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Plutonium in North Atlantic Ocean Organisms; Ecological Relationships

by

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ABSTRACT

A considerable series of North Atlantic Ocean organisms have been analyzed, by radiochemistry and alpha spectrometry, for fallout plutonium 239-240 and 238. Organisms from the near-shore environment have been selected to show the effects, on plutonium uptake, of variations in feeding habits, association with sediment or with absorptive surfaces, and of trophic level. In general plutonium concentrations are higher in organisms feeding on sediment or on surfaces than in those drawing largely on the water itself. There is some evidence that plutonium concentrations are higher in organisms of higher trophic levels.

Introduction

The world oceans have been the recipient of quantities of artificially produced plutonium isotopes. The element, released during nuclear weapons testing, and as ^{238}Pu derived from the SNAP-9A burnup in 1964, was distributed worldwide in the atmosphere. More locally plutonium isotopes are being introduced to the aquatic environment from nuclear fuel reprocessing installations (Kelleher 1969, Mitchell 1969, Templeton and Preston 1966), detected periodically in air, water and soil samples near plutonium fabrication facilities (Anon. 1970), and released following accidents involving nuclear devices (Aarkrog 1970).

We need only to acknowledge the growing use of plutonium in the nuclear industry to realize there is a need to understand the behavior, fate and transfer of plutonium within the aquatic environment.

With the exception of numerous papers discussing the physiological and toxicological properties of plutonium, however, little attention has been given to plutonium uptake in nature (Olafson and Larson, 1963) other than mammal and plant studies conducted at nuclear test site facilities. This has been largely because it is considered that the major hazard from plutonium comes from inhalation and it has been shown that uptake of plutonium by plants is low and further uptake by man through the gut is low. A recent report (Romney, et al 1970), however, shows levels of ^{239}Pu persisting in soils, plants and small mammals indigenous to Nevada fallout areas ten years after testing. The small mammals maintained in contact with the residual environmental contamination showed high levels of ^{239}Pu in the G. I. tract. Romney, et al (1970) refer to an article by Brechbill who found that cattle grazing near the Project Rollercoaster site had high levels of ^{239}Pu in bone tissue and significant levels in the spleen and lungs. The results indicate that ingestion was the major route through which ^{239}Pu entered the cattle. Deposition in the bone and other

tissue appears to be a continuing process and because of the extremely slow turnover rate in the body increased levels in body tissue occur with time. Olafson and Larson (1963) in 1961, in fact, urged caution in drawing conclusions regarding ecological implications of plutonium and questioned whether the data then available showed concentrations that were only the beginning of a continuously rising trend.

The behavior and concentration of plutonium in the marine environment was relatively neglected until Pillai, et al (1964), finding significant concentration of ^{239}Pu in several marine species, suggested a need for more information on the biogeochemistry of plutonium in the sea. We have examined several facets of the distribution of plutonium in the ocean and present here our results from studies of the concentration of plutonium in marine invertebrates of the North Atlantic. Many of the species for which data is reported are part of the human food chain. Data from our investigations of plutonium levels in aerosols, sea water, some sediment and biological samples will be used when applicable but have been reported elsewhere (Bowen, et al, 1971, Wong et al 1971, Wong et al 1970a). An account of our study of ^{239}Pu levels in marine sediments is being prepared. We here refer to some of this data as unpublished results. Changes of ratio of ^{239}Pu to ^{90}Sr and/or ^{137}Cs are of interest in assessing relative enrichments in the biota and biogeochemical behavior of these radioisotopes. Some data for the latter two isotopes are presented as well.

Methods

Samples processed for the measurement of specific radionuclides include large invertebrates and plankton. The invertebrate, principally benthic samples, were collected by a variety of methods from New England coastal regions during 1970-71. The sargassum weed was collected, by dip netting, in the southern Sargasso Sea and

other seaweed samples collected by hand or net from New England shores. Figure 1 shows the locations, referred to by number in Table 1, of samples obtained from the region of Cape Cod, Massachusetts.

Post collection treatment of the samples has been described previously (Wong, et al 1971). In brief, the samples are dissected and weighed. In all cases, after wet weight determination, the samples were dried at 105° C to constant weight and then cremated in Pyrex beakers in a muffle furnace. Incineration was begun in a cool muffle, the temperature slowly raised to 450-475° C and held at this point until ashing was complete. Stable carriers of other radioelements of interest, in addition to ^{236}Pu, were added to the ash in known amounts to serve as radiochemical yield monitors. The shells and/or tissue ash were dissolved in HNO_3-HCl or HNO_3 and plutonium, in addition to strontium and cesium, in some cases, was isolated radiochemically from the samples by procedures described by Bowen, et al (1971), Wong et al (1970b) and Wong (1971b). ^{90}Sr and ^{137}Cs were detected on low level beta detectors (Noshkin and De Agazio, 1966) by methods in use at this laboratory for a number of years and described by Wong et al (1970b). Plutonium was electro deposited on stainless steel discs and determined by alpha particle spectrometry (Bowen et al 1971). Absolute disintegration rates of ^{239}Pu and ^{238}Pu were obtained by correcting the counting data by the chemical recovery, determined from the ^{236}Pu content of the processed sample, and correcting for detector efficiency. The latter was determined by analyzing standards of known ^{239}Pu content, supplied us by the U.S. Atomic Energy Commission, Health and Safety Laboratory, New York. The identifications of the standards are: HASL ^{239}Pu (.001-XXIII); HASL-^{239}Pu (.001-XII); and HASL ^{239}Pu (.003-M). Determination of possible reagent contamination is part of our standard procedure for the assay of any radioisotope. ^{239}Pu and ^{238}Pu were below detection limits in all reagent blanks.

Analtyical Results

Plutonium 239 concentrations, as well as $^{238}Pu/^{239}Pu$, $^{137}Cs/^{239}Pu$ and $^{90}Sr/^{239}Pu$ ratios, measured in the samples analyzed to date, are presented in Table 1. All values are expressed in dpm per 100 kg wet weight of sample. Concentration data reported for some organisms in two previous publications (Wong, et al 1971 and 1970a) have been corrected and reproduced in Table 1 as well. Correction of our previous results was necessary due to a discrepancy in the detector efficiency previously used. All values have been normalized to the standards described above.

Several composite samples were separated into two fractions to check the recovery of ^{239}Pu by both wet and dry ashing techniques. No significant difference in concentration was found. The ashing procedure used was considered reliable since no loss of Pu occurred during ashing or no refractory compounds were formed which were insoluble or lost in subsequent chemical treatment.

The alpha spectrometers currently used have resolutions of 40-60 KeV at 5.30MeV, dependent on sample thickness, insufficient to resolve completely any activity from isotopes such as ^{228}Th ($E\alpha$ =5.43 and 5.34 MeV) and ^{238}Pu ($E\alpha$=5.50 and 5.46 MeV). Since the quantity of ^{238}Pu in the samples was extremely low (<10% of ^{239}Pu) great care had to be taken to insure complete removal of thorium from the plutonium prior to the electrodeposition. Although extreme care was taken with all samples, a number of samples were contaminated with traces of ^{228}Th. The contamination problem was only uncovered by analyzing in detail the entire alpha spectrum. A peak, for example, was observed at 6.29 MeV and attributed to the decay of ^{220}Rn; a granddaughter of ^{228}Th. Re-solution of several suspected samples; decontamination of Th; redeposition and reanalysis confirmed the suspicion that ^{238}Pu was overestimated. ^{238}Pu data is presented only for samples having no detectable contamination.

In Table 2 are set out the means and ranges of ^{239}Pu concentration factors. The concentration factor is the ratio between the activity (dpm) per unit mass fresh (wet weight) organism or tissue and the concentration in the same mass of "average North Atlantic sea water" from Bowen, et al (1971). The value of the latter is 0.14 dpm/100 kg.

Dashes are used in the tables to show either that the sample so indicated is still undergoing analysis, that it was lost, that the data was suspect, or that isotopic analysis was not performed.

The reported error is ± 1 standard deviation from the counting data.

Discussion

Reference to Tables I and II shows ^{239}Pu is readily detectable in all samples analyzed and concentrated by many marine species to levels significantly above those found in sea water. These results are consistent with the observation of Pillai, et al (1964) and Wong, et al (1970). ^{238}Pu in those samples where reliable values were established, is less than 10% of the ^{239}Pu activity.

$^{238}Pu/^{239}Pu$

The ratio of the two Pu isotopes in fallout has increased since 1966 due to the release of 17 kilocuries of ^{238}Pu, to the atmosphere, by the disintegration of a SNAP-9A power source in 1964. However, the large background level of ^{239}Pu ensured that the ratio of the two isotopes in cumulative deposition has not significantly changed. The cumulative deposition in Tokyo, Japan, between 1958 to 1966 (Miyake, et al 1970) resulted in a $^{238}Pu/^{239}Pu$ value of 0.037. The totals deposited in Tokyo at the end of April 1969 were 0.97 mc/km^2 ^{239}Pu and 0.042 mc/km^2 ^{238}Pu (Miyake, et al 1970) resulting in a ratio of 0.043 in cumulative plutonium deposited. The latter value agrees well with the ratio, 0.045, we have found in three, 0-3 cm depth section of marine sediment samples taken from shallow depths in Buzzards Bay and

Vineyard Sound, Mass. during 1970 (unpublished data). The quantities and ratios of plutonium isotopes in the sediments are, therefore, indicators of total delivery. This observation supports previous arguments (Bowen, et al 1971) that ^{239}Pu is rapidly depleted by active processes in the water column. This same conclusion is reached by assessment of the $^{137}Cs/^{239}Pu$ and $^{90}Sr/^{239}Pu$ values in marine sediments. The sediment samples described above contained ^{137}Cs, ^{90}Sr and ^{239}Pu in the ratios: $^{137}Cs/^{239}Pu = 4.2$ and $^{90}Sr/^{239}Pu = 0.5$. These values, compared to the mean value of 123 and 98 in sea water (Bowen, et al 1971) show Pu is being removed much more rapidly to sediments than the more soluble Cs and Sr radioisotopes. Due to rapid depletion of plutonium from the water column, the ratio of $^{238}Pu/^{239}Pu$ in water should, with time, increase in value. This effect should be particularly noticeable in near shore areas uncontaminated by local introduction. Six selected samples of water from Woods Hole harbor collected during 1970 contained plutonium having a ratio, $^{238}Pu/^{239}Pu$, of 0.12 (unpublished data). We use the above information as an aid in deciphering the origin of plutonium in some marine invertebrates.

Invertebrates

Of the invertebrates analyzed, the mussel, clam, oyster and scallop are filter feeders, subsisting on tiny organisms and organic detritus removed from suspension. ^{239}Pu concentration in the mussel body averages 51 dpm/100 kg, a level 300 (range 250-350) times that in sea water. The concentration factor is very similar to that found for Mytilus californianus (mean value=260; range: 230-290) by Pillai, et al (1964) and that we found in the brown or horse mussel from Cape Cod. The similar concentration factor found, for a species separated by both time and environment, suggests Mytilus, for the most part, regulates its plutonium content in relation to the concentration in the water rather than other sources such as the sediments. This argument is, in addition, supported by noting the relative high value of the $^{238}Pu/^{239}Pu$ ratio in both the body and shell; a value more typical of the ratio found in the water than in the sediment. The blue mussel were found attached to

rocks or piles well removed from bottom sediments. If the food, rather than the water itself, is the principal source of plutonium, the mussel must feed on recently formed suspensions within the water column rather than on aged resuspended or bottom detritus. These results suggest Mytilus is an excellent biological indicator for plutonium in assessing environmental aquatic levels. The similar ratio in both body and shell also indicates plutonium is rapidly exchanged across the mantle, the site involved in both food collection and shell formation. The shell has, on the average, 64% more plutonium than the same weight of body. It is significant to note that the levels of ^{239}Pu formed by Pillai, et al (1964) in Mytilus, (collected in 1964) were less than 1/2 those of our 1970 samples. Fallout plutonium concentrations in the mussel have increased a factor of two in six years whereas deposition, as recorded in Tokyo (Miyake, et al 1970) has increased only by 25%.

The starfish, in each case, were collected as they were feeding on the mussel beds. The ^{239}Pu concentration, although different in each sample, was about four times that found in the mussels upon which each was feeding. The $^{238}Pu/^{239}Pu$ ratio is the same as the average mussel value. Carey (1969) found, from studies of ^{65}Zn levels in Oregon coastal starfish, that the prey, rather than the environment, was the main route through which this radioisotope was transferred to the starfish. The similar $^{238}Pu/^{239}Pu$ ratio and the similar enrichment found in each starfish support this conclusion also for plutonium. Plutonium, it is concluded, shows increasing concentration along this simple food chain.

The marine worm contained the highest concentration of plutonium found in the marine invertebrate so far analyzed. The worm, Nereis?, is a non-selective deposit feeder and ingests quantities of surface sediment. The surface sediments in the Bay are significantly higher in Pu than the water, averaging 61 dpm/kg (unpublished data). The low $^{238}Pu/^{239}Pu$ ratio found in the worm is similar to that of the sediment and leads one to conclude most of the plutonium found in the creature is derived from

material deposited on the sea floor.

The moonshell, a carnivorous snail, feeds on smaller snails and other molluscs. The $^{238}Pu/^{239}Pu$ value in its body is low, similar to sedimentary material, and the ^{239}Pu content is among the highest levels found. We are inclined to believe most of the plutonium found in this predator originates in the sediment and finds its way up the food chain.

The brittle star, it is thought, derives its food from particulate matter on the sea floor. The animal has a relatively high content of ^{239}Pu but the ratio of the two isotopes suggests a source of plutonium other than the sediments. It is possible that the brittlestar preferentially selects from the organic detritus more recently deposited material, having by inference a higher Pu isotope ratio.

The oyster was the marine invertebrate with the lowest body content of plutonium. The samples analyzed were transplanted from Long Island Sound to a commercial bed in Waquoit Bay 6 months prior to collection. The present bed is in two feet of water and contains more sand than the parts of the Bay from which, for example, the scallops were obtained. Beach sand, we find, contains on the average twenty times less ^{239}Pu than silty deposits (Wong, et al 1970a, 1971a). Due to the rapid flushing of the shallow bed, little suspended detritus finds its way to the bottom. Both of these environmental factors could account for the lower levels of Pu found in the oysters.

The concentration of ^{239}Pu in the body of the scallop is on the average slightly higher than its relative, the mussel. The plutonium isotope ratio in both body and shell suggests a sedimentary source for the element. The adductor muscle, that part commonly consumed in large quantities by man, contains levels of plutonium significantly lower than the rest of the body. As in higher order animals (Wong, et al 1971a) plutonium is not highly concentrated in muscle tissue. This confirms the

observation of Ward (1966) who found the concentration factor for Pu in lobster shell to be two orders of magnitude higher than in the meat.

The sponge was dredged from the scallop bed in Waquoit Bay. It contained high levels of ^{239}Pu and the isotope ratio is between the lowest value observed in deposit feeders and the higher values, typical of the suspension feeders and their predators. This ratio is not unreasonable since the sponge typically feeds on suspended material but quantities of benthic residue enter the sponge either as occluded detritus or by burrowing worms who use the sponge as host.

The benthic invertebrates tend to have lower $^{137}Cs/^{239}Pu$ concentration values than do the mussels or other organisms feeding principally on suspended material. Discounting major discrimination between the isotopes, this again reflects the feeding habits of the creatures and the source of radioisotopes incorporated into their bodies. It was mentioned earlier that the average $^{137}Cs/^{239}Pu$ value in water is 123 while the ratio in surface sediments is 4.2.

The marine sargassum, principally fluitans and S. Natans, is the optimum Pu-collector so far analyzed, much more efficient than even the inshore plants sampled. The weed is a true surface water scavenger. Paar (1939), who conducted the most recent (1933-35) extensive population survey reported that no significant amount of weed is noted even 1.5 meters below the ocean surface. We are at present unable to explain the apparent high affinity of the sargasso weed for plutonium but if, as it has been found for ^{144}Ce (Simek, et al 1969), surface adsorption plays a relatively insignificant part in the accumulation of radionuclides by sargassum, the plant must be rapidly fixing the radionuclide internally. There appears to be no obvious colleration of concentration with either collection date or location. With the exception of the 1965 sample, the ratio of the plutonium isotopes increases, although not systematically, with time. The Pu concentration may be more closely correlated with short term changes in delivery patterns to the ocean surface rather than with water concentration.

Considering that the weed may cover considerable water passage through areas of the Sargasso Sea and outside of this area to regions as far north as 40° N latitude this could result in, depending on the abundance and annual production and depletion rate, considerable recycling of Pu from one oceanic area to another. Paar's (1939) estimate, based on 1933, 1934 and 1935 collections, of the standing crop of Sargasso Weed in the Sargasso Sea is roughly 1.5×10^3 kg (wet weight)/km^2. Using an average value of 46 dpm ^{239}Pu/kg as a representative concentration over the years, 1965-70, the yearly crop is able to effectively remove 32 nCi/km^2. Since the area of the Sargasso Sea is 5.2×10^6 km^2, there are 0.2 curies of ^{239}Pu in the yearly crop of Sargasso weed which is, according to Paar, self-generating over the year. Death occurs either by moving to marginal zones of the ocean during midwinter and very early spring, or by being washed to shore. Lower water temperature may be the cause of degeneration. This quantity of activity, or a major fraction of it, moving into confined basins, such as the northwest part of the Gulf of Mexico where considerable quantities of weed are found locally along the beaches, can result in significant concentrations of ^{239}Pu in the respective water mass due to death and degeneration. The effect will be continuous since each new yearly crop will add to the amount scavenged and displaced.

Conclusions

The plutonium content of the marine invertebrates analyzed is variable and dependent upon the environment from which they feed. The isotope ratio, $^{238}Pu/^{239}Pu$, offers some clues as to the environmental source of the element found in the animals.

Significant concentration of ^{239}Pu is found in marine vegetation. The Sargasso weed may cycle considerable quantities of ^{239}Pu to specific near-shore regions.

Mussels appear to be excellent biological indicators of plutonium. Over the last six years the concentrations of plutonium in Mytilus have doubled. This increase is greater than one would predict from land deposition.

Plutonium has been shown to be increased in concentration in starfish, predator of blue mussel, and it may generally increase in other food chains as well.

By far, of all the marine invertebrates, the largest levels of plutonium are found in benthic organisms and those predators feeding on benthic organisms. Scallop mussel tissue contains less plutonium than either its body or shell. In every case, where both body and shell were analyzed, the average concentration of ^{239}Pu in the shell exceeded that in the body. The shell may be the major repository of plutonium in these organisms. Marine invertebrates represent an important vector transferring environmental plutonium into human food chains.

When the much higher relative biological effectiveness of alpha versus beta or gamma radiation is considered, plutonium isotopes now contribute more than either ^{90}Sr or ^{137}Cs to the artificial radiation of marine invertebrates.

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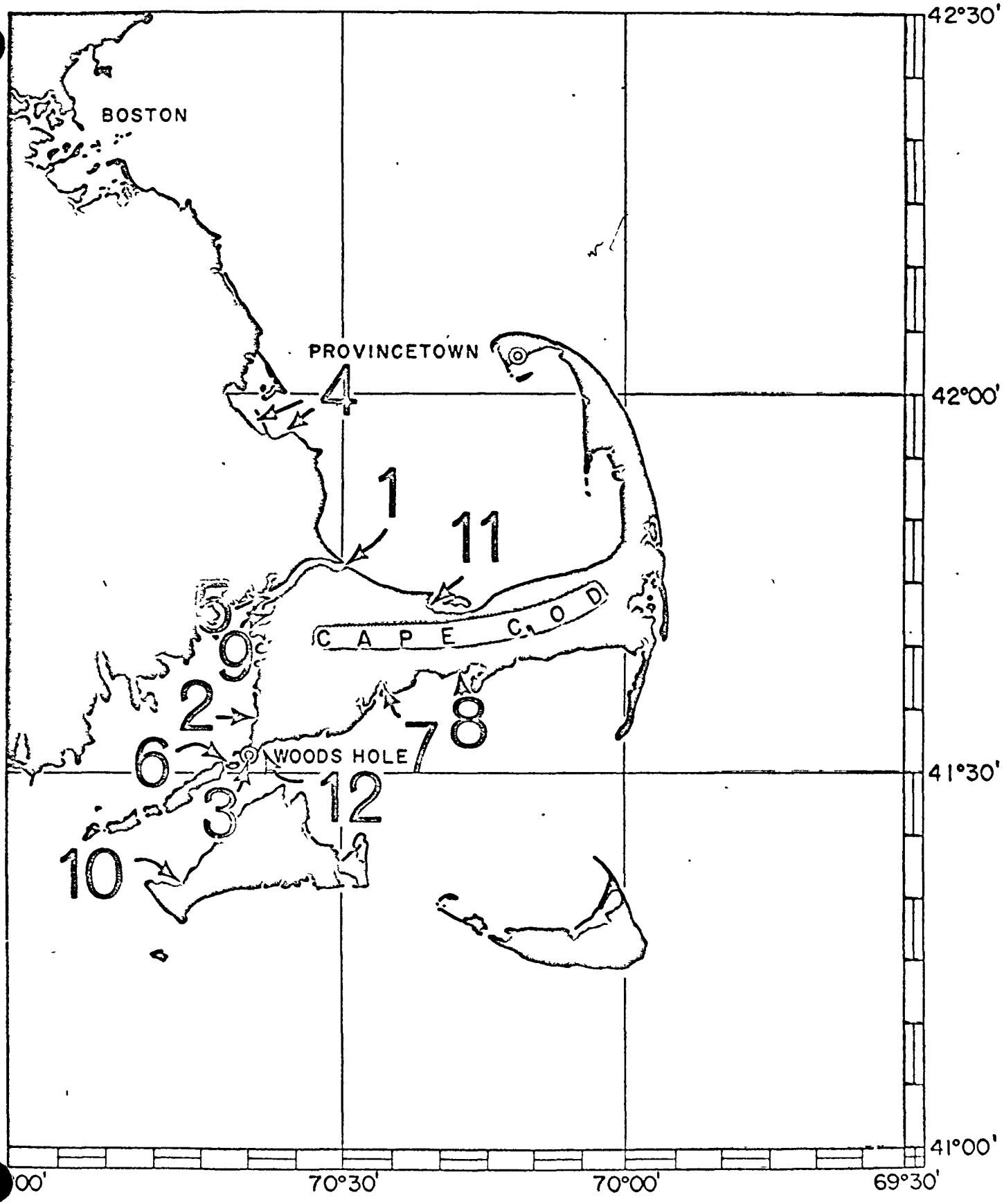
Fig. 1. Location of samples, referred to in Table I, collected from the area of Cape Cod, Massachusetts.


BOSTON
PROVINCETOWN
CAPE COD
WOODS HOLE
1
2
3
4
5
6
7
8
9
10
11
12
42°30'
42°00'
41°30'
41°00'
70°30'
70°00'
69°30'

RADIOISOTOPE DATA FOR MARINE ORGANISMS

TABLE I

	Map (Fig. 1) Ref.	Collection date	Tissue Analyzed	Tissue Wet wt. (kg)	dpm/100 kg (wet weight) Pu^{239}	$\frac{Pu^{238}}{Pu^{239}}$	$\frac{Cs^{137}}{Pu^{239}}$	$\frac{Sr^{90}}{Pu^{239}}$
...vertebrates								
...(Mytilus edulis)								
...Canal, East End	1	6-70	body	1.76	51±19[a]	--	15 ± 6	0.8± .4
...Canal, East End	1	6-70	body	0.94	56± 8[a]	.099	18 ± 3	1.4± .9
...Beach	2	3-70	body	0.37	62±11[a]	---	---	---
...Beach	2	6-70	body	0.54	56± 8[a]	---	19 ± 3	2.1±0.8
	3	7-70	body	0.75	36± 5[a]	---	21 ± 3	4.1±1.5
...Beach, Conn.		11-70	body	0.63	97±11	---	---	---
...meteor	4	3-71	body	0.43	55± 8	---	---	---
...Beach	2	3-70	shell	0.48	89±14[a]	---	---	---
...Beach, Conn.		11-70	shell	0.78	98± 8	.081	---	---
...Canal, West End	5	11-70	shell	0.51	92±14	.095	1.1± 1.0	---
...(Modiolus modiolus)	6	6-70	body	0.97	64± 6[a]	.071	15 ± 2	1.9± .8
...(Nucula arenaria)	6	6-70	body	0.28	83±17[a]	---	27 ± 6	---
...(...minica)	7	10-70	body	0.52	19± 5	---	---	---
	7	10-70	body	0.51	31± 5	---	20 ± 7	---
...medians)	8	10-70	a.m.*	0.53	2± 1	---	600 ±300	---
	7	10-70	a.m.*	0.61	7± 2	---	---	---
	9	10-70	body	1.04	131± 6	.041	---	---
	8	10-70	body	0.63	87± 8	.057	6 ± 1	---
	7	10-70	body	0.66	78± 8	---	---	---
	7	10-70	shell	0.51	115±11	.058	---	---
...ascina)	7	10-70	body	1.81	27± 2	---	---	---
	7	10-70	shell	0.43	56± 6	.093	1.8±1.0	---
...(...a heros)	10	10-70	body	0.42	126± 9	.046	4.7± .8	---
	10	10-70	shell	0.46	131± 9	---	0 ±1	---
...forbesi)								
...East End	1	6-70	body	1.40	220± 8[a]	.085	1.2± .2	11.3± .5
	3	7-70	body	1.77	167±20	.090	---	12.6± .6
...oidea)	7	11-70	body	0.54	145±15	.090	---	---
	11	10-70	body	0.61	778±42	.056	1.2± .2	---
...delicata)	7	11-70	body	0.41	399±51	.074	1.5± .3	---
...(...opossum sp.)		4-70		0.32	1990±75[a]	.051	---	1.0± .4
		7-69		0.76	911±35	.092	---	
		11-68		0.22	624±30	.026	4.4±0.8	---
		5-66		0.25[b]	1070±47	.030	1.0± .5	0.6± .2
		2-66		0.05[b]	18500±62	.035	---	---
		12-65		0.14[b]	4500±32	.057	---	---
...agile)	3	11-70		0.64	39± 7	---	6.1±2.3	---
	3	2-71		0.41	139±22	---	10.3±2.8	---
	4	3-71		0.51	126±13	---	---	---
	4	3-71		0.51	301±20	---	---	---
	4			0.39	76±37	---	---	---
	12	3-71		0.47	20± 8	---	---	---

...tor muscle

a = corrected data from Wong et al (1971).
b = estimated wet weight from dry weight.

CONCENTRATION FACTORS OF PU 239 FOR MARINE INVERTEBRATES

TABLE II

Organism	Tissue	Mean	Range
Blue Mussel	body	300(7)[b]	250-350
	shell	490(3)	470-520
Brown Mussel	body	340(1)	
Soft Shell Clam	body	440(1)	
Oyster	body	130(2)	100-160
Scallop	adductor	24(2)	10-37
	body	520(3)	410-690
	shell	600(1)	
Whelk	body	140(1)	
	shell	300(1)	
Moonshell	body	660(1)	
	shell	690(1)	
Starfish	body	1020(1)	
Brittle Star	body	760(1)	
Marine Worm	body	4100(1)	
Sponge	body	2100(1)	
Sargasso Weed		2.1×10^4(6)	$.3\text{-}10 \times 10^4$
Other Sea Weed		6.2×10^2(6)	$1\text{-}16 \times 10^2$

b = number of samples averaged