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EARLY SURVEILLANCE AROUND COASTAL NUCLEAR INSTALLATIONS

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ABSTRACT

Several biological systems have been used to detect extremely small amounts of radioactivity emanating from nuclear reactors that discharge directly into marine environments. Special attention has been given to plutonium.

Inorganic absorbing systems, including certain ferrocyanides, provide convenient and sensitive means for monitoring the caesium radionuclides in sea water. These may well be used for monitoring automatically. Generally, the absolute concentrations of radiocaesium can be determined by comparisons with the concentrations of the natural nuclide, caesium-133, that is present in the marine environment at a level that frequently may be predicted.

Selected plants have been found most useful, when samplings are normalized against surface area. Certain brown algae are useful for monitoring plutonium, especially nuclide ratios. Silver and cobalt nuclides accumulate preferentially in red algae and marine grasses.

Surveillance by means of marine animals is sometimes more sensitive, but proper sampling and interpretations are much more difficult. Several of the common molluscs may be used effectively as monitors of cobalt, silver and other nuclides.

Larger fish, whenever their exposure pattern can be estimated, may be exploited for monitoring various metal nuclides through the use of selected organs.

Marine biological systems are frequently such sensitive indicators that account must be made of contributions from many sources, even from global fallout more than ten years old. Successful biological surveillance of a given nuclear installation generally requires comparisons of nuclide ratios at several distances using consistent tissues at consistent seasons.

INTRODUCTION

Many large nuclear installations are expected to appear along the sea coasts where large populations are growing. Planning disposal systems that will minimize hazards and economic losses requires thorough understanding of the probable behaviors of several classes of radionuclides that may find their way into coastal waters. Fortunately, some useful information concerning environmental responses can be learned through careful studies made long before pollutions have reached levels of any reasonable concern to human welfare. Early information of this sort, however, requires the use of highly specialized equipment and techniques not generally employed by those interested only in satisfying existing legal restrictions concerning the use of the environment. Moreover, coordinated efforts of biologists, chemists and physicists familiar with oceanographic problems are required.

This report will describe some small changes that have been observed following installation about 5 years ago of a 400 MW coastal nuclear power plant at San Onofre, California, about 70 Km north of Scripps Institution of Oceanography. Nuclide levels of four different radioactive elements observed at San Onofre will be compared with contemporary coastal and oceanic background levels which largely have depended, in a more or less complex way, on global fallout. A few comparisons with levels reported for other coastal installations will be made; however, most attention at Scripps has been given to determining which nuclides associated with nuclear power production are accumulating in which marine species, and in developing extremely sensitive methods for following radioactive changes in the several marine niches along coasts that are relatively openly exposed to the ocean.

EXPERIENCES WITH RADIOACTIVE SILVER NUCLIDES

The changing concentrations of two silver nuclides attributable to global fallout have, since 1964 [1-5], been studied at Scripps in numerous oceanic and coastal samples. Because of these background records, it has been possible to identify extremely small silver nuclide anomalies in marine samples collected near the San Onofre Power Plant. Anomalous silver nuclide ratios have been found near this plant, and also higher absolute concentrations of silver-110m relative to concentrations found in comparable biological samples collected 70 Km away from the plant, i.e., near La Jolla.

Table I illustrates that at least three different marine species, that accumulate silver nuclides readily, can be collected in the vicinity of this power plant, and also at points some distance away, so as to be useful for searching for local anomalies. The more common brown algae were found to be not useful for silver studies; however, a red alga, a mollusc, and a surf grass were found to be effective and convenient.

Figure 1 and Table II illustrate how much more sensitive and definitive are comparisons made between silver nuclide ratios rather than between absolute concentrations. It should be noticed in Figure 1, that, in several types of reference samples collected between 1970 and 1973, silver-110m/silver-108m ratios between 1 and 3 were measured. However, biological specimens collected near San Onofre exhibited a ratio of about 200.

It should be noted in Table II that extremely high absolute concentrations of both silver nuclides sometimes may be accumulated in organisms that apparently have experienced no other artificial nuclide source but that of global fallout. Yellowfin tuna caught in the central N. Pacific sometimes contain nuclide ratios near unity but absolute concentrations many times higher than other oceanic fish.

These small traces of silver generally were determined by simply drying selected tissues and use of a summing-coincidence (NaI) gamma spectrometer [6], or a two-dimensional gamma spectrometer [4], or both. It was later found that radiosilver traces may be rapidly and quantitatively electroplated from plant or animal tissues after a silver carrier has been added to a slurry ground in a Waring Blendor [7]. This form of concentrating might be especially advantageous for those wishing to concentrate silver nuclides to the small volumes that are most suitable for germanium-lithium type of gamma spectrometers.

COBALT NUCLIDES

Several gamma emitting cobalt radionuclides also are useful for distinguishing the very earliest traces of power plant nuclide waste from fallout backgrounds. This may be seen from Table I where cobalt-60 and cobalt-58 concentrations in samples collected near San Onofre and others collected 70 Km away at La Jolla (Scripps Institution) are compared. In the case of cobalt, as with silver, red algae and green surf grasses were found to accumulate higher concentrations than do brown algae; and in general, more intense cobalt concentrations may be found in molluscs, but these are more difficult to collect routinely.

Coincidence-gamma spectrometers similar to those used for radiosilver determinations were also used for small traces of cobalt. No chemical separation is needed but drying or ashing adds much to convenience and sensitivity. Where larger fish are available, it is often profitable to dissect and count activity only in the liver tissue. The half-life of cobalt-58 (72 d) is a convenient one; when normalized against cobalt-60, the cobalt-58 activity provides a sensitive indicator ratio. It is necessary, of course, to collect control samples from other areas exposed only to global fallout. Moreover, it is necessary to repeat collections of control samples frequently enough to keep up to date the record of the global fallout background. Experience has shown that the oceanic biosphere readily detects new inputs of nuclear fallout, especially silver and cobalt nuclides, and since 1968, relatively large new fallout inputs of zinc-65 have been detected in the ocean by inspection of biological concentrators [8].

CAESIUM NUCLIDES

Consideration of caesium nuclides can hardly be avoided; however, surveillance techniques different from those that succeed with the transition metals must be adopted. The long life of caesium-137 and its persistence in upper layers in the ocean [8] provide for large-scale normalization against the changing global fallout background, especially its large scale and long range aspects. On the other hand, the short life of caesium-134 (2.1 yr) and the presence of this nuclide in power reactor wastes may give evidence of a change in coastal disposal rates.

Table III illustrates the wide range of caesium-137/caesium-134 ratios that have been observed, varying from about 1.4 in samples collected at San Onofre to greater than 100 in recent oceanic waters exposed only to fallout.

No biological system has been found that accumulates caesium as effectively as those that may be exploited for collecting large samples of silver, cobalt and plutonium nuclides. Biological accumulations of caesium seldom are more than 100 times the concentration in sea water. Moreover, one must normalize the surveillance samples with care, because the alkalies vary considerably within apparently identical biological samples. Normalization against potassium rather than against sample weight appears most appropriate; for example, Figure 2 illustrates the parallel behavior of caesium and potassium in different tissues of an oceanic tuna.

Several inorganic agents are available for selectively concentrating caesium nuclides from sea water to levels > 50,000 times the levels in sea water. When ratios of gamma nuclides are desired, the direct gamma measurements introduced by Boni [11] of caesium absorbed from sea water onto granular ferrocyanides are effective. When absolute levels of environmental caesium concentration are required, however, normalization against natural caesium collected on the absorber [12] can be used and is especially effective for detecting extremely small traces of radiocaesium.

Large samples of caesium nuclides can be collected routinely by pumping large volumes of water through granular absorber beds. This monitoring can be done automatically at points near a nuclear installation.

PLUTONIUM MARINE POLLUTION STUDIES

The first measurements of plutonium in sea water and marine organisms made by Pillai at Scripps in 1964, disclosed the fact that many marine plants had strong affinities toward this element [13]. This suggested the surveillance of an environment by analysis of samples of its plants. In fact, the levels of the minute traces of plutonium-238 in local waters were established by measuring the plutonium-238/plutonium-239 ratio in the ashes of giant brown algae. This was much simpler than precipitating plutonium directly from 500-litre samples of sea water as was done by Miyake in 1969 [14]. However, Miyake did not have available in the open Western Pacific suitable algae for conveniently concentrating his samples.

Interest in possible coastal plutonium contaminations increased when it became clear just how much of this nuclear material was being used and how much larger would be the amounts to be transported when its use for fuel was fully exploited. Inspections were made of several convenient marine organisms [15,16] toward selecting effective plutonium monitors. Plutonium concentrations were found in numerous marine species, but the concentrations varied widely and were difficult to describe and compare because of the many morphological, chemical, and physical differencies amongst marine species. Neither sample normalization by wet weight, by dry weight, nor by ash weight was satisfactory. Some extreme cases were recognized however. The lowest concentrations (~ 0.001 pCi wet Kg) were generally found in muscle tissues, especially in large fish; much higher concentrations were found on large algae and surf grasses especially on the thinner blades, 0.2 to 2 pCi/wet Kg for example, in bulk samples of brown algae [15,16]. When it became evident

that plutonium accumulated upon surfaces of many different organisms, the concentrations at several depths below the surface were quantitatively compared in typical specimens of some of the giant brown algae that are convenient to collect along the California coast [16] at 10-60 meter depths beyond the surf zone. It was found that thin layers (0.2 mm) scraped from outer plant surfaces often had plutonium concentrations more than 200 times higher than what was found deep inside the plant. Clearly then samples must somehow be normalized with reference to surface areas. Fortunately, in many plant species, major synthesizing blades of fairly uniform thickness may be collected, so that areas may be estimated from sample weights. Moreover, this normalization problem largely disappears whenever only the nuclide ratios in the environment must be determined by biological samplings.

Even when only nuclide ratios are required, there are several choices that may be made toward obtaining the most sensitive estimates of the more recent environmental conditions through sampling plant tissues. Amongst the factors that are involved are the age and growth rate of the tissues. Growth rates of few species have been studied, however, the giant brown alga Macrocystis pyrifera is of commercial importance and some aspects of its growth therefore have been recorded. It grows near the San Onofre power plant and also grows several hundred miles to the north and south. The plants grow in "forests" beyond the surf line, sending up numerous slender stipes 15-30 meters long tying together a hundred or more blades, each buoyed by a small bulb. The distal blades grow extremely rapidly, and the oldest, deepest, blades are seldom more than six months old. In a recent study at Scripps [17], radioactive concentrations on numerous blades along stipes were compared. At distances of about 1/2 meter from the growing (distal) end of the stipe to about 10 meters nearer to the older (holdfast) end, the surface concentrations of plutonium-239 were found to increase at a fairly uniform rate from 1.0×10^{-17} curies/cm^2 to 4.0×10^{-17} curies/cm^2. Since all of the leaves, on the portions of stipe that were studied, had developed within a period estimated at about 120 days, it may be concluded that plutonium-239 deposited at a fairly uniform rate of about $3.0 \times 10^{-17}/120$ or 2.5×10^{-19} curies/day/cm^2 over the blade surfaces.

Since typical blades are 0.4 mm thick, there are 50,000 cm^2 of area/wet Kg of wet blade sample; therefore, $2.5 \times 10^{-19} \times 5 \times 10^4 \cong 1.3 \times 10^{-14}$ curies are accumulated per day per Kg of wet sample.

Young (ten-day old) blades having 1.0×10^{-17} curies/cm^2 or 0.5 pCi/Kg have commonly been observed. Moreover, older blades with roughly 4 times higher concentrations might have been selected where the most recent environmental conditions were not the main objective.

Recent clean sea water samples collected near Scripps may be expected to contain about 6×10^{-4} pCi plutonium-239/litre and about 3×10^{-5} pCi plutonium-238/litre. Thus sampling 1 Kg of mature Macrocystis kelp blades corresponds to sampling $4 \times 0.5/6 \times 10^{-4}$ or about 3300 litres of sea water. For this reason, only about 100 g of wet leaves are now used at Scripps for monitoring coastal plutonium-239, and larger samples are needed only when the less abundant plutonium-238 nuclide is to be determined with great precision.

Table IV lists a few plutonium-239/plutonium-238 ratios determined near the San Onofre power plant and elsewhere for comparison. Some useful

comparisons are afforded by this data were collected during early procedural development studies. However, demonstration of an anomalous plutonium ratio near the power plant has not yet been made. Nevertheless, very re ntly developed techniques now appear to make tests of this sort possib e and also relatively simple to carry out.

DISCUSSION

Relatively few people who must develop coastal pollution surveillance methods will be afforded the convenience of a giant alga for their studies. However, there are various other effective marine concentrators although few have been studied in any detail as indicator organisms for specific elements. A great deal more must be learned even about the algae and coastal grasses, how fast they grow, which nuclides collect selectively in which species, and at what periods of their life cycles.

It was of interest to find, soon after it was noted that plutonium deposited from sea water rigorously proportional to areas of interfacial surfaces that were exposed, that another alpha emitting radionuclide accumulated proportionally in many of the same surfaces, but at a much higher rate. This is, on one hand, a nuisance requiring careful chemical purification of the plutonium before counting is done. On the other hand, this second nuclide, natural polonium-210, is so abundant and so easy to purify and analyze that its trends and its distributions in the marine environment can suggest some of the probable behavior of the much-harder-to-measure plutonium nuclides. For example, the activity of polonium-210 has, in studies of hundreds of brown algal blades, been found to deposit upon a unit area 230±50 times as intensely as plutonium-239 deposits, whether the blade is 10 days or 120 days old. Similar parallel deposition rates of polonium and plutonium are evident, although still not fully studied, in connection with several other sea water/biota interfaces common in the marine world. Why two, so unlike, elements should behave this way in the ocean is philosophically exciting to some of us. Other people too may discover that there is a convenient way for studying growth and deposition rates; measuring polonium-210 is a relatively simple and easy way of learning something about the behavior of extremely small traces of plutonium.

A warning should be given, however, that no further simplicity in the polonium-210/plutonium-239 ratio has been observed further up in the trophic web. Plutonium usually concentrates very little in tissues of higher organisms. Edible muscular portions of fish concentrate plutonium activities only 2 or 3 times the levels in the sea [13] while on thin algal blades the wet weight concentration may be a thousand-fold higher [15,16]. On the other hand, many tissues of higher organisms build up exceedingly high concentrations of the natural alpha emitter polonium-210. Fortunately, most of these tissues are seldom eaten by humans. For example, an extreme concentration of polonium-210 has been observed in tissues of the little studied and almost never eaten "pyloric caecal mass" of one of the large oceanic fish [20]. Here the polonium-210 activity has been found to accumulate 2.2 million times the typical sea water level; and the radioactive dosage burden (from 5 million volt alpha particles released inside these tissues) is suspected to be the highest ever encountered in bulk tissues of an organism living in a truly natural environment.

CONCLUSIONS

Extremely early signs of radioactive pollutions associated with coastal nuclear installations can be detected and identified at levels just above the contemporary backgrounds caused by global fallout. Special techniques are required, however.

Careful studies of the biological concentrators present in the environment as to growth habits and as to behavior toward specific elements at trace levels may uncover factors leading to extremely sensitive--and sometimes also convenient--surveillance procedures.

Many of the environmental factors needed for developing early surveillance methods are of a type much needed for understanding the ultimate capacities of the coastal environments that may be used for disposing much of the nuclear wastes of the future.

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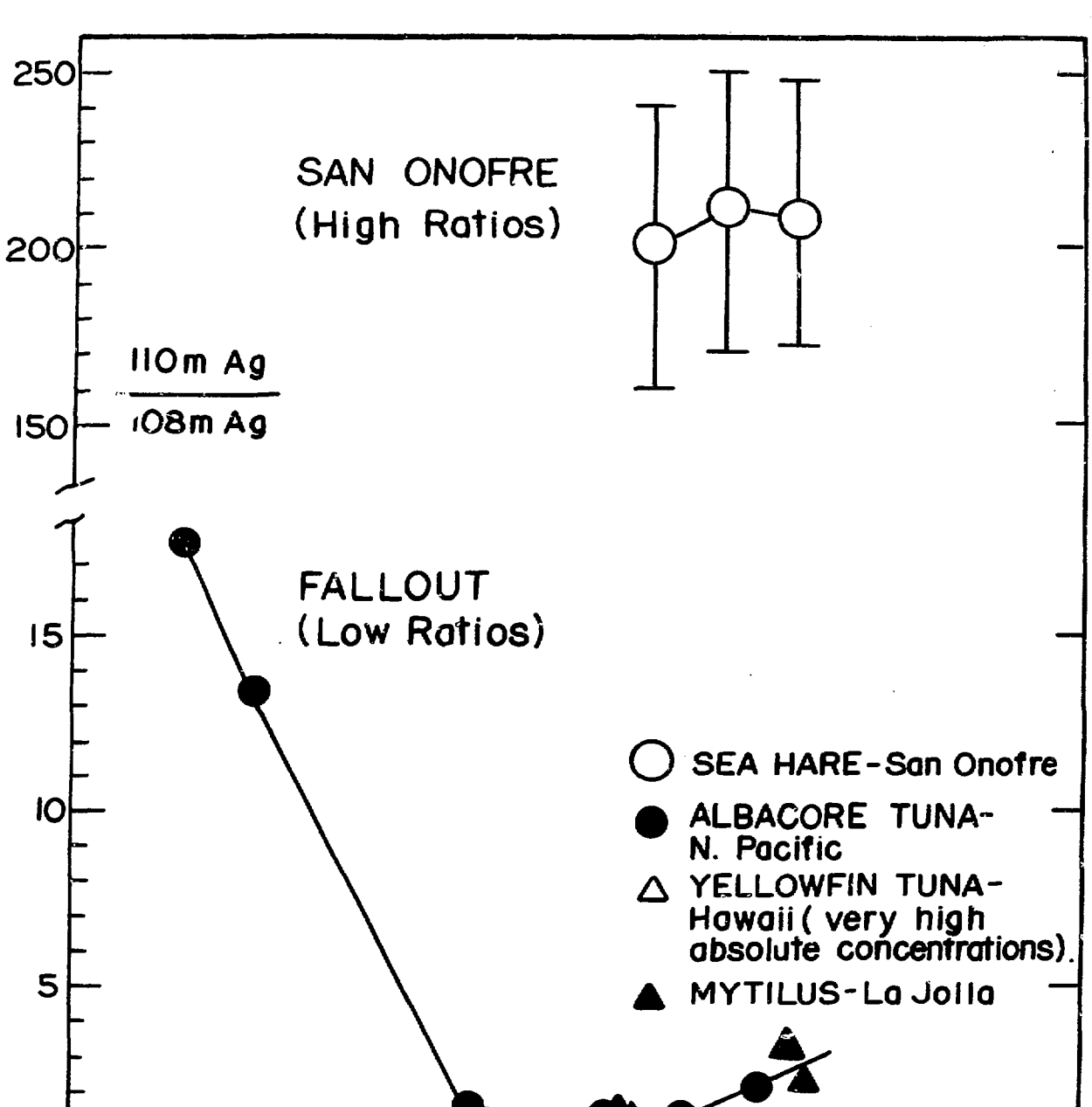

Figure 1. Distinctive silver nuclide ratios found near a coastal nuclear power plant at San Onofre, California. The much different ratios in fallout for different years and in different biological samples are shown. See Table II.

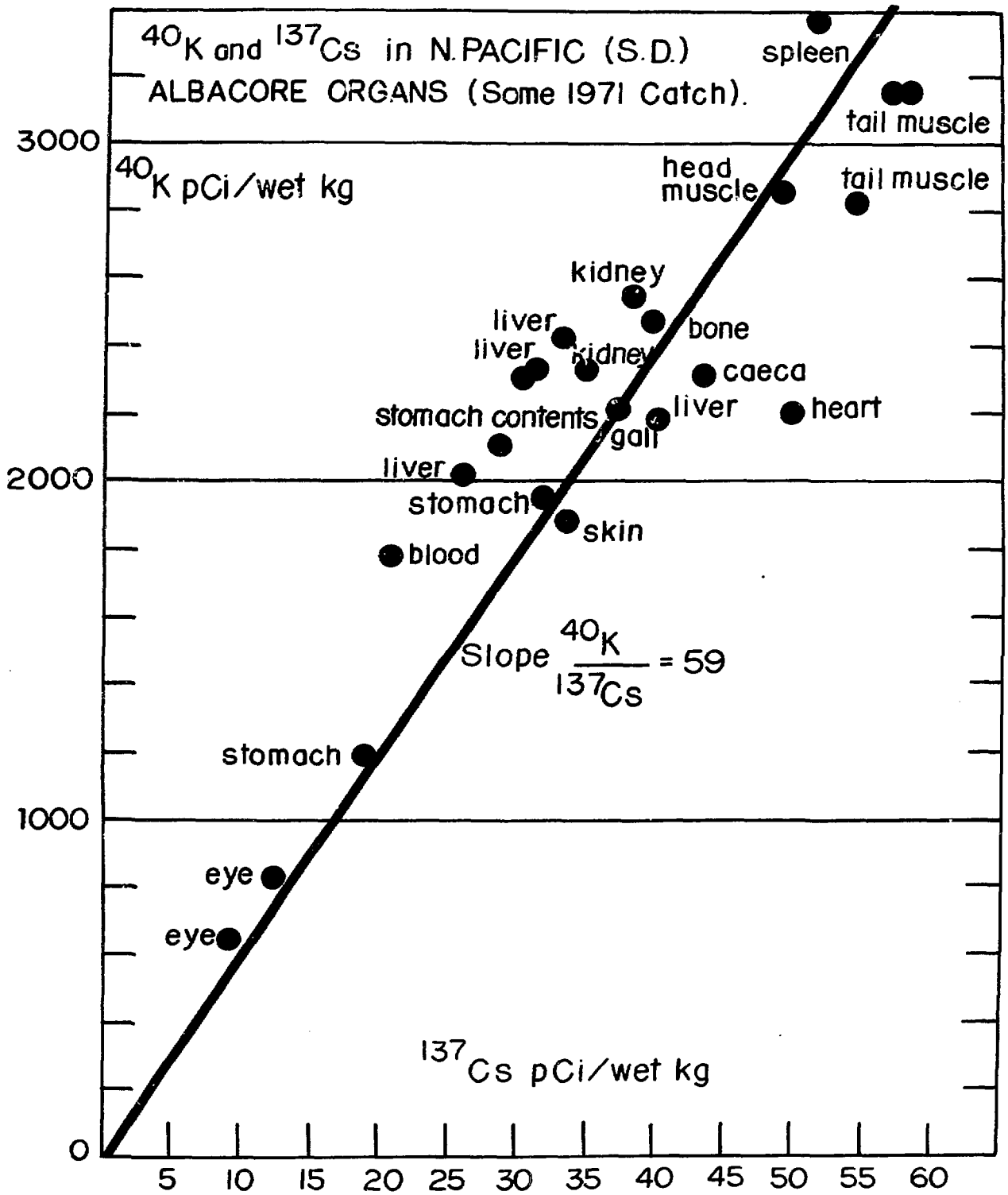

Figure 2. Ranges of concentrations of caesium-137 and potassium-40 in a large oceanic fish Thunnus alalunga. The parallel behavior of these two nuclides suggest the usefulness of potassium for normalizing biological samples to be assayed for traces of radiocaesium. In the ocean, natural caesium is strictly proportional to potassium.

Table I. Small coastal disposals of artificial radioactivities from the San Onofre Nuclear Power Plant evidenced by relatively high concentrations in selected marine organisms collected locally.[1]

Sample	Date Collected	pCi/Kg wet weight[2] ^{60}Co (5.3 yr)	^{58}Co (72 d)	^{110m}Ag (253 d)
Sea hare (Aplysia californica) a shell-less mollusc collected near San Onofre	12/12/70	85	2260	114
	3/10/71	63	734	78
	6/22/71	16	87	16
	10/ 4/71	45	125	49
	1/ 7/72	7	12	12
	12/18/72	34	64	195
collected near La Jolla	6/22/71	2	< 1	< 1
Agar Agar (Gelidium sp.) a red alga collected near San Onofre	12/12/70	24	313	35
	3/10/71	5	44	6
	11/ 2/71	4	11	13
collected near La Jolla	6/22/71	5	< 1	< 1
Surf grass (Phyllospadix sp.) a flowering plant collected near San Onofre	3/10/71	31	426	42
	11/ 2/71	21	42	25
	1/17/72	9	15	14
	12/18/72	58	160	85
collected near La Jolla	3/10/71	9	< 1	< 1

[1] For comparison, concentrations are given for the same species collected at La Jolla, Ca., 70 Km away.

[2] Counting errors of measurements are 10% or better. Half-lives of these three artificial nuclides are given in parentheses. Other nuclides also have been detected.

Table II. Silver-110m/Silver-108m ratios at San Onofre and elsewhere.

Location	Date	Species		pCi/Kg wet ^{110m}Ag	pCi/Kg wet ^{108m}Ag	$^{110m}Ag/^{108m}Ag$
San Onofre	12/70	sea hare	3	120	0.6	200
	10/71	Aplysia calif.	3	49	(~0.23)	213
	12/72	(whole)	3	195	0.9	217
San Diego	7/64	albacore tuna	26	81	4.6	17.6
(100 miles	7/65	Thunnus alalunga	26	100	7.5	13.3
west)	7/68	(livers)	30	8.7	5.9	1.5
	7/70		59	5.8	4.6	1.3
	7/71		76	4.9	4.2	1.2
	7/72		60	10.1	4.8	2.1
Hawaii	9/70	yellowfin tuna Thunnus albacares (livers)	2	123	113	1.1
La Jolla	10/70	mussel	30	0.7	0.5	1.4
	1/73	Mytilus calif.	30	1.9	0.6	3.2
	2/73	(whole)	30	1.5	0.7	2.1

Table III. Caesium-137/Caesium-134 ratios at San Onofre and elsewhere.

Location	Date	Sample	$^{137}Cs/^{134}Cs$
Pacific Ocean[1]	1971	open ocean sea water	> 100
San Onofre, Ca.	7/6/71	discharge water	1.4
Indian Point, N.Y.[2]	1970	fish	1.0
United Kingdom[3]	11/70	coastal sea water	7.0

[1] Typical 1971 sea water near California had about 0.3 pCi/ℓ ^{137}Cs.
[2] Power reactor on Hudson River estuary [9].
[3] Measured at Scripps for the IAEA sea water reference sample intercalibrations, 1971 [10].

Table IV. Plutonium-239/Plutonium-238 ratios at San Onofre and elsewhere.

Location	Date	Sample	pCi/Kg wet[1] ^{239}Pu	pCi/Kg wet[1] ^{238}Pu	$^{239}Pu/^{238}Pu$
San Onofre	3/71	surf grass	0.27±0.02	0.015±0.005	18±6
San Onofre	5/73	*Phyllospadix sp.* brown algae	1.33±0.06	0.074±0.021	18±5
La Jolla	5/73	*Macrocystis sp.* brown algae	1.50±0.05	0.068±0.016	22±5
La Jolla[2]	11/71	*Pelagophycus sp.* brown algae	2.16±0.15	0.12±0.04	18±6
La Jolla[2]	7/71	*Pelagophycus sp.* Calif. sea water	0.0006±0.0002	-	-
La Jolla[3]	4/64	brown algae	0.45±0.02	0.015±0.005	30±10
La Jolla[3]	4/64	*Eisenia sp.* Calif. sea water	0.0004±0.0001	-	-
Irish Sea[4,7]		"sea weed"	2230±50	330±10	6.8±0.3
40°N[5]	12/64	HASL soil, before SNAP accident			38
40°N[5]	1970/71	HASL soil, after SNAP accident			27±4
40°N[6]	1971	HASL surface air samplings			1 to 13

[1] One counting error (± one standard deviation).
[2] See Ref. [16].
[3] See Ref. [13].
[4] Measured at Scripps for the IAEA sea weed reference sample intercalibrations 1972 (technical report).
[5] See Ref. [18].
[6] See Ref. [19].
[7] Assuming 10:1 wet weight/dry weight.