Radioactivity and its Relationship to Oceanic Food Chains

Charles Osterberg, William G. Pearcy, and Herbert Curl, Jr.

Department of Oceanography
Oregon State University

ABSTRACT

Gamma-ray spectra of some primary producers (single-cell plants), filter-feeding herbivores, and carnivores, assigned to trophic levels I, II, and III–V, respectively, were prepared from marine samples taken in the Pacific off Oregon during 1961–1962. These organisms had been exposed in their natural environment to both fission products from fallout and neutron-induced radionuclides from reactors on the Columbia River. Comparisons of spectra of organisms from different trophic levels, determined from stomach contents and the literature, show that Zr^{95}-Nb^{95} and Ce^{141} were concentrated by primary producers and herbivores but not by carnivores. Cr^{51} was abundant in only filtered samples (primary producers). Mn^{54}, Co^{60}, and Cs^{137} were found in only herbivores and carnivores. Zn^{65} was found in every marine organism examined. We conclude that the abundance of Zr^{95}-Nb^{95} and Ce^{141}, in particular, may be useful in marine trophic-level studies. Peaks due to these fission products are greatly reduced in spectra of predaceous animals as compared with spectra of herbivores.

Introduction. The oceans receive a substantial share of radioactive fallout resulting from nuclear testing because of their large area and the drainage they receive from the continents. Prevailing westerly winds carry tropospheric fallout from nuclear tests in northeastern Asia across the Pacific Ocean to North America. Levels in the environment are normally quite low, but certain fission products are accumulated by filter-feeding zooplankton (Osterberg 1962b, 1963).

Radionuclides on the Pacific coast of America are also introduced by nuclear reactors at Hanford, Washington. Many trace elements in the Columbia River water used to cool the reactors are activated by the intense neutron flux (Nelson 1961). These induced radionuclides are returned to the river, and ultimately portions of them enter the ocean.

The presence of radioisotopes in the ocean off the Oregon coast and the supply of nekton and plankton available from our midwater-trawl program have made it possible to investigate the presence of both fallout and neutron-induced radionuclides in oceanic food chains. The gamma-ray spectra of marine organisms from different trophic levels are compared to determine which radionuclides are passed through food chains and which are discriminated against. Assignment of an organism to a particular trophic level is, in most cases, based on studies of stomach contents, supplemented with references from the literature.

Methods. Phytoplankton and detritus were collected by passing surface seawater through a 5″ membrane filter (0.65 μ) plus glass fiber prefilter (Gelman Instrument Company). The filters were ignited in an open crucible to destroy the membrane filter, and the residue was placed in a muffle furnace for an hour at 500°C and then ground with mortar and pestle before packing into counting tubes.

Macroplankton and micronekton were collected in a six-foot Isaacs-Kidd midwater trawl towed for 30 minutes from 200 m depth to the surface. Plankton and nekton samples were freeze-dried after preservation in formaldehyde. Large or oily samples were further concentrated by ashing in a muffle furnace. Dried and/or ashed concentrates of the entire animals,[1] including digestive tracts, were then packed into counting tubes (Falcon Plastics, Item # 2001). For analysis, tubes containing the prepared samples were placed in the well of the 5″×5″ NaI(TI) primary crystal of the Hanford, Washington, total-absorption anticoincidence spectrometer (Perkins et al. 1960). Counting time was 30 minutes, with 30-minute background subtracted.

Results.

Trophic Level Relationships. The first trophic level in the pelagic environment consists of single-cell plants. The second trophic level consists of filter-feeding herbivores. Carnivores compose the higher trophic levels. Most food relationships are not simple food chains but are more often complex webs. Since feeding animals are opportunists, the uncertainties of diet are great, particularly in the case of large predators. Nevertheless, food patterns do exist and some division into trophic levels is possible.

Trophic Level i. Phytoplankton and detritus were trapped on a membrane filter through which surface seawater was passed. Most animals were removed by prefiltration through a # 6 mesh net.

Trophic Level ii. *Euphausia pacifica.* Euphausiids may feed on phytoplankton, small crustaceans such as copepods, or detritus (Ponomareva 1954, MacDonald 1927, Marshall 1954). Although Ponomareva noted that *E. pacifica* occasionally fed on crustaceans, this euphausiid is primarily a filter-

[1] Salps and, of course, tuna liver were exceptions. Only the opaque interior "nucleus" (or digestive tract) of the salp was used, since the transparent outer portion was found to be low in radioactivity (Osterberg et al. 1963).

feeding herbivore. Setae of the thoracic legs of our adult *E. pacifica* are about 20–40 μ apart; thus, the filtering apparatus is equipped to collect most marine diatoms.

Calanus cristatus, *Salpa* spp., and *Clio pyramidata* are pelagic zooplankton that feed on suspended particles, principally phytoplankton. *Calanus* spp. are mainly herbivores (Marshall and Orr 1955), as are the cavolinid pteropods (Marshall 1954, Yonge 1926). *Salpa* spp. are indiscriminate feeders and principally phytoplankton grazers (Marshall 1954, Yount 1958, Foxton 1961).

TROPHIC LEVEL III. *Pasiphaea pacifica.* This carid prawn is particularly common near the mouth of the Columbia River. Stomachs of 31 individuals were examined. Contents consisted of fragments of animals that were apparently dismembered and masticated before ingestion. Chitinous remains of crustaceans were noted, including mandibles and eyes similar to those of *Euphausia pacifica.* Several cephalopod beaks were also found. These observations, like those on other oceanic prawns (Chace, 1940), indicate that adult *P. pacifica* are carnivores.

Sergestes similis. Only a few observations on the feeding habits of this mesopelagic prawn have been made previously (Barham 1956). Our examination of the stomachs of these common animals revealed that they are capable of ingesting whole zooplankton. Entire copepods were noted as well as fragments of larger euphausiids and prawns. Several fish scales were also present.

Lampanyctus leucopsarus. Thirty-four stomachs of this lanternfish, the dominant mesopelagic fish taken in midwater-trawl collections off Oregon, were examined. It feeds largely on euphausiids, calanoid copepods, and amphipods.

Tactostoma macropus. Stomachs of 52 specimens of this stomiatoid fish were examined. Many were empty. Euphausiids and sergestid prawns occurred most frequently, but about half the total combined stomach contents by volume was due to the presence of several lanternfish. *T. macropus* appears to be intermediate between trophic levels III and IV.

TROPHIC LEVEL III–V. *Thunnus alalunga.* Several hundred migrating albacore tuna were captured during the summer of 1962, from 25 to 50 miles off the northern Oregon coast, and stomachs of 62 of the tuna were examined. Most of the stomachs were empty or less than one-quarter full. Cephalopods composed about 75 % of the bulk of the stomach contents, fish about 18 %, and crustaceans about 5 %.

Radioanalyses. Gamma-ray spectra of organisms from several trophic levels taken at the same time and same location are shown in Fig. 1. Trophic level I is represented by a membrane filter through which surface seawater has been passed. Although chlorophyll *a* was present (1.56 mg/m^3), the data do not indicate the percentage of particulate organic matter. The low amount of potassium-40 suggests that only a small quantity of inorganic material

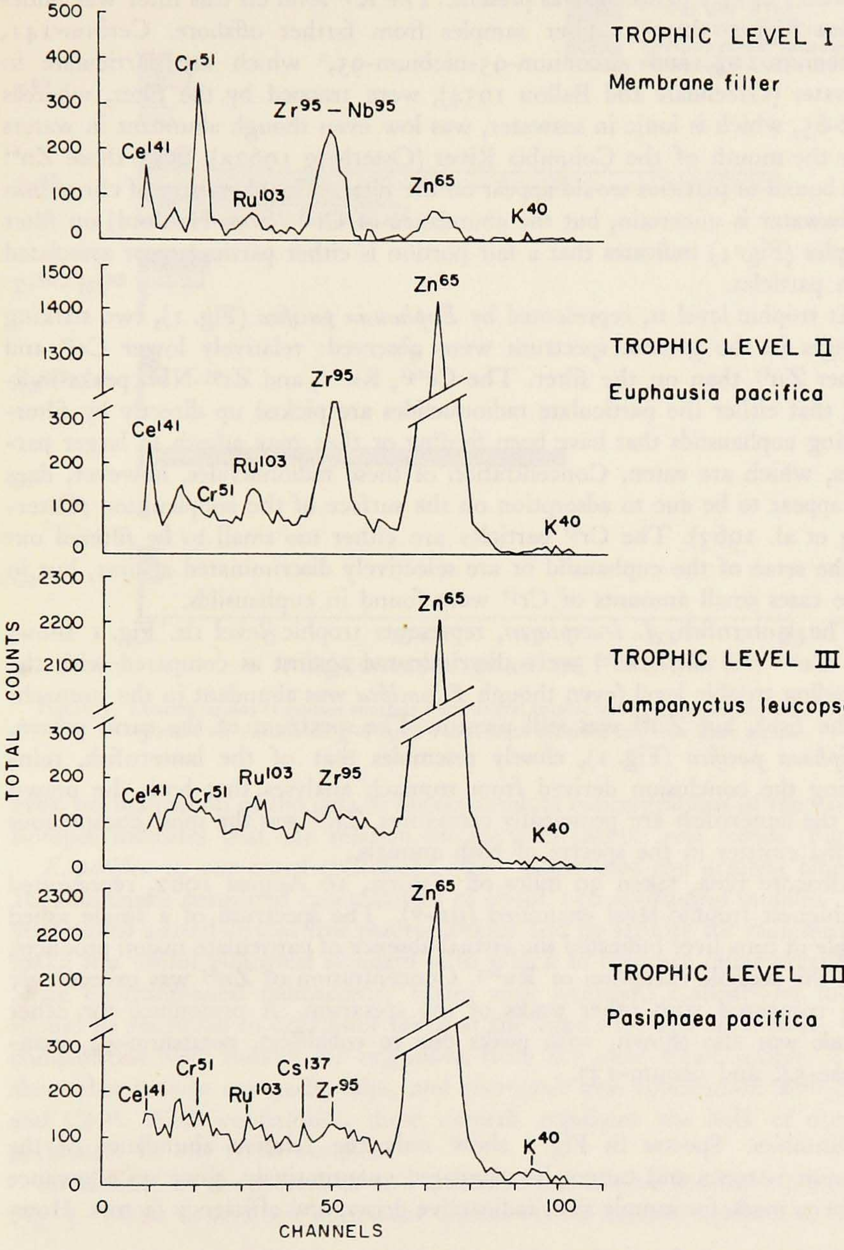

Figure 1. Comparison of gamma emitters from several trophic levels. Organisms for all four spectra were collected 15 miles off Astoria on 5–6 April 1962. All trophic-level-II-and-III animals were from the same trawl sample.

(probably as clay particles) was present. The K^{40} level on this filter was nonetheless higher than in filter samples from farther offshore. Cerium-141, ruthenium-103, and zirconium-95-niobium-95,[2] which are particulate in seawater (Greendale and Ballou 1954), were trapped by the filter, whereas zinc-65, which is ionic in seawater, was low even though abundant in waters near the mouth of the Columbia River (Osterberg 1962a). Only those Zn^{65} ions bound to particles would appear on the filter. The chemistry of chromium in seawater is uncertain, but the abundance of Cr^{51} (from Hanford) on filter samples (Fig. 1) indicates that a fair portion is either particulate or associated with particles.

At trophic level II, represented by *Euphausia pacifica* (Fig. 1), two striking changes in the gamma spectrum were observed: relatively lower Cr^{51} and higher Zn^{65} than on the filter. The Ce^{141}, Ru^{103}, and Zr^{95}-Nb^{95} peaks indicate that either the particulate radionuclides are picked up directly by filter-feeding euphausiids that have been feeding or they may adsorb to larger particles, which are eaten. Concentration of these radionuclides, however, does not appear to be due to adsorption on the surface of the zooplankton (Osterberg et al. 1963). The Cr^{51} particles are either too small to be filtered out by the setae of the euphausiid or are selectively discriminated against, but in some cases small amounts of Cr^{51} were found in euphausiids.

The lanternfish, *L. leucopsarus*, represents trophic level III. Fig. 1 shows that Ce^{141} and Zr^{95}-Nb^{95} were discriminated against as compared with the preceding trophic level (even though *E. pacifica* was abundant in the stomachs of the fish), but Zn^{65} was still present. The spectrum of the carid prawn, *Pasiphaea pacifica* (Fig. 1), closely resembles that of the lanternfish, reinforcing the conclusion derived from stomach analyses that both the prawn and the lanternfish are principally carnivores. Zn^{65} was the most conspicuous gamma emitter in the spectra of both animals.

Albacore tuna, taken 30 miles off Astoria, 10 August 1962, represented the highest trophic level examined (III–V). The spectrum of a single ashed sample of tuna liver indicated the virtual absence of particulate fission products, with the possible exception of Ru^{103}. Concentration of Zn^{65} was exceedingly high compared with other peaks of the spectrum. A preference for other cations was also shown, with peaks due to cobalt-60, potassium-40, manganese-54, and cesium-137.

Discussion. Spectra in Fig. 1 show only the relative abundance of the different isotopes and cannot be compared quantitatively, since no allowance has been made for sample size, radioactive decay, and efficiency factors. How-

[2] Our figures generally show the peak due to Zr^{95}-Nb^{95} simply as Zr^{95}, but the techniques used do not permit a separation of Zr^{95} from its daughter, Nb^{95}. There is also some uncertainty with regard to Ru^{103} and Ru^{106}, but our evidence indicates a preponderance of Ru^{103} in these samples. No attempt was made to differentiate between Ce^{141} and Ce^{144}.

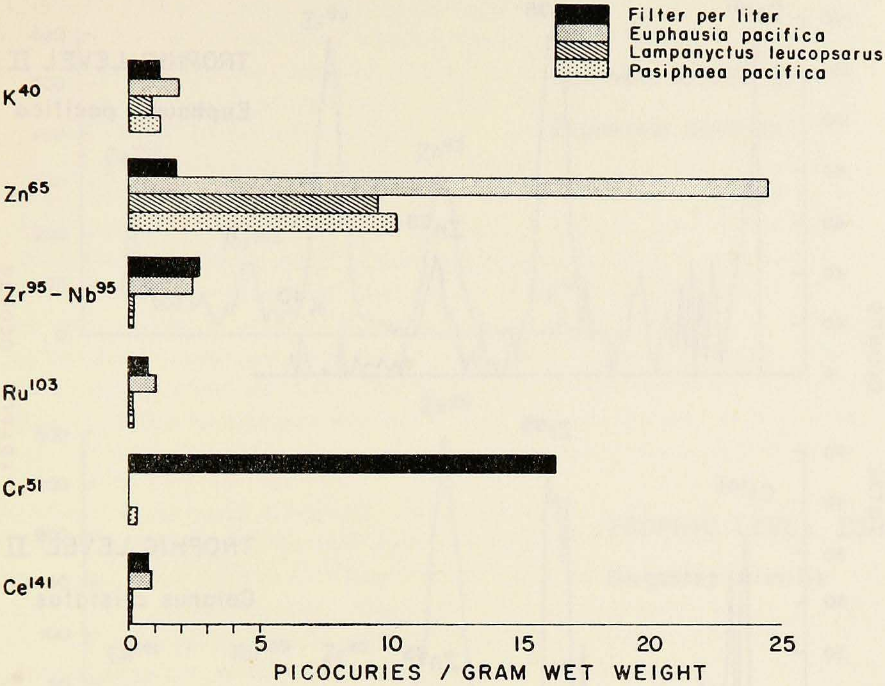

Figure 2. Concentrations of gamma emitters from several trophic levels. All organisms were collected 15 miles off Astoria on 5–6 April 1962. Note different units for filter sample.

ever, normalization of the data to obtain absolute concentrations of the various isotopes indicates that the relative spectra demonstrate real trends (Fig. 2).

E. pacifica in our trawl collections often contributed the greatest biomass. Its abundance permitted radioanalyses of about 150 euphausiid samples. The year-round availability of this macroplankter and its affinity for radionuclides make it a useful biological standard with which to compare other organisms. Since environmental radioactivity varies with location,[3] comparisons ideally should be restricted to organisms taken at the same time and same place. Our comparisons were usually for organisms from the same trawl sample. They show that certain copepods, salps, and pteropods also concentrate Zr^{95}-Nb^{95} and Ce^{141}. With euphausiids, these animals represent the bulk of oceanic herbivores in our trawl samples.

Similarities in the spectra of two animals from trophic level II are seen in Fig. 3. These spectra are somewhat typical of our oceanic herbivores. That

3 Short-period variations in fission-product levels in euphausiids from a single location are small. Nine consecutive tows, made over a period of eight hours, 50 miles off Newport, 11–12 April 1962, show the following averages and standard deviations: Zr^{95}-Nb^{95}, 13.6 ± 1.2 picocuries/g, and Ce^{141}, 17.5 ± 2.8 pc/g, dry weight.

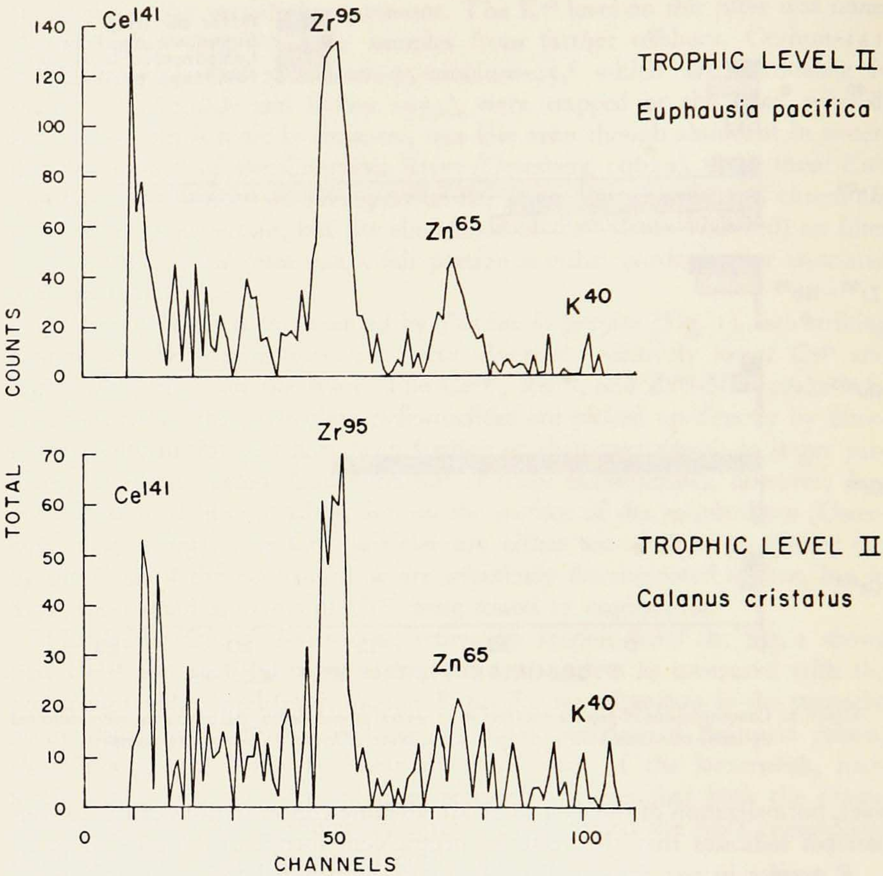

Figure 3. Comparison of spectra of euphausiids and copepods from the same sample, collected 105 miles off Astoria on 6 June 1962. The scatter in the lower spectrum is due to small sample size.

is, prominent peaks due to Zr^{95}-Nb^{95} and Ce^{141} appear in the spectra of salp, copepod, pteropod, and euphausiid samples taken in late 1961 and throughout 1962.

When spectra of organisms from higher trophic levels are compared with the spectrum of *E. pacifica*, the details described for Fig. 1 form a general pattern; i.e., in every case there was a reduction in the Zr^{95}-Nb^{95} and Ce^{141} peaks in predators relative to those observed in euphausiids. This discrimination was noted in the dozen or so instances when direct comparison of spectra from the same sample was possible, and it was reinforced by a large number of analyses of high trophic-level marine organisms that invariably were low in fallout peaks. This experience prompted us to consider *Sergestes similis* as a

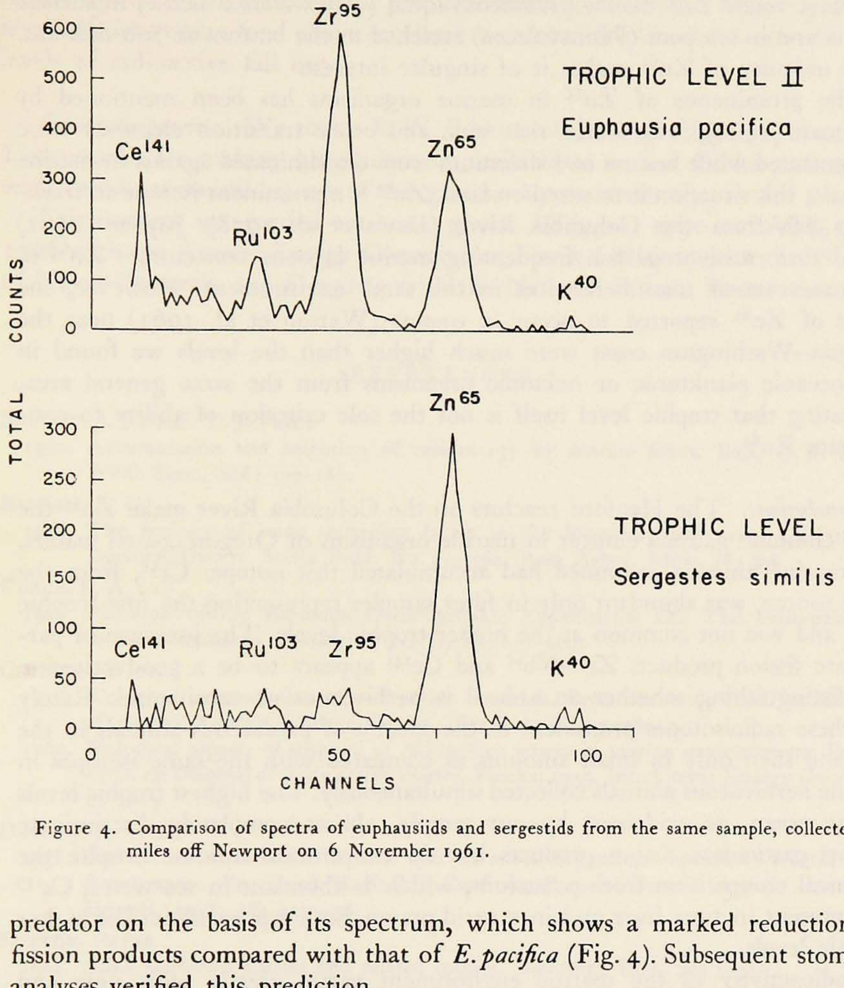

Figure 4. Comparison of spectra of euphausiids and sergestids from the same sample, collected 25 miles off Newport on 6 November 1961.

predator on the basis of its spectrum, which shows a marked reduction in fission products compared with that of *E. pacifica* (Fig. 4). Subsequent stomach analyses verified this prediction.

Ruthenium-103, presumably an anion (Lowman 1960), was the only obvious noncationic species regularly found in trophic levels III–V. It was present in the tuna liver and has been reported (Kaye and Dunaway 1962) to occur in the liver of a cotton rat, *Sigmodon hispidus*, but Chipman (1960) observed little uptake of Ru^{106} in digestive tracts of menhaden (*Brevoortia tyrannus*). Cesium-137, which we also found in tuna liver, has been observed to concentrate slowly in some marine fish (Chipman 1958, Baptist and Price 1962). Pendleton and Hanson (1958) have shown that Cs^{137} is particularly concentrated at higher trophic levels.

The affinity for zinc by all trophic levels, with the possible exception of trophic level I, makes Zn^{65} by itself a poor indicator of feeding relationships.

We have found Zn^{65} in the predaceous squid (*Onychoteuthis banksi*) in surface waters and in sea pens (Pennatulacea) attached to the bottom at 700 fathoms. This ubiquity of Zn^{65} makes it of singular interest.

The prominence of Zn^{65} in marine organisms has been mentioned by Lowman (1960), who stated that zinc and other transition elements were concentrated while cesium and strontium were discriminated against in marine animals; this situation is reversed on land. Zn^{65} is a prominent isotope in fresh-water fish from the Columbia River (Davis et al. 1958). Joyner (1962) found that carnivorous fish frequenting marine lagoons concentrate Zn^{65} to a greater extent than herbivores in the same environment. However, the levels of Zn^{65} reported to occur in oysters (Watson et al. 1961) near the Oregon–Washington coast were much higher than the levels we found in any oceanic planktonic or nektonic organisms from the same general area, indicating that trophic level itself is not the sole criterion of ability to concentrate Zn^{65}.

Conclusions. The Hanford reactors on the Columbia River make Zn^{65} the most common gamma emitter in marine organisms of Oregon coastal waters. All marine animals examined had accumulated this isotope. Cr^{51}, from the same source, was abundant only in filter samples representing the first trophic level and was not common at the higher trophic levels. The presence of particulate fission products Zr^{95}-Nb^{95} and Ce^{141} appears to be a good criterion for distinguishing whether an animal is herbivorous or carnivorous. Rarely are these radioisotopes prominent in the spectra of predaceous animals in the sea, and then only in small amounts as compared with the same isotopes in marine herbivorous animals collected simultaneously. The highest trophic levels in the ocean, as evidenced by our sample, almost completely discriminate against particulate fission products but do concentrate cations. Despite the chemical competition from potassium, which is abundant in sea water, Cs^{137} was present in tuna liver and in a carid prawn, both representatives of higher trophic levels.

Radioactivity in the marine environment varies greatly with time and location. Very likely changes in stable trace elements also occur, although comparable data do not exist. Future work on marine food chains should include both measurements, so that specific activities can be determined. However, these local differences are minimized in our data by intercomparison of organisms from the same trawl sample.

We conclude that particulate radioactive fallout is concentrated at the second trophic level by filter-feeding plankton but that very little of this radioactivity is present in the animals commonly utilized in the diet of man. On the other hand, neutron-induced Zn^{65} is more likely to enter the human food chain, but it is one of the more innocuous radioisotopes since it decays principally by electron capture and emits few ionizing particles. The discrimination against particulate fission products tends to make the higher trophic levels of the marine food chain excellent sources of food in the event of high levels of radioactive fall out.

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