(Reprinted from Nature, Vol. 262, No. 5571, pp. 745–748, August 26, 1976)

Plutonium levels in Kwajalein Lagoon

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Reported plutonium levels in fish from both Kwajalein and Enewetak lagoons, suggest that Kwajalein Lagoon contains significantly more plutonium in its environment than would be expected from worldwide fallout levels alone, although quantities of plutonium greater than fallout concentrations have not been detected in the lagoon water. If there is no reason to reject the published&h data, then individuals on Kwajalein Atoll who supplement their diet with foods from the local marine environment may have plutonium body burdens similar to the low levels predicted for individuals on similar diets at Enewetak Atoll.

KWAJALEIN ATOLL (9°N 167°40′E) is located in the western (Ralik) chain of the Marshall Islands. In the same island chain and some 300 miles to the north-west is Enewetak Atoll (1 I °20′N 162°20′E), one of the United States nuclear testing sites during the 1940s and 1950s. It has been reported' that no immediate debris from the nuclear tests at Enewetak and Bikini (another Pacific test site) was deposited at Kwajalein. Measurements during I972 and I973 between 5 and 15°N, over a wide range of longitudes, indicated concentrations of between 0.22 and 0.44 pCi m^{-3} for $^{239,240}Pu$ in Pacific surface water[2,3]. The mean $^{239,240}Pu$ concentration in the surface waters of Kwajalein and other Pacific atoll lagoons located in this latitude band, therefore, would be expected to be similar to the surface oceanic level (0.34 $\pm$ 0.11 pCi m^{-3}).

This is not true of Enewetak Lagoon. In late 1972, the average measured concentration of $^{239,240}Pu$ in the lagoon surface water[3] was 39 pCi m^{-3}, approximately 100 times the level predicted from worldwide fallout. Clearly, some components of the atoll contaminated by fallout debris during the tests are contributing substantial amounts of $^{239,240}Pu$ to the lagoon water masses. The difference in $^{239,240}Pu$ concentrations between the two lagoons is a direct reflection of the activity levels in the environments of the two atolls. It also follows that if plutonium uptake in living organisms is expressed in terms of a concentration factor (the concentration of plutonium in the tissue of the marine organism divided by its concentration on an equivalent weight basis in the surrounding water), invertebrates, fish, or other marine organisms can be useful as indicator species of the level of environmental contamination.

Schell and Watters[4] have given $^{239,240}Pu$ concentrations in various organs of selected marine organisms collected at Enewetak and Kwajalein atolls. They concluded, however, that the plutonium and americium concentrations in the convict surgeon fish from Enewetak Atoll, for example, are not significantly higher than those measured at the control station, Kwajalein Atoll. The mantle and muscle tissues of a clam (*Tridacna* sp.) collected in the south-eastern region of Enewetak Atoll were, moreover, found to contain only one-sixth as much plutonium as those of a *Tridacna* sp. collected at Kwajalein Atoll[4]. On the other hand, the viscera and kidney of the same Enewetak Atoll clam had higher concentrations than those of the Kwajalein Atoll specimen.

In an assessment of the possible plutonium dose to man from food and other environmental pathways, Wilson et *al.*[5] compiled, in the form of lognormal median concentrations, all the plutonium survey data collected at Enewetak Atoll during 1972 and 1973 (ref. 1). The concentration values for all the fish muscle samples taken are reproduced here in Table 1, together with similar lognormal mean concentrations compiled from the same survey[1] data for fish bone and gut from Enewetak Atoll and muscle, bone, and gut concentrations, also from the same survey data[1], for fish collected off three islands of Kwajalein Atoll (W. R. Robison, private communication). Wilson *et al.*[5] found no significant differences in mean plutonium concentrations among four fish groups, which included, among others, planktonic and detritus feeders, grazing herbivores, bottom-feeding carnivores, and pelagic carnivores. Accordingly, they based their predictions of exposures to plutonium from ingestion of marine foods on the mean concentration of plutonium in the muscles of all the fish taken from the lagoon of Enewetak Atoll. Significantly, however, the $^{239,240}Pu$ in the lagoon water ranged from 1 to 96 pCi m^{-3} at the fish sampling sites'.

Radionuclide concentration independent of environment?

The data of Schell and Watters[4], together with those of Table 1, show that average levels of $^{239,240}Pu$ in fish bone, muscle, and gut from Enewetak Atoll are similar to, or even lower than, those of the fish indigenous to the control station, Kwajalein Lagoon. There are several possible important conclusions that can be derived from these data. The first is that fish collected for consumption by man will contain, on average, essentially the same concentrations of plutonium radionuclides regardless of the source or level of plutonium in the local environment. Obviously, such a conclusion would greatly affect future plans for releasing low-level transuranics to the marine environment. Furthermore, it would force us to concede that the concept of a plutonium concentration factor for fish is meaningless. Also, it conflicts with a large body of plutonium concentration data for Atlantic fish species that derive their plutonium body burdens from worldwide fallout levels in the Atlantic Ocean. For example, for a number of Atlantic species, including bottom feeders, water-column feeders, and large predators[6-8], a lognormal median of all available bone concentration data is only 1 x 10^{-4} pCi g^{-1} (wet or dry), 900 times less than that for the bones of the Kwajalein fish (Table 1). A similar large discrepancy remains when the concentrations in the muscle of the Atlantic and Kwajalein fish are all normalised to an equivalent weight basis (wet or dry).

Plutonium levels in the Atlantic waters (where some of the fish were caught) range only between 0.2 and 1 .1 pCi m^{-3}, according to Bowen *et al.*[8]. Calculating a concentration factor from the data for Atlantic fish and water and using a value of 0.4 pCi m^{-3} as the assumed mean plutonium level from fallout in the Kwajalein Lagoon, provides values of between 0.2 and 1.0 10^{-4} pCi g^{-1} for the bone of fish for this lagoon. These values are orders of magnitude lower than the lognormal median

Table 1 Lognormal median concentrations of $^{239,240}Pu$ in fish tissues (pCi g^{-1} dry weight) collected at Enewetak and Kwajalein atolls*

	Enewetak Atoll (all samples)		Kwajalein Island		Kwajalein Atoll Meck Island		Enewetak Island	
All fish species								
Bone	0.038	(24)	0.086	(3)				
Muscle	0.013	(123)	0.023	(11)	0.01	(6)	0.53	(3)
Gut	0.45	(6)	0.051	(3)			0.42	(2)
Surgeonfish								
Muscle	0.028	(28)	0.02	(3)			0.96	(1)
Gut	0.019	(26)	0.03	(2)			0.43	(1)
Mullet								
Muscle	0.014	(25)						
Gut	0.75	(19)						
Goatfish								
Muscle	0.008	(21)						
Gut	0.093	(18)						
All other fish								
Bone	0.038	(24)	0.086	(3)				
Muscle	0.009	(49)	0.024	(8)	0.01	(6)	0.40	(2)
Gut	0.25	(44)	0.14	(1)			0.41	(1)

*See refs 1 and 5.

†Values in oarentheses are numbers of samnles analysed. Average muscle and gut values in pCi g^{-1} dry weight can be converted to average pCi g^{-1} wet weight by dividing by 3.5.

concentrations in Table 1. This discrepancy cannot be accounted for by any possible differences related to trophic levels or feeding habits. A similar calculation also yields large discrepancies between predicted and measured concentrations for fish muscle.

Excess plutonium at Kwajalein

An alternative explanation to account for these discrepancies would be that Kwajalein Lagoon contains significantly more plutonium in its environment than would be expected from worldwide fallout levels alone. To test this possibility, 55 l unfiltered water samples were collected during May and June of 1975 from the locations shown in Fig. 1. During June, two samples were also collected outside the Atoll in the north equatorial surface waters, to provide information on the plutonium levels in the open ocean in this region. Unfortunately, our schedule did not allow time to collect fish on Kwajalein Atoll.

The water samples were analysed for $^{239,240}Pu$ and ^{137}Cs using the methods described in refs 9 and 10. Our analytical results are shown in Table 2. Although the lagoon was not sampled in great detail, the data are sufficient to show that the average $^{239,240}Pu$ concentration in Kwajalein Lagoon (0.45 ± 0.21 pCi m^{-3}) is nearly the same as the mean for the surface water of the ocean in the area, and that it agrees reasonably well with the levels previously predicted from worldwide fallout.

At the time the Kwajalein water samples were being processed we were also participating in an intercomparison exercise with Woods Hole Oceanographic Institution to determine

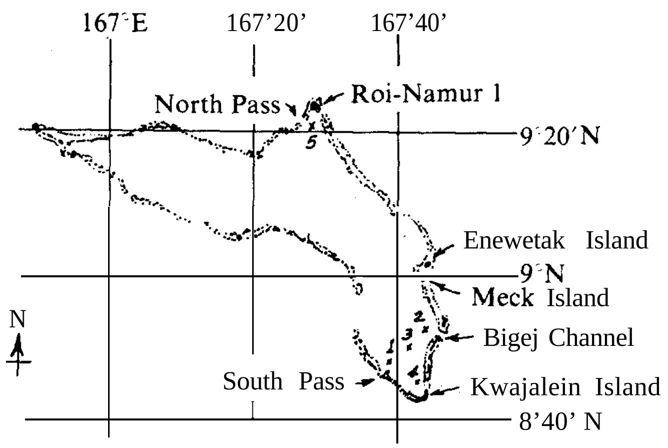

Fig. 1 Locations of sampling sites (see Table 2).

$^{239,240}Pu$ levels in replicate surface-water samples from one location in the North Atlantic. The mean value for eight samples from Woods Hole (V. T. Bowen, unpublished) was 0.63 ± 0.16 pCi m^{-3}, whereas ours for 10 samples was 0.70 ± 0.30 pCi m^{-3}. This analytical agreement (as well as that in other national and international intercomparisons that we have completed) lends a measure of confidence to our data. In addition, these comparative data show that, as expected from worldwide depositional data", average plutonium levels are somewhat lower in Kwajalein Lagoon than in

Table 2 Concentrations of $^{239,240}Pu$ and ^{137}Cs (pCi m^{-3}) in seawater in Kwajalein Lagoon and two locations in north equatorial waters

Station*	Depth (m)	Collection date	$^{239,240}Pu$	^{137}Cs	$^{239,240}Pu/^{137}Cs$
1	Surface	5/10/75	0.33 (20)†	137 (3)	0.0024 (20)
1	44	5/10/75	0.87 (13)	144 (5)	0.0060 (14)
2	Surface	5/08/75	0.29 (27)	131 (5)	0.0022 (27)
3	Surface	5/08/75	0.26 (24)	127 (3)	0.0020 (24)
3	47	5/08/75	0.33 (25)	129 (3)	0.0026 (25)
4	Surface	5/08/75	0.54 6)	129 (4)	0.0041 (16)
5	Surface	6/14/75	0.52 it 8)	132 (4)	0.0039 (18)
10°26'N 166°31'E	Surface	6/15/75	0.36 (32)	132 (3)	0.0027 (32)
11°16'N 165°45'E	Surface	6/15/75	0.53 (23)	143 (4)	0.0037 (23)

*Kwajalein stations are shown in Fig. I.

†Values in parentheses are the lo counting errors expressed as percentages of the listed values.

North Atlantic surface waters. The average $^{239,240}Pu/^{137}Cs$ ratio of 0.0032 for Kwajalein Lagoon is also in good agreement to the average value for open ocean, in contrast to the ratio of 0.074 12 in the Bikini and Enewetak lagoons[3]. This illustrates that both the $^{239,240}Pu$ and the ^{137}Cs content of the lagoon are derived from worldwide fallout.

We are still without an obvious explanation for the discrepancy in the fish-tissue data; that is, why should the Kwajalein fish, in a region of the equatorial Pacific contaminated only by worldwide fallout, have significantly higher body burdens of plutonium than fish in the North Atlantic which is also contaminated only with worldwide fallout? Furthermore, why do the Kwajalein Atoll fish have the same or even higher body burdens than the fish at Enewetak Atoll, where higher levels of plutonium are found in the lagoon? And why were the concentrations in Enewetak Lagoon fish not correlated with those in the specific local environment where they were sampled, or with feeding habit or trophic level? A cursory search of the literature surprisingly revealed only one other relevant fallout report-the plutonium level in the body of a single marine fish in fallout-contaminated waters of the Pacific[12]. The plutonium concentration in this sample and the computed concentration factor are closer to those from the North Atlantic fish than to those from Kwajalein. On the other hand, plutonium levels in tissues from catfish from Trombay[13], in plaice from the vicinity of Windscale[14], and in several species from areas of Thule[15] are higher than those in the tissues of the North Atlantic fish, and close in value to those from Kwajalein and Enewetak. But as the last three sets of data are from areas known to have local plutonium contamination from reprocessing plants (and one nuclear accident), they support our argument that levels of plutonium in fish reflect local environmental levels.

To sum up, it appears that available plutonium data from fish tissues are inconsistent. If we accept the available Enewetak and Kwajalein Atoll data as correct, then we can only conclude that the availability to all fish and invertebrates of plutonium from fallout, or for that matter from any local source, depends more on the type of environment than on the plutonium levels in that environment. The data could also suggest that coral atolls may have specific biogeochemical processes that regulate the availability of plutonium regardless of levels in the environment or in food. On the other hand, if we assume that concentrations in fish and invertebrates are proportional to those in the environment, then we can only conclude that many of the atoll fish data from the laboratories involved in analytical programmes[1,4] are in error. Since there is no other evidence at this time to refute the analytical results from the atolls, we can only urge caution in applying concentration factors measured in one marine area to predict approximate plutonium levels in fish from other marine areas. In addition, it seems, on the basis of the published data[1,4] and the summary assembled in Table 1, that individuals on Kwajalein Atoll who supplement their diet with foods from the local marine environment may have plutonium body burdens similar to the low levels predicted[5] for individuals at Enewetak on similar diets.

This work was performed under the auspices of the US Energy Research and Development Administration.

Received April 14; accepted June 26, 1976.

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Printed in Great Britain by Henry Ling Ltd., at the Dorset Press, Dorchester, Dorset