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PLUTONIUM CONCENTRATIONS IN FISH AND SEAWATER FROM  
KWAJALEIN ATOLL

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## Abstract

A followup study has been made to assess the concentrations of  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$  in the marine environment of Kwajalein Atoll. Fish collected from the Atoll in 1972 had body burdens of plutonium that were substantially higher than concentrations in similar species from locations contaminated only with global fallout. Our recent results, however, indicated that Kwajalein lagoon seawater contained levels of plutonium more similar to global fallout levels found in north equatorial Pacific surface waters. No satisfactory explanation for the reported plutonium levels in fish from Kwajalein collected in 1972 could be deduced from the available data. The highest plutonium concentrations reported for the 1972 reef species of fish could expose man, through ingestion of marine foods, to a dose rate as high as 25% of the (3 mrad/yr over 70 yrs) proposed EPA guideline for annual total transuranic dose rate to bone. Our present results show the dose rate from the marine food pathway is nearer to 0.005% of the recommended EPA value and are consistent with the view that Kwajalein Atoll contains plutonium concentrations that are expected from global fallout.

The magnitude of the plutonium levels reported in fish collected from Kwajalein lagoon during 1972 was excessively high and these results appear to be inconsistent with other environmental data from the lagoon.

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## Introduction

In a previous report (No 76) describing the concentrations of specific radionuclides detected in the lagoon water of Kwajalein Atoll (9°N 167°40'E), it was concluded that the concentrations of  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$  were similar to the global fallout concentrations detected in north equatorial Pacific surface waters. However, samples of fish collected from Kwajalein Atoll in 1972 (Ne 73) contained concentrations of  $^{239+240}\text{Pu}$  that were higher than levels in fish from other global regions contaminated only with global fallout debris. Another study had reported (Sc 75) that the concentrations of plutonium in convict surgeon fish collected from regions of Enewetak Atoll, one of the Pacific nuclear test sites used by the U.S. between 1948 and 1958, were not significantly different than levels in fish from the control station, Kwajalein Atoll. This was unexpected because during 1972, 1974 and 1976 the average concentration of  $^{239+240}\text{Pu}$  in Enewetak lagoon water (No 78a) was 60 to 100 times higher than the 1975 average Kwajalein lagoon water concentration of  $0.45 \pm 0.21 \text{ pCi/m}^3$  (No 76). One is compelled to submit, based on these data, that the use of a concentration factor to relate plutonium concentrations in marine organisms to environmental concentrations seems highly questionable. Based on these 1972 plutonium concentration data (Ne 73) for eviscerated whole fish and the average concentration in 1975 Kwajalein lagoon seawater, the calculated concentration factors for the Kwajalein fish would range from  $2 \times 10^3$  to  $6 \times 10^5$ . These values are orders of magnitude larger than other reported (Ne 75, No 78b, He 75, No 72) concentration factors for plutonium in fish.

The  $^{239+240}\text{Pu}$  concentrations of eviscerated and whole reef fish from Enewetak Island of Kwajalein Atoll in 1972 ranged from 230-960 pCi/kg dry

ingestion pathway is sufficient reason to warrant a re-examination of the concentrations of plutonium at Kwajalein and to resolve some of the questions raised from the previous report (No 76). Concentrations of plutonium in water, fish and sediment in recently collected samples from Kwajalein and north equatorial surface waters are discussed and compared to previously reported data.

### Sample Collections and Analytical Results

During 1976, 50 liter unfiltered seawater samples were obtained from lagoon locations 1, 5, 7, 8 and 10 shown in Fig. 1. Stations 1 and 5 duplicate locations sampled during 1975. Between 1975 and 1977 surface seawater samples were collected at several open ocean locations within the area of the Northern Marshall Islands. Plutonium and cesium were separated from the water samples and the concentrations of  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$  were determined using published methods (Wo 78, Wo 71). Our analytical results are shown in Table 1 along with 1975 lagoon seawater concentrations previously reported (No 76). In a few samples where the  $^{238}\text{Pu}$  concentrations were above our detection limits, the  $^{238}\text{Pu}/^{239+240}\text{Pu}$  ratios were computed and are given in Table 1. For comparative purposes, concentrations in surface seawater samples collected in the north equatorial Pacific since 1972, from the cited references, are also compiled in Table 1. At one ocean surface station where the particulate material collected on a 1  $\mu\text{m}$  filter was analyzed, the concentration of  $^{239+240}\text{Pu}$  associated with the filtered material represented 13% of the total concentration in the water.

Eighteen mullet (14 female and 4 male), Crenimugil crenilabis, were collected on October 21, 1976 from the lagoon reef off Enewetak Island at Kwajalein Atoll. The standard length of the fish varied from 265 to 386 mm. Reef fish collected from this island in 1972 (Ne 73) had the highest reported plutonium body burdens at Kwajalein Atoll. Mullet are primary consumers and represent species belonging to the second trophic level. Guary and Fraizier (Gu 77) maintain that concentration factors for

counted on Ge (Li) detectors to determine the concentrations of  
emitting radionuclides associated with the tissues. Each pooled sample was  
then radiochemically processed (Wo 71) to isolate plutonium. The dry

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being processed. Recent concentration data and concentration factors of plutonium determined in mullet tissue from several islands of Enewetak Atoll (No 78b) will be used to estimate the expected concentration in the missing tissues from Kwajalein fish.

$^{95}\text{Zr}$  was detected by gamma spectrometry only in the mullet viscera and stomach contents. The  $^{95}\text{Zr}$  in the mullet gut probably originates from recently ingested material contaminated with fresh fallout debris from the Chinese atmospheric nuclear test conducted on 26 September 1976. The concentration in the viscera and stomach contents were  $68.5 \pm 13.0$  and  $307 \pm 32$  pCi/kg wet weight, respectively, on the day of collection.

$^{137}\text{Cs}$  was above detection limits only in the snapper muscle tissue and its concentration was  $11.9 \pm 1.5$  pCi/kg wet weight.

Grab samples of lagoon reef sediment (0-11 cm) off Enewetak Island, Kwajalein Atoll were obtained in early 1976. The average concentration of  $^{239+240}\text{Pu}$  in 9 sediment samples was  $6.2 \pm 1.3$  pCi/kg dry weight and the mean  $^{238}\text{Pu}/^{239+240}\text{Pu}$  ratio was  $0.032 \pm 0.008$ . The estimated quantity of  $^{239+240}\text{Pu}$  in this sediment to a depth of 11 cm, is  $1.4 \pm 0.5$  mCi/km<sup>2</sup>.

This value falls within the range of fallout plutonium inventories ( $1.4 - 3.0$  mCi/km<sup>2</sup>) that we have determined in water columns from samples collected during 1973 in the north equatorial Pacific between 4° and 16° N latitude.

open ocean is slightly lower than the lagoon mean concentration but it is within the statistical uncertainty associated with the value.

The concentrations of  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$  in surface water collected in 1975 and 1976 at station 5 were essentially identical during the two years. Unlike the levels at station 5, the  $^{239+240}\text{Pu}$  concentrations at station 1 were different. Bottom water from station 1 in 1975 and 1976 surface water contained plutonium levels which were higher than the lagoon average. Station 1 is approximately 100 yards from the remains of the Prinz Eugen, a WW II German cruiser that was used as a target vessel during the Able and Baker nuclear events held at Bikini in 1946. The vessel was towed to Kwajalein after the tests and capsized on the lagoon shelf. Fish collected near the vessel in 1947 showed trace amount of radioactive material in the organisms (Hi 62). It is possible that even after 31

years, the remains of the Eugen are still contaminated and small quantities of plutonium are continuously remobilized to the water column near this source. However, if this should be the case, the amount remobilized is not in sufficient quantities to alter the average lagoon water concentration above fallout background levels.

Since 1972 the average surface seawater fallout concentrations of  $^{239+240}\text{Pu}$  in the north equatorial ocean have varied by no more than a factor of 2. The variations in surface concentrations are due to differences in the rates of new atmospheric fallout to the sea surface and to processes moving plutonium laterally and to depths in the water mass. We can assume that variations in the concentrations of plutonium no larger than those encountered in the open ocean since 1972, occurred in the lagoon water mass.

To summarize, the mean water concentration of  $^{239+240}\text{Pu}$  in Kwajalein lagoon during 1975 and 1976 was slightly higher, but not outside the statistical uncertainty associated with the average global fallout levels in northern equatorial surface waters. 1975 and 1976 north equatorial surface concentrations of plutonium were similar to the levels detected during 1972.  $^{238}\text{Pu}/^{239+240}\text{Pu}$  ratios measured in the 1976 water samples from the ocean and lagoon were similar in value which indicates that the radionuclides were derived from the same source. The upper 11 cm of sediment from one area of the lagoon contains a plutonium inventory within the range of the fallout inventories measured in the water columns of the north equatorial Pacific. We conclude that global fallout is the principal source of plutonium and cesium radionuclides found in the Kwajalein lagoon environment.

wet/dry and ash/dry weight ratios of the stomach contents and sediment were identical. The similarity in plutonium concentrations, isotopic ratio and weight ratios indicate that the stomach contents of the mullet consists largely of benthic sedimentary material. The stomach content of the snapper, on the other hand, was found to contain a much lower concentrations of plutonium, more typical of the levels associated with the flesh of marine organisms.

<sup>95</sup>Zr was detected in the viscera and stomach content samples of the mullet collected on 21 October 1976. The radionuclide was introduced to the aquatic environment sometime after 26 September 1976. Generally low <sup>95</sup>Zr concentration factors for marine fish species are measured in

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laboratory experiment which shows that  $^{95}\text{Zr}$  is poorly metabolized (Po 66). Since less than a month elapsed between the time the  $^{95}\text{Zr}$  was introduced to the environment and the time the fish were caught, and in view of the apparent low metabolism of  $^{95}\text{Zr}$ , it is reasonable to assume that most of the  $^{95}\text{Zr}$  measured in the viscera sample is associated with the intestinal contents. If so, this data could be used to estimate the amount of the intestinal contents in the viscera sample which in turn allows us to estimate the concentration of plutonium associated with the internal organs associated with the dissected viscera sample. We first calculate the amount of the contents in the GIT using the  $^{95}\text{Zr}$  specific activity of the separated stomach contents. Of the 638 dry grams of viscera sample separated from the 18 fish, 326 grams of the material could account for the total  $^{95}\text{Zr}$  measured in the viscera. The weight of the viscera without the computed gut contents is then 312 dry grams and has a wet/dry weight ratio of 4.39. The viscera plus computed gut contents contained a total of 2.92 pCi of  $^{239+240}\text{Pu}$ . 326 dry grams of contents at 5.00 pCi/kg (stomach content concentration) should contain 1.63 pCi of  $^{239+240}\text{Pu}$  leaving 1.29 pCi of plutonium associated with the viscera tissues and organs.  $^{239+240}\text{Pu}$  associated only with the wet viscera is then estimated from the above arguments to be 0.94 pCi/kg. This comparative analysis shows that a significant fraction of the plutonium measured in unseparated "viscera" sample is associated with the contents in the GI tract.

Although the muscle and gill samples of the mullet were lost during processing, the concentration of plutonium can be estimated from other data. An extensive analysis was recently completed to assess plutonium and

other radionuclide concentrations in mullet collected from 13 different islands of Bikini and Enewetak Atolls (No 78b). Representative lagoon water samples were also collected for analyses. Concentration factors for plutonium in the organs and tissues of mullet were computed from the analytical results (No 78b). Excellent agreement was found between predicted and measured concentrations in fish at Bikini using only concentration factors generated from Enewetak data and the measured water concentrations at Bikini. The average concentration factor for plutonium in wet mullet bone was  $139 \pm 54$  (No 78b) and the average concentrations of plutonium in the muscle, skin and gill tissues relative to the bone were 0.19, 2.6 and 6.2, respectively, on a dry weight basis. Using these relative values, the average measured Kwajalein lagoon seawater concentration of 0.54 fCi/l, and the wet/dry weight ratios in Table 2, we estimate that the wet Kwajalein mullet bone and skin tissue should contain  $^{239+240}\text{Pu}$  concentrations of 0.075 and 0.21 pCi/kg, respectively. The measured concentrations of plutonium in these tissues were 0.072 and 0.29 pCi/kg. The close agreement between the measured and predicted values indicates that there is a great deal of validity in the concept of a concentration factor and we feel justified in applying this predictive model to estimate the mullet muscle and gill concentrations at Kwajalein. We estimate that the muscle and gill concentrations should have been 0.0034 and 0.12 pCi/kg wet weight, respectively.

Recent measured concentrations of plutonium in coastal California Rockfish (Sebastes sp.) muscle and bone (No 78c) provides additional support to these arguments and suggests further that there may be little

variation in the concentration factor for plutonium in fish muscle and bone tissues for species from different trophic levels or global locations. Rockfish from the area near the Farallon Islands collected in 1977 contained  $^{239+240}\text{Pu}$  associated with the muscle and bone at concentrations of  $0.008 \pm 0.003$  and  $0.043 \pm 0.004$  pCi/kg wet weight respectively (No 78c). The average concentration in the upper 100 m of seawater was  $0.7 \pm 0.3$  fCi/l (No 78c). Using the concentration factors from the Marshall Island results and the average Farallon water concentration, the predicted plutonium in the muscle and bone would have been  $0.010 \pm 0.004$  and  $0.10 \pm 0.04$  pCi/kg. These concentrations are in reasonable agreement with the measured values.

From the total weights of the tissues and concentrations of  $^{239+240}\text{Pu}$  listed in Table 2 and the assumptions developed above, it is possible to reconstruct concentrations in any assembly of tissues and organs to compare with the 1972 concentration data.

Table 3 summarizes measured plutonium concentrations in Kwajalein Atoll reef fish from 1972 (Ne 73, Sc 75) and concentrations in the whole, eviscerated, and viscera samples collected in 1975 and 1976. The concentration of  $^{239+240}\text{Pu}$  in the 1972 dry eviscerated reef fish from Kwajalein Atoll range from 4 to 960 pCi/kg. Our measurements show concentrations in eviscerated reef fish 22 to 5000 times lower than these values. The concentration in dry eviscerated snapper is 440 times lower than the concentration measured in the 1972 snapper. Concentrations in 1972 dried viscera samples ranged from 9-430 pCi/kg. Our reconstructed viscera concentrations are orders of magnitude less than this range of

rates to bone of man, from ingested plutonium via the marine food pathway at Kwajalein, are closer to 0.005% rather than 10-25% of the EPA recommended guidelines.

The concentrations of plutonium in the tissues of mullet and snapper were not as different as we would have expected from results of previous studies and models (Gu 77, Li 77, Bo 76). The estimated plutonium concentration in mullet muscle and the measured concentration in the snapper are identical within experimental error as are the concentrations associated with the bone. The skin of the snapper has associated with it slightly higher concentrations of plutonium than mullet but, on the other

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hand, the estimated mullet gill concentrations are higher than levels measured in the snapper. The largest difference in concentrations are associated with the viscera samples. However, if the estimated plutonium in the intestinal contents of the mullet are subtracted from the measured total viscera concentration, only 0.94 pCi/kg wet would be associated with the mullet viscera. This is roughly a factor of two higher than the concentration associated with the snapper viscera. The concentration of plutonium associated with the contents in the snapper stomach was small. Although the intestinal tract of the snapper is small relative to the stomach, the ingested material in the intestine tract does contribute some weight to the viscera sample. If we assume this weight to be half the sample weight, as was the estimated weight for the contents in the mullet GI tract, the concentration of  $^{239+240}\text{Pu}$  associated only with the snapper viscera would be equivalent to the concentration in the mullet viscera.

Concentrations of  $^{239+240}\text{Pu}$  in the eviscerated samples of mullet and snapper are essentially identical whereas the average concentration in the whole mullet, on either a wet or dry weight basis, is 4 to 5 times higher than the concentration in the entire snapper. We would agree with previous studies that there is a difference in the concentration of plutonium as a function of trophic level and feeding habits only when concentrations in the entire fish are compared. However, if only concentrations in the eviscerated fish tissues are considered, the difference is insignificant.

Apparently mullet ingest more plutonium associated with their food items than the snapper, but the amounts of  $^{239+240}\text{Pu}$  concentrated in the muscle, skin or bone in fish from either the first or second trophic level

fish as the trophic level of the species increases, the slope of the gradient is strongly influenced by the concentrations associated with material in the GIT and not the quantities absorbed.

Locations which leads us to believe that there is a great deal of validity  
in the concept of a concentration factor for estimating concentrations of  
plutonium in fish.

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TABLE I  
 CONCENTRATIONS OF  $^{239+240}\text{Pu}$  AND  $^{137}\text{Cs}$  ( $\text{pCi m}^{-3}$ ) IN SEAWATER FROM KWAJALEIN LAGOON  
 AND IN SURFACE SEAWATER AT LOCATIONS IN THE NORTH EQUATORIAL PACIFIC

Kwajalein Lagoon <sup>a</sup> Stat. No. and Depth Sampled (m)	Collection Date	$^{239+240}\text{Pu}$	$^{137}\text{Cs}$	$^{238}\text{Pu}/^{239+240}\text{Pu}$	Reference to Previously Reported Data
1-Surf.	5/10/75	0.33 (20) <sup>b</sup>	137(3)		No 76
1-44	5/10/75	0.87 (13)	144(5)		No 76
2-Surf.	5/08/75	0.29 (27)	131(5)		No 76
3-Surf.	5/08/75	0.26 (24)	127(3)		No 76
3-47	5/08/75	0.33 (25)	129(3)		No 76
4-Surf.	5/08/75	0.54 (16)	129(4)		No 76
5-Surf.	6/14/75	0.52 (18)	132(4)		No 76
1-Surf.	4/17/76	0.72 (44)	133(3)		
5-Surf.	4/18/76	0.50 (12)	135(3)		
7-Surf.	4/17/76	0.53 (22)	139(3)		
7-38	4/17/76	0.57 (34)	149(3)	0.11(50)	
8-Surf.	4/17/76	0.57 (16)	132(4)	0.16(70)	
10-Surf.	10/23/76	0.37 (26)			
N. Equat. Pacific					
14°00'N180°00'W	10/72	0.35 (12)	170(12)		No 74
12°00'N170°00'W	10/72	0.44 (16)	140(14)		No 74
12°53'N173°28'E	11/73	0.22 (15)	131(4)		
05°53'N172°09'W	12/73	0.29 (66)	114(2)		
04°34'N179°02'E	12/73	0.47 (46)	122(4)		
10°26'N166°31'E	6/75	0.36 (32)	132(3)		No 76
11°16'N165°45'E	6/75	0.53 (23)	143(4)		No 76
10°00'N165°33'E	4/76	0.28 (40)	152(3)		
10°33'N164°07'E	4/76	0.39 (50)	142(4)		
10°47'N163°35'E	4/76	0.32 (15) ("soluble" fraction) 0.046(15) ("particulate" fraction)		0.16(15)	
10°00'N165°33'E	10/76	0.20 (10)	132(3)		
09°59'N165°24'E	5/77	0.25 (23)	128(3)	0.30(50)	

<sup>a</sup>Kwajalein station locations are shown in Fig. 1

<sup>b</sup>Values in parentheses are the  $1\sigma$  counting error expressed as percentage of listed value.

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TABLE 2

$^{239+240}\text{Pu}$  Concentrations in Tissues of Fish from Kwajalein Atoll

	Muscle	Bone	Gill	Viscera*	Skin	Stomach Contents	Testes	Ovary
<u>Mullet-Crenimugil crenilabris</u>								
Tissue dry weight (gm)	1100	348	59.4	638	690	46.2	3.7	88.7
wet/dry weight ratio	4.00	1.78	4.27	2.77	1.67	1.21	3.07	2.70
$^{239+240}\text{Pu}$ (pCi/kg wet weight)	lost	0.072(8)	lost	1.65(3)	0.29(8)	4.13(3)	1.14(18)	0.18(27)
<u>Snapper-Lethrinus kallopterus</u>								
Tissue dry weight (gm)	243	87.4	7.3	24.4	69.3	9.0		
wet/dry weight ratio	4.12	2.07	3.49	3.87	1.69	4.57		
$^{239+240}\text{Pu}$ (pCi/kg wet weight)	0.003(30)	0.098(17)	<0.001	0.58(8)	0.46(10)	<0.003		

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\* Viscera samples contain: intestines, stomach, liver and other digestive organs minus the contents of the stomach.

TABLE 3  
Comparative Concentrations of  $^{239+240}\text{Pu}$  in Reef Fish from Kwajalein Atoll

Year Collected	Island	Organism	Tissue	wet/dry weight Ratio	$^{239+240}\text{Pu}$ pCi/kg dry	Reference
1972	Enewetak	Convict surgeon	Eviscerated	3.54	960±80	Ne 73
"	"	"	Viscera	5.26	430±30	"
"	"	Damselfish	Entire	3.09	230±120	"
"	"	Parrotfish	Viscera	2.95	410±20	"
"	Kwajalein	Convict surgeon	Eviscerated	3.54	10±1	"
"	"	"	"	3.54	11±5	"
"	"	"	Viscera	5.26	9±1	"
"	"	"	"	5.26	110±10	"
"	"	Grouper	Entire	3.37	9±1	"
"	"	Snapper	Eviscerated	3.41	80±5	"
"	"	Halfbeak	Entire	4.57	540±10	"
"	"	Rabbitfish	Eviscerated	3.91	4±1	"
"		Convict surgeon	Eviscerated		20±3	Sc 75
"		"	Viscera		40±14	"
1975	Kwajalein	Snapper	Entire		0.29	
			Eviscerated		0.18	
			*Viscera		1.64	
1976	Enewetak	Mullet	Entire		1.22	
			Eviscerated		0.19	
			*Viscera		4.12	

\*"Viscera" concentration is reconstructed from stomach contents and separated viscera samples which are the 1972 definition of "Viscera" sample.

## Table Captions

Table 1- Concentrations of  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$  ( $\text{pCi m}^{-3}$ ) in Seawater from Kwajalein Lagoon and in Surface Seawater at Locations in the North Equatorial Pacific.

Table 2-  $^{239+240}\text{Pu}$  Concentrations in Tissues of Fish from Kwajalein Atoll.

Table 3-Comparative Concentrations of  $^{239+240}\text{Pu}$  in Reef Fish from Kwajalein Atoll.

## Figure Captions

Fig. 1.-Sampling Locations in Kwajalein Lagoon.

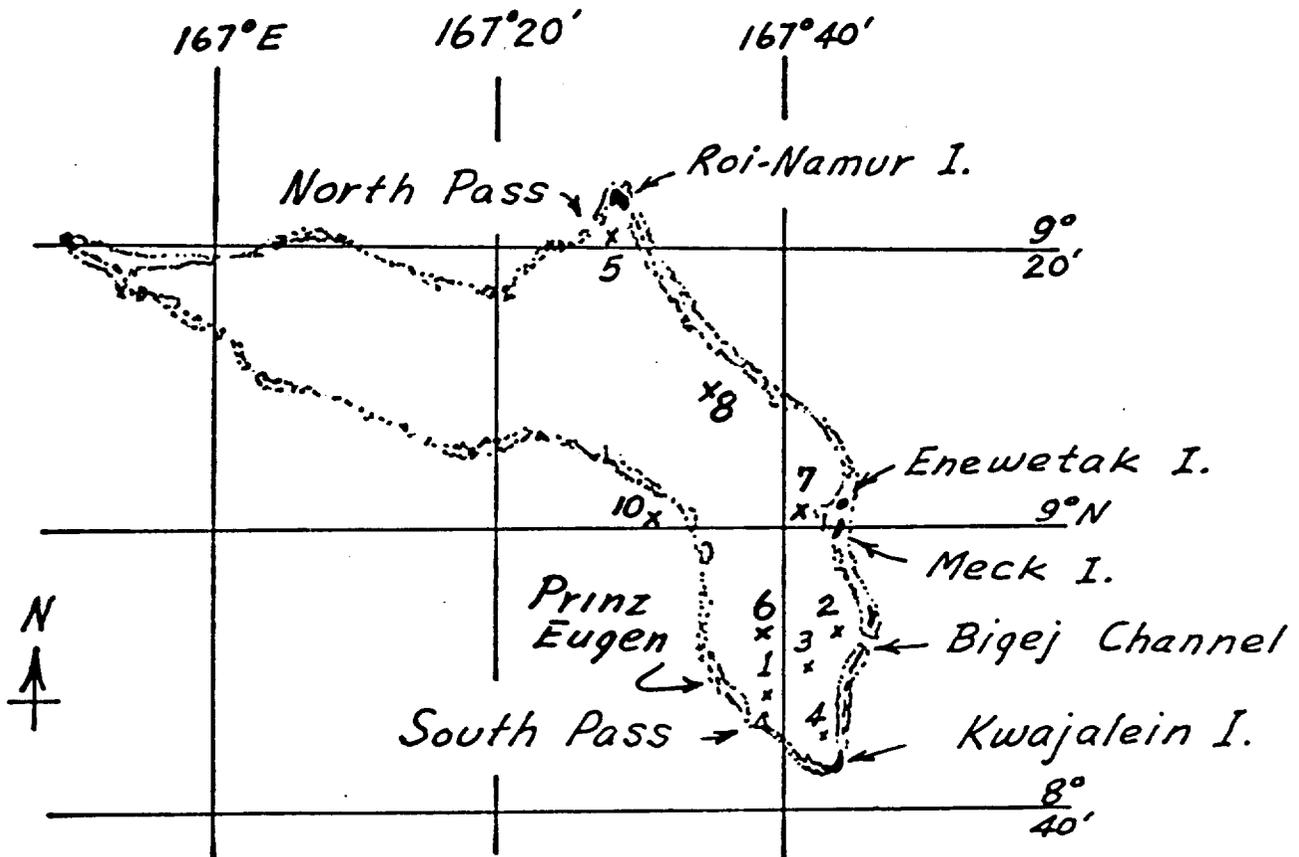


Fig. 1 Sampling locations in Kwajalein lagoon

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Fig. 1

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