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CONTRIBUTIONS FROM THE ALPHA EMITTER,
POLONIUM-210, TO THE NATURAL RADIATION ENVIRONMENT
OF THE MARINE ORGANISMS

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ABSTRACT

In describing the natural background radiations in the marine environment, emphasis in the past has been given to contributions from potassium and cosmic rays; much less has been said about the alpha emitters. However, there is evidence that many marine organisms accumulate natural alpha emitters to levels that might be contributing substantially to their burden from ionizing radiation. There are increasing opportunities for alpha emitters to enter the ocean because of the increasing use of plutonium-239. Distributions of polonium-210 are of interest because it has been found that this natural nuclide accumulates to relatively high levels in some of the same marine ecosystems that accumulate plutonium effectively. The progress of the natural nuclide is technically much easier to follow, yet its distributions might serve to give some warning as to the probable fate of plutonium.

Concentrations of polonium-210 in a variety of marine organisms are compared and also the radioactive burdens that may be inferred from bulk tissue samples. Some difficulties of interpretations are discussed and the need for more detailed measurements in specific organs and tissues. Some inferences about polonium-210 from lead-210 measurements also are presented.

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INTRODUCTION

Both polonium-210 and plutonium-238 were, about ten years ago, given serious consideration as sources of auxiliary power for certain space craft. At that time neither of these had been measured in seawater or in marine organisms. This was of some concern because most of the large rockets in this country are launched at stations near the coast. It was imperative to acquire some knowledge of the contemporary environmental conditions in the coastal waters that might be changed by an accident to a rocket carrying one of these nuclides. One major rocket testing station is located at Point Mugu only 200 miles north of La Jolla. Therefore preliminary studies of plutonium in the marine environment were carried out at the Scripps Institution [1], and these were followed by a brief survey of concentrations of polonium-210 in coastal seawater and in a few marine organisms [2].

These early marine studies of polonium-210 included the inspection of a few marine organisms to establish the concentration magnitudes at a few distinct trophic levels and were made in the hope of finding one or more readily harvestable marine species which might accumulate enough polonium-210 in their tissues to justify their use as indicators of changes of polonium-210 in the environment. It was found in these first measurements that polonium-210 generally accumulated effectively in many of the same species that had been observed to build up highest concentrations of plutonium. For example, the brown algae, several species of which are easy to gather, and some species of which are harvested commercially in southern California, were in general found to be sensitive indicator-species, often concentrating plutonium and polonium to levels more than 1000 times the levels found in the sea. However, the muscular tissues of those marine fishes which were inspected exhibited much smaller amounts of these two elements.

After collaborating in the preliminary studies of polonium-210 at Scripps, one of the present authors (TMB) continued his survey of a wide variety of marine organisms for polonium-210, and its precursor lead-210, at Oregon State University, and presented his experimental data in support of a thesis in oceanography [3] describing distributions of lead-210 and also emphasizing the part played by polonium-210 in the marine environment. His samples were collected mostly in 1968 in coastal waters near Oregon, and included samples from several depths. In several instances, he encountered polonium-210 concentrations that were somewhat higher than any that had been measured directly in a marine species living in a natural environment.

The disintegration of polonium-210 gives rise to internal bombardments by alpha particles, each carrying an energy of more than 5 MeV. Therefore, it became of interest to consider how much this radioactivity contributes to the total ionizing dose that was experienced by marine organisms living under various natural conditions. In a much earlier study, made at a time when hardly any alpha concentrations had been reported for marine tissues [4], the contributions from alphas had not been added to the other burdens expected from cosmic rays and from external and internal beta and gamma sources. Nevertheless, it was pointed out that the natural radioactive burdens should

vary greatly within the marine environment, depending both upon the physical size of the organism and also upon the location in the sea of its habitat. As examples, the direct effects of cosmic rays become negligible at depths below 100 meters; and the central organs of the large fishes are shielded by intervening tissues from beta particles coming from the sea. It is convenient here to make use of these earlier environmental comparisons; however, it should be anticipated that difficulties will be encountered when we attempt to compare the effects of alpha emitters also.

APPARENT RANGE OF POLONIUM-210 CONCENTRATIONS

It is important to understand how widely polonium-210 concentrations vary in the environment. Even when measured in the usual way in bulk biological samples, the concentrations vary greatly; and when special tissues are selected, the range is still more extreme.

Table I collects together some data available from internal reports [2] or from an academic thesis [3] prior to 1968, comparing polonium-210 concentrations in bulk samples of several unlike marine organisms. The concentrations labeled (F) were made at Scripps using an alpha spectrometer; those labeled (B) were measured by Beasley with a ZnS(Ag) phosphor coupled to a photo-multiplier following chemical reductions and plating procedures that have been reported elsewhere [3]. It should be pointed out that only some of the higher tissue concentrations are presented here; many tissues have been encountered that accumulated little polonium-210 relative to what is in seawater.

ENERGIES AVAILABLE FROM POLONIUM-210

Only crude dose comparisons can be hoped for with the available data. It is convenient, and it appears conservative, to compare doses in terms of energy that is available and presumably might be absorbed in the same tissues that are being inspected. Therefore, the average dose rates in millirads/year have been computed for each concentration (Table I); Table II presents doses computed for a slightly different list of samples.

Since natural doses coming from external sources of radiation fall into several categories, this is taken into account in Table II. Wherever possible, with the limited number of measurements available, marine organisms are grouped into ecological domains experiencing distinct levels of radiation coming from external sources. For each organism, the total radiation expected from all other sources, excluding alpha sources, are listed along with the energies that might be added by the polonium-210 activities that have been measured in bulk tissues. In the righthand column are listed the estimated doses that might be added by the polonium-210 activities relative to the doses from all other natural radiations.

The sketch presented by Table II is made somewhat more interesting by the addition of some alpha burden data reported for humans [5]. This makes it apparent that the alpha burdens experienced by some marine animals may be high relative to some that have been reported for man. However, this last comparison is a dangerous one since the alpha distributions must be known in much more detail before the effects of alpha accumulations can be evaluated properly.

Wide variations in polonium-210 concentrations that have been reported in marine species are difficult to interpret. Some of these extremes can be attributed to known extremes in the environment. For example, in the case (listed in Tables II and III) of the benthic worms that live in sediments, high concentrations of radium usually are present. On the other hand, there have been some extreme concentrations that are very difficult to

attribute to anomalous environmental conditions. For example, there is little to suggest why extremely high concentrations have been found in certain small oceanic fish, e.g., lanternfish (Myctophidae) collected at 0-200 meter depths. One guess might be that this species has selected food containing extreme concentrations of polonium-210.

Some recent measurements have been made at Scripps to determine whether the extreme concentration in the 0-200 meter sample of lanternfish listed in Table I was typical. In this instance, polonium was estimated indirectly through some lead-210 measurements. Concentrations of lead-210 found in four year old samples of lanternfish from northern California are listed in Table III. In these old samples, only the long-lived lead-210 precursor of polonium-210 remained. Both nuclides enter the sea together [6] as summarized by Figure 1, and both nuclides may accumulate in the fish. However, after a few months, only lead-210 can remain in a dead specimen. The lead concentration measurements are listed in Table III along with some rough estimates of the polonium-210 concentrations that may have been present in the living fish. In this instance, a ratio of polonium-210/lead-210 of 45 was assumed, a value that has been found in certain other marine tissues. The polonium-210 concentrations, estimated in this manner, range between 2000 and 10,000 dpm/wet Kg, which are not inconsistent with the earlier measurement 7030 that has been listed in Table I for the same species. The value of the ratio of polonium-210/lead-210, used for this estimate may, of course, be too high; however, ratios as high as 54 have been observed in connection with several midwater species [3]. Moreover, Table IV illustrates some high values of this ratio that have been observed recently at Scripps for organs of large fish and marine mammals.

The high ratios of polonium-210/lead-210 that frequently are observed suggest that much of the polonium reported for bulk marine tissues has entered as "unsupported" polonium and has not resulted from the decay of accumulated lead-210. This was suspected first in plankton by Cherry [7], and later in larger organisms by Beasley [3] and by Shannon and Cherry [8]. Holtzman [9] found a polonium-210/lead-210 ratio of 44 in kidney tissue of a whale. On the other hand, the polonium-210/lead-210 ratio in some other marine tissues have been somewhat less than 10; in some cases both the lead and the polonium concentrations being high and in other cases both being low. For example, the stomach contents of a Rex sole (Glyptocephalus sp.) were found to contain 5800 dpm polonium-210/Kg and 1900 dpm lead-210/Kg; the stomach contents of a rock fish (Sebastes sp.) had only 200 and 38 respectively [3].

The partition of polonium-210 from lead-210 in the biosphere is perhaps best illustrated by some recent measurements [10] of concentrations in tissues of elk kelp summarized in Table V. The outer layers of this giant algae are intense concentrators of polonium and lead (and also of plutonium); however, the ratio of polonium-210 to lead-210 is close to unity similar to what has been reported for seawater [11]. These measurements illustrate one case where polonium-210 is excluded in preference to lead-210 since the polonium-210/lead-210 ratio was found to be less than unity for inner layers of this alga.

CONCLUSION

It is apparent now that alpha radiations, especially those from polonium-210 cannot be ignored in studies of radiation in the natural marine environment. However, it should also be evident that few of the measurements that now are available give information about local concentrations of alpha emitters. Because the range of alpha particles are extremely short, it is impossible to say what burdens they are contributing to critical tissues.

Even when the objective is only the monitoring of changes in the environment, much more care should be taken in sampling organisms.

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Table I. Some polonium-210 concentrations found in seawater and marine organisms of the northeast Pacific (1965-1968).

Depth (meters)	Sample	Reference ^a	Concentration dpm/Kg wet ^b	"Dose rate" added by 210Po mrads/year ^c
<10	Pacific Coastal Surface seawater--Scripps Pier collected 1966	F	0.082	0.0037
<u>Southern California Coast Collected 1965-1966</u>				
0-100	Squid, <u>Loligo</u> sp. (Hepatopancreas tissues)	F	330	15
0-100	Bonito (muscle)	F	22	1
0-100	<u>Sardo chiliensis</u> Albacore (muscle)	F	300	14
	<u>Thunnus alalunga</u>			
<u>Marine Plants, Southern California Coast Collected 1965-1966</u>				
-	Brown alga (kelp)	F	380	17
0- 3	<u>Eisenia arborea</u>	F	<0.2	<0.009
0- 10	Phytoplankton (mostly dinoflagellates, from "red tide" July 1965)	F		
<u>Selected Marine Organisms, Several Depths, Oregon Coast 1968</u>				
<1	<u>Lepas</u> sp. (whole)	B	3490	160
0- 10	Oysters (soft tissue)	B	1040	47
0- 10	Copepods, <u>Acartia clausii</u>	B	890-2490	30-110
0- 10	Mixed zooplankton	B	550	25
50-100	<u>Sagitta elegans</u>	B	1660	75
50-100	<u>Euphausia pacifica</u>	B	640	29
0-200	Lanternfish	B	7030	320
	<u>Tarletonbeania crenularis</u>			
100-200	Lanternfish	B	460	21
	<u>Tactostoma maropus</u>			
400-600	Lanternfish	B	600	27
	<u>Lampanyctus ritteri</u>			
<u>Animals Living Near Sediments, Oregon Coast 1968</u>				
	Polychaete worms (benthic unidentified sp.)	B	17,800	800
	Sea cucumber	B	810	36
	<u>Paleopatides</u> sp.			

a: F = Folsom, Pillai and Beasley [2]; B = Beasley [3]; b: counting errors were less than $\pm 10\%$; c: it is taken that mrads/year = $0.045 \times$ dpm polonium-210/Kg. Relative biological effectiveness (RBE) of the alpha particle is disregarded here.

Table II. Apparent doses added by polonium-210 in selected marine organisms.^a

Type of organism and its environment ^b	Sum of estimated dose rates from all external sources and internal potassium ^b mrad/year	Estimated dose rate added by alphas of ²¹⁰ Po in the tissues ^c mrad/year	Estimated fraction of dose rate added by ²¹⁰ Po ^d %
<u>Man</u>			
Over granite, 10,000' elevation	207 ^e	12 ^f	5
Over granite, sea level	142	12	7
Over sedimentary rock level	75	12	15
Over the ocean level	52	12	20
<u>Larger marine organisms^g</u>			
Near sea surface	64		
Lanternfish caught, 0-200 meters ^h	--	320	500
Barnacle (<i>Lepas</i>) surface ⁱ	--	160	240
Deep in the sea (>100 meters)	30		
Lanternfish caught, about 800 meters ^j	--	110	370
<u>Smaller marine organisms^k</u>			
Near sea surface	39		
Copepods (0-10 meters)	--	20-50	50-130
In seawater (>100 meters deep)	>5		
(very small organisms not sampled here)	--	--	--
In sediments (>100 meters deep in the sea)	40-620		
Polychaete worm from bottom sediments ^l	--	800	80-200

FOOTNOTES

- a: Organisms having high concentrations were selected for each domain.
- b: These are estimates reported by Folsom and Harley in domains they selected in 1957 [4].
- c: Dose is given in terms of energy only, no weight was given to account for any special effectiveness to alphas. For humans, the reports of Holtzman [5] were used, for marine organisms those of Beasley [3].
- d: This ratio is energy added by alphas relative to all other radiations.
- e: Cosmic doses estimated for mid-latitudes.
- f: Human alpha dose estimates includes some contributions from radium also.
- g: Large here means thick enough to stop most of the internal ⁴⁰K betas.
- h: *Tarletonbeania crenularis* (whole organism).
- i: *Lepas anatifera* from glass floats (whole organism).
- j: *Lampanyctus leucopsarus* caught at 800 meters (whole organism).
- k: Small here means thin enough to lose substantial beta energy of ⁴⁰K to the surroundings.
- l: These worms are not small relative to beta ranges, but live intimately with sediments containing radium.

Table III. Lead-210 concentrations in lanternfish (*Myctophidae*)^a.

Sample No.	Collection ^b		No. of specimens analysed	210Pb conc. dpm/g		Calculated ^c "unsupported" 210Po, dpm/Kg wet
	Location	Date		Dry	Wet ^d	
350	37°N, 123°W	May 1950	1	340±70	68±14	3,000
353	37°N, 125°W	May 1950	3	230±60	45±12	2,000
354 ^e	41°N, 127°W	Mar-Aug 1950	6	580±80	103±15	4,600
351	41°N, 127°W	July 1951	1	940±170	220±40	10,000

a: *Tarletonbeania crenularis* from the North Pacific; b: samples donated by Dr. John MacGregor, National Marine Fisheries Center, La Jolla, Calif.; c: assuming an "unsupported 210Po"/210Pb ratio of 45, similar to what was found in fresh albacore livers; d: preserved in formalin; e: composite of two samples, two collected in March plus four collected August 1950.

Table IV. Polonium-210/lead-210 ratios in marine vertebrates (samples collected in 1971 near San Diego).

Sample No.	Sample	210Po/210Pb
28	Albacore ^a - liver	45±7
29	heart	4±1
30	spleen	24±1
31	kidney	20±1
52	Killer whale, ^b muscle	19±3
60	Dolphin - liver	48±5
61	blubber	34±2
65	spleen	39±5
69	muscle	30±3

a: muscle tissues typically contain less 210Po; for example, about 300 dpm/Kg compared with 20,000 representative of liver tissues; b: in this case only an aquarium animal was used.

Table V. Polonium-210/lead-210 ratios in various tissues of giant brown alga, *Pelagophycus porra* (elk kelp) collected on San Diego coast, 1971.

Description of Sample	$^{210}\text{Po}^a$	$^{210}\text{Po}/^{210}\text{Pb}^b$
Bulb layer, 0-0.2 mm from outer surface	2600	1.17
0-1 mm from outer surface	120	1.15
1-3 mm from outer surface	2	0.03
3-5 mm from outer surface	2	0.02
Blade section (~1 mm thick) near tip	100	2.58
(~2 mm thick) near stem	50	1.19

a: Wong, Hodge and Folsom (in press 1972) [10]; b: unpublished measurements by Kai Wong, 1972 [12].

Figure 1. Model suggesting paths by which polonium may come into the marine biosphere, from data of Goldberg [6].