SCCWRP #0006

JUNE 1972

MUSSELS AND BARNACLES AS INDICATORS OF THE VARIATION OF MANGANESE-54, COBALT-60, AND ZINC-65 IN THE MARINE ENVIRONMENT

D. R. YOUNG

T. R. FOLSOM

To be presented at the IAEA Symposium on the Interaction of Radioactive Contaminants with the Constituents of the Marine Environment Seattle, Washington 10-14 July 1972

*

SOUTHERN CALIFORNIA COASTAL WATER RESEARCH PROJECT SCCVPP A LOCAL GOVERNMENT AGENCY FOR MARINE ECOLOGICAL RESEARCH MUSSELS AND BARNACLES AS INDICATORS OF THE VARIATION OF MANGANESE-54, COBALT-60, AND ZINC-65 IN THE MARINE ENVIRONMENT

D. R. YOUNG Southern California Coastal Water Research Project Los Angeles, California, U.S.A.

T. R. FOLSOM Scripps Institution of Oceanography La Jolla, California, U.S.A.

ABSTRACT

The intertidal byssal mussel Mytilus californianus and the oceanic gooseneck barnacle Lepas anatifera are efficient indicators of spatial and temporal changes in levels of three radiometals in the marine environment. Lepas specimens collected from the northeastern Pacific during the first half of 1964 demonstrated oceanic-to-coastal ratios of manganese-54, cobalt-60, and zinc-65 fallout from the 1961-62 thermonuclear tests of approximately 3:1, 4:1, and 2:1, respectively. Cesium-137 in the surface layers of the two sectors (whose centers lie about 1,500 and 300 km west of San Diego) also showed a 3:1 oceanic-to-coastal enhancement of fallout from this source. Mytilus specimens collected during 1963-64 along the northeastern Pacific Coast showed a fairly uniform distribution of manganese-54 and cobalt-60 between latitudes 46 and 29° north, but dramatically reflected the point source of Hanford-produced zinc-65 emanating from the mouth of the Columbia River. Relatively high zinc-65 concentrations, possibly related to this source, were detected in Mytilus from northern Baja California, Mexico--1,900 km to the south.

Except for a late-1963 peak in manganese-54 concentrations in southern California <u>Mytilus</u>, the ecological half-times of the three nuclides observed in the invertebrates between mid-1963 and late 1964 at specific intertidal, coastal, and oceanic platforms were consistent with radioactive decay rates. This suggests an approximate equilibrium between input and removal of these radiometals in the mixed layer.

Mussel and barnacle tissue-to-seawater enrichment factors for the three muclides exceed 1,000. In <u>Mytilus</u> 4 to 16 cm in length, size variations had no significant effect on nuclide concentration, but 70% of the cobalt-60 and zinc-65 soft-tissue radioactivity was located in the kidneys and digestive glands of this organism.

Bay mussels (<u>M. edulis</u>) taken from the coastal surf zone showed concentrations similar to those in <u>M. californianus</u>, but <u>M. edulis</u> in two nearby bays had lower values. Intertidal coastal mussels and gooseneck barnacles recently collected from 14 California stations indicate average 1971 "baseline concentrations" of manganese-54, cobalt-60, and zinc-65 of 0-5, 0-4, and 0-11 pCi/wet kg, respectively.

MUSSELS AND BARNACLES AS INDICATORS OF THE VARIATION OF MANGANESE-54, COBALT-60, AND ZINC-65 IN THE MARINE ENVIRONMENT

INTRODUCTION

The radioisotopes manganese-54, cobalt-60, and zinc-65, which enter the marine ecosystem from fallout and other sources, are difficult to measure in seawater, even when very large samples are processed. In view of the recent renewals of atmospheric testing of nuclear devices and the growing utilization of nuclear power, it is desirable to be able to detect and follow these neutron-induced isotopes in some other component of the marine environment. The study described in this paper suggests that the intertidal mussel <u>Mytilus californianus</u> and the surface-feeding oceanic barnacle <u>Lepas anatifera</u> reflected spatial and temporal differences of manganese-54, cobalt-60, and zinc-65 in the surface layer of the northeastern Pacific Ocean during 1963 and 1964. Thus, these organisms may be useful as indicators of the variability of the three radiometals in marine waters.

For more than a decade, a study of radioactive fallout in the Pacific Ocean has been in progress at Scripps Institution of Oceanography [1, 2]. In the spring of 1963, samples of both a phytoplankton bloom ¹ and the phytoplankton-feeding mussel <u>M. californianus</u> from Scripps pier, La Jolla, first indicated the presence of manganese-54 in southern California waters [3]. Zinc-65 also was measured in the mussel samples; following the development of a specialized spectrometer, cobalt-60 levels in the mussels were determined as well [4, 5]. The principle source of these radiometals is believed to have been the large atmospheric nuclear tests of 1961-62.²

Seawater samples were also taken in the 1963 survey. Although we were able to measure manganese-54 in 200-liter samples, zinc-65 was undetectable in samples as large as 1,000 liters [3], and a 2,000-liter sample did not yield any measurable cobalt-60.

This earlier study convinced us that the most practical method of investigating the distribution of these three radiometals in the northeastern Pacific Ocean would be to utilize the concentrating ability of selected marine organisms, such

^{1.} Mostly dinoflagellates, predominantly Gonyaulax polyedra.

^{2.} The time of activation of two other neutron-induced radiometals, silver-110m and silver-108m, that also appeared in the marine biota following these tests has been determined to be 1961-62 from the activity ratios observed in various samples [6,7,8].

as <u>M. californianus</u>. The intertidal gooseneck barnacle <u>Pollicipes</u> <u>polymerus</u> and the oceanic, surface-living gooseneck barnacle <u>Lepas</u> <u>anatifera</u> were also found to accumulate these isotopes to easily measurable levels. As <u>M. californianus</u> ranges from the middle of Baja California, Mexico (approximately 30° north) to the Gulf of Alaska (approximately 60° north) [9], and <u>L. anatifera</u> is found on flotsam throughout the northeastern Pacific, efforts in the program described in this paper were centered on these two organisms.

e 5

SAMPLING METHODS

Collections of the coastal mussel <u>M</u>. <u>californianus</u> were made from four different regions of the northeastern Pacific coast in 1963-64; logistical problems required that each region be studied at a different season. The first region, the southern California coastline, was sampled at nine stations (shown on Figure 1) between Point Dume and National City during spring of 1963. Efforts in northern California (the second region) were concentrated at Bodega Head (Figure 2), about 80 km north of San Francisco; six collections were made here between June and August 1963.

In the summer and fall of 1963, <u>M. californianus</u> collections were made in Oregon (the third region) at four stations south of the Columbia River (Figure 2).³ In early 1964, five stations (Figure 1) off northern Baja Califor-

3. These samples were taken in conjunction with a zinc 65 loss-rate experiment that has already been reported [10].







Figure 2. Four regions studied, and representative values (pCi/wet kg soft tissue) of manganese-54, cobalt-60 and zinc-65 in Mytilus californianus from each region. The data for each station in the Oregon region are given so that possible coastal gradients can be examined.

nia, the fourth region, were sampled for this species. To investigate concentration changes at at least one station over this time interval, <u>M. californianus</u> specimens from Scripps pier were analyzed periodically between spring 1963 and fall 1964. Collections of the bay mussel <u>M. edulis</u> also were made periodically from Scripps pier and from nearby Mission and San Diego Bays.

Collections of the oceanic gooseneck barnacle <u>L</u>. <u>anatifera</u> were first made in the spring of 1963 from a floating instrument platform anchored 10 km off La Jolla. Subsequent samples were obtained there during the winter, summer, and fall of 1964. A similar time series was obtained for <u>L</u>. <u>anatifera</u> collected from another floating instrument platform anchored at Ocean Station "November," approximately 2, 500 km southwest of La Jolla. In addition, a number of <u>L</u>. <u>anatifera</u> samples were collected during 1964 from anchored floats and drifting material in the northeastern Pacific between the California coast and Ocean Station "November."

ANALYTICAL PROCEDURE

In preparing the <u>Mytilus</u> and <u>L</u>. <u>anatifera</u> specimens for radioassay, only the soft tissues of the mussels were taken, while the entire gooseneck barnacles were utilized. The mussel samples, which usually ranged between 8 and 14 cm in length, were separated from the shells in the field or upon return to the laboratory. After draining off excess seawater, the tissues were placed in a cylindrical polyethylene container 10 cm in diameter and preserved with a small quantity of Formalin. The barnacle samples, which ranged between 2 and 10 cm in length, were drained and preserved in the field; thus, the entire sample, along with the preservative, was transferred to the assay container. Typical sample sizes for <u>Mytilus</u> and <u>L</u>. anatifera were 300 and 100 wet g, respectively.

The gamma-ray spectra were obtained by first counting the sample for 1,000 min on a 5-by 4-in. NaI (Tl) crystal in conjunction with a 200-channel pulse-height analyzer. Corresponding backgrounds were subtracted electronically, and corrections were made for varying sample height. The manganese-54 and zinc-65 signals were determined from the areas of their photopeaks (0.84 MeV and 1.11 MeV, respectively). The cobalt-60 concentrations generally were determined by drying or ashing the sample, along with the preservative fluid, and counting the residue on a specially-designed summing-coincidence gamma-ray spectrometer [11]. (By utilizing the cobalt-60 sum peak at 2.50 MeV, we were able to avoid interferences from zinc-65 and potassium-40 and significantly increase our capability for measuring cobalt-60 in the biological samples.)

RESULTS

Figure 2 gives representative concentrations of manganese-54, cobalt-60, and zinc-65 observed in the entire soft tissues of <u>M. californianus</u> collected between April 1963 and February 1964 along the northeastern Pacific Coast.⁴ The coastline stations sampled have been grouped into

^{4.} In this report, all nuclide concentrations are corrected for radioactive decay to the collection date: The physical half-times used for manganese-54, cobalt-60, and zinc-65 were 291 days, 5.2 years, and 244 days, respectively.

four regions: Oregon, northern California (Bodega Head), southern California, and northern Baja California. The region means, and their standard deviations and 95% confidence intervals, have been given for all but the Oregon group--the data for this region were not averaged so that possible gradients downcurrent from the Columbia River could be examined.

The concentrations of the three radiometals observed in whole <u>L</u>. <u>anatifera</u> specimens collected from floating objects in the northeastern Pacific are plotted in Figures 3 through 5. The crosses indicate anchored substrates, and the dots indicate drifting substrates; most of the values for anchored substrates are based on two or more replicate samples. The samples from Sectors I, IIA, and IIB were collected during January 1964, May to September 1964, and June to August 1964,⁵ respectively. (For comparison, previously reported [12] cesium-137 concentrations in surface water samples collected from the same general areas are plotted in Figure 6; this figure is discussed in the following section.)

Figure 7 illustrates the time series data for manganese-54 and zinc-65 in <u>L. anatifera</u>. Time series data for all three radionuclides in <u>M. californianus</u> are presented in Figure 8.

Table I gives statistical upper limits (with 95% confidence) of manganese-54, zinc-65, and cobalt-60 concentrations in 1971 samples of <u>M.</u> <u>californianus</u> and <u>P. polymerus</u> from throughout the Southern California Bight. These upper limit values are based on 2 sigma of the counting background beneath the photopeak and are corrected for radioactive decay to the collection date.

DISCUSSION

The circulation of the waters in the northeastern Pacific appears to have had an important bearing on the distribution of manganese-54, cobalt-60, and zinc-65 in the mussels and barnacles surveyed in this study. The North Pacific Current flows eastward between latitudes 40 and 50° north--the latitudes of highest radioactive fallout--until it reaches the North American continent. Much of the flow is then deflected south to form the California Current [13,14]. At approximately latitude 46° north, the Columbia River empties into the Pacific; this river receives nuclear reactor cooling water at Hanford, Washington, and has been a significant source of radioactivity to the northeastern Pacific for many years [15,16,17]. Thus, near its point of origin, the California Current is subject to radioactive inputs from two sources, atmospheric fallout and Columbia River effluent.

The current itself is a relatively wide, slow moving body of water. Sverdrup and Fleming [18] have estimated that more than 90% of the southeastern transport occurs in the top 300 m and within 800 km of the coast. The mean speed of the current is 15 cm/sec--about 13 km/day.

As shown in Figure 1, after passing southern California, the California Current swings shoreward opposite northern Baja California. Part of the flow then turns northward inshore of the southerly flow, entering a complex

^{5.} The values at 30° N, 140° W (Ocean Station "November") are averages of March and September 1964 collections.







•

Figure 4. Concentrations of cobalt-60 (pCi/wet kg) in whole Lepas anatifera collected during 1964 from floating anchored (+) and drifting (*) substrates in the northeastern Pacific.



Figure 5. Concentrations of zinc-65 (pCi/wet kg) in whole Lepas anatifera collected during 1964 from floating anchored (+) and drifting (9) substrates in the northeastern Pacific.



Figure 6. Comparison of cesium-137 values (pCi/100 liters) in surface waters of the northeastern Pacific before and after the nuclear weapons tests of 1961-62. After Folsom, et al., 1968, figs. 1.1 and 1.3.

~

Collec	etion			Nuc	lide Co (pCi/v	ncentr wet kg)	ations	
	Lat.		54 ₁	Mn	60	Co	65	Zn
Station	(0N)	Date	<u>М.с.</u>	<u>P.p</u> .	<u>M.c</u> .	<u>P. p.</u>	<u>M</u> .c.	<u>Р.р.</u>
Bodega Hd.	380151	24 Jul	3.7	4.6	3.5	4.1	9.7	12.0
Pt. Sal	340451	23 Oct	-	7.7	-	8.3	-	17.3
San Miguel I.	י34 ⁰ 00י	23 Jun	3.9		3.5	-	10.5	_
Anacapa I.	34000'	21 Jun	4.3	4.4	3.5	3.5	11.1	11.0
Santa Monica	34 ⁰ 00'	10 Sep	3.1	3.8	3.0	3.7	6.1	9.7
Pt. Vincente	33045'	6 Sep	3.1		3.0	-	6.1	-
Seal Bch.	330451	8 Sep	3.1	_	3.0	_	6.1	-
Newport Bch.	33040'	25 Jun	-	7.2	-	6.4	-	19.3
Santa Barbara I.	33030'	21 Jun	-	6.0	-	5.1	-	16.2
San Clem. Bch. ^a	33030'	13 Jun	7.1	5.2	3.5	5.7	9,9	12.0
San Nicholas I.	33 ⁰ 15'	29 Jun	3.8	4.7	3.5	4.2	10.1	12.1
Oceanside	33 ⁰ 15'	22 Oct	-	5.1	-	5.7	-	12.1
San Clemente I.	330001	16 Jun	-	7.4	-	5.5	-	21.2
Punta Banda	32000	13 Nov	3.1	3.1	3.3	3.3	6.9	6.9
	Ave	rage	3.9	5.4	3.3	5.0	8.5	13.6

100

Table I. Upper limits (with 95% confidence) of 1971 concentrations of manganese-54, cobalt-60, and zinc-65 in <u>Mytilus</u> californianus soft tissues and whole <u>Pollicipes</u> polymerus from intertidal stations in California and Baja California.

a. San Clemente Beach P. polymerus collected 26 Oct 71.



Figure 7. Time series of manganese-54 and zinc-65 concentrations in samples of whole Lepas anatifera from floating instrument platforms anchored 2,500 and 10 km southeast of La Jolla, California (T_{E 1/2} = estimated ecological half-time).



N 6

< 3

ħ

Figure 8. Time series of manganese-54, zinc-65, and cobalt-60, in soft tissue samples of <u>Mytilus californianus</u> from Scripps pier, La Jolla. The estimated linear fits (solid lines) for manganese-54 and zinc-65 plots yielded the ecological half-times (T_E 1/2) shown. For cobalt-60, the mean value is indicated (dotted line).

system called the Southern California Gyre; the remaining portion of the surface current continues south [14].

Below a depth of about 150 m, the water off northern Baja California is flowing northwest. This flow, called the California Undercurrent, travels adjacent to the continental slope at speeds of up to 25 cm/sec and has a temperature and salinity characteristic of water of the equatorial Pacific [19]. The California Undercurrent normally passes beneath the Southern California Gyre, and Emery [20] has proposed that, at 200 to 300 m, 50% of the water in the Southern California Bight is southern water and 50% is northern water.

In the late fall and winter, a northward-moving surface flow, called the Davidson Current, appears inshore, north of Point Conception. This current, identified as southern in character by Sverdrup and Fleming [18], may be part of the California Undercurrent. The Davidson Current continues along northern California within 150 km of the coast, but it disappears from the surface in spring, when the strong winds from the northeast begin [21]. These winds continue into summer and tend to speed up the California Current and to extend the southeasterly flow of water to the coast. However, during certain periods, there may be an insufficient component of the wind toward shore at some points along the coast; in such cases, upwelling of water from depths of 200 to 300 m may occur [14].

During the early 1960's, zinc-65 was one of the predominant gammaemitters found in marine organisms from California Current waters near the mouth of the Columbia River [22, 23]. Most of the zinc-65 in waters off Washington and Oregon at that time has been attributed to Columbia River effluent [16, 24], which transported approximately 25,000 Ci/mo of radiozinc to the sea [25]. The dramatic accumulation of zinc-65 over other radionuclides in local marine biota, which has been well documented in the past [15, 16, 26], was again observed in our study: Concentrations of zinc-65 in Oregon mussels were two and three orders of magnitude higher, respectively, than manganese-54 and cobalt-60 concentrations. In addition, the two- to three-order of magnitude difference between zinc-65 levels in Oregon mussels and concentrations in those from the California regions also points to the Columbia River as the principle source of zinc-65 in Oregon waters at this time.

Although the averages of manganese-54 and cobalt-60 were essentially the same in mussels from both southern California and Baja California, there were distinct differences in the zinc-65 values for these two regions. Radiozinc concentrations below the U.S.-Mexico border showed an increase, significant at the 95% confidence level, above those in southern California specimens.⁶ Highest zinc-65 levels were found in samples from Punta Banda (116 pCi/wet kg), Punta Calaveras (106 pCi/wet kg), and Punta Colnett (122 pCi/wet kg)--points on the section of the northern Baja California coast where the California Current swings shoreward. These data suggest that the mainstream of the current carried anomalously high concentrations of zinc-65, possibly from the Columbia River.

^{6.} Such comparisons are based on the assumption that the concentration values are normally distributed about their means.

The Bodega Head mussels also seemed to be exhibiting the impact of the water circulation characteristics of the Pacific Coast. The low absolute concentrations of manganese-54 and cobalt-60 in the northern California samples, which were collected between June and August 1963, may have been a reflection of the influence of the Davidson Current or of upwelling from the California Undercurrent; either of these flows would expose the Bodega Head mussels to water from the eastern Equatorial Pacific, where nuclear fallout levels are significantly lower than in the northern latitudes [12]. However, zinc-65 concentrations in these mussels were relatively high compared to the values for the other two nuclides, suggesting a radiozinc source other than nuclear fallout: The northern California mussels, like the Oregon specimens, may have been exposed to Columbia River zinc-65 through the California Current.

Concentrations of the three nuclides in the L. anatifera surveyed (Figures 3 through 5) also suggest relatively high zinc-65 levels in the California Current. Because crustaceans and mollusks generally accumulate trace elements to differing degrees, the absolute levels of the three radionuclides in L. anatifera and M. californianus are not comparable. But a comparison of the levels of each nuclide in the barnacles from Sector I stations (Figures 3 through 5) is pertinent.⁷ Manganese-54 (Figure 3) and cobalt-60 (Figure 4) concentrations at the three stations in the mainstream of the California Current (121 to 123° west longitude) were essentially the same as those for the inshore station off San Diego, which is in the Southern California Gyre. However, zinc-65 concentrations in barnacles from the three stations in the California Current were significantly higher than those for the samples from the inshore station, again indicating that the mainstream of this major current carried anomalously high zinc-65 levels southward along the Pacific Coast.

On a larger scale, Figures 3 through 5 show a distinct geographic difference in the <u>L.</u> anatifera concentrations of all three radionuclides. Average of the values in Sectors I, IIA, and IIB (given in picocuries per wet kilogram in the following table) reveal an increase toward the west:

	<u>Coastal Sector</u>	Oceanio	<u>Oceanic Sector</u>		
	r	<u>IIA</u>	IIB		
Manganese-54	上 550	1,260	2,450		
Cobalt-60	20	50	110		
Zinc-65	240	310	670		

These oceanic-to-coastal enhancements in the barnacle nuclide concentrations could be representative of higher concentrations of the radionuclides in waters to the west. An alternate hypothesis is that some quality of the oceanic water increased the ability of <u>L</u>. <u>anatifera</u> living there to accumulate the three trace radiometals. Unfortunately, we know of no 1964 measurements of seawater concentrations of manganese-54, cobalt-60, or zinc-65 in the oceanic and coastal sectors in guestion. However, we did make a

^{7.} These values are averages of two to four samples collected during January 1964.

few measurements of fallout cesium-137 in surface water samples collected during 1964, both off the southern California coast (Sector I) and between 125 and 140° west longitude (Sector II); these concentrations are plotted in Figure 6. For comparison, cesium-137 concentrations in surface seawater samples obtained in a 1960-61 survey are also shown in this figure.⁸

The major source of the manganese-54, cobalt-60, and zinc-65 in the barnacles collected in 1964 is believed to have been the 1961-62 nuclear tests. In contrast, there was a significant cesium-137 baseline level in the surface waters of the North Pacific in 1959-61 that was a result of the nuclear tests that occurred prior to 1959 [12]. To obtain an estimate of the relative amounts of 1961-62 nuclear fallout in seawater in the two sectors during 1964, we have subtracted the average 1960-61 cesium-137 concentration for each sector from the average 1964 value for that sector (values are in picocuries per liter):

	<u>Coastal Sector</u>	Oceanic Sector	
	<u>I</u>	<u>II</u> III	
1964 Cesium-137	е — 0.25	0.60	
1960-61 Cesium-137	0.15	0.25	
Net Cesium-137	0.10	0.35	

The net cesium-137 values were then used to obtain an approximate oceanicto-coastal ratio of 1961-62 fallout in the surface waters. In the following table, these values are compared with concentrations of manganese-54, cobalt 60, and zinc-65 in <u>L. anatifera</u> from Sectors I and II on Figures 3 through 5 (values for Sectors IIA and IIB on the figures have been averaged into a single Sector II value for ease of comparison):

	Coastal	<u>Oceanic</u>	Oceanic-to-Coastal
	T	II	II:I
	Е ———	W	
Manganese-54	550	1,860	3.4
Cobalt-60	20	80	4.0
Zinc-65	240	490	2.0
Cesium-137	0.10	0.35	3.5

Because of spatial and time differences and the relatively few data available, the above comparison can only be considered approximate. However, it does indicate that oceanic-to-coastal ratios of fallout nuclides from the 1961-62 nuclear tests in <u>L. anatifera</u> and seawater samples were in reasonable agreement. This in turn lends further support to our hypothesis that the zinc-65 gradients observed in <u>L. anatifera</u> and <u>M. californianus</u> in southern and Baja California waters were a reflection of anomalously high zinc-65 concentrations in the mainstream of the California Current, possibly a signature of the Columbia River.

Thus, the distributions of manganese-54, cobalt-60, and zinc-65 observed in <u>M. californianus</u> and <u>L. anatifera</u> during 1963-64 seem to be directly related to known patterns of water circulation and nuclear inputs in the North-

^{8.} In plotting these concentrations, we have not separated Sector II into Parts A and B because of the relatively few data available.

eastern Pacific. This aspect of our findings has particular relevance to present studies of pollution in southern California coastal waters, as it suggests that some of the contaminants observed in marine waters immediately to the south and west of the highly populated southern California region may have in fact emanated from areas much farther to the north.

The data discussed above suggest that L. anatifera and M. californianus are useful indicators of spatial changes in seawater radiometal concentrations in surface layers of the ocean. These organisms may also be efficient indicators of temporal changes in surface waters. Marine mollusks and crustaceans are reported to respond relatively quickly to changes in environmental concentrations of the three radiometals; biological half-times generally fall within the range of a few days to a few months [10, 27-3]]. Time series data on manganese-54 and zinc-65 in L. anatifera specimens collected in 1963-64 from stations 2,500 and 10 km off La Jolla are shown in the semilogarithmic plots of Figure 7. Figure 8 presents semilogarithmic plots of the levels of manganese-54, zinc-65, and cobalt-60 in M. californianus collected from Scripps pier, La Jolla, during this time interval. Estimated linear fits to these plots have been drawn (solid lines); such fits assume an exponential loss rate. (The data on cobalt-60 in the mussels were too scattered to allow estimation of a loss rate--we have simply indicated the mean value with a dotted line.) Using these lines, ecological half-times (the average time that it takes for the observed concentration of the radionuclide in a population in an undescribed environment to decrease by half) have been estimated. The majo observation from these data is that, between mid-1963 and the end of 1964, the ecological half-times of these radionuclides in oceanic and nearshore L. anatifera and in intertidal M. californianus were roughly consistent with a model of a surfacelayer ecosystem in which the nuclide input and output rates are equal and the reservoir concentrations are governed mainly be radioactive decay. The one exception appears to be manganese-54 in L. anatifera from Ocean Station "November," where the estimated ecological half-time of about 700 days is 2-1/2 times the decay rate of manganese-54. However, to the first order, the radioactive decay model described above is sufficient to explain the data.

ENRICHMENT FACTORS

14

A reliable determination of the average <u>M. californianus</u> and <u>L. anatifera</u> enrichment factors (relative to seawater concentration) for the three radiometals would require extensive seawater measurements; as indicated above, this was not possible in our study. However, a very few measurements of surface water from Scripps pier during the summer of 1963 suggested that the concentrations of manganese-54, cobalt-60 and zinc-65 in the inshore water of the Southern California Gyre were ~0.06, <0.005, and <0.01 pCi/liter, respectively. The manganese-54 concentrations were obtained from more than 10 actual determinations, but the cobalt-60 and zinc-65 maxima were based on only one and three large samples, respectively, and are order-ofmagnitude estimates only. Nevertheless, by dividing these estimates into the corresponding average radiometal levels observed during this period in <u>M. californianus</u> and <u>L. anatifera</u> specimens from Scripps pier and the coastal float anchored 10 km off La Jolla, we obtained average enrichment factors on the order of 1,000 to 10,000 or greater. These factors are consistent with biological enrichment values obtained from data on the general marine levels of manganese, cobalt, and zinc summarized by Goldberg [32] and Bowen [33]. Such data illustrate again the advantage of using indicator organisms to monitor trace level radiometals in marine ecosystems.

EFFECT OF MUSSEL SIZE

To determine the degree to which variations in the average size of mussels collected at different stations would influence observed radionuclide distributions, four length classes (4, 8, 11, and 16 cm) of <u>M</u>. <u>californianus</u> were collected at Bodega Head in July 1963. The entire soft tissues were assayed for manganese-54, cobalt-60 and zinc-65 concentrations. ⁹ Some data on the effects of size were also obtained in two collections (in mid-1963 and early 1964) at Punta Banda, Baja California; during the first collection, 15- and 30-cm mussels were grouped and assayed, while 5- and 15-cm groups were assayed in the second collection.

In the Bodega Head mussels, only zinc-65 showed a possibly significant trend with size--the 16-cm mussels showing somewhat less than 50% higher levels than the 4-cm samples. In the Punta Banda mussels, only cobalt-60 showed a possibly significant increase with size; again, the longest mussels had less than 50% higher levels than the smallest. As the majority of <u>M. californianus</u> collected for our coastal distribution study ranged between 8 and 14 cm in length, size differences could not have introduced any important bias into our results.

DISTRIBUTIONS IN MYTILUS TISSUES

To examine the distribution of manganese-54, cobalt-60, and zinc-65 in the soft tissues of <u>M. californianus</u>, individuals 16 cm in length were collected from Bodega Head in July 1963 and dissected into five soft tissue groups; percentages of the whole soft tissue nuclide concentrations associated with each tissue group are given in the following table:

		Nuclide				
	Wet Wt.	Con	<u>centrations</u>	ations (%)		
Tissue	<u>(%)</u> <u>54_{Mn}</u>		60 _{Co}	$\frac{65_{Zn}}{2}$		
Kidnevs	4	12	23	52		
Digestive Glands	12	25	45	20		
Gonads	23	21	6	8		
Ctenidia	12	8	10	5		
Musculature	49	34	16	15		

As the table shows, 70% of the cobalt-60 and zinc-65 present in the whole soft tissues of the mussels occurred in the kidneys and digestive glands. Thus, capability for detecting these two nuclides could be increased significantly by analyzing only the kidneys and digestive glands from a large number of specimens.

^{9.} The I-sigma relative counting error at the levels observed was approximately ±15%.

A separate analysis revealed that manganese-54 was easily measured in the shells of these mussels (although cobalt-60 and zinc-65 were not); shells had the same manganese-54 concentrations as the digestive glands. As mussel shell mass is greater than soft tissue mass, shell material, which is easily isolated, may be the most useful fraction of the mussel in studies to determine the presence of manganese-54 in marine surface waters.

47章

MYTILUS CALIFORNIANUS VS. M. EDULIS

In a geographical survey, it is often necessary to utilize another species of the indicator organism being studied. To determine if the bay mussel <u>Mytilus edulis</u> was a suitable substitute for <u>M. californianus</u> in a radiometal survey, a study of these two organisms was conducted from spring 1963 to spring 1964. Specimens of both species from Scripps pier, as well as specimens of <u>M. edulis</u> from nearby Mission and San Diego Bays, were analyzed for manganese-54, cobalt-60, and zinc-65. The table below summarizes the results with ratio averages for the three nuclides:

	Ratio Average and Standard			
Ratio	Deviation of the Mean			
Description	54Mn	60 _{Co}	65Zn	
<u>M.</u> <u>edulis</u> at Scripps pier to <u>M.</u> <u>calif.</u> at Scripps pier	1.3±0.2	1.1 ± 0.1	0.8 ± 0.1	
<u>M.</u> <u>edulis</u> at Mission Bay to <u>M. calif.</u> at Scripps pier	0.4 ± 0.1	0.5 ± 0.2	<0.3	
<u>M. edulis</u> at San Diego Bay to <u>M. calif.</u> at Scripps pier	0.6 ± 0.1	0.3±0.1	<0.3	

As the table indicates, there was generally good agreement between the concentrations of the nuclides in samples of M. edulis and M. californianus collected from Scripps pier, indicating that the behavior of M. edulis toward the three radiometals is essentially the same as that of M. californianus and that data on the two organisms may be interchanged or overlapped where necessary. This finding is important because, although M. californianus is distributed over more than 3,000 km of coastline in the northeastern Pacific, M. edulis is found along many coastlines of the world ocean.

However, this investigation showed that, for data to be interchangeable, all specimens must be collected from the coastal surf zone: The values for all three nuclides were significantly lower in <u>M. edulis</u> from the two bays than in the coastal specimens of both species.

1971 BASELINE LEVELS IN MUSSELS AND BARNACLES

During 1971, we again surveyed the distribution of manganese-54, cobalt-60, and zinc-65 in the intertidal zones of the Southern California Bight, using mussels and barnacles as indicator organisms. Assays were performed on whole

soft tissues of the byssal mussel <u>M</u>. <u>californianus</u> and on whole specimens of the gooseneck barnacle <u>Pollicipes</u> <u>polymerus</u> from 12 southern California stations; samples were also taken at Bodega Head in northern California and at Punta Banda in Baja California.

No artificial radioactivity was detected in any of the samples. In Table I, we have listed the upper limits (with 95% confidence) of the nuclide concentrations at each station. These data show that, on the average, 1971 "baseline concentrations" of the three nuclides in these two indicator organisms fell in the following ranges:

	54_{Mn}	60Co	65_{Zn}
<u>M. californianus</u>	0-4	0-3	0-8
P. polymerus	0-5	0-5	0 - 14

ACKNOWLEDGEMENTS

We wish to thank Prof. C. L. Osterberg and Prof. J. W. Hedgpeth, Oregon State University, Prof. J. D. Isaacs and Barbara Edwards, Scripps Institution of Oceanography, and the Commander Western Area, U.S. Coast Guard, for assistance in obtaining <u>Mytilus</u> and <u>Lepas</u> samples. The assistance of Deirdre Van Hofwegen and Robin Simpson, Southern California Coastal Water Research Project, is gratefully acknowledged.

This research was supported by the U.S. Atomic Energy Commission, the U.S. Office of Naval Research, and the Southern California Coastal Water Research Project.

REFERENCES

- [1] FOLSOM, T. R., MOHANRAO, G. L., WINCHELL, P., Fall-out cesium in surface sea water off the California coast, Nature 187 (1960) 480.
- [2] FOLSOM, T. R., SREEKUMARAN, C., "Some reference methods for determining radioactive and natural cesium for marine studies," Reference Methods for Marine Radioactivity Studies, Intl. Atm. Energy Agency, Vienna (1970)129.
- [3] FOLSOM, T. R., YOUNG, D. R., JOHNSON, J. N., PILLAI, K. C., Manganese-54 and zinc-65 in coastal organisms of California, Nature 200 (1963) 327.
- [4] FOLSOM, T. R., YOUNG, D. R., EDWARDS, B. A., "Concentrations of fallout observed in selected marine organisms of the Pacific Ocean," Section VIII, Observations for 1964 Related to Fallout Traces in the Ocean and its Organisms, (FOLSOM, T. R., Ed.), Marine Environment Radioactivity Studies Group, Scripps Inst. of Oceanog., Univ. of Calif., La Jolla, Calif., (1965).
- [5] FOLSOM, T.R., YOUNG, D.R., Silver-110m and cobalt-60 in oceanic and coastal organisms, Nature 206 (1965) 803.
- [6] FOLSOM, T.R., GRISMORE, R., YOUNG, D.R., Long-lived gammaray emitting nuclide silver-108m found in Pacific marine organisms and used for dating, Nature 227 (1970) 941.
- [7] GRISMORE, R., FOLSOM, T.R., YOUNG, D.R., Silver-108m dating in marine organisms, Nature 234 5828 (1971) 347.
- [8] GRISMORE, R., FOLSOM, T.R., HODGE, V.F., YOUNG, D.R., A study of the radio-silver signature of the 1961-62 nuclear weapons testing period, Trans. New York Acad. Sci. (in press 1972).
- [9] FITCH, J.E., Common Marine Bivalves of California, Fish Bull. No. 90, State of Calif., Dept. Fish & Game (1953).
 [10] YOUNG, D.R., FOLSOM, T.R., Loss of zinc-65 from the California
- sea-mussel Mytilus californianus, Bio. Bull. 133 (1967) 438.

- [11] FOLSOM, T.R., YOUNG, D.R., FINNIN, L.E., "Sum-coincidence gamma-ray spectrometry in tracing cobalt-60 and silver-110m in marine organisms", Radioisotope Sample Measurement Techniques in Medicine and Biology, Intl. Atm. Energy Agency, Vienna (1965) 57.
- [12] FOLSOM, T.R., MOHANRAO, G.J., PILLAI, K.C., SREEKUMARAN, C., "Distribution of cesium-137 in the Pacific", USAEC Rep. HASL-197, New York (1968) I-95.

感觉 1 成

台

. 9

ė,

- [13] SVERDRUP, H.U., JOHNSON, M.W., FLEMING, R.H., The Oceans, Prentice-Hall, New Jersey (1942).
- [14] JONES, J.H., General Circulation and Water Characteristics in the Southern California Bight, So. Calif. Coastal Water Res. Proj. Tech. Rep., Los Angeles (1971).
- [15] WATSON, D. G., DAVIS, J. J., HANSON, W. C., Zinc-65 in marine organisms along the Oregon and Washington coasts, Science 133 (1961) 1826.
- [16] OSTERBERG, C., PATTULLO, J., PEARCY, W., Zinc-65 in euphausiids as related to Columbia River water off the Oregon coast, Limnol. & Oceanog. 9 (1964) 249.
- [17] PERKINS, R.W., NELSON, J.L., HAUSHILD, W.L., Behavior and transport of radionuclides in the Columbia River between Hanford and Vancouver, Washington, Limnol. & Oceanog. 11 (1966) 235.
- [18] SVERDRUP, H.U., FLEMING, R.H., The waters off the coast of southern California, Bull. Scripps Inst. Oceanog. <u>4</u> (1941) 261.
- [19] WOOSTER, W. S., JONES, J. H., California undercurrent off northern Baja California, J. Mar. Res. 28 (1970) 235.
- [20] EMERY, K. O., The Sea Off Southern California, Wiley, New York (1960).
- [21] REID, J. L., Jr., Physical Oceanography of the Region near Point Arguello, Univ. of Calif., IMR 65-19 (1965).
- [22] OSTERBERG, C., SMALL, L., HUBBARD, L., Radioactivity in large marine plankton as a function of surface area, Nature <u>197</u> 4870 (1963) 883.
- [23] PEARCY, W.G., OSTERBERG, C.L., Vertical distribution of radionuclides as measured in oceanic animals, Nature 204 4957 (1964) 440.
- [24] PEARCY, W.G., OSTERBERG, C.L., Depth, diel, seasonal, and geographic variations in zinc-65 of midwater animals off Oregon, Int. J. Oceanol. & Limnol. 1 2 (1967) 103.
- [25] OSTERBERG, C., CUTSHALL, N., CRONIN, J., Chromium-51 as a radioactive tracer of Columbia River water at sea, Science 150 (1965) 1585.
- [26] WATSON, D.G., DAVIS, J.J., HANSON, W.C., Interspecies differences in accumulation of gamma emitters by marine organisms near the Columbia River mouth, Limnol. & Oceanog. 8 2 (1963) 305.
- [27] BRYAN, G. W., Concentrations of zinc and copper in the tissues of decapod crustaceans, J. mar. biol. Ass. U.K. 48 (1968) 303.
- [28] FOWLER, S. W., SMALL, L. F., DEAN, J. M., Experimental studies on elimination of zinc-65, cesium-137, and cerium-144 by euphausiids, Marine Biology 8 (1971) 224.
- [29] KUENZLER, E.J., "Elimination of iodine, cobalt, iron, and zinc by marine zooplankton," Symposium on Radioecology, Second National Symposium, Ann Arbor, Michigan, CONF-670503 (1967) 462.
- [30] SHIMIZU, M., KAJIHARA, T., HIYAMA, Y., Uptake of ⁶⁰Co by marine animals, Rec. Oceanog. Works Jap. <u>10</u> 2 (1970) 137.
- [31] BRYAN, G.W., WARD, E., The absorption and loss of radioactive and non-radioactive manganese by the lobster, Homarus Vulgaris, J. mar. biol. Ass. U.K. 45 (1965) 65.
- [32] GOLDBERG, E.D., "Biogeochemistry of trace metals," Chapt. 12, Treatise on Marine Ecology and Paleoecology, (HEDGPETH, J.W., Ed.), Geol. Soc. America Memoir 67 1 (1957).
- [33] BOWEN, H. J. M., Trace Elements in Biochemistry, Academic Press, London (1966).