sample, the amount of unsupported ^{210}Po lost by decay and the ingrowth of new ^{210}Po from ^{210}Pb decay must be computed from the counting data to arrive at the initial activity of ^{210}Po in a sample.

In samples where both ^{210}Po and ^{210}Pb are determined, the ingrowth-decay corrections to the date of collection are straightforward. However, both radionuclides were determined in only 28% of the samples processed for this study. The mean values for the ^{210}Pb/^{210}Po activity ratios measured in these (28%) different samples are shown in Table 2. Concentrations of ^{210}Po are greater than the ^{210}Pb precursor in the flesh, liver and the viscera of fish and in marine invertebrates. The enrichment of ^{210}Po in these tissues and organs from fish and invertebrates collected from other global locations has been noted previously (Cherry and Shannon, 1974). In fish bone and vegetation samples, the ratio of the two radionuclides is not readily distinguished from unity and in many cases the ^{210}Pb can exceed the ^{210}Po concentration (Noshkin et al., 1984).

The majority of the samples were processed within 0.5–90 days of collection so that applicable ingrowth and decay corrections were relatively small in most cases. However, counting corrections are applied to the results in the remaining samples (where only ^{210}Po was determined) by using the following method. The mean value of the ^{210}Pb/^{210}Po ratio (R) in a comparable type of sample from Table 2 is used with the measured ^{210}Po activity (Po_m) at time t (days between collection and separation from ^{210}Pb) in Eq. 1 to estimate the initial concentration of ^{210}Po (Po_i) in the respective sample.

$$Po_m = [\lambda_2/(\lambda_2 - \lambda_1)] * (RPo_i) * (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + Po_i * e^{-\lambda_2 t} \quad (1)$$

λ_1 and λ_2 are the decay constants in days for ^{210}Pb and ^{210}Po, respectively. This procedure was

Table 2
Mean activity ratios of ^{210}Pb/^{210}Po in different samples from the Marshall Islands

Sample type	Number of measurements	Mean value ratio	Standard deviation
Flesh of fish			
Surgeon fish	7	0.31	0.09
Mullet	5	0.17	0.14
Trophic III fish	9	0.04	0.04
All fish	21	0.16	0.14
Liver of fish			
Surgeon fish	2	0.36	0.29
Other fish	5	0.06	0.05
All fish	7	0.14	0.19
Bone of fish			
Surgeon fish	5	0.70	0.36
Other fish	8	0.94	0.32
All fish	13	0.84	0.34
Viscera of fish			
All fish	5	0.02	0.01
Invertebrates			
Soft parts-all	4	0.014	0.014
Vegetation			
All	9	0.92	0.46

tested on several samples where the concentrations of both radionuclides were measured. Using this method, a difference of no more than two percent is found between the measured and estimated value. This correction technique is used with the remaining fresh samples to compute the ^{210}Po concentration at the time of sample collection.

4. Results

4.1. ^{210}Po and ^{210}Pb intake with local foods

The results from the different processed samples are summarized in Table 3. The number of samples of each type analysed is shown along with the mean concentration and range in activity levels.

Table 3
Summary of ^{210}Po and ^{210}Pb concentrations in samples from the Marshall Islands

	^{210}Po Bq/kg wet wt.			^{210}Pb Bq/kg wet wt.		
	No. of samples	Range in values	Mean value	No. of samples	Range in values	Mean value
Marine samples						
Flesh of fish (common names)						
Reef species						
Unicorn	1		0.6	1		0.2
Triggerfish	1		1.6			
Rabbitfish	1		3.8			
Surgeon fish	13	0.4–25.2	4.5	7	0.1–7.0	2.1
Neomyxus	7	5.4–20.2	11.4	3	1.3–4.7	2.7
Crenimugil	11	5.9–25.9	12.2	4	0.1–1.1	0.4
All trophic II Reef Fish	34	0.4–25.9	7.9	15	0.1–7.0	1.6
Bonefish	7	4.5–15.0	6.4	2	0.32–0.62	0.5
Flagtail	2	16.3–20.7	18.5	1		0.2
Goatfish	27	8.5–37.7	20.2	6	0.15–0.63	0.3
All trophic III reef fish	36	4.5–37.7	17.4	9	0.15–0.63	0.4
Parrotfish	3	2.5–4.5	3.7	3	0.1–0.11	0.1
All trophic IV reef fish	3	2.5–4.5	3.7	3	0.1–0.11	0.1
Flesh of all reef fish	73	0.4–37.7	12.5	27	0.1–7.0	1.0
Pelagic species						
Grouper	1		0.4			
Ulua	4	6.6–38.1	17.9			
Jack	1		24.4			
Snapper	4	0.7–3.1	2.2			
Rainbow runner	2	17.0–28.9	22.9			
Mackerel	3	3.7–4.5	4.0			
Bonito	4	21.5–53.3	36.9			
Flesh of all pelagic fish	19	0.4–53.3	16.4			
Flesh of all fish	92	0.4–53.3	13.3	27	0.1–7.0	1.0
Liver of fish						
All fish	13	80–1020	515.0	9	5.2–132	38.0
Bone of fish						
All fish	20	43–800	152.8	13	45–444	128.2
Viscera of fish[a]						
All fish	8	100–5370	1725.0	7	3.0–41	26.8

Table 3 (*continued*)

	^{210}Po Bq/kg wet wt.			^{210}Pb Bq/kg wet wt.		
	No. of samples	Range in values	Mean value	No. of samples	Range in values	Mean value
Content of Viscera[b]						
All fish	23	53–5185	827.0	5	6.3–22	10.3
Invertebrates						
Flesh of clams						
Tridacna squamosa	6	29–70	55.7	2	0.8–2.4	1.6
Flesh of marine Crustacea						
Grapus tenuicrustatus	1		9.0			
Panulinus penicillatus	1		11.2			
Terrestrial samples						
Flesh of marine feeding birds						
Sterna sumatrana	1		31.0			
Sula leucogaster	3	27.3–56	34.8	1		0.1
Viscera of marine feeding birds						
All species	4	102–217	149.3	1		1.5
Eggs of marine feeding birds						
All species	3	20–88	46.0	1		1.0
Domestic chicken						
Flesh	1		0.3			
Viscera	2	0.25–1.0	0.6			
Eggs	1		0.0			
Liver	1		1.1			
Flesh of land crabs						
Birgus latro	12	15–69	40.8	2	0.1–0.3	0.2
Coenobita perlatus	1		23.0			
Vegetation samples						
Breadfruit (pulp)	18	0.01–0.07	0.03	1		0.02
Breadfruit (skin)	10	0.06–0.23	0.13			
Pandanus (pulp)	8	0.02–0.19	0.13	2	0.02–0.2	0.11
Coconut meat	18	0.01–0.29	0.08			
Coconut juice	8	0.002–0.02	0.01	1		0.00
Copra meat	2	0.03–0.09	0.06	1		0.03
Copra juice	2	0.009–0.04	0.03	1		0.01
Papaya pulp	5	0.009–0.03	0.02	1		0.01
Pumpkin pulp	3	0.00–0.026	0.01	1		0.06
Banana fruit	3	0.03–0.04	0.03	1		0.03
Lime juice	3	0.01–0.022	0.02	1		0.00
All vegetation samples	80		0.06	10		0.03

Concentrations reported on date of collection.
[a] Includes stomach; small and large intestine; and pyloric caecum.
[b] Includes contents of stomach; and/or intestines

With the tropical climate and the lack of refrigeration on most atolls, fresh local food is consumed immediately, or within 1 or 2 days of collection. Therefore the concentrations of the radionuclides in the food items on the day of ingestion are essentially equal to the measured concentrations on the date of sample collection. Relevant mean concentrations of ^{210}Po and ^{210}Pb

in foods from Table 3 are transferred to columns 2 and 3 in Table 4. The concentrations in Table 4 are multiplied by the respective value for local food intake in both the IA and IUA diets from Table 1 to estimate the daily intake of the two radionuclides.

4.2. Comparative data

Table 5 has several sets of important comparative results from different regions within the Marshall Islands. It shows that the mean level of ^{210}Po in the flesh of fish, in coconut crabs, and in vegetation from all atolls are very comparable, which rules out the possibility of contamination or contribution from a local source at any atoll. Concentrations of ^{210}Po in surface sea water from Enewetak and Bikini lagoons also are no different from levels determined in Kwajalein lagoon or in surface water outside the atolls in the North Equatorial Pacific Ocean. These concentrations are also similar to the mean value (see Table 5), compiled from data generated by others, in surface water from the 0–15°N latitude band (Cherry and Heyraud, 1988).

In the most recent growth sections of a living coral from Bikini the concentration of ^{210}Pb accumulated by the organism averaged 7.4 ± 1.1 Bq/kg (Noshkin et al., 1975). The average con-

Table 4
Local foods and ^{210}Po, ^{210}Pb intake per day from the Marshallese diet model

Local food	Concentration		Imported Food available (IA)		Imported Food unavailable (IUA)	
	^{210}Po (Bq/kg)	^{210}Pb (Bq/kg)	^{210}Po (Bq/day)	^{210}Pb (Bq/day)	^{210}Po (Bq/day)	^{210}Pb (Bq/day)
Reef fish	12.5	1.0	0.30	0.02	0.54	0.04
Pelagic fish	16.4	2.6[a]	0.29	0.05	0.77	0.12
Marine crab	9.0	0.1[a]	0.02	0.00	0.09	0.00
Lobster	11.2	0.2[a]	0.04	0.00	0.19	0.00
Clams and trochus	55.7	1.6	0.36	0.01	1.94	0.06
Coconut crab	40.8	0.2	0.12	0.00	0.53	0.00
Octopus	20[b]	1.2[c]	0.09	0.01	0.50	0.03
Turtle	16.4[d]	2.6[d]	0.07	0.01	0.09	0.02
Turtle eggs	16.4[e]	2.6[e]	0.15	0.02	1.92	0.30
Chicken flesh	0.3	0.3[f]	0.00	0.00	0.01	0.01
Chicken liver	1.1	1.1[f]	0.01	0.01	0.01	0.01
Chicken gizzard	0.6	0.6[f]	0.00	0.00	0.00	0.00
Chicken eggs	0.0	0.0[f]	0.00	0.00	0.00	0.00
Pork	0.3[g]	0.3[f]	0.01	0.01	0.01	0.01
Local bird flesh	33.0	0.1	0.09	0.00	0.44	0.00
Bird viscera	149.0	1.5	0.24	0.00	0.69	0.01
Bird eggs	46.0	1.0	0.07	0.00	0.52	0.01
Terrestrial vegetation	0.06[h]	0.03[h]	0.02	0.01	0.04	0.02
Water and water products	0.02[i]	0.02[i]	0.02	0.02	0.01	0.01
Total			1.89	0.17	8.31	0.65

[a]Estimated from appropriate ^{210}Pb/^{210}Po values in Table 2.
[b]No data for local sample. Value for *Octopus* from Guary et al. (1981).
[c]No data for local sample. Value for *Octopus* assumed identical to squid from Takata et al. (1968).
[d]No data for local sample. Value assumed equal to pelagic fish flesh.
[e]No data for local sample. Value assumed equal to flesh concentration.
[f]No data. Value assumed equal to ^{210}Po concentration.
[g]No local sample. Value assumed equal to chicken flesh concentration.
[h]Mean concentration for pandanus, breadfruit, forms of coconuts, papaya, squash, pumpkin and banana.
[i]Concentration in rainwater from Turekian and Cochran (1981) applies to all water/water products consumed.

Table 5
Some ^{210}Po comparative results[a]

Sample type	Concentration Bq/kg				
	Equatorial Pacific	Control Sites[b]	Bikini Atoll	Enewetak Atoll	Rongelap Atoll
Surgeonfish flesh		3.6 ± 4.5 (4)	2.3 ± 2.5 (8)		
Surgeonfish flesh[c]			1.6–9.4 (4)		
Mullet flesh		15.1 ± 7.8 (4)	10.7 ± 0.4 (9)	8.0 ± 3.6 (3)	16.6 ± 5.1 (2)
Goatfish flesh		16.8 ± 5.4 (10)	24.3 ± 8.1 (11)	17.0 ± 6.2 (5)	
Pelagic fish flesh			26.0 ± 15.7 (4)	14.6 ± 14.6 (14)	
Coconut crab flesh			30 ± 19 (3)	41 (1)	44 ± 13 (7)
Breadfruit pulp		0.041 ± 0.013 (3)	0.030 ± 0.019 (16)	0.026 (1)	0.01 (1)
Coconut pulp		0.044 ± 0.001 (3)	0.078 ± 0.066 (8)	0.11 ± 0.11 (7)	
Coconut juice		0.015 ± 0.006 (2)	0.005 ± 0.002 (5)	0.005 (1)	
Surface Seawater					
mBq/kg	1.15 ± 0.08 (2)		1.23 ± 0.10 (3)	1.12 ± 0.12 (7)	
mBq/kg (0–15°N)[d]	1.17 (25)				

[a] Number of samples in parenthesis; concentrations reported in fresh weight.
[b] Results for samples collected from Kwajalein, Pohnpei and Majuro.
[c] Results from Nevissi and Schell (1975).
[d] Mean concentration reported in surface sea water from 0–15° N latitude (Cherry and Heyraud, 1988).

centration reported in recent sections from different species of coral collected from regions of the Atlantic, Pacific and Indian oceans is 7.6 ± 3.7 Bq/kg (Shen and Boyle, 1987). The level in the Bikini coral is in good agreement with the mean concentration in coral from other global locations where no local sources of contamination are encountered.

In 20 samples of surface sediment (0–2 cm) collected recently inside Bikini lagoon, the average concentration of ^{210}Pb is 49 ± 31 Bq/kg which falls within the range and is somewhat less in value than 73 ± 20 Bq/kg reported by Beasley (1969) in samples of sediment (soil data not considered) from Bikini lagoon.

This additional comparative data defends the earlier conclusion that all ^{210}Pb (along with its granddaughter, ^{210}Po) detected in environmental samples from any of the atolls in the Marshall Islands is naturally occurring and further shows that the mean concentration associated with any identical environmental component is the same at all atolls. Therefore the concentrations can be used to assess uptake and dose to individuals with comparable diets inhabiting other islands in the Pacific.

4.3. Estimated concentrations of ^{210}Po and ^{210}Pb in imported foods

Concentrations of ^{210}Po and ^{210}Pb have been determined only in local food items from the Marshall Islands. Concentrations in the imported foods identified in Table 1 also must be estimated for a complete description of the dietary intake of the radionuclides.

Mean values for the concentration of ^{210}Pb are estimated for the foods listed in Table 1 (except for canned fish) from information provided for Japanese, USA, and UK diets (Takata et al., 1968; Morse and Welford, 1971; Smith-Briggs et al., 1986) and are shown in Table 6. In the USA and UK the average concentration of ^{210}Pb in marine foods is ~ 0.15 Bq/kg while in Japan the mean level from all fish analysed for the diet survey is computed to be 4.3 Bq/kg. The average value from these 3 studies would be 1.5 Bq/kg. However, according to the Marshallese diet survey, the preferred canned fish are tuna and mackerel. The mean concentration of ^{210}Pb in these fish, determined from the Japanese dietary data, is 0.8 Bq/kg. This value is similar to the mean concentration for ^{210}Pb in the muscle of fish (1.0 Bq/kg) determined in this study. We arbitrarily

Table 6
Imported foods and ^{210}Po, ^{210}Pb intake per day from the (IA) Marshallese diet model

Imported food	^{210}Po		^{210}Pb	
	Bq/kg	Bq/day	Bq/kg	Bq/day
Bread	0.096	0.01	0.096	0.01
Pancake-cake	0.096	0.01	0.096	0.01
Rice	0.042	0.01	0.042	0.01
Potatoes	0.032	0.00	0.032	0.00
Sugar	0.041	0.00	0.041	0.00
Canned meat	0.041	0.01	0.041	0.01
Canned chicken	0.041	0.00	0.041	0.00
Canned fish	1.500	0.22	0.800	0.12
Juice	0.042	0.02	0.042	0.02
Carbonated drinks	0.007	0.00	0.007	0.00
Powdered milk	0.040	0.00	0.040	0.00
Evaporated milk	0.040	0.01	0.040	0.01
Noodles (pasta)	0.032	0.00	0.032	0.00
Total		0.29		0.19

select 0.8 Bq/kg to represent the concentration of ^{210}Pb in any imported canned fish.

In the UK there is a deficiency found for ^{210}Po, relative to ^{210}Pb, in off-shelf samples of bread, cereal and sugar (Smith-Briggs et al., 1986). This deficiency can also be expected in similar foods from any country that exports goods to the Marshall Islands. However, it will be assumed that sufficient time will elapse between the collection (packaging) of these items by exporting countries and delivery to the Marshall Islands to ensure that ^{210}Po will have grown into equilibrium with ^{210}Pb by the time the foods are eaten. Therefore the ^{210}Po concentration in all imported foods, except for canned fish, shown in Table 6 is assumed to be equivalent to the ^{210}Pb concentration.

Data from Pentreath (1977) and the concentration ratios shown in Table 2 indicate there is a large initial excess of ^{210}Po in the flesh of fish. However, if there is a time lapse between collection and ingestion, any excess ^{210}Po will be reduced by radioactive decay and some amount of ^{210}Po will grow in from the decay of ^{210}Pb. We assume a time of 1 year is not unreasonable for processed fish in cans to reach a dinner table in the Marshall Islands. If the original ratio of ^{210}Pb/^{210}Po in freshly canned mackerel or tuna is 0.16 (mean value from Table 2 for flesh) and the concentration of ^{210}Pb is 0.8 Bq/kg (see above), then the concentration of ^{210}Po in the canned fish after one year is 1.5 Bq/kg. We use 1.5 Bq/kg as the value for ^{210}Po in imported fish but acknowledge that the actual amount of ^{210}Po associated with any canned fish delivered to the Marshall Islands will vary and, in part, depend on the efficiency of industrial processing and commercial transport.

4.4. Estimated concentrations of ^{210}Po and ^{210}Pb in drinking and household water

Table 1 shows that ~ 1.0 kg of water, in different forms, is consumed daily. Rainwater is the preferred and main source of water for drinking and cooking even if a good groundwater supply is available. A variety of cisterns are encountered in the Marshall Islands that store rainwater collected from residence or municipal roof catchment systems. Turekian and Cochran (1981) determined the concentration of ^{210}Pb in rainwater on Enewetak during 1979. The mean concentration was 0.022 Bq/kg. Rainfall and ^{210}Pb concentrations can change from year to year. There is no established program to monitor ^{210}Pb in rain so that this concentration is assumed to apply to both ^{210}Po and ^{210}Pb in annual collections of rainwater used for drinking anywhere in the Marshall Islands during past, present, and future years. This is a reasonable value for drinking water since it compares well with concentrations reported in other sources of municipal water. It is approximately half the mean level for ^{210}Pb (0.04 Bq/kg) reported in UK drinking water (Maul and O'Hara, 1989) and leads to a daily intake of both radionuclides which is ~ 2 times the 0.018-Bq average from USA community drinking water supplies (Cothern et al., 1986).

5. Discussion

5.1. ^{210}Po in the environmental samples

Concentrations of ^{210}Po measured in the flesh of species of fish from the lagoons at Marshall Island atolls were generally higher than reported concentrations in flesh (and other tissues) of different species from northern European waters (Camplin and Aarkrog, 1989). It is reported that in these waters 'concentrations of this nuclide in fish tend to be relatively low and rarely greater than 10 Bq/kg' (Camplin and Aarkrog, 1989). The mean concentration in the flesh of 9 of the 17 species of reef and pelagic fish collected from the Marshall Islands is greater than 10 Bq/kg.

The Marshall Island fish results can only be directly compared (species with species) with one previous study (Nevissi and Schell, 1975) where it was determined that the average concentration of ^{210}Po in the flesh of surgeonfish collected from Bikini in 1972 was between 1.6 and 9.4 Bq/kg wet weight. Several months elapsed between the collection and analysis of these samples. Only ^{210}Po was extracted and measured, so these values represent (according to the authors) lower and upper limits if, first, the ^{210}Po was derived entirely from the decay of ^{210}Pb in the sample or, second, little ^{210}Pb was present and the ^{210}Po measured was the true concentration present at the sampling time. The mean concentration of ^{210}Po we find in the flesh of surgeonfish is 4.5 Bq/kg, a value that falls between the limits given by Nevissi and Schell (1975). These comparative results are shown in Table 5.

We supplied the IAEA Marine Environment Laboratory (MEL) in Monaco several replicate terrestrial and marine samples from the Marshall Islands. Independent determinations of ^{210}Po were made on these samples. The results agreed with our measurements of ^{210}Po levels in the flesh of fish, in invertebrates, and in samples of terrestrial vegetation.

It appears that the mean concentration of ^{210}Po in the flesh of many fish from lagoons of coral atolls in the equatorial Pacific is generally higher than the mean level of ^{210}Po encountered in different species of fish from colder, northern European waters.

There are distinct differences in the mean concentration of ^{210}Po among species of the same trophic levels as seen, for example, in Table 3 between mullet and surgeonfish (trophic level 2); between bonefish and goatfish (trophic level 3); and among the larger pelagic carnivores. Cherry et al. (1989) suggest that the differences in body burdens of ^{210}Po may be traced to differences in the type of food consumed. Feeding habits of reef species from the same trophic levels are very different. The main source for ^{210}Po accumulated by fish is believed to be the food chain (Pentreath, 1977; Cherry et al., 1989); therefore it is not unreasonable that levels in different food may influence the levels of ^{210}Po noted among tissues of different species of fish. Note in Table 3 that the concentration associated with the contents removed from the viscera varies significantly, confirming that there are large differences in the amount of ^{210}Po with the material ingested by fish.

In spite of finding comparable mean concentrations in the different species of fish from the different atolls, levels in individual fish of the same species can vary significantly as indicated by the range of 0.4–25.2 Bq/kg for ^{210}Po encountered, for example, in the flesh of surgeonfish (Table 3). Pentreath et al. (1979) and others (Cherry and Shannon, 1974) have noted similar large variations in flesh concentrations within species. These differences are interesting observations but are not yet explained on a quantitative basis.

Unlike fish, the concentrations of ^{210}Po in the flesh of crabs and clams from the Marshall Islands are comparable with the levels measured in tissues of mollusca and crustacea collected from the UK and elsewhere (Pentreath and Alington, 1988; Rollo et al., 1992).

Concentration factors for ^{210}Po in muscle to that in filtered sea water have been calculated using a mean value of 1.15 mBq/l (see Table 5) for ^{210}Po in seawater. In reef species, values range from 0.5×10^3 for unicorn fish to 2×10^4 for goatfish. Values for pelagic species span a comparable range from 0.4×10^3 to 3.7×10^4. The concentration factors for the edible parts of mollusca and crustacea are 48×10^3 and 9×10^3, respectively. The mean value computed for flesh from all fish in the Marshall Islands is 1.2×10^4. This concentration factor is two times larger than the mean value computed for muscle of epipelagic teleosts (sardines, mackerel, tuna, etc.) from the Atlantic (Carvalho, 1988).

Terrestrial vegetation samples from the atolls are low in both ^{210}Po and ^{210}Pb. The radionuclides are not effectively transferred to any of the terrestrial food crops, hence organisms feeding only on vegetation are expected to contain low concentrations of ^{210}Po.

Nesting seabirds found on land rely on the marine rather than the terrestrial environment for food as seen in Table 3 from the relatively high ^{210}Po levels in the flesh and viscera. A squid was also identified among the gut contents of one bird, an observation that confirms the source of food for these birds. Considerable ^{210}Po is also found associated with the eggs of seabirds. The chicken, as well as the eggs of this bird, are low in ^{210}Po reflecting a diet of terrestrial food, as anticipated.

The relatively high levels of ^{210}Po associated with the flesh (body and claw) from the Coconut crab, *Birgus latro*, from Rongelap and Enewetak were, at first, considered anomalous. It was assumed that the animal always foraged for food, low in ^{210}Po, in the terrestrial environment. However, Reese (1987) indicates that the crab can often be found eating animal (dead fish) or vegetable remains as well as fruit and probably bird eggs and is readily attracted to almost any kind of human food. Two crabs, having relatively high levels of ^{210}Po associated with the flesh (both claw and body), were collected from the island of Enidrik at Bikini Atoll. This island has no coconut trees and the crabs were captured while in the act of eating a whole seabird. Seabirds must now also be considered part of the diet. The organs and tissues of seabirds and fish are high in ^{210}Po while levels in terrestrial vegetation are low. Therefore, to account for the relatively high body burdens of ^{210}Po we concluded that crabs preferred marine foods rather than terrestrial foods. However, new data proved this conclusion wrong. Two additional crabs were obtained from the island of Bikini at Bikini Atoll. This island has no nesting seabirds. The level of ^{210}Po found in the flesh removed from these crabs averaged 0.46 Bq/kg, 2 orders of magnitude lower than the average level in muscle from crabs residing on the island of Enidrik with nesting seabirds. The crabs from Bikini Island must subsist mainly on a terrestrial diet, showing that the animals are true opportunistic scavengers of any marine or terrestrial foods. ^{210}Po appears to be a good diet-indicator for the types of foods recently consumed by

Table 7a
^{210}Po and ^{210}Pb intake from the Marshallese diets

	Total intake		^{210}Po		^{210}Pb	
	kg/day	kg/year	Bq/day	Bq/year	Bq/day	Bq/year
Imported and local foods available (IA) for consumption						
Imported food	1.99	726	0.29	106	0.19	69
Local food	1.33	485	1.89	690	0.17	62
Total	3.32	1211	2.18	796	0.36	131
Only local foods available (ILA) for consumption						
Local food total	1.54	560	8.31	3033	0.65	237

Table 7b
Comparative results from some other countries[a]

	^{210}Po (Bq/year)	^{210}Pb (Bq/year)
USA	22	19
Germany	62	62
USSR	54	84
Argentina	18	
Japan	176	230
India	21	
Special cases (Arctic dwellers)		
Canada	1351	
Finland	932	116
Alaska	1351	135
USSR	540	540

[a]Data from Holtzman (1980).

the species. The concentrations in these later samples from Bikini Island are not included among the values used to generate the average listed in Table 3.

5.2. Intake of ^{210}Po and ^{210}Pb associated with local and imported foods

The annual intake of ^{210}Po and ^{210}Pb is computed from the dietary information provided in Tables 1, 4 and 6 and is shown in Table 7a. Also shown, for comparison, in Table 7b are values of annual intake from other countries, abstracted from the review by Holtzman (1980). The average annual intake of ^{210}Po and ^{210}Pb in the current (IA) diet is higher than amounts ingested with foods elsewhere in the world outside the Arctic. The UNSCEAR (1988) shows that the average annual intake of ^{210}Po and ^{210}Pb in diets from 'normal areas' is 40 Bq in both cases. The estimated annual intake of ^{210}Po in the Marshall Islands is 20 times greater than this value and the ^{210}Pb intake is 3 times greater. Eighty-seven and seventy-four percent of ^{210}Po and ^{210}Pb, respectively, in the total Marshall Island diet is derived from the local and imported aquatic foods, including seabirds.

Total food intake associated with the IUA diet is 46% of the amount in the IA diet but ingestion of ^{210}Po and ^{210}Pb is seen to be approximately 4 and 2 times greater in the IUA diet. Intake of ^{210}Po with foods in the IUA diet exceeds the quantity ingested with any Arctic diet shown in Table 7b. The IUA diet represents the minimum amount of local food necessary for survival. Intake of ^{210}Po with food was probably higher in previous generations at the atolls when only local food was available for consumption. Changing lifestyle from a domestic food gathering society to one relying on imported foods has resulted in a significant reduction in the dietary intake (and in the corresponding dose) of the two naturally occurring radionuclides.

5.3. Dose models

A preliminary dose estimate from ingestion of ^{210}Po associated with local Marshall Island reef fish was provided by Robison et al. (1987) using the guidelines recommended in ICRP 30 (ICRP, 1979). There are other recommended guidelines (Kendall et al., 1987; Eckerman et al., 1988) based on criteria in ICRP 30. The conversion factors recommended in these publications have also been used with concentrations of ^{210}Po in different foods (Smith-Briggs et al, 1986; Pentreath and Alington, 1988) to estimate dose to adults from ingestion. However, during the last few years there have been a number of changes suggested for the gut uptake factor and the tissue weighting factors for ^{210}Po and ^{210}Pb.

ICRP 60 (ICRP, 1991b) recommended significant changes in tissue weighting factors that resulted in a reduction of the numerical value for the dose coefficients previously used in ICRP 30. Phipps et al. (1991) updated NRPB data on dose per unit intake based on these new ICRP recommendations and Rollo et al. (1992) used these updated values in a recent assessment of ^{210}Po dose to individuals in the UK from sea food consumption.

A significant factor in calculating the dose from ingestion of ^{210}Po and ^{210}Pb is the choice of a gut transfer factor. Holtzman (1980) discussed early work which suggested that the intestinal absorption of ^{210}Po ingested with food could be several times the value of 0.1 used in ICRP 30 or in ICRP 60. Interestingly, Hunt and Alington (1993) point out that the value of 0.1, the recommended value until recently, was based only on a single case of oral administrated ^{210}Po as an inorganic salt to a volunteer in 1950 and some supplementary data for rats. An expert group convened by the Nuclear Energy Agency (NEA) reviewed the existing data and recommended a value of 0.3 be used for the gut transfer factor of Po and that it be increased to 0.3 from 0.2 for ^{210}Pb (Phipps et al., 1991). The NRPB (Phipps et al, 1991) considered the value of 0.3 for ^{210}Po to be over-cautious and recommended the continued use of 0.1. Hunt and Alington (1993) conducted a series of recent experiments with human volunteers and demonstrated that the gut absorption factor for ^{210}Po could be increased to ~ 0.8.

The ICRP is re-evaluating the ingestion dose coefficients for ^{210}Po and ^{210}Pb and is now recommending 2.3×10^{-6} Bq/Sv for ^{210}Po and 1.5×10^{-6} Bq/Sv for ^{210}Pb as the values for the adult effective dose per unit intake (K.F. Eckerman, personal commun.).

Table 8 lists the different dose conversion factors and Table 9 shows the differences in committed effective adult dose using both the IA and IUA diets and some of the suggested models. There is an order of magnitude difference between the lowest and highest value for committed effective dose from ingestion of ^{210}Po using the different factors. It is therefore impractical to compare dose from ingestion of the radionuclide with other values published in the literature. We believe the latest recommendations (E.F. Eckerman, personal commun.) from the ICRP are the best currently available to estimate dose from ingestion. Using this model the annual combined effective dose from ^{210}Po and ^{210}Pb ingested with foods in the IA diet is ~ 2 mSv (200 mrem). The IUA diet leads to an annual effective dose of ~ 7.3 mSv (730 mrem).

The average annual effective dose from natural background sources in most areas of the world is 2.4 mSv (UNSCEAR, 1988). The major contribution (> 60%) to this natural exposure is from radon. Exposure to radon is insignificant in the

Table 8
Some recent guidelines and recommendations for dose from ingestion of ^{210}Po and ^{210}Pb

Source	^{210}Po		^{210}Pb	
	Gut absorption factor (f_1)	Dose/unit intake Sv/Bq	Gut absorption factor (f_1)	Dose/unit intake Sv/Bq
ICRP 30 (ICRP, 1979, 1981)	0.1	4.4×10^{-7a}	0.2	1.36×10^{-6a}
Kendal et al., (1987)	0.1	4.3×10^{-7a}	0.2	1.4×10^{-6a}
Eckerman et al., (1988)	0.1	5.1×10^{-7a}	0.2	1.45×10^{-6a}
NEA (1988)[d]	0.3		0.3	
ICRP 61 (ICRP, 1991)[c]	0.1	2.2×10^{-7b}	0.2	1.0×10^{-6b}
Phipps et al. (1991)	0.1	2.1×10^{-7b}	0.2	8.6×10^{-7b}
Phipps et al. (1991)	0.3	6.2×10^{-7b}	0.3	1.3×10^{-6b}
Hunt and Alington (1993)	0.8			
Eckerman (Pers. Commun., 1993)	0.5	2.3×10^{-6b}	0.3	1.5×10^{-6b}

[a]Committed effective dose equivalent.
[b]Committed effective dose.
[c]Committed effective dose computed from Annual Limits of Intake.
[d]Discussed in Phipps et al. (1991).

Table 9
Committed effective dose for adults from intake of ^{210}Po and ^{210}Pb in the Marshallese diet using different dose models shown in Table 8

Dose coefficients[a]	Diet[b]	f_1 values		Intake (Bq/year)		mSv/year[c]		
		^{210}Po	^{210}Pb	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb	Total
Mean of 1, 2, 3	IA	0.1	0.2	796	131	0.37	0.18	0.55
Mean of 1, 2, 3	IUA	0.1	0.2	3033	237	1.39	0.33	1.72
Mean of 1, 2, 3 with 4	IA	0.3	0.3	796	131	1.11	0.27	1.38
Mean of 1, 2, 3 with 4	IUA	0.3	0.3	3033	237	4.17	0.50	4.67
Mean of 5, 6a	IA	0.1	0.2	796	131	0.17	0.12	0.29
Mean of 5, 6a	IUA	0.1	0.2	3033	237	0.65	0.22	0.87
6b	IA	0.3	0.3	796	131	0.52	0.18	0.70
6b	IUA	0.3	0.3	3033	237	1.95	0.33	2.28
5 with 7	IA	0.8	0.2	796	131	1.40	0.13	1.53
5 with 7	IUA	0.8	0.2	3033	237	5.34	0.24	5.58
8	IA	0.5	0.3	796	131	1.83	0.20	2.03
8	IUA	0.5	0.3	3033	237	6.98	0.36	7.34

[a] Numbers are from column 1, Table 8.
[b] Diet IA is for imported and local food available; diet IUA is for imports unavailable.
[c] Committed effective dose or committed effective dose equivalent. Multiply by 100 to convert dose values to mrem/year.

Marshall + Islands because of the maritime conditions, low concentrations in soil of the parent radium radionuclide, and because of the open, outdoor lifestyle of the Marshallese people (Robison et al., 1987). The external dose from terrestrial radiation and cosmogenic radionuclides is very low (0.02 mSv/year) so that most of the natural background dose is due to the external cosmic radiation and food ingestion pathways. The dose from cosmic radiation is ~ 0.22 mSv/year (Gudiksen et al., 1976) and naturally occurring ^{40}K contributes 0.18 mSv/year to the internal dose (Robison, et al. 1987). Including the dose from ingestion of ^{210}Po and ^{210}Pb, the total effective dose from natural background sources in the Marshall Islands is, like other areas of the world, also 2.4 mSv. However, unlike continental areas, 83% of the annual background dose is presently derived from ingestion of ^{210}Po and ^{210}Pb associated with indigenous food.

It is suggested that the contribution to the natural background effective dose experienced by other global societies from ingestion of ^{210}Po (and ^{210}Pb) should be re-evaluated, especially for consumer groups with high intake of different seafoods.

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