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Radionuclides

Artificial Radionuclides in Marine Organisms in the Northeast Pacific Ocean off Oregon, U. S. A.

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INTRODUCTION

Marine organisms modify the distribution of radioisotopes in the oceanic ecosystem. Various vital processes of plants and animals concentrate elements. The distributions of the biota are often independent of their watery environment; they may actively migrate vertically or horizontally or may remain in place while the water moves past them. In the case of plankton, the concentrating organisms passively move with the currents.

The Pacific Ocean off Oregon is ideally suited for research on the biological fate of artificial radionuclides. A constant supply (steady-state) of low level radionuclides is transported into the ocean environment from the Columbia River, a point source. These radionuclides are derived primarily from the atomic works at Hanford, Washington, where the waters of the Columbia River are used to cool the reactors. Radioactive elements are induced from the trace elements in the river water as it passes through the high neutron flux within the reactors [1] . The cooling water is returned to the river , and many of the radioisotopes ultimately reach the sea. Approximately 900 curies per day were discharged into the Pacific Ocean during the period 1961-1963 [2] [3] [4].

The Columbia River enters the northeast Pacific Ocean at the boundary between Oregon and Washington. Its discharge of fresh water into the ocean, the Columbia River plume, varies in position from season to season [5] [6] . Because the direction of wind stress changes from northerly in the winter to southerly in the summer, the plume is found near shore to the north in the winter and off Oregon to the southwest in summer. In addition, when coastal surface waters are blown offshore by the summer winds, deeper water upwells in its place forming a band of upwelled water between the plume and the coast of Oregon.

Radionuclides from atomic testing and the resulting worldwide fallout of fission products have also been present at low levels off the northwest coast of the United States during the period of study.

Induced radionuclides from the Hanford reactors and fission products from fallout have been detected in a variety of animals in the northeast Pacific Ocean as far as 490 km off the coast of central Oregon and to depths of 2860 m. Zinc-65 is biologically the most important induced radionuclide entering the ocean from the Columbia River. It occurs in phytoplankton, zooplankton, and nekton in the upper layers of the water column [7] [8] [9] [10] [11]. Zinc-65 and other man-made radioelements have also been detected in the sediments [12] and in bottom invertebrates [13] [14].

Marine animals must be efficient feeders to survive. Filter-feeding organisms must process large volumes of water to obtain enough energy from living and non-living organic particulate material for maintenance, growth, and reproduction. Benthonic deposit-feeding invertebrates must pass large quantities of sediment through their guts to provide enough organic material to support life processes. Carnivores feed on both the above animal groups and ingest protoplasm that has already incorporated elements collected from large volumes of seawater or sediment. Organisms, then, are mechanical concentrators and integrators of elements, or their radioisotopes, used in life processes. The radioactively-tagged elements present in the sea off Oregon and Washington make this area ideal for a study of the transfer of elements through the food web.

MATERIALS AND METHODS

Numerous stations off Oregon and Washington were sampled during a four-year period to provide planktonic, nektonic, and benthonic organisms from a range of depths and environments for radioanalysis. The large zooplankton and nekton were collected by an Isaacs-Kidd Midwater Trawl, the benthonic organisms by a 7-m semi-balloon shrimp trawl, and sediment by a 0.1-m^2 Smith-McIntyre bottom grab. Phytoplankton and detritus were collected by passing seawater through a membrane filter.

Animal samples were preserved in 10% neutral seawater-formalin or deep-frozen on board ship. The samples were usually sorted to species in the laboratory. They were then counted and dissected into major tissue levels or organ systems, when possible. The samples were prepared for radioanalysis by drying to a constant weight at 65° C in a drying oven, ashing at approximately 550° C in a muffle furnace, grinding carefully with a mortar and pestle, packing into 15-cc plastic tubes, and sealing the tubes with a cork and paraffin wax. When the number and volume of a species were low, whole organisms were packed in the tubes. Total samples of mixed species were sometimes counted in a polyethylene bottle on top of the crystal. Gamma-ray emissions were counted with a 512 channel Nuclear Data ND-130 Gamma-ray Spectrometer for 100 or 400 minutes.

To measure artificial radioactivity in the sedimentary environment of the benthos, we have taken two approaches. One method used a bottom grab with hinged top flaps to obtain the top 1 cm of sediment for radioanalysis in the laboratory. The sediment was dried, and a standard volume in a 15-cm plastic petri dish was counted on top of the crystal. The other method was to use an *in situ* gamma-ray probe in shallow water [15].

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RESULTS

The amount of radionuclides from fallout and Columbia River sources varied in both time and space off the coast of Oregon. Amounts varied among species and among animal feeding types. Variations were noted in both the pelagic and benthonic organisms.

Geographic Variations

Induced radionuclides flowing into the sea from the Columbia River have been found to vary geographically both in pelagic and bottom animals. Highest levels of zinc-65 in fauna were generally found near the mouth of the Columbia [7][14][16].

The picture of ^{65}Zn concentrations in plankton and nekton in relation to geographic variations is a complex one, probably affected by advection with currents and by lateral and vertical motion of the organisms. Osterberg, et al. [7] found the highest levels of radiozinc in the euphausiid Euphausia pacifica, near the mouth of the Columbia River in the spring, but further south off Newport in the summer after the plume had extended to the south. The latter animals probably had been in the plume waters for longer periods of time and had accumulated higher amounts of radionuclides than euphausiids found near the river's mouth. Throughout the year off Oregon, radiozinc concentrations in E. pacifica remained higher than in the same species beyond the influence of the Columbia River. Samples of this euphausiid from Alaska and California waters were much lower in zinc activity [7].

The amount of zinc-65 in bottom invertebrate organisms at the same depth generally decreases with increasing distance from the mouth of the Columbia River (Table 1). The decrease in radioisotope concentrations with distance from the river is not surprising. The animals involved generally remain in one spot with the river plume flowing overhead in the surface waters. Those closer to the river would receive larger amounts of radioisotopes for longer periods of time. This differs from the plankton and nekton which are at least partially transported in the plume itself.

Increasing distances offshore to the west produce a rapid reduction in radiozinc concentrations in bottom animals. Bathymetric effects greatly reinforce the general decrease with distance.

In contrast to the localized geographic distribution of Columbia River radioisotopes, fission products from worldwide fallout from atomic testing were fairly broadly distributed. Geographic differences usually were not apparent in ecologically similar animals off Oregon [13] [17].

Bathymetric Variations

The distribution and concentration of the induced radionuclides in the plume vary markedly with depth. Generally, deep-living mesopelagic fishes have lower concentrations of zinc-65 than shallow-living species (Figure 1). However, Pearcy [18] and Pearcy and Osterberg [9] have shown that the pattern of zinc-65 concentrations in animals collected with the midwater trawl is a complex one, varying both daily and seasonally. This changing pattern is related to the diurnal vertical migrations of animals and to the shifting position of the Columbia River plume.

Radiozinc in pelagic animals is affected by seasonal variation in the upper 150 m. Highest values of 2 pCi (wet preserved weight) were found

Table I

Zinc-65 in *Allocentrotus fragilis* (sea urchin)

Distance from Columbia River (km)	Depth (m)	Date	Zinc-65 pCi/g ash-free dry wt) (with standard deviation)		
			body	gonad	test
59	200	8/28/65	89.8 ± 7.5	75.0 ± 3.3	0.0
90	200	8/28/65	119.2 ± 6.8	64.3 ± 2.1	0.0
122	200	8/28/65	48.3 ± 3.4	38.0 ± 1.6*	0.0

*counting error

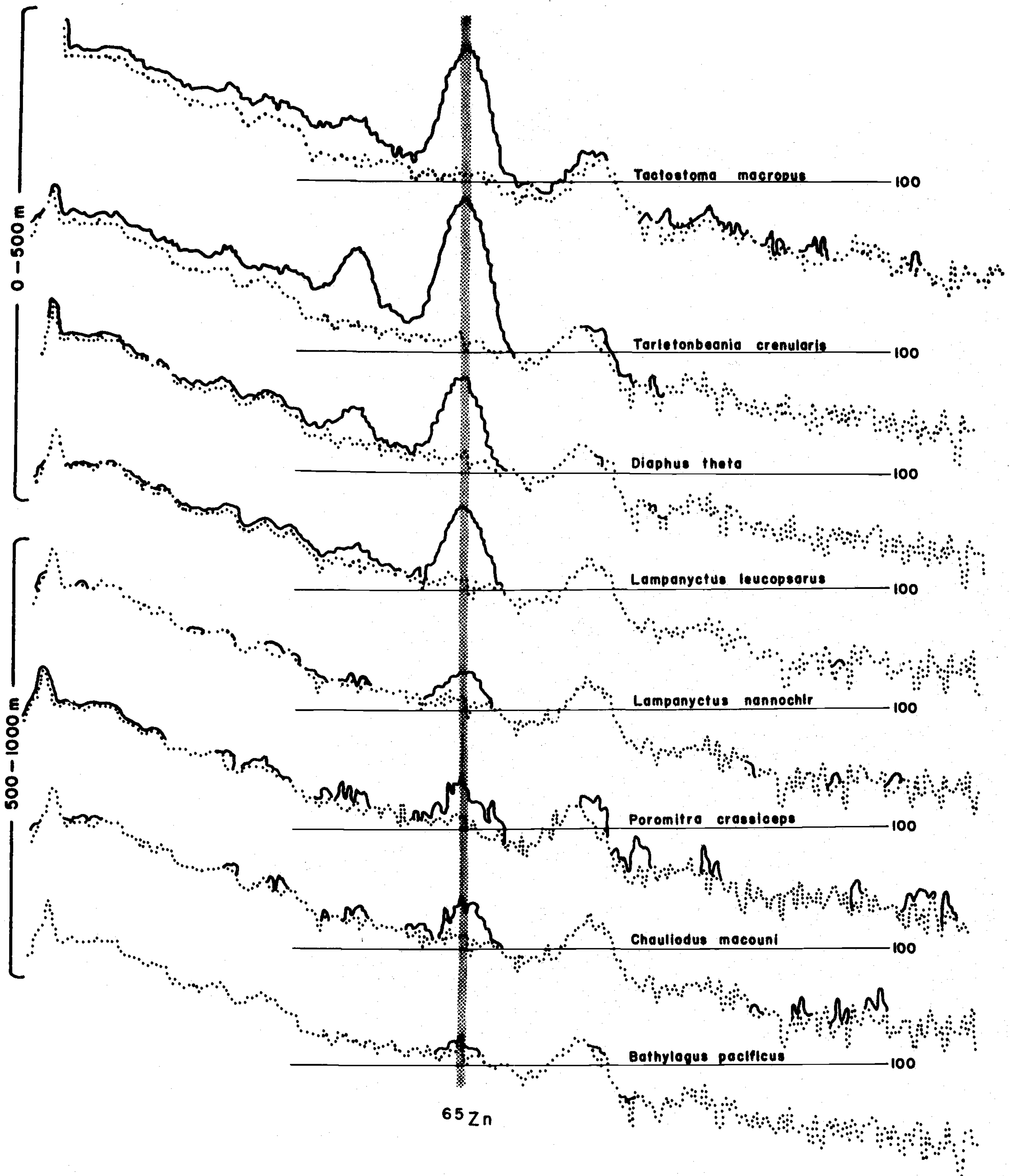

Fig. 1 Gamma spectra of mesopelagic fishes. The upper four species are shallow species compared to the lower four species. The ordinate indicates the total number of counts on a log scale.

during the summer, and lowest values of 0.2 pCi were found in mid-winter in the upper water layers. In depths of 150-500 m, the same seasonal pattern was evident, but damped; below 500 m, the activity was generally less than 0.2 pCi/g with little or no seasonal change. Hence, in the winter, ^{65}Zn concentration in the animals was fairly uniform within the entire 1000 m water column, while in the summer, significantly higher amounts of ^{65}Zn were found in organisms in the upper 150 m (Figure 2).

Vertical migrations affect the average concentration of ^{65}Zn in organisms, particularly in surface waters. The average ^{65}Zn activity was usually higher for animals collected during the night than for those collected during the day (Figure 2). This increase correlates with a diurnal change in species composition also evident at the surface. Consistent day-night differences in the amount of zinc-65 were not apparent at mid-depth or in deep water, however.

Concentrations of ^{65}Zn in the bottom fauna are related to bathymetry. Below about 600 m depth, the ^{65}Zn concentrations in all fauna drops off precipitously. Asteroids with the same feeding habits show this characteristic decrease of radiozinc concentrations in the animals with depth (Figure 3). The amount of ^{65}Zn decreases from 32.1 ± 1.3 pCi/g (ash-free dry weight) at 100 meters depth to about 3.6 ± 0.5 pCi/g at 800 m. Below 800 m ^{65}Zn generally remains at a low or undetectable level; however, it is sometimes found in fauna at the deepest (2860m) and most distant (266 km) station from the river's mouth.

In contrast to the bathymetric distribution of ^{65}Zn and other induced radionuclides originating from the reactors at Richland, Washington, fission products in fallout during the winter of 1961 were more concentratrated in the surface than in mesopelagic organisms [9]. Surface animals (0-150 m) contained nine times as much zirconium-95 - niobium-95 as the deeper forms.

However, ^{95}Zr-^{95}Nb were found in bottom animals in about the same amounts per unit weight in both shallow and deep water [13]. Yet, the bottom animals radioanalyzed are deposit-feeding, and the presence of the relatively short half-lived radioisotopes in the deep fauna suggested rapid sinking. The rate of sinking for very small fallout particles in the sea would be expected to be very slow from purely physical processes (Stoke's Law). It was suggested that filter-feeding zooplankton in the surface waters were compacting ingested phytoplankton along with adsorbed fission products into faecal pellets which, being much larger, fell to the bottom at a much faster rate than the minute, individual fallout particles. A comparison of the decline of fallout fission products in the air [19] and of fission products in the deep bottom fauna at 2860 m depth shows very

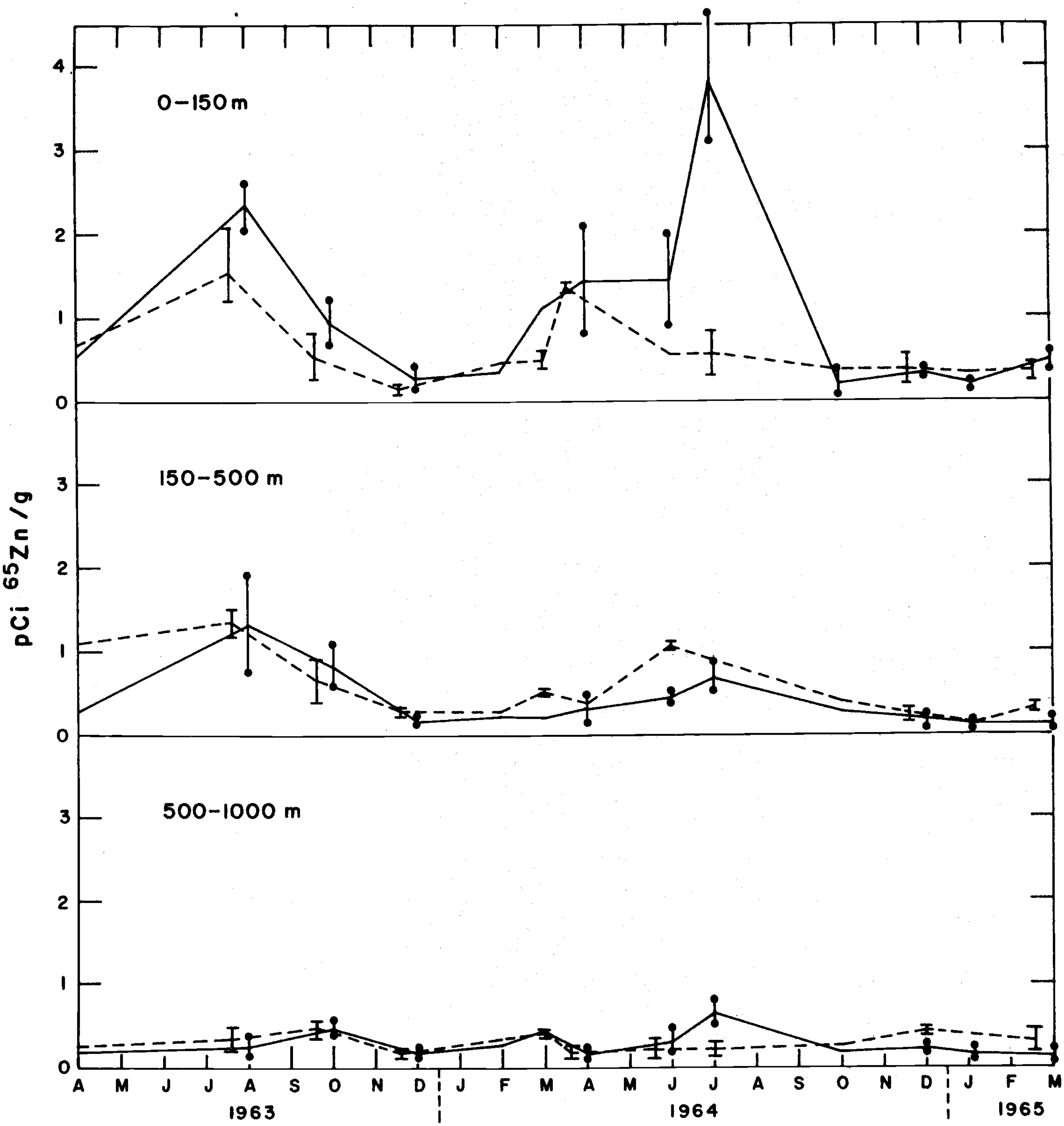

Fig. 2 Variations of zinc-65 (in picocuries/g, wet weight) in midwater trawl collections from the depths. The solid line connects averages for night collections, and the dotted line connects averages of daytime collections. Range of values is also indicated.

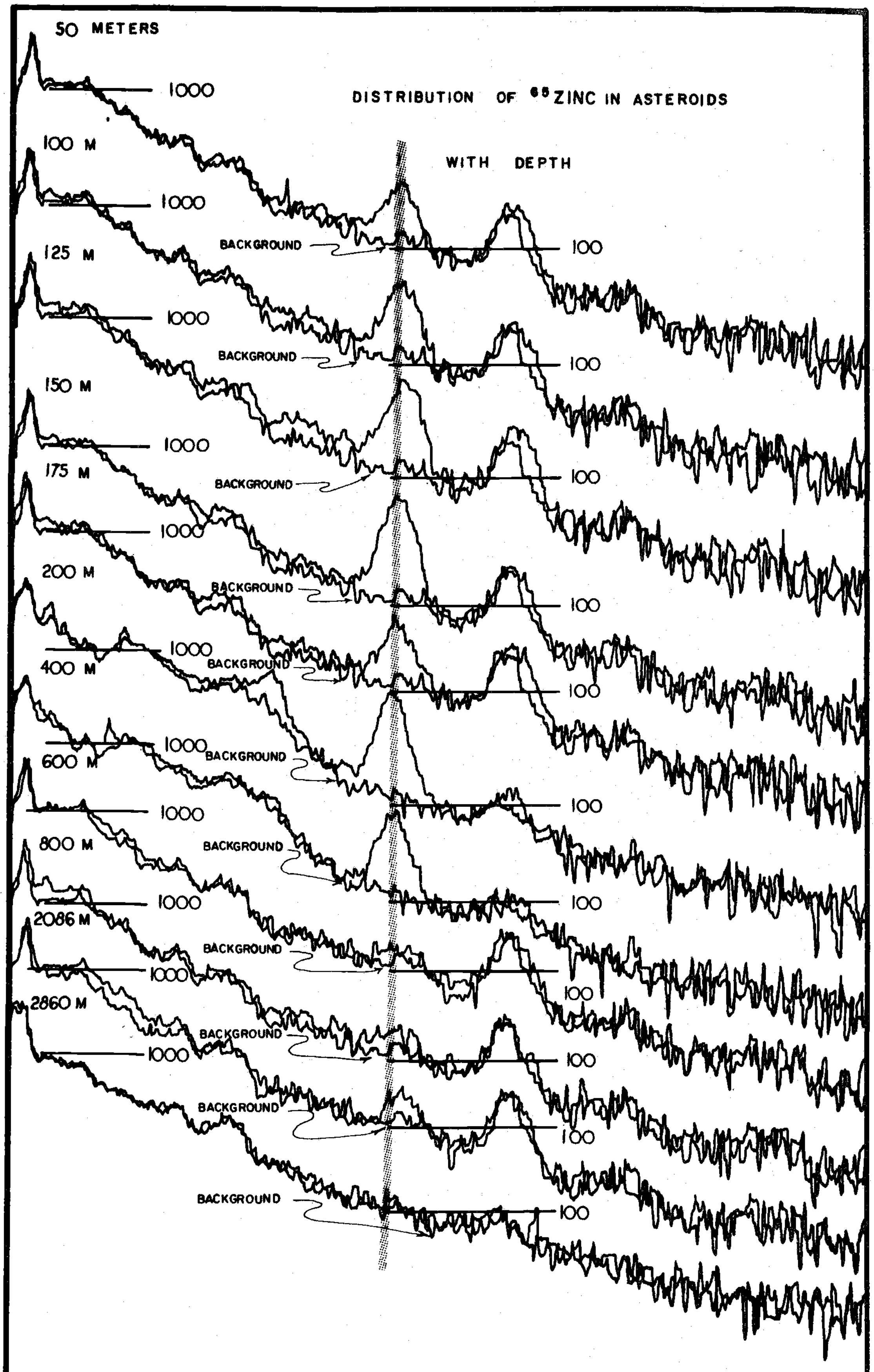

Fig. 3 Eight normalized gamma-ray spectra for starfish from various depths. Note the zinc-65 peak and its decrease with depth. All data from 50 to 800 meters from June 1963 - 1965. The sample from 2086 meters is from August 1963 and that from 2860 is from May 1963. The ordinate indicates the total number of counts in a log scale.

Table II

Seasonal changes in Zinc-65 in shrimp, *Pandalus jordani*

Date	Depth (m)	(Zinc-65 pCi/g ash-free dry wt. with standard deviation)
2/7/65	245	8.6 ± 0.5
6/4/65	175	44.3 ± 0.9
10/24/65	200	17.7 ± 0.7

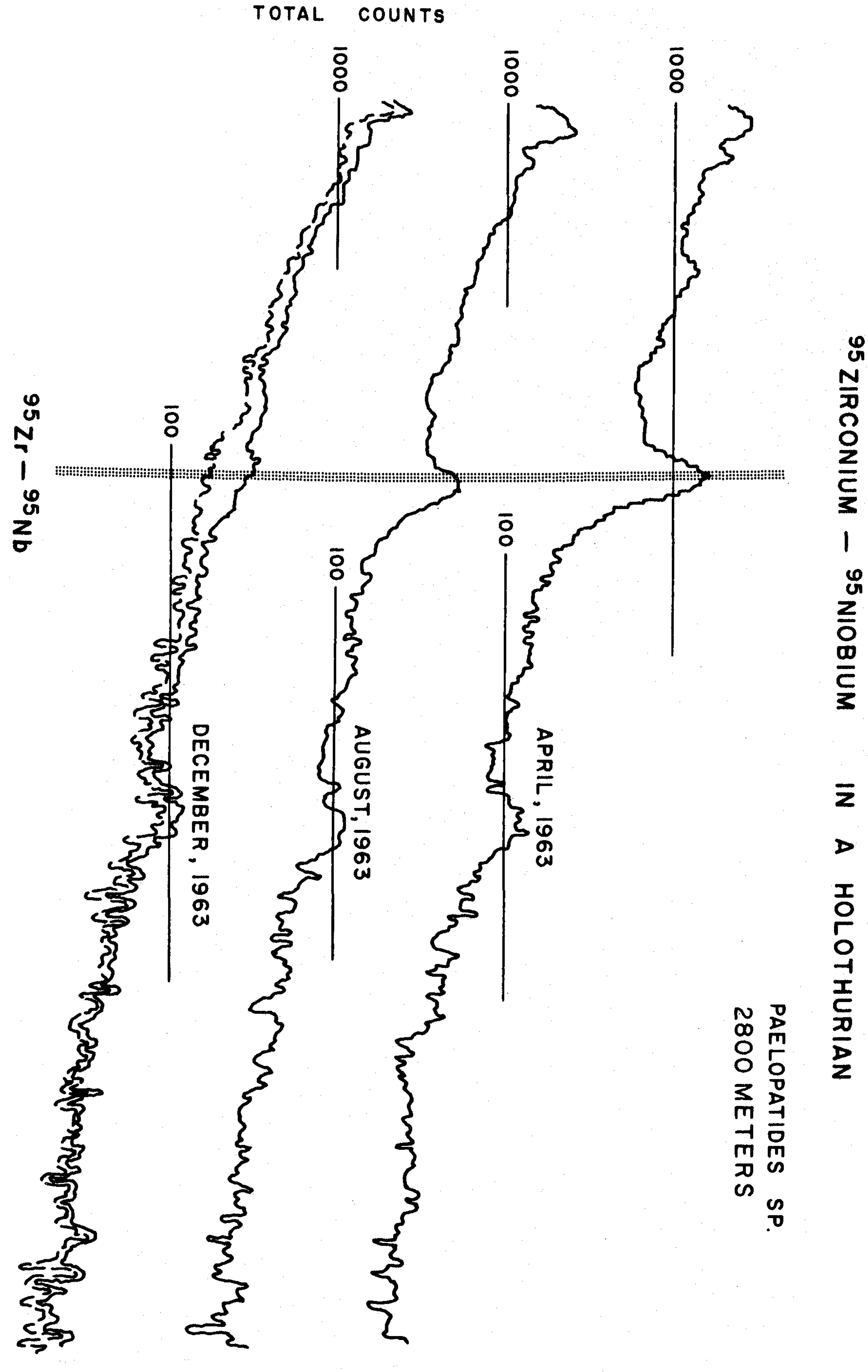

Fig. 4 Weight normalized gamma-ray spectra for deep sea cucumber, (Paelopatides sp.). Total number of counts are indicated on a log scale.

little lag. The rapid decrease of the fission products in the air during the latter part of the summer of 1963, after the cessation of atmospheric testing, was repeated a short time later in the bottom fauna at 2860 m depth, 106 km off the central Oregon coast (Figure 4).

Seasonal Variations

The amount of artificial radionuclides in the marine biota off Oregon is intimately linked to seasonal growth and reproductive changes of the organisms and to the geographic position of the Columbia River plume. Cycles in ^{65}Zn in the fauna are suspected to respond to both types of cyclical phenomena. To date, only the effect of the seasonal shift of the plume position has been demonstrated to be of major importance.

During the summer, surface animals from a single station off Oregon showed greater amounts of radiozinc than deep water fauna (Figure 2). The Columbia River plume usually includes this station during summer months. During the winter, on the other hand, when the plume is oriented to the north off the Washington coast, the station does not show the effects of the plume, and the amount of ^{65}Zn is about the same in both surface and deep animals.

Seasonal changes in the distribution patterns of induced radionuclides in bottom animals are similar to those in the pelagic fauna (Table II). Further work will be necessary to draw conclusions, however. The summer maximum of ^{65}Zn concentrations appears to come at a later time of year in animals from greater depths [14]. A seasonal increase in the total radiozinc in the gonad of sea urchins has been noted, though it has not been shown yet whether this increase is relative with respect to other organs or whether it reflects an increase in the total amount of ^{65}Zn in the organism.

Variations within the Food Web

Position of animals in the food chain affects both the induced radionuclide and fission radionuclide concentrations in the pelagic and bottom fauna [8] [14].

Osterberg, Pearcy, and Curl [8] demonstrated that ^{65}Zn increases up the food chain in the pelagic environment relative to the fission products, ^{95}Zr-^{95}Nb, but an absolute increase is not always apparent. Zinc-65 was found in every organism analyzed, but zirconium and niobium were concentrated mainly by primary producers and herbivores, and not by carnivores. Fission products from fallout were less evident in predaceous pelagic animals than in herbivores.

The same relationships between feeding habits and trophic level and the abundance of radionulides are evident in the bottom invertebrates, though the food webs are usually shorter. During the period when they were detectable, ^{95}Zr-^{95}Nb were found only in sediment-feeding organisms while the amount of ^{65}Zn generally increased in predatory organisms. The fission products were probably in the gut of the organisms. The simple food chain illustrated in figure 5 shows little or no radiozinc in the sedimentary environment of the ophiuroids (Ophiura sp.), but the efficient, sediment-feeding brittle stars have concentrated a significant amount of ^{65}Zn. This genus of ophiuroids is known for its deposit-feeding habit [20] [21]. Gut content analyses have shown that the top carnivore, Luidia foliolata, feeds predominately on this genus of brittle stars in this area (Carey, unpublished data). In this case the predaceous animal has less radiozinc per unit weight than the deposit-feeding forms, but our previous work has shown that among closely related animals, the predator has a higher concentration of ^{65}Zn than the deposit-feeder living on the organic material in the sediment [12].

Variations between the Organisms and their Environment

Off central Oregon we have found the amount of man-made radionuclides to be very low in water and sediment, the abiotic environment of the animals. The radionuclides may be too low to measure, but readily detected in organisms from the same place collected at the same time. Osterberg [22] has noted there is little similarity between the gamma-ray spectra of the benthic invertebrates and those of the sediments in, or on, which they are living.

DISCUSSION

The distribution in the fauna of fission products, e. g. ^{95}Zr-^{95}Nb and neutron induced radionuclides, e. g. ^{65}Zn, are basically different through time and space. Radiozinc varies both geographically and bathymetrically with time and is found in significant concentrations thoughout the trophic structure of the pelagic and bottom faunal assemblages. Fallout radionuclides, on the other hand, were found in greatly reduced amounts in carnivores, and were widely distributed geographically in all fauna and with depth in the benthos.

The contrasting distributions of ^{65}Zn and ^{95}Zr-^{95}Nb in space, time, and organisms can be explained by biological and physical means. Zinc, being a biologically important element [23] , is concentrated by the biota in surface waters, and radiozinc off Oregon and Washington is primarily associated with a point source, the surface-located Columbia River plume. The extensive seasonal position shift of the ^{65}Zn-rich plume waters explains

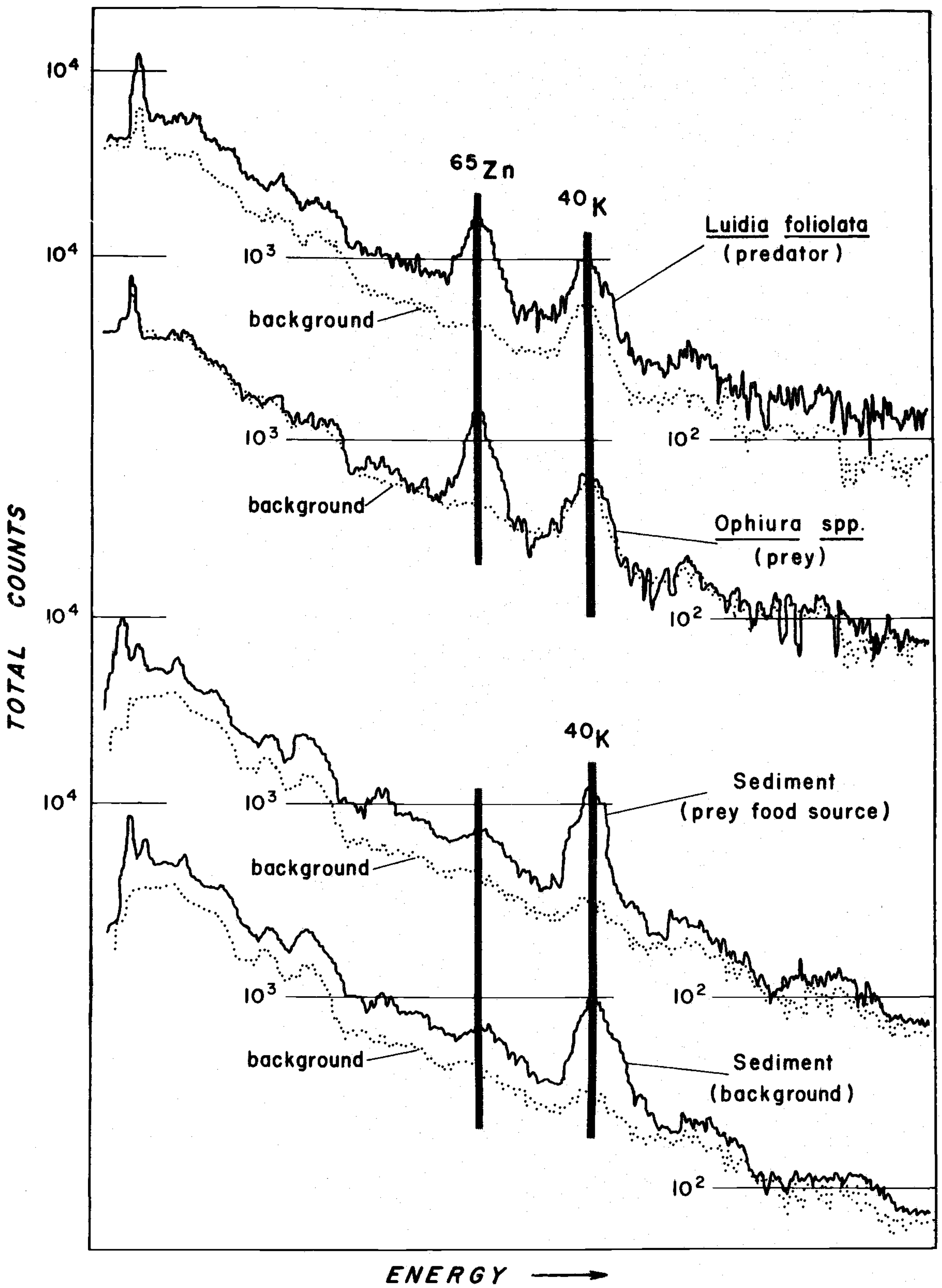

Fig. 5 A comparison of the gamma-ray spectra of components of a benthonic food chain. From top to bottom the spectra represent: (1) a predaceous asteroid, Luidia foliolata; (2) the major prey, an ophiuran, Ophiura spp.; (3) sediment from the environment of the two echinoderms; and (4) sediment from a similar environment beyond the influence of the Columbia River Plume. The top two spectra have been weight normalized. The bottom two spectra are not significantly different. The peak in the zinc region in the sediment spectra is probably due to Compton scatter.

many of the seasonal phenomena described. Furthermore, ^{65}Zn has a long biological half-life and is, therefore, retained for relatively long periods of time by the fauna. Zirconium, on the other hand, is not important to biological functions [24] and is not retained by the organisms for any significant period of time. Also, fallout is more or less uniform over the area that we have studied. Though ^{95}Zr-^{95}Nb concentrations in pelagic animals may be higher in the surface waters, they do seem to pass through the pelagic food chain fairly quickly and thus can be found in about the same concentrations in both shallow and deep benthonic invertebrates.

Radioecological studies in another marine environment with a continuously controlled radioactive discharge from a point source have been reported by Mauchline, Taylor, and Ritson [25] and Mauchline and Taylor [26]. The liquid effluent from the Windscale Works of the United Kingdom Atomic Energy Agency is discharged from a pipeline 2.5 km offshore. In contrast to the radionuclides from the Hanford reactors, those produced at Windscale are primarily fission products. The distribution and concentration of these radioactive elements have been studied in a population of the carnivorous thornback ray (Raia clavata) and beach organisms in the vicinity of the effluent. The guts and gut contents of the rays were generally much more radioactive than the internal organs and tissues. Fission radionuclides were not accumulated in the rays to any great extent. There appeared to be a correlation of radioisotope concentration in the intertidal environment with position in the food chain; animals in higher trophic levels had smaller concentrations of radionuclides. Although the physiology of different taxonomic groups may have more effect than feeding relationships [25] [27], these results are similar to those found for pelagic organisms off the Oregon coast.

Lowman [28] has pointed out that there are many biological factors affecting the uptake of radioisotopes in marine organisms. Feeding habits are just one. The degree to which an isotope is taken up by an animal also depends on the amount of stable isotope present, and on other biological factors.

Further research is necessary to understand fully the cycling of elements in the marine ecosystem in the northeast Pacific Ocean off Oregon. Measurement of stable elements in the organisms will hopefully give us a truer picture of the rates and routes of the cycling of these elements through the food web.

SUMMARY AND CONCLUSIONS

1- ^{65}Zn is found in all groups of animals in the pelagic and benthic environments, in higher concentrations in some animal groups than others. The highest levels measured, however, are still very much below hazard levels.

2- Highest concentrations of ^{65}Zn in the fauna usually were found near the Columbia River, a point source. However, pelagic organisms further to the south showed high concentrations during the summer.

3- Bathymetric, seasonal and diurnal effects on the distributions of radionuclides in the biota are interconnected, particularly in the macroplankton and nekton. ^{65}Zn stratified in the pelagic organisms in the summer, was uniformily distributed with depth in the winter. ^{65}Zn concentrations were higher in the macroplankton and nekton in surface waters at night. ^{65}Zn decreases with depth in the benthos.

4- ^{95}Zr-^{95}Nb, ubiquitous in geographic distribution, were found at all depths, though in the pelagic fauna they were in higher concentration in surface waters.

5- ^{65}Zn was found throughout the food chain, while ^{95}Zr-^{95}Nb decreased rapidly after the first two trophic levels.

6- Man-made radionuclides, in very low concentrations in water and sediments of the marine environment off central Oregon, are readily detected in the fauna. Organisms are important determiners for the distribution and concentration of radionuclides in the marine ecosystem.

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FIGURES

Fig. 1 Gamma spectra of mesopelagic fishes. The upper four species are shallow species compared to the lower four species. The ordinate indicates the total number of counts on a log scale.

Fig. 2 Variations of zinc-65 (in picocuries/g, wet weight) in midwater trawl collections from the depths. The solid line connects averages for night collections, and the dotted line connects averages of daytime collections. Range of values is also indicated.

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