RADIOANALYSIS OF OCEANIC ORGANISMS IN THE PACIFIC OCEAN OFF OREGON

by

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Progress Report

1 March 1963 through 29 February 1964

Submitted to the

U.S. Atomic Energy Commission

Contract AT(45-1)1750

Approved:

Wayne V. Burt, Chairman
Department of Oceanography

Reference 64-2
March 1964

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I INTRODUCTION

This report covers the first full year of operation of our gamma-ray spectrometry facility. A part of the contract period was devoted to calibration of the instrument, and therefore, a brief report on this procedure is included.

Demands on the spectrometer grew rapidly during the year and soon exceeded the capacity of the single instrument. Some projects were therefore restricted.

Priorities were established on the following basis:

(1) Atomic Energy Commission research (Osterberg, Carey, Pearcy) with samples probably containing Cr^{51}, because of the short half-life.

(2) Other AEC research.

(3) Thesis research.

The instrument was programmed 24 hours per day five days a week on AEC research but was generally free on weekends for student use for thesis research.

Instrumentation

An ND-130AT 512 channel spectrometer was used in conjunction with shielded 5x5-inch NaI(Tl) Harshaw crystal, with 3/4x3-inch well. Readout was by IBM typewriter, Tally tape, Tektronix oscilloscope and Houston X-Y recorder. Voltage to the photomultiplier was furnished by a John Fluke Model 412 Power Supply. Readout tapes were folded, placed into special envelopes (Dresser Products, Inc.) along with IBM readout and X-Y graph, and filed for reference.

Each Monday morning a new 100-minute background was prepared using 12 cc of distilled water. A count of Cs^{137} and Co^{60} sources was made, and charted to check for changes in sensitivity. Once a month, linearity tests were made, and backgrounds compared for subtle contaminations. In practice, calibration was checked before each new sample was counted. Installation of a refrigeration unit minimized drift.

Data reduction has been by spectrum stripping for the "sum" peaks of scandium-46 and cobalt-60 when present, with successive stripping by IBM 1620 computer for K^{40}, Zn^{65}, Zr^{95}-Nb^{95}, Ru^{103},

Cr^{51}, and Ce^{141}. This simple program is antiquated and will be replaced shortly by a "least squares" program. The latter has proven quite successful in other laboratories. Impetus for our change-over is mostly based on the disappearance of Zr^{95}-Nb^{95}, Ru^{103}, and Ce^{141} from our spectra, and the appearance of Cs^{137}, Ru^{106}, Ce^{144} and Mn^{54}. This requires a change in computer program, which will be made in March when the laboratory is dismantled and moved to the new Oceanography Building. This move will require recalibration, and the computer program will be developed for the reconstructed lead pit in the new laboratory.

II CALIBRATION

Charles Osterberg and Norman Cutshall

The Nuclear Data ND130A pulse height analyzer and 5x5" well-type scintillation detector have been calibrated for 10 gamma emitters, and Compton correction factors have been determined for six of these.

Efficiency Factors

Solutions (12 ml) of Co^{60}, Sc^{46}, Ba^{140}-La^{140}, Zn^{95}, Zr^{95}-Nb^{95}, Ru^{103}, I^{131}, Cr^{51} and Ce^{141} were sealed in individual 15-cc counting tubes and analyzed 13 June 1963 at Hanford Laboratories. Absolute activities in disintegrations per minute were calculated for the standards using Hanford efficiency factors. A 12-cc solid standard source for K^{40} was prepared and its absolute activity determined. Standards were counted at Oregon State University on 17 June 1963 and limits of summation were arbitrarily selected so as to include most of the photopeak area. Oregon State University efficiency factors were determined as follows:

$$\text{Efficiency factor} = \frac{\text{disintegrations per minute at Hanford} \times e^{-\lambda t}}{\text{counts per minute in photopeak}}$$

where $e^{-\lambda t}$ is a correction factor for decay. Standard master punched tapes were prepared for each solution.

Compton Correction Factors

Correction factors for the Compton continuum were determined as follows:

A = counts in photopeak (above background)
B = counts in area to be corrected (above background)
Compton correction factor = B/A

For example, the Zn^{65} standard spectrum contains approximately 1 count between the limits of summation for Ru^{103} for every 5 counts in the Zn^{65} photopeak. Thus, the correction factor for Zn^{65} in the Ru^{103} region is about 0.2 (i.e., 0.203; see Table 1).

A Fortran program was written for the IBM 1620 that would reduce raw data to absolute activity for K^{40}, Zn^{65}, Zr^{95}-Nb^{95}, Ru^{103}, Cr^{51}, and Ce^{141}, these six radionuclides being most common in our samples. All sample spectra are carefully examined before reduction to determine the presence of interfering gamma emitters. If interferences are present, as for example, Sc^{46} in Columbia River filter samples, these are measured and their effect removed by spectrum stripping before IBM reduction is completed.

Table 1. Table of Efficiency Factors and Compton Correction Factors Oregon State University 5x5-NaI(Tl) Well Crystal

Radionuclide:	K^{40}	Zn^{65}	Zr^{95}-Nb^{95}	Ru^{103}	Cr^{51}	Ce^{141}
Limits of summation (channels)	136-154	105-116	69-81	45-54	29-35	12-15
Efficiency factor	44.1	6.57	2.28	4.02	14.5	2.37
Compton correction factor:						
K^{40}		0.116	0.182	0.184	0.130	0.150
Zn^{65}			0.171	0.203	0.114	0.061
Zr^{95}-Nb^{95}				0.0775	0.0922	0.0533
Ru^{103}					0.057	0.053
Cr^{51}						0.048

The percent average deviations for five determinations of each efficiency factor is 1 percent, and the percent average deviation for correction factors is 1.7 percent.

III STUDIES BY LABORATORY PERSONNEL

A Analysis of Natural Terrestrial Waters

Norman Cutshall

Rain and river water were analyzed with filtration and precipitation methods to determine effects of terrestrial fallout on the marine environment. Osterberg et. al. (1963) show that marine sediments near the mouth of the Columbia River have fallout activities clearly related to distance offshore. The implication that the Columbia River has delivered much of the fallout in these sediments is obvious. If the fraction of marine fallout activity delivered by rivers is appreciable, the total activity and constitution of fallout in onshore versus offshore waters could be quite different.

Several possibly pertinent mechanisms are easily recognizable:

(1) Readily soluble fallout may be less likely to settle out of flowing streams and thus may be more readily carried to rivers by surface runoff and to the ocean by rivers.

(2) On the other hand, readily soluble fallout may more easily be sorbed to suspensoids and thus be more likely to settle out.

(3) Assimilation of fission products by biota may produce variations in the composition of suspensoids with biologically important radionuclides being preferentially retained and other radioelements rapidly excreted in particles larger than those originally present.

(4) Suspensoids caused to flocculate by sea water may provide effective scavenging of fission products upon introduction into the ocean. The relative importance of these and more subtle mechanisms is not so readily seen. The purpose of this phase of investigation was to begin an evaluation of the relative importance of the various limbs of the terrestrial fallout budget (Fig. 1).

Procedures

Filters. Natural waters were pumped through 0.65 micron membrane filters with glass fiber prefilters (Gelman Instrument Co.) and both filters were analyzed in the well of the 5" crystal. Filtration was continued until either the flow was diminished to virtually nothing (river samples), or until the sample was exhausted (rain samples). The volume of filtrate was measured and data are reported in picocuries per liter. Volumes of samples were characteristically about 20 liters.

Precipitates. A few of the Columbia River samples and rain water samples were subjected to $Fe(OH)_3$ scavenging precipitations intended to remove anions and colloids from solution. Consecutive precipitations on selected samples showed that one carefully made precipitate quantitatively

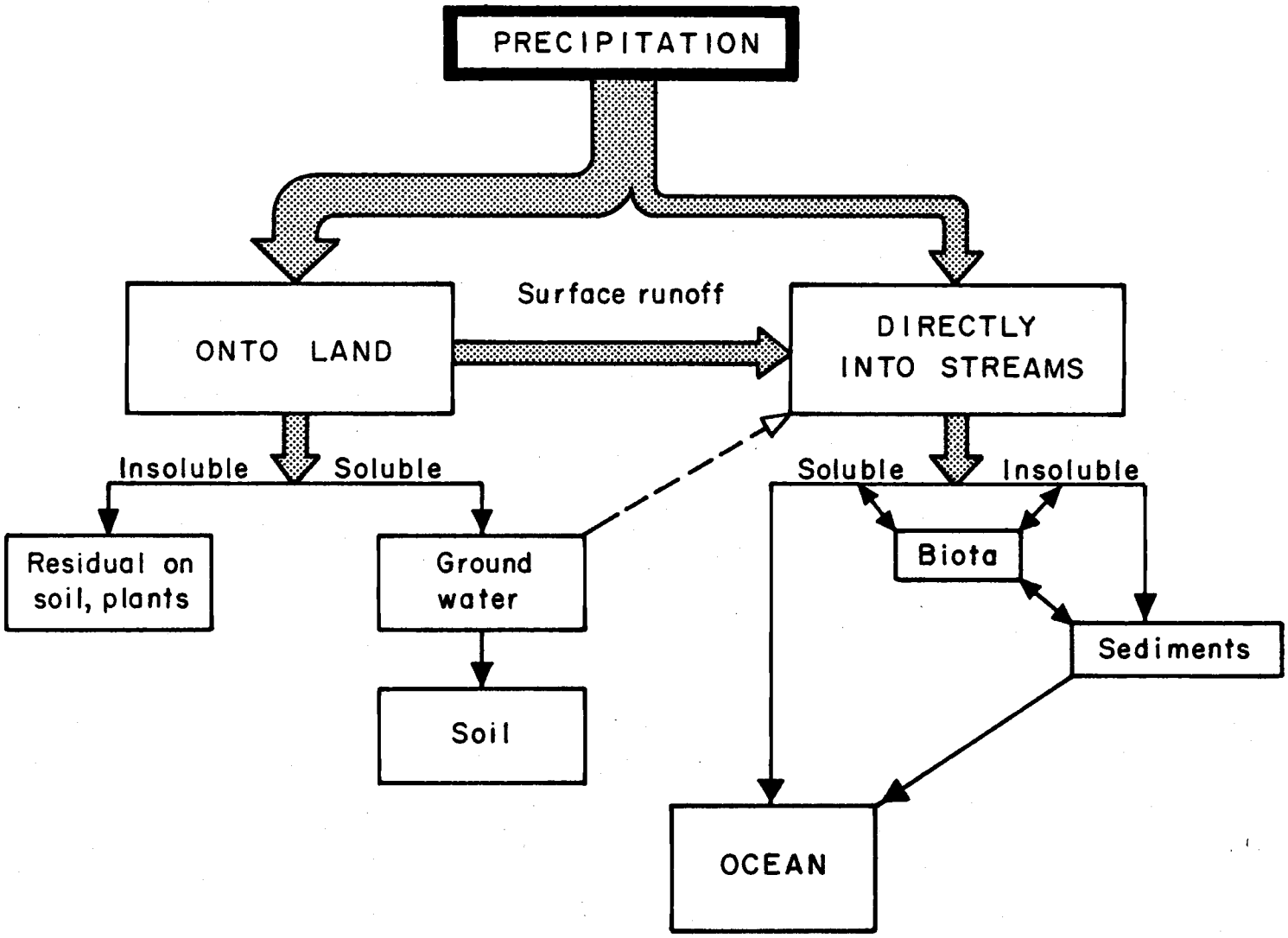

Figure 1. Terrestrial Fallout Budget.

removed fission products associated with colloids (first precipitate > 100 x second precipitate). Precipitates were filtered with Whatman #42 filter paper and counted.

Ion Exchange. Several filtrates were treated with ion exhange resins (Dowex 50 and Dowex 1) to concentrate ionic radionuclides.

Rain Water Analyses. Rain water, collected with a polyethylene-lined container, was filtered through 0.65 micron filters. Filters were counted and these data are presented in Table 2. Values in Table 2 represent minima since a considerable fraction of the fallout in rain is capable of passing through these filters. Activity delivered per square mile was calculated by multiplication of the sum of Zr^{95}, Ru^{103}, and Ce^{141} activity by the total number of liters falling on each square mile during each month. These values are probably reliable as to order of magnitude, but uncertainty in the fraction of activity actually retained on the filters prevents a more precise estimate.

Table 2. Mean monthly fallout in rainfall.

Month	Mean activity, picocuries/liter				Corvallis rainfall, inches**	Sum of Zr^{95}, Ru^{103}, and Ce^{141}, curies/mi^2
	Zr^{95}-Nb^{95}	Ru^{103}	Ce^{141}	Sum		
Feb 1963	244	30	105	379	5.23	0.13
Mar	256	32	64	352	6.30	0.16
Apr	450	51	167	668	4.64	0.20
May	222	24	45	291	3.95	0.10
Jun	304	64	116	484	0.96	0.03
Nov	17	6.2	8.9	31.1		
Dec	8	3.1	7.1	18.2		
Jan 1964	7	2.5	4.2	13.6		

**From U.S. Weather Bureau, Climatological Data by Sections, 1963.

By January 1964, Zr^{95}-Nb^{95} activity had decreased by a factor of about 40, or slightly more rapidly than simple radioactive decay calculations would predict. Probably the rapid decrease results from two factors: (1) dilution of fallout in the stratosphere, (2) much of the activity delivered during the period January-April 1963 may have resulted from rapid transfer of Russian fallout across the tropopause in northerly latitudes. Decay studies of Ru^{103} and Ce^{141} are complicated by the presence of one-year Ru^{106} and 285-day Ce^{144}, respectively. The values of Ce^{141} and Ru^{103} reported for dates after June 1963 are probably in error since correction for the longer-lived isotopes was not made. It should be mentioned that the error in Ce^{141} is predominantly from Ce^{144} and the error in Ru^{103} is predominantly from Ru^{106}. This means that although the absolute activity is incorrect,

the relative chemistry is still valid since the contamination in each case is isotopic with the element being measured.

Ferric hydroxide precipitates were made in filtrates of many of the samples taken from November 1963--January 1964. It was found that about twice as much total activity passed through the filters as was retained. The fraction of ruthenium associated with particles too small to filter was found to be higher than the corresponding fraction of Zr^{95}. This distribution probably reflects the "fractionation effect" wherein large particles are relatively more enriched in Zr^{95} than are small particles. The behavior of $Ce^{141-144}$ during filtration and precipitation was erratic.

Anion exchange resin shows no marked ability to concentrate the activity in rain. Acidification of the filtrate with sulphuric acid and immediate addition of Ba^{++} was found to quantitatively remove $Ce^{141-144}$ but only a slight amount of Zr or Ru. Whether Ce^{+3} was leached from the colloidal fallout or the entire particles solubilized is now under study.

Natural Radioactivity in Rain Water. Rain samples taken during periods of extremely heavy rainfall and filtered immediately were found to contain 27-minute Pb^{214} (RaB) and its daughter, 20-minute Bi^{214} (RaC). It is possible that 3.8-day Rn^{222} may be dissolved in rain and only its decay products filtered out, or perhaps only the decay products themselves are present in rainwater. This will be investigated in the near future.

B River Water Analyses

Norman Cutshall and Lauren Larsen

Various Oregon rivers were sampled by filtration during 1963 (Table 3). Readily detectable levels of fission products were found during the months January-April, but no fission products were detected in December. Within a given region the activity of suspensoids was approximately proportional to the amount of rainfall (Fig. 2). Activity appeared to increase in the order: Coast region < Willamette Basin < Eastern Oregon (Table 4). The volume of rainfall in these regions increases in the order: Eastern Oregon < Willamette Basin < Coast Region. Evidently, the level of activity is not a simple function of volume of rainfall when different regions are compared.

It was found that the cast iron pump used in these experiments concentrated fallout on rust inside the pump. When the Metolius River was sampled at its source, a total of 0.06 picocuries/liter of Zr^{95}-Nb^{95} + Ru^{103} + Ce^{141} was found on the filter. * If contamination of samples

*The Metolius River in Central Oregon emerges from underground through a porous rock stratum and should be free of fallout at the sample site.

Table 3. List of Rivers Sampled.

Region	River	Number of samples
Coast	Nehalem	2
	Nestucca	2
	Siletz	2
	Alsea	1
	Siuslaw	2
	Coquille	1
	Rogue	1
	Umpqua	2
Willamette Basin	Clackamas	2
	Yamhill	2
	North Fork Santiam	2
	South Fork Santiam	2
	Mary's	2
	McKenzie	2
	Willamette	31
Eastern Oregon	Deschutes	4
	Metolius	1
	Snake	2
	Burnt	2
	John Day	6
	Owyhee	2
	Malheur	2
Other	Columbia	23

Table 4. Average Activity of River Waters Tabulated by Month and Region

	Picocuries/liter		
	Zr^{95}-Nb^{95}	Ru^{103}	Ce^{141}
January 1963			
Coastal Region	0.36	0.09	0.27
Willamette Basin	0.85	0.32	0.59
Eastern Oregon	0.54	0.20	1.4
February 1963			
Willamette Basin	7.0	1.9	1.3
March 1963			
Coast Region	2.6	0.59	0.41
Willamette Basin	14.9	5.3	3.4
Eastern Oregon	4.1	2.5	1.2
April 1963			
Willamette Basin	5.9	2.0	1.2

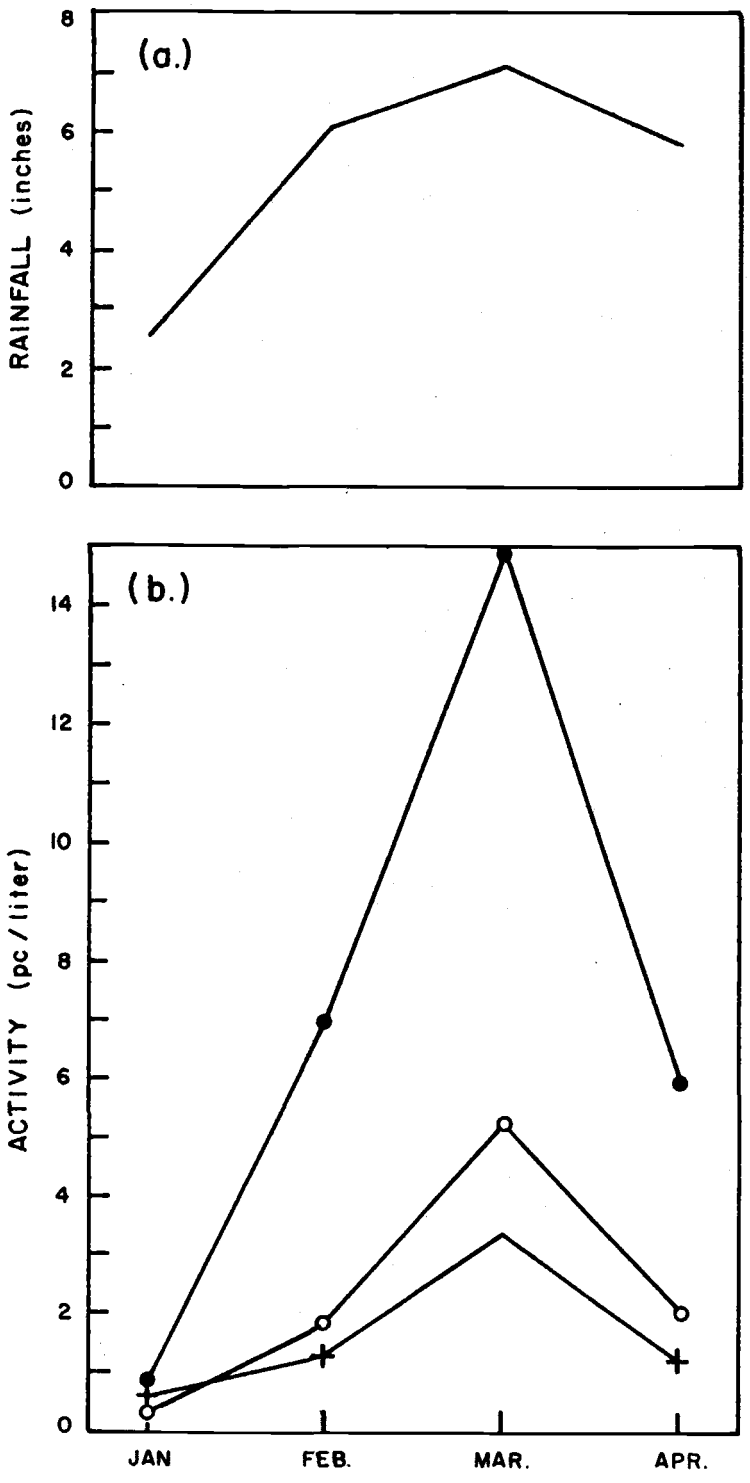

Figure 2. (a) Total monthly rainfall in the Willamette Basin, Jan. - April 1963.
(b) Average monthly radioactivity (in picocuries/liter) in rivers in the Willamette Basin, Jan. - April 1963.

by erosion of rust from the pump was constant from sample to sample, contamination was small compared with levels observed in most filters. If contamination were highly variable, as would be the case if large flakes of rust were responsible, then division of the filter should not result in equivalent division of activity. Several sample filters were cut into quarters and the quarters analyzed separately. Division of the filters caused equivalent division of activity. This indicates that no great fraction of activity resulted from large flakes of rust or from individual fallout particles. In spite of the apparently negligible level of contamination from the cast iron pump, it was replaced by a stainless steel pump in an attempt to completely eliminate cross contamination of samples. River water pumped with the stainless steel pump is not detectably contaminated by residue from previously filtered rain samples.

Mechanisms of Introduction of Fallout into Rivers

Fallout is probably introduced into rivers by way of two major pathways: (1) direct precipitation into rivers and (2) surface runoff of rain water. (We shall represent these by DP and SR.) It is unlikely that water passing through the soil (groundwater) in its rain-to-river cycle will carry any fallout with it. The fraction of rain entering a river by DP should be nearly independent of volume of rainfall, while that fraction entering rivers via SR should be a highly variable function of rate of rainfall, volume of rainfall and degree of saturation of soil. A plot of the ratio of fallout activity in rivers to that in rainfall versus the volume of rainfall should have a slope proportional to SR and an extrapolated intercept proportional to DP (Fig. 3). The observed nature of these graphs is somewhat different from that of the theoretical plot. Apparently the fallout entering rivers via DP was negligible in comparison to that entering via SR during the period for which data were obtained. Furthermore, a rainfall of about 5-1/2 inches per month appears to have been necessary to cause appreciable fallout to enter rivers in the Willamette Basin via surface runoff.

It is recognized that the above treatment is a gross oversimplification of the problem at hand. For instance, we have assumed that the volume of water entering rivers via groundwater is constant. Consequently, these estimates of the relative importance of DP and SR as pathways for the transport of fission products must be regarded as first approximations. It is notable that during January 1963, when rainfall was only 2.57 inches (or less than half that required for reasonable contribution from surface runoff), fallout activity appearing in rivers was lower by an order of magnitude than during the ensuing three months. Interesting although it is, this phase of investigation has been suspended because fallout levels in river waters are no longer high enough to be discernible.

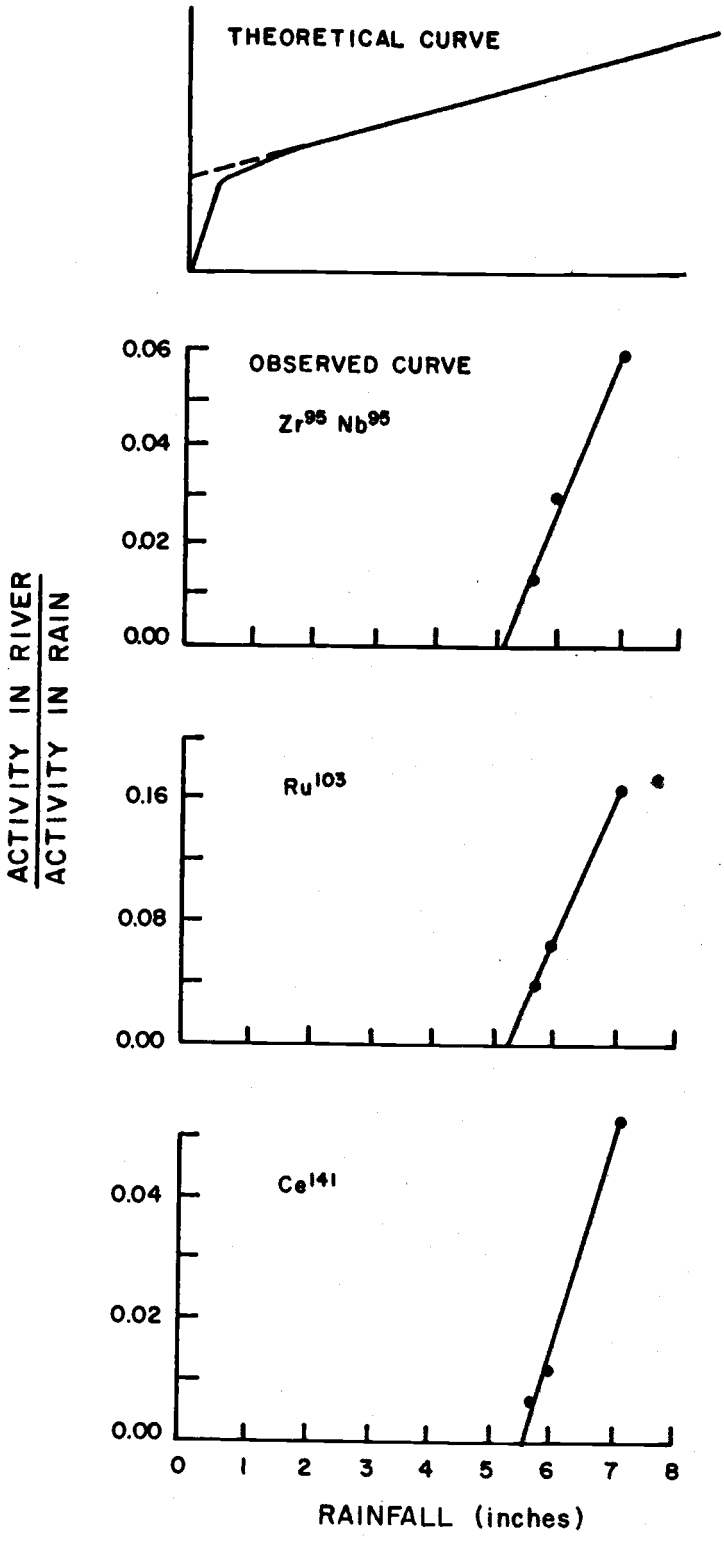

Figure 3. Theoretical and observed curves of ratios of radioactivity in river: RA in rain versus rainfall in inches.

C Miscellaneous Observations on Fallout

Norman Cutshall

The roof of the six-floor Physics-Chemistry Building yields relatively pure, high activity fallout samples. Principal gamma-emitters detected were Zr^{95}-Nb^{95}, Ru^{103}, and Ce^{141}. Lesser amounts of Ru^{106}-Rh^{106}, and Ce^{144}-Pr^{144} were also identified. Chemical separation allowed the characterization of Cs^{137} and Mn^{54}. (The gamma-ray energies of these latter two are so similar to Zr^{95}-Nb^{95} that their positive identification must involve chemical separation.) Fallout collected from the roof was only sparingly soluble in concentrated H_2SO_4, aqua regia or EDTA.

It was observed that steel wool suspended in the Willamette River during May 1963 adsorbed fallout, presumably on rust. The adsorption of Ru^{103} was relatively greater than Zr^{95}-Nb^{95} sorption. It is thought that this preferential sorption resulted from the distribution of Zr^{95}-Nb^{95} and Ru^{103} on large and small fallout particles, respectively, rather than chemical specificity of the fission products themselves. It is physically reasonable to expect smaller particles to be sorbed more readily than large particles. As reported in the section on rain water, small particles are higher in Ru^{103} as compared to Zr^{95}-Nb^{95} than are large particles.

Summary

Corvallis rainfall during the early months of 1963 deposited fission products in total monthly quantities equivalent to at least 5 percent of the estimated level of natural surface radioactivity. (Arnold and Martell, Scientific American, September 1959, estimate natural surface activity as 2 curies/square mile.)

A fraction of this activity (at least 1 to 10 percent) was carried into Oregon rivers, mostly via surface runoff. The degree of sedimentation of fallout within the rivers is not known. Suspensoids within the Columbia River appear to be carried at about 1/3 the rate of colloidal or ionic materials. The degree of flocculation of radioactive materials upon introduction of river waters into the ocean remains to be seen. The decrease in fission product activity following the curtailment of atmospheric testing enforces the direction of investigation into other phases of the terrestrial fallout budget. For instance, Cs^{137} is readily detectable in Oregon deer flesh. Concentration and retention of radioactivity from fallout by biota may cause division of geochemical pathways.

D In Situ Measurement of Gamma Emitters in the Columbia River

Mark I

C. Osterberg, D. Jennings, and N. Cutshall

Reactor effluent discharged into the Columbia River from the Hanford Atomic Products Operation contains more than 60 different radionuclides. The only three gamma emitters with significant activity near the river mouth are chromium-51, neptunium-239, and zinc-65 (Watson, et. al., 1963). In order to determine the feasibility of detecting Cr^{51} in situ in the river, an experiment was carried out at the United Fish Co-op Warehouse dock in Astoria on 11 October 1963.

A 3x3-inch NaI(Tl) Crystal was mounted in a 4-inch styrene pipe and insulated against thermal and mechanical shock (Fig. 4). The probe was then mounted on a float and placed in the river. The crystal was placed three feet below the surface of the water, and spectral data were taken for a 15-hour period to determine whether any tidal fluctuations could be detected. Consecutive 40-minute counts were taken from 0100 to 1600, and readout from the ND-130 AT Analyzer was by IBM typewriter and Tally punch-tape. * In addition to spectral data, salinity, temperature, conductivity, and pH were recorded at the time of each readout.

Three consecutive spectra (each of 40 minutes duration) show that Cr^{51} contributed the most obvious peak, although there is evidence of both Zn^{65} and K^{40}, with lesser peaks probably from fallout (Fig. 5). Considering the environmental conditions, stability was remarkable, i.e., the three spectra coincide almost perfectly. Absence of drift was gratifying.

Figure 6 shows Cr^{51} and salinity plotted over the time of the experiment. It can be seen that a decrease in salinity is accompanied by an increase in Cr^{51} activity in the river and vice versa. This is to be expected since a decrease in salinity indicates a decrease of sea water per volume. As the relative amount of high salinity sea water decreases, the amount of river water increases. River water is rich in Cr^{51}, so as salinity of water decreases, Cr^{51} content increases.

The graph of temperature and Cr^{51} versus time (Fig. 7) shows a direct relationship between Cr^{51} and water temperature. River water was warmer than the ocean water and also had a higher content of Cr^{51}; thus, the more river water present, the higher the temperature and the greater the Cr^{51} activity.

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* We gratefully acknowledge the assistance of Dave Smith of Nuclear Data, whose loan of an ND-130AT 512 channel analyzer complete with readout attachments made this trial possible.

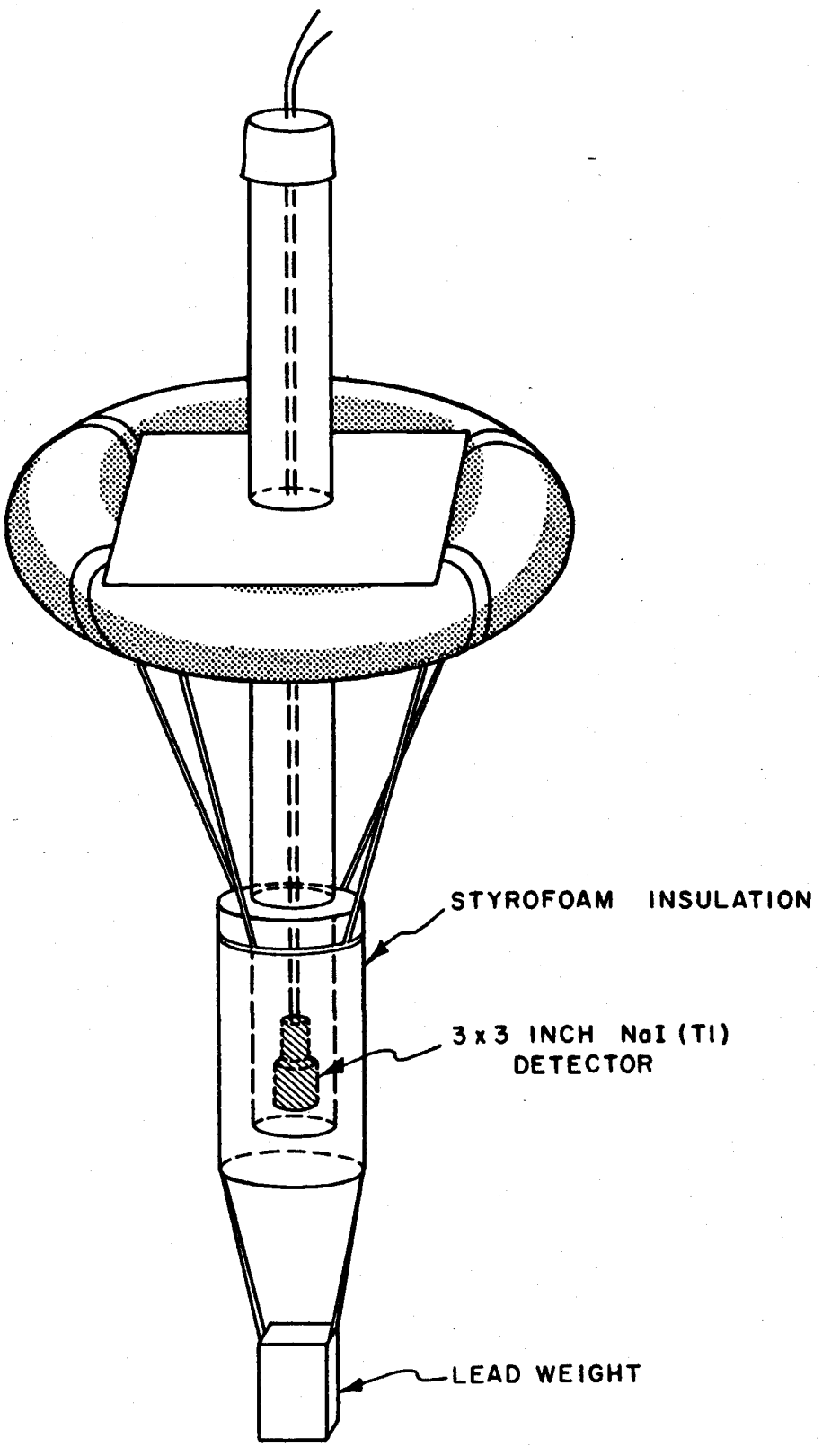

Figure 4. Mark I In situ gamma-probe.

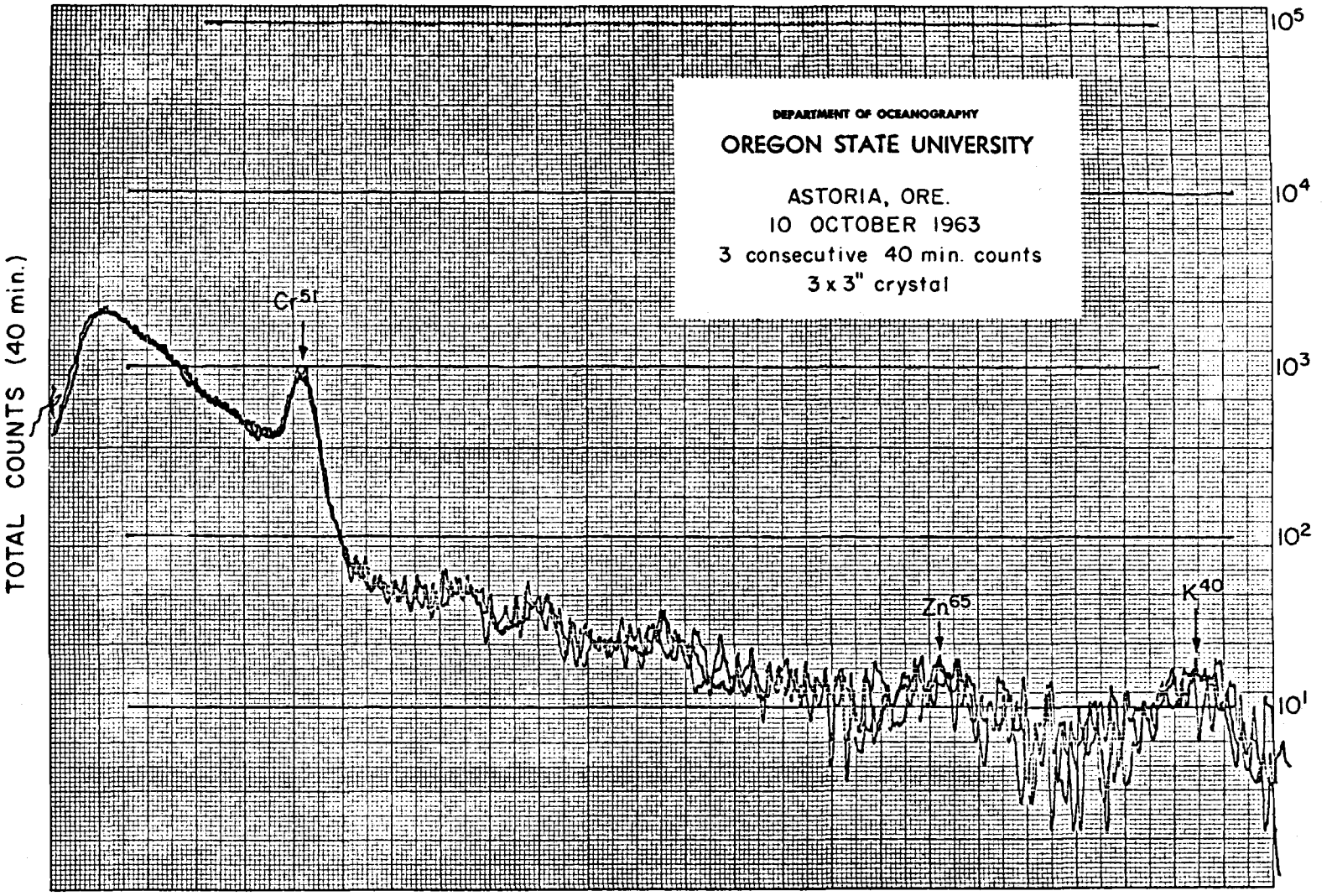

Figure 5. Three consecutive 40-minute spectra, made with in situ probe. Note relative freedom from drift. Astoria, Oregon. 10 October 1963.

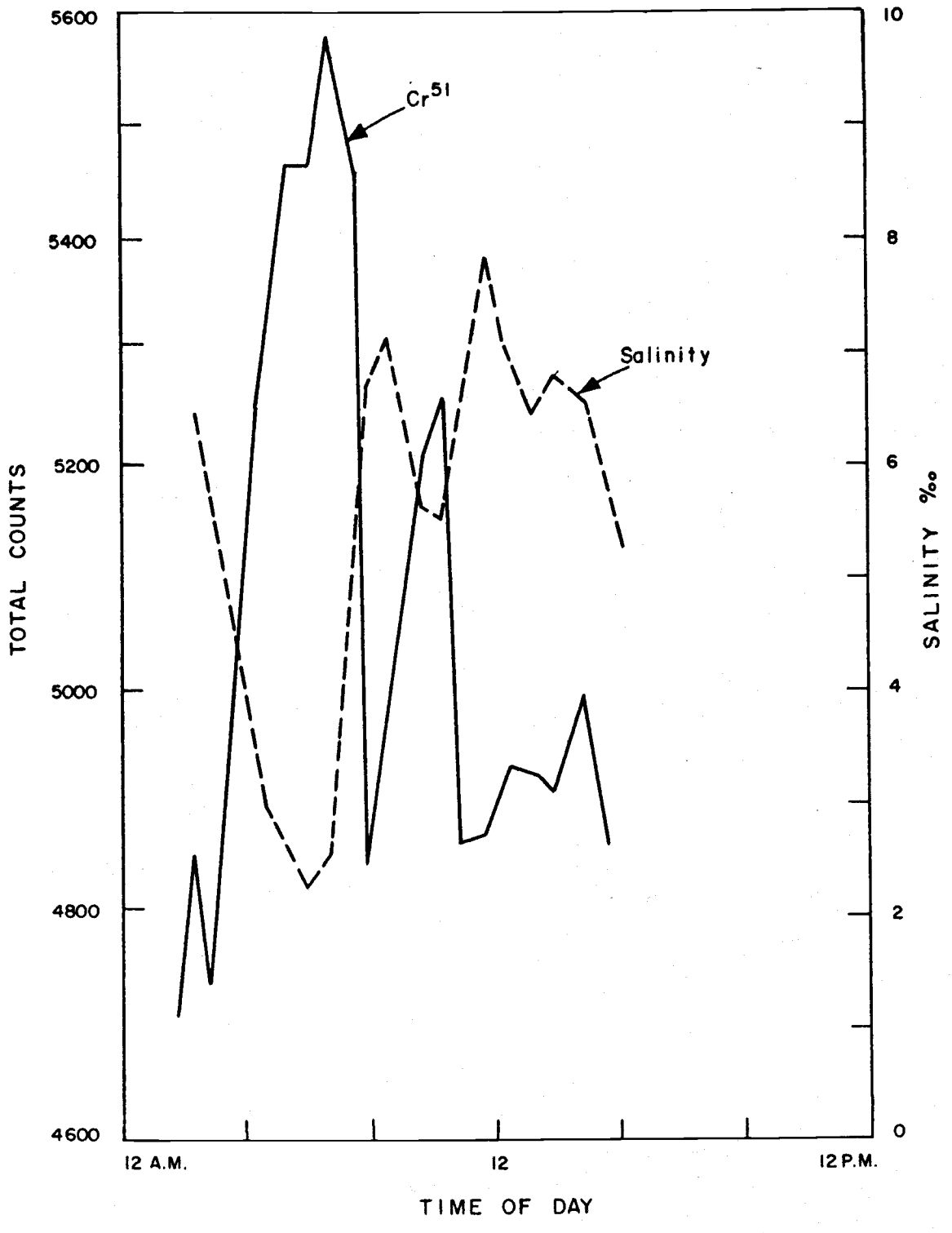

Figure 6. Comparison of chromium-51 and salinity versus time. Astoria, Oregon. 10 October 1963.

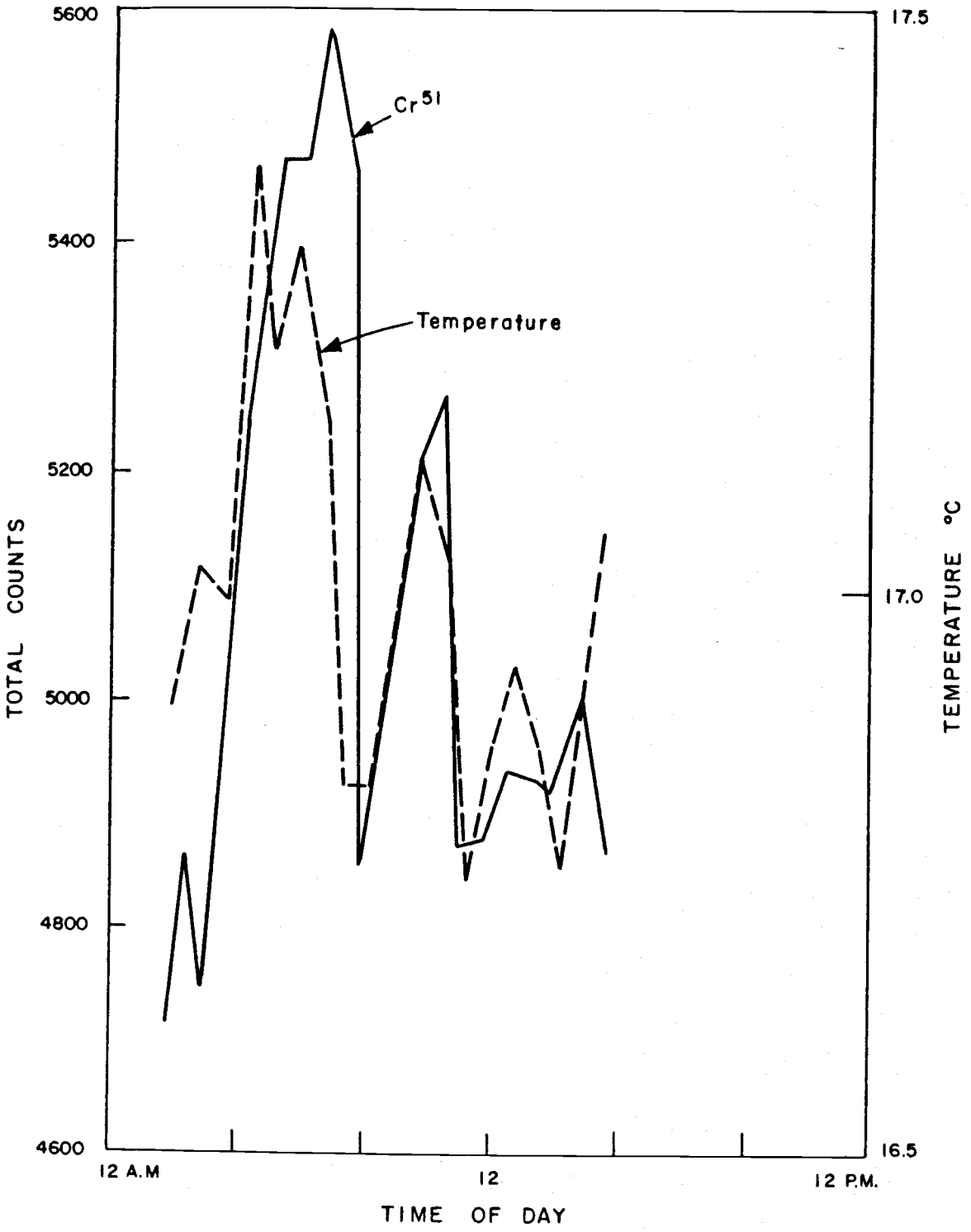

Figure 7. Comparison of chromium-51 and temperature, versus time. Astoria. 10 October 1963.

A comparison of salinity with pH was also made which shows that they coincide quite well, except for a few unexplainable points. The sea water has both higher salinity and pH than the river water so that pH increases as salinity increases, and decreases as salinity decreases. However, pH measurements are of doubtful value because of the poorly poised nature of river water.

We cannot expect to be able to tell much about the complex movements of the Columbia River tides and currents from 15 hours of observation at a shore station. However, the ability of our detector to follow small tidal fluctuations by changes in Cr^{51} content indicated that the sensitivity of the instrument was sufficient to warrant more sophisticated studies. Therefore, a second experiment was designed. A report on this, in which the in situ probe was mounted on a boat, follows.

Mickey Mouse I*

Following the preliminary study at Astoria, in which it was found that in situ measurements of Cr^{51} and Zn^{65} are feasible, more extensive in situ measurements were made up the Columbia River to gain more knowledge of the budget of radionuclides in the river. The trawler GARDA MARIE was chartered and a 6-day cruise from 18-23 December 1963 was undertaken from Astoria to Hood River and back to Astoria. On the up-stream leg of the cruise, biological samples were taken with an otter trawl at about 20-mile intervals and some sediment samples were obtained. In addition, magnetometer readings were taken to observe the earth's field. At Hood River, the in situ probe was mounted on the side of the boat and connected to an ND-130 AT Analyzer-computer stored in the hold. Power for the analyzer was supplied by a 5 kw gasoline generator. On the way down the river, stations were chosen at approximately 15-mile intervals. At each station, a 40-minute count was taken with the probe and river water was filtered through a 0.65 micron Millipore filter. The filters were counted later in the laboratory. Also, three 400-minute counts were taken while anchored overnight. A Co^{60} standard was held near the probe several times each day to check calibration.

Table 5 displays data from both the probe and filters. The remarkable agreement of probe and filter results from duplicate samples taken at Hayden Island gives us confidence in our data. A further confirmation of the reliability of the data was observed when the ship was taken up into the Willamette River at Portland. Very low levels of Cr^{51} and Zn^{65} were seen on the filters, and the spectra from the probe showed no measurable Cr^{51} or Zn^{65}.

The Cr^{51} and Zn^{65} counted on the filters are measures of the Cr^{51} and Zn^{65} associated with particulate matter. The activity of these radionuclides observed with the probe gives the total activity in the

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* A report of this cruise is being prepared and will be submitted for possible publication.

Table 5. In situ and Filter Data.

	Station	Distance from mouth of Columbia (mi)	Probe*		Filters	
			Cr^{51} x cnst.	Zr^{65} x cnst.	Picocuries/l	Picocuries/l
1.	Hood River	130	78.0	4.42	127.5	6.1
2.	Above Bonneville Dam	112	68.8	4.30	89.7	11.6
3.	Below Bonneville Dam	110	65.0	3.70	97.4	12.5
4.	Rooster Rock	95	61.2	3.80	102.4	12.1
5.	Government Island	85	56.9	3.77	89.6	11.6
6.	Interstate Bridge	76	41.9	2.70	83.6	9.6
7.	Hayden Island	75	52.0	3.05	107.6	12.9
	Hayden Island Duplicate	75	51.8	3.09	107.9	12.8
8.	Campbell Lake	65	37.5	2.70	68.1	7.7
9.	Walker Island	51	20.2	0.90	40.5	4.4
10.	Puget Island	38	28.3	2.03	167.6	13.5
11.	Harrington Point	21	31.2	1.80	90.3	6.0
12.	Astoria Dock	11	37.2	1.32	78.9	3.1
13.	Buoy 14	4	25.7	0.95	76.5	2.0
14.	Willamette Buoy 6				4.9	0.4
15.	Willamette Swan Island				2.4	0.2
					6.3	0.0

* Probe data are expressed in units of area of the photopeak, which are proportional to picocuries per liter. Absolute units cannot be used since the detector has not been calibrated for an "infinite" sample."

water, both in associated form and a non-associated form. The ratio of the activity of a nuclide on the filter to the activity of that nuclide as measured with the probe gives the percent of association of the radionuclide with particulate matter.

Figure 8 shows percent of Cr^{51} associated with particles plotted against distance from the mouth of the Columbia River. It can be seen that the Cr^{51} gradually continues to increase its association with particulate matter all the way from Hood River to the mouth of the river.

Figure 9 shows percent of Zn^{65} associated with particles versus distance from the mouth of the Columbia River. The Zn^{65} increases its degree of association from Hood River down to about 20 miles above the mouth where it comes into contact with salt water. The percent of association then decreases toward the mouth. The points on Figures 9 and 10 at the 130-mile station are not accurate since the in situ counting was done in the center of the channel and filter data were taken in slow moving water near the dock where we observed less activity on the filters than in the channel. The data point at 38 miles is anomalous since the volume of water filtered at this station was low, and apparently an error occurred in the estimation of the volume of water. Data points for the percent of Zn^{65} associated are somewhat more scattered than the Cr^{51} data due to the greater inherent inaccuracy of reducing the Zn^{65}, which has much less activity in the river. However, the data do indicate that the Zn^{65} becomes dissociated from particles when it comes in contact with sea water. This corroborates the findings of Vern Johnson and Norm Cutshall who have treated sediments from the Columbia River with sea water in our laboratory and have observed the dissociation of about 20 percent of the Zn^{65} from particles (see elution curves in Johnson's paper).

The conclusion drawn is that sea water will cause some Zn^{65} to dissociate from particulate matter while Cr^{51} does not exhibit this tendency to dissociate.

The map (Fig. 10) shows where spectra were taken. The nearest station to the Hanford reactors was Menaloose Island, between Hood River and The Dalles. The spectrum from this location (Fig. 11) shows considerably more Zn^{65} than one made in salt water below Astoria (Fig. 12). The Cr^{51} peak is a prominent one at both locations, however. A spectrum made in the center of the channel at Swan Island on the Willamette River gives no evidence of Zn^{65} or Cr^{51}, although K^{40} is present with the possibility of some fallout (Fig. 13).

Overnight (400-minute) counts were made at three locations in the river. Maximum resolution was not attained, probably due to variations in voltage from our portable generator. Figure 14 is a spectrum made at the dock at Hood River, while the spectrum in Figure 15 was made while anchored in the center of the channel at Walker Island below Longview, Washington.

River water temperature varied between 5 and 6°C, but reached 7.1°C in the estuary. Presumably metabolic activity in organisms was

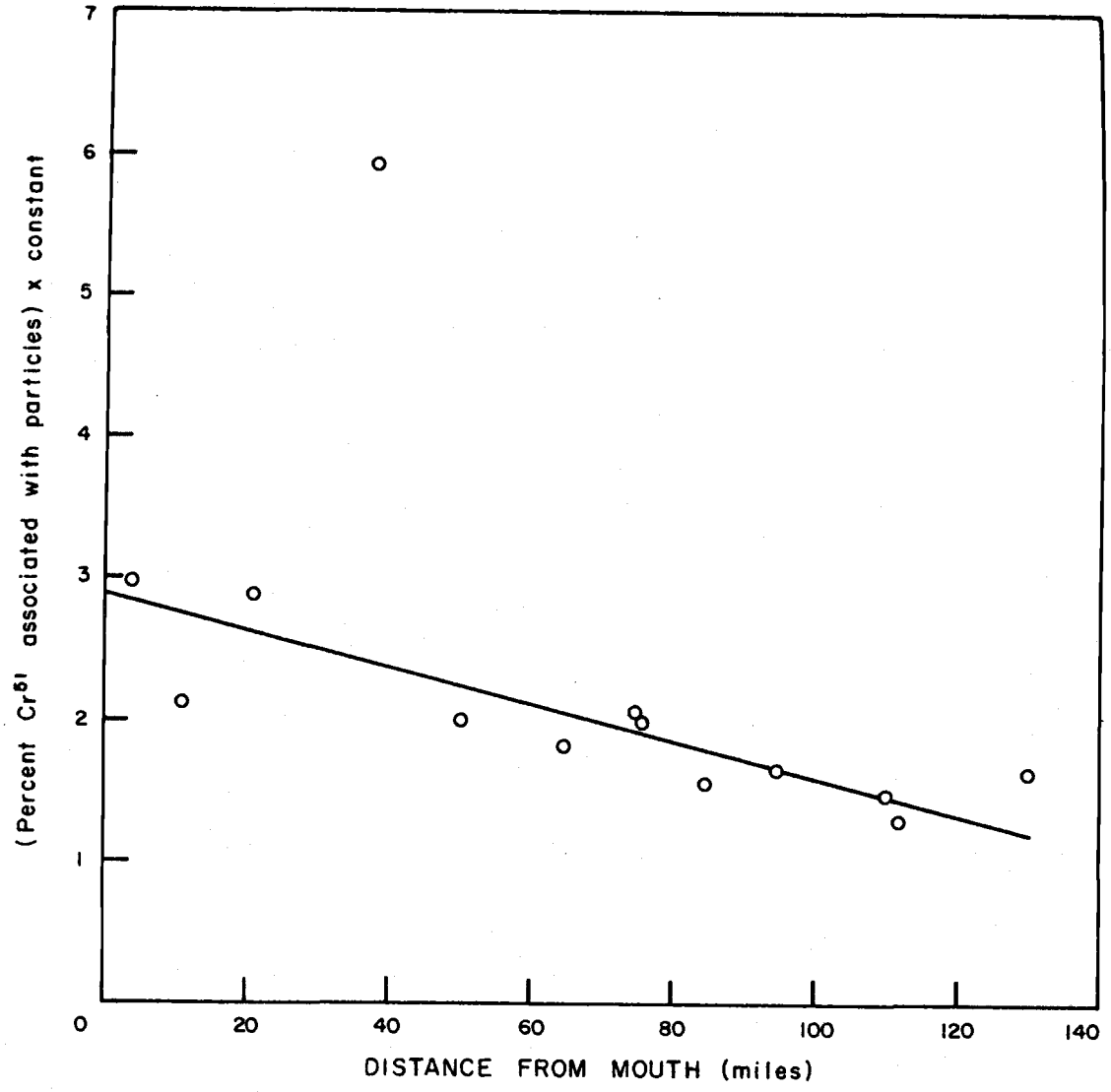

Figure 8. Percent Cr^{51} associated with particles (x constant) versus distance from mouth of the Columbia River.

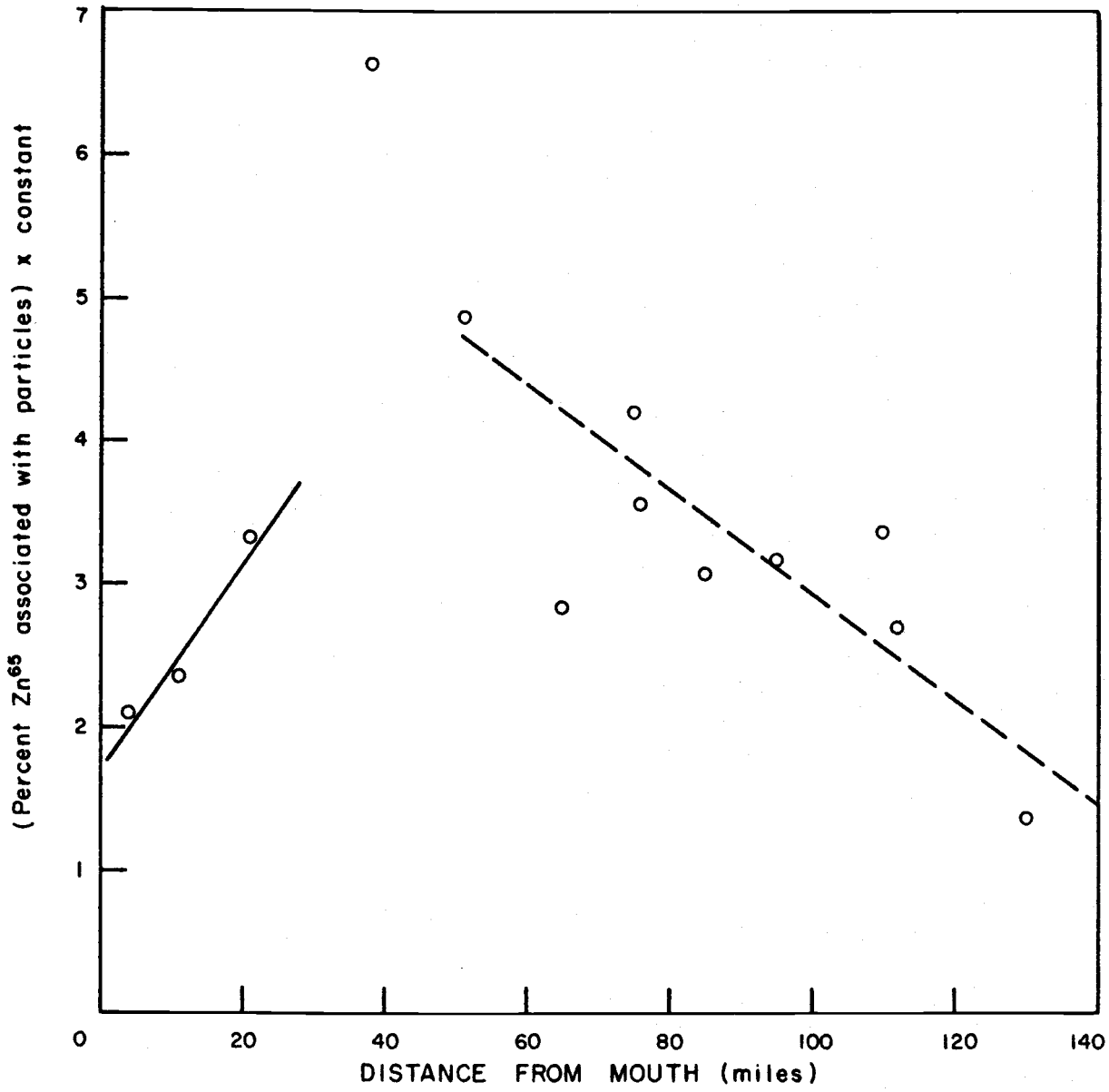

Figure 9. Percent Zn^{65} associated with particles (x constant) versus distance from the mouth of the Columbia River. Note change in ratio near mouth.

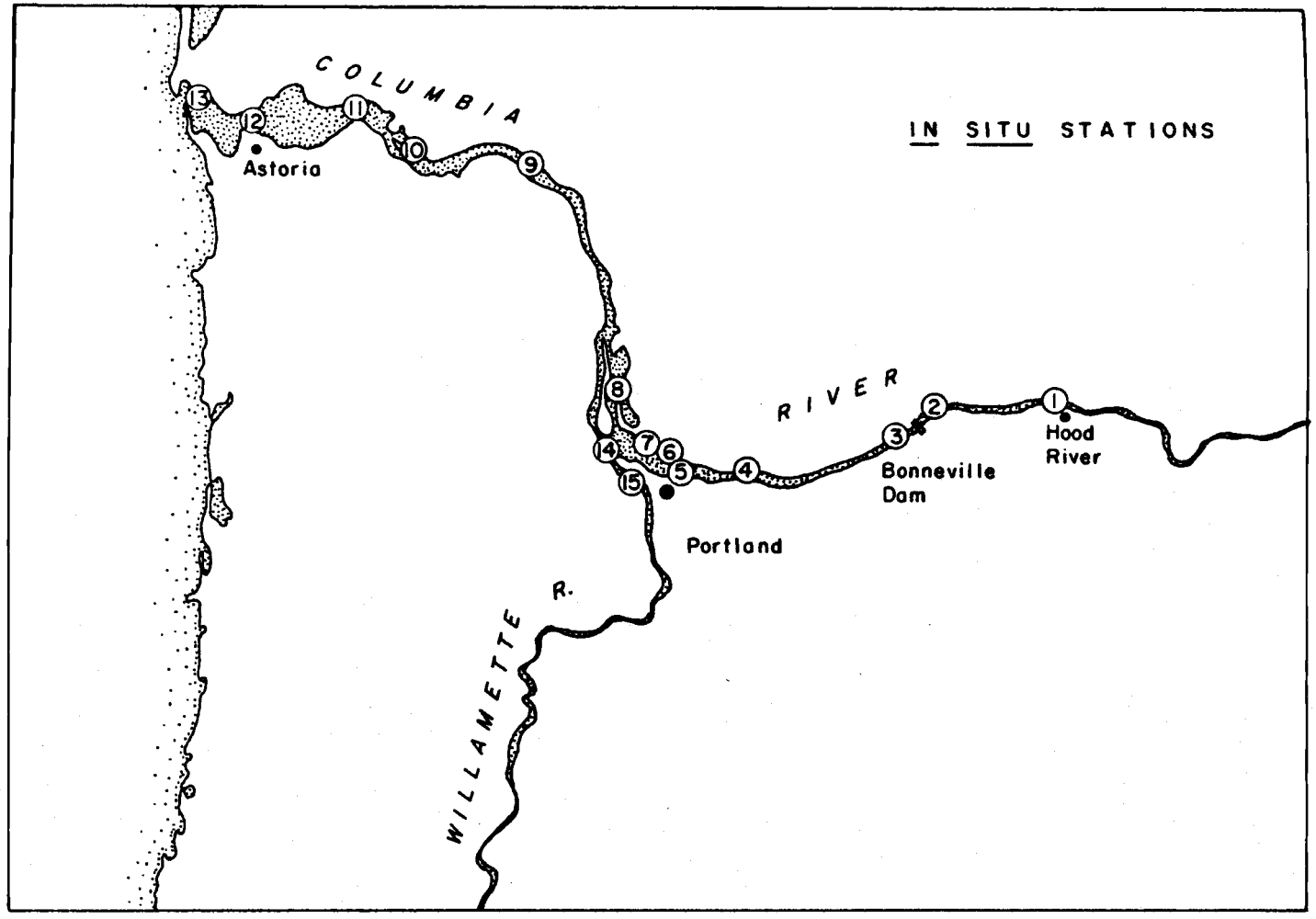

Figure 10. Map of Columbia River, showing in situ and filter stations, 18-23 December 1963.

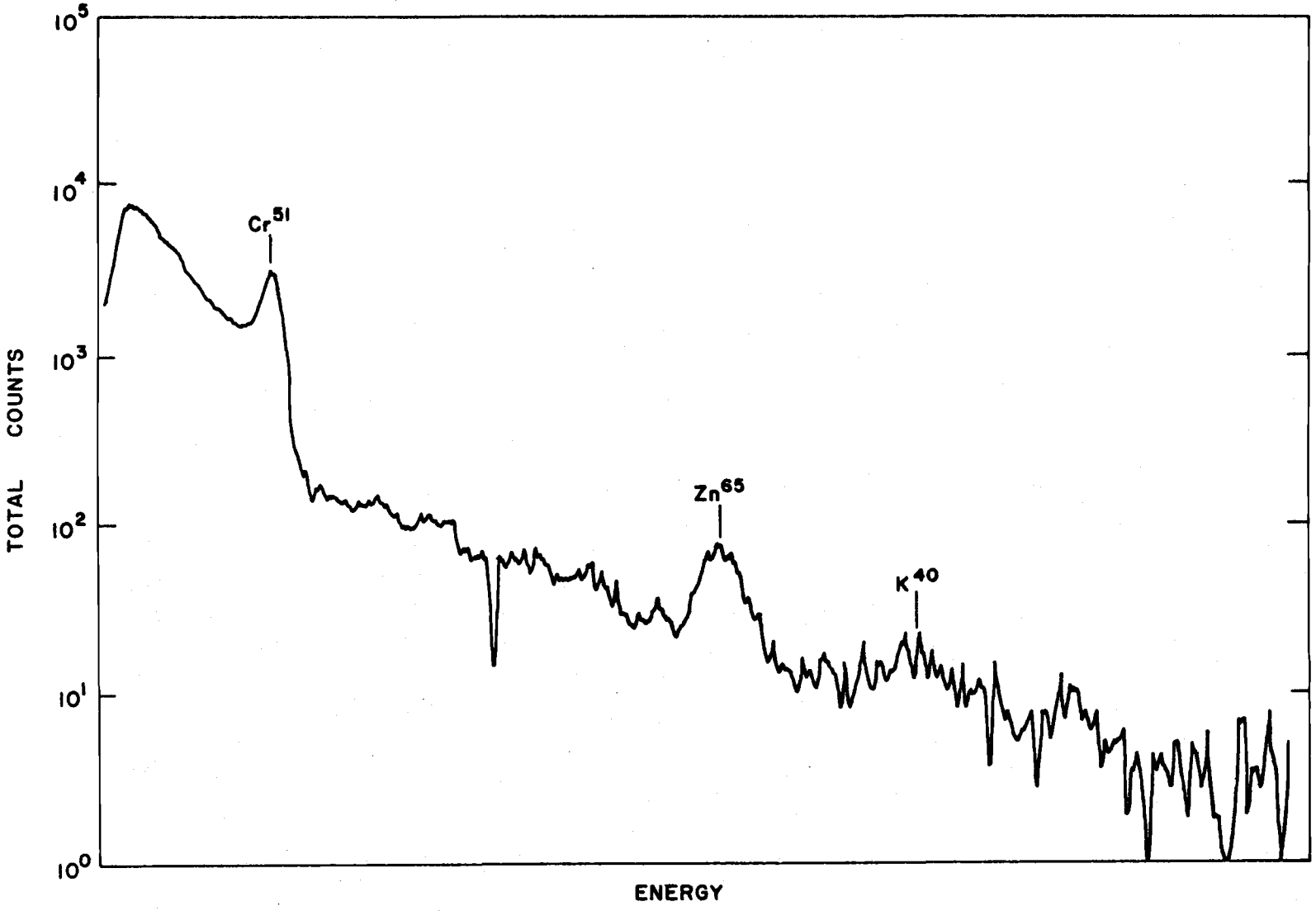

Figure 11. In situ spectrum above Bonneville Dam. 40 minute count.

Figure 12. *In situ* spectrum near mouth of Columbia River. 40 minute count.

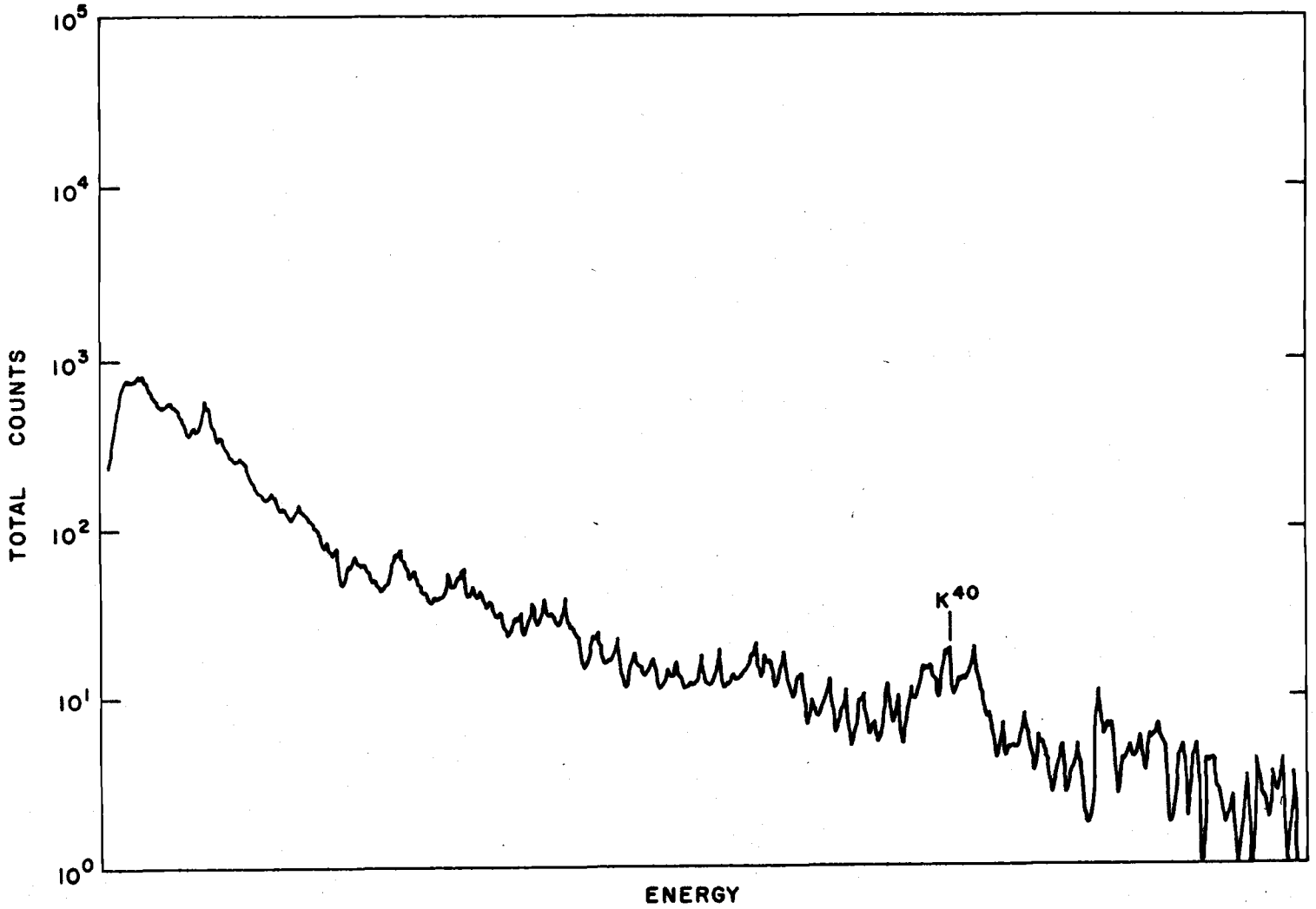

Figure 13. In situ spectrum off Swan Island in the Willamette River. 40 minute count.

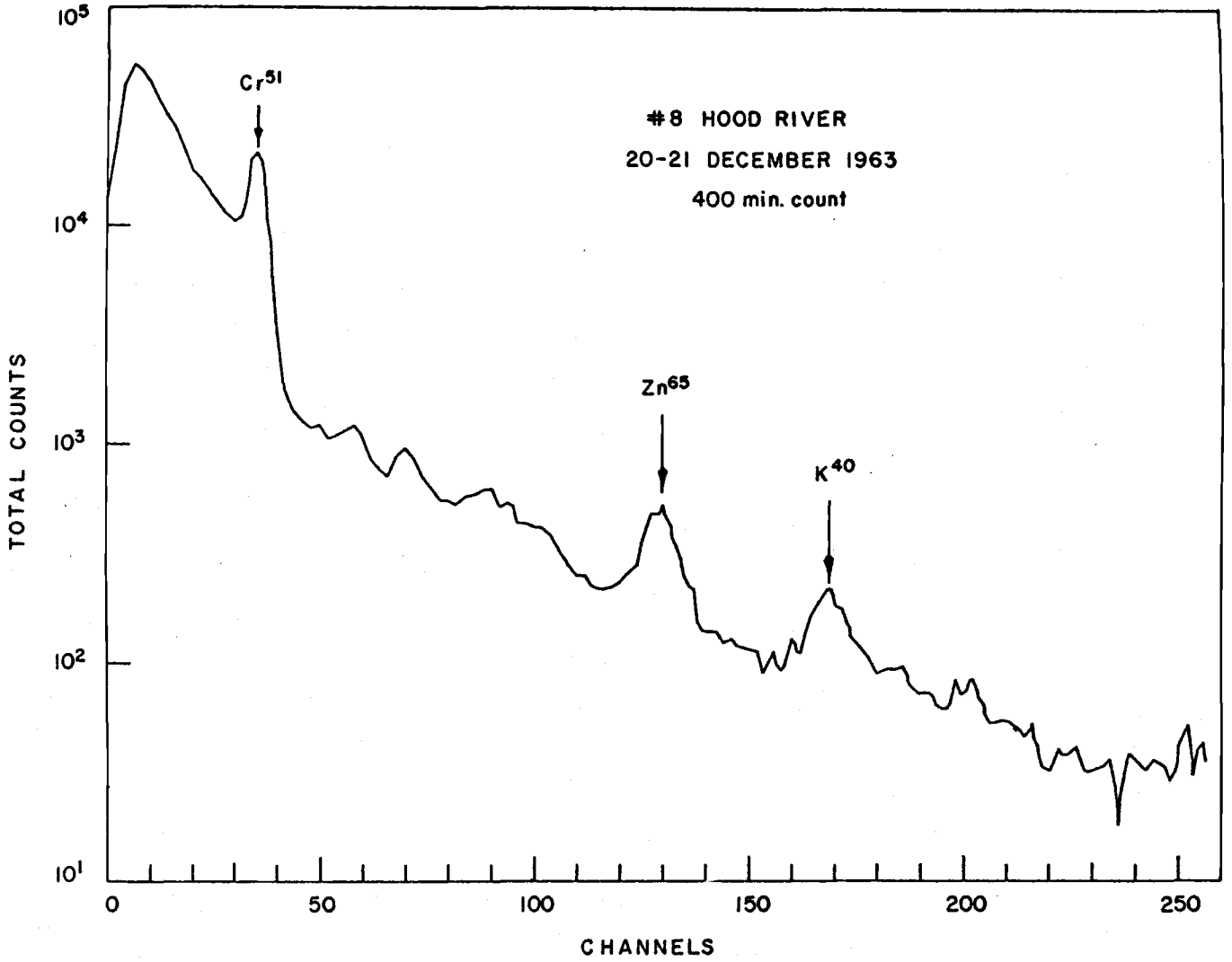

Figure 14. In situ spectrum at dock in Columbia River at Hood River, Oregon. Overnight (400 min.) count.

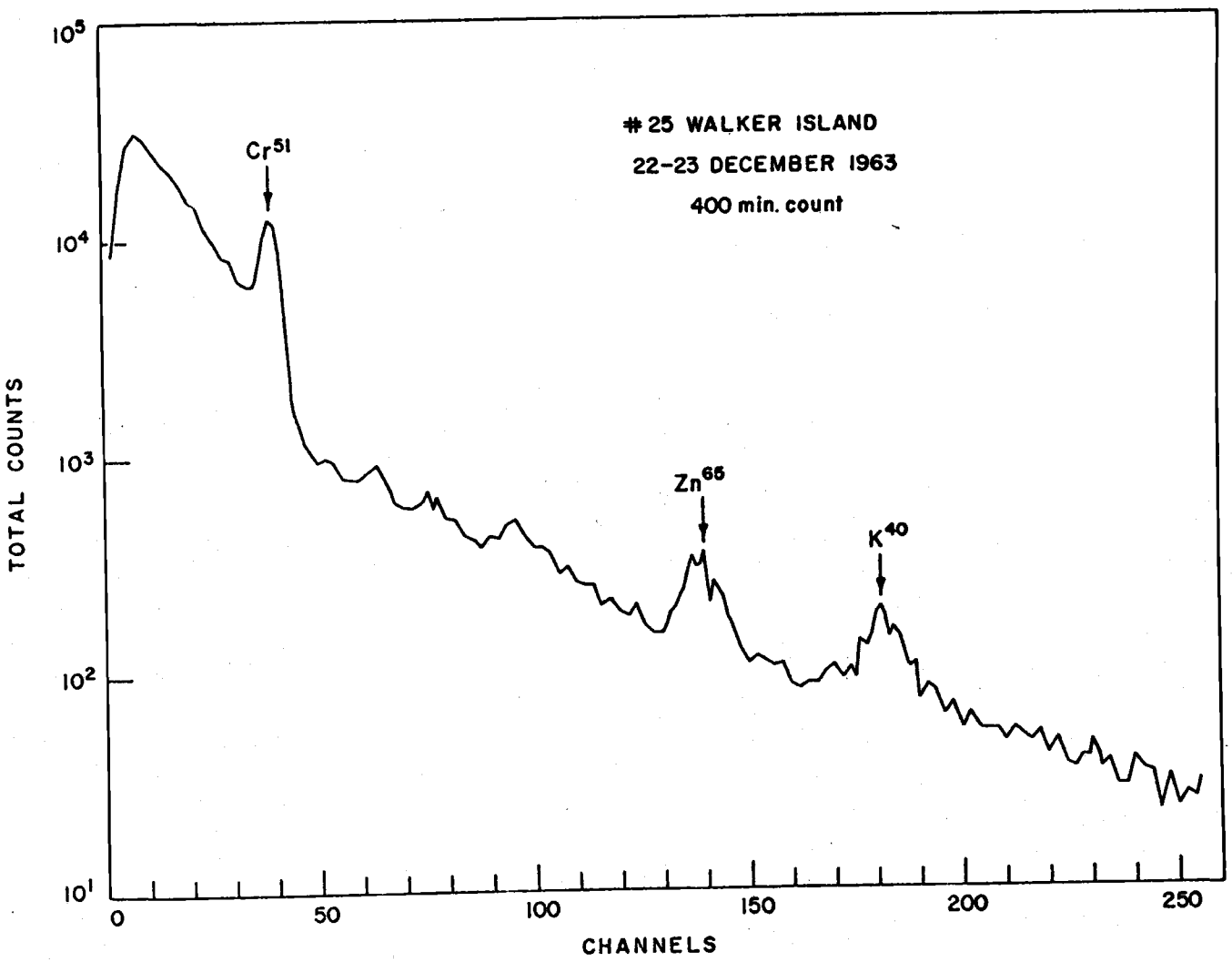

Figure 15. In situ spectrum in channel near Walker Island in Columbia River, about 14 miles above Astoria, Oregon. Overnight (400 min.) count.

reduced compared with the Mark I studies, when temperature exceeded 17° C. A summer cruise would probably show changes from the December pattern. These changes, as related to temperature, should give an indication whether biological (temperature sensitive) or physical processes (relatively insensitive to temperature) predominate.

E. Columbia River Filter Samples

Norman Cutshall and Charles Osterberg

Filter samples of Columbia River water were obtained in July 1963 during the sediment sampling trip and in December 1963 in conjunction with *in situ* probe studies. Activity of fission products during July was minor in comparison to Cr^{51} and Zn^{65}. Fission products were not detected in the samples taken in December.

July Series

Ferric hydroxide precipitations were made in the July filtrates to determine distribution of radioactivity between filterable and non-filterable (colloidal and ionic) fractions. Average of five samples showed 1/3 of Zr^{95}-Nb^{95} was retained by filters. This fraction is almost identical to that observed in rain water. Apparently Zr^{95}-Nb^{95}, which becomes associated with particles after introduction into rivers, is removed from suspension into sediments.

Zn^{65} was characteristically more than 90 percent filterable (i.e., removed by filters) while 80 percent of Cr^{51} ordinarily passed through the filters. Experiments made in our laboratory indicate that Cr^{51} on bottom sediments is not exchangeable. If this is also true for Cr^{51} fixation on suspensoids, one may calculate a minimum transport rate for suspensoids by assuming the change in activity of Cr^{51} radioactivity on suspensoids with distance is due only to radioactive decay. A similar calculation may be made for non-filterable Cr^{51} and for Zn^{65}. These values are tabulated in Table 6. Loss of Cr^{51} or Zn^{65} by means other than radioactive decay or dilution by other rivers will cause transport rates calculated in this way to be lower than the true value (A number of tributaries flow into the Columbia.). It is significant that the more biologically active element of the two (Zn^{65}) has a lower apparent transport rate. It is also interesting that the apparent transport rate of filterable activity is significantly lower than non-filterable activity. This is suggestive that flushing rates for non-particulate contaminants will be faster than for particulate wastes.

Table 6. Apparent Transport Rates

	July		December
	Non-filterable	Filterable	Filterable
Cr^{51}	16 mi/day	6 mi/day	6 mi/day
Zn^{65}	2 mi/day	0.6 mi/day	0.6 mi/day

December Series

Filter samples were taken between Hood River, Oregon, and the mouth of the Columbia during the in situ probe voyage. Activities of Cr^{51} and Zn^{65}, are tabulated in Table 7. Apparent transport rates for suspensoids were similar to those for the July series (Table 6). The ratio of Cr^{51}/Zn^{65} for these samples is shown in Figure 16. This ratio does not decrease with time as might be expected from simple consideration of half-lives (Cr^{51}--27.8 days, Zn^{65}--245 days). The lower apparent transport rate of Zn^{65} combines with its lower decay rate to produce a fairly constant Cr^{51}/Zn^{65} ratio within the river. The marked rise in the Cr/Zn ratio within the estuary (lower 20 miles) is thought to be due to displacement of exchangeable Zn^{65} from suspensoids by Na^{+} and Mg^{++} in sea water.

Table 7. Radioactivity on Filters

Location	Miles from river mouth	Miles from reactors	Particulate picocuries/liter Cr^{51}	Zn^{65}	Cr/Zn
Hood River	130	200	127.5	6.1	20.9
Above B. Dam	112	218	89.7	11.6	7.73
Below B. Dam	110	220	97.4	12.5	7.79
Rooster Rock	95	235	102.4	12.1	8.45
Gov't Island	85	245	89.6	11.6	7.74
Int'st Br.	76	254	83.6	9.6	8.71
Hayden Island	75	255	107.6	12.9	8.33
Duplicate	75	255	107.9	12.8	8.34
Campbell Lake	65	265	68.1	7.7	8.85
Walker Island	51	279	40.5	4.4	9.20
Puget Island	38	292	167.6	13.5	12.4
Harrington Pt.	21	309	90.3	6.0	15.1
Astoria	11	319	78.9	3.1	25.4
Buoy 14	4	326	76.5	2.0	38.2

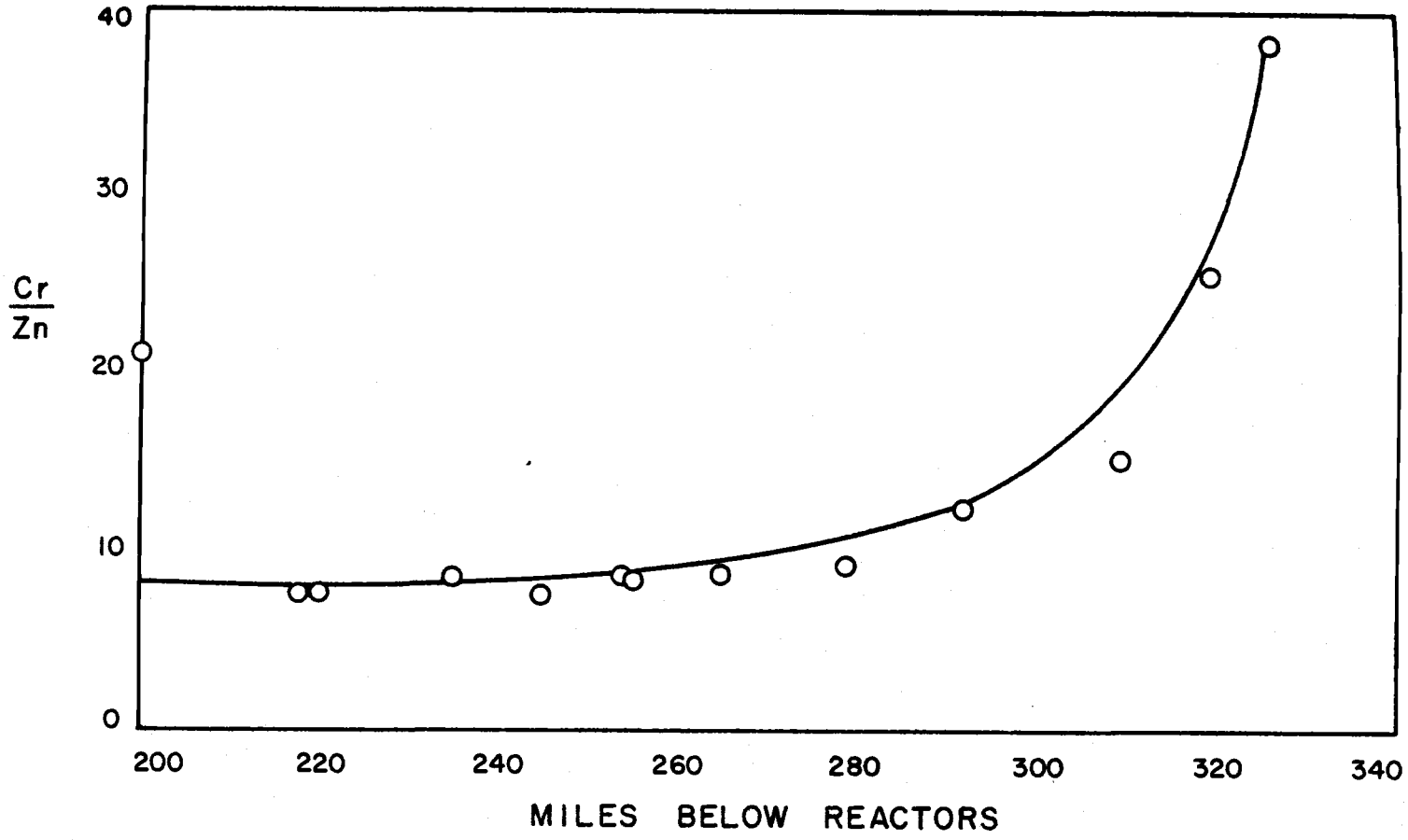

Figure 16. Cr^{51}: Zn^{65} (on filters) versus distance from the nuclear reactors at Hanford. Note change in ratios at estuary (about 300 miles below reactors).

F Radionuclides in Migratory Albacore Tuna

Charles Osterberg and Lauren Larsen

Gamma-ray spectra of livers of migratory albacore tuna (Thunnus alalunga) show that fish arriving early in the summer off Oregon contain manganese-54, potassium-40, and zinc-65. Fish taken three weeks later, farther north, contain much more Zn^{65}, but less Mn^{54} and K^{40}, per unit weight. Increase in Zn^{65} can be attributed to the low level steady state amount of this isotope maintained in the Pacific Ocean by Hanford Laboratory reactors on the Columbia River. Interpretation of the behavior of Mn^{54} and K^{40} is more difficult, however. At no time were levels sufficiently high to be considered a health hazard, and extremely sensitive equipment was required for their measurement.

All fish were taken by lines by the R/V ACONA while at cruising speed (8 to 9 knots) between hydrographic stations off Oregon. Digestive tracts were either frozen or preserved in formalin. In the laboratory, livers were removed, freeze-dried, ashed, placed in 15 cc plastic tubes, and counted in the well of a 5x5-inch NaI(Tl) crystal. Signal from the detector was resolved with an ND-130A 512-channel gamma-ray spectrometer.

Uncertainties exist concerning the migratory pathways followed by albacore. The fish are associated with warmer water, generally appearing quite suddenly in mid-summer about 50 miles off southern Oregon, apparently going north. They persist for a short time, and then disappear suddenly in early fall. Department records for the past several years show the earliest appearance off Oregon of albacore in July, and the latest to be in September. However, our records are sparse since albacore sightings are incidental to the main purpose of our cruises.

Origin of Mn^{54} is likewise uncertain. Although not a fission product, it is found in fallout. Levels of Mn^{54} may be higher in parts of the ocean. The long (291 days) half-life makes it quite possible that areas adjacent to previous nuclear tests may still contain local concentrations of Mn^{54}. Albacore feeding in these areas could accumulate, and perhaps retain, relatively high levels of Mn^{54}.

The spectra (Fig. 17) show that the livers of albacore caught in July contained much more Mn^{54} and K^{40}, but less Zn^{65} than those caught in August. These are composite samples, and all individual samples show this trend. Reversal in Mn^{54} and Zn^{65} abundances occurred in less than a month. September data are only slightly different from those of August.

Loss of Mn^{54} may result from a lower specific activity in coastal waters; i.e., Mn^{54} associated with fallout may undergo isotope dilution if stable manganese should be more abundant in Oregon waters. Therefore, biological turnover would tend to favor the replacement of Mn^{54} with stable manganese, and levels of Mn^{54} would drop accordingly. This theory would seem correct, were it not for a similar decrease in K^{40}. Background levels of K^{40}, the naturally radioactive isotope of

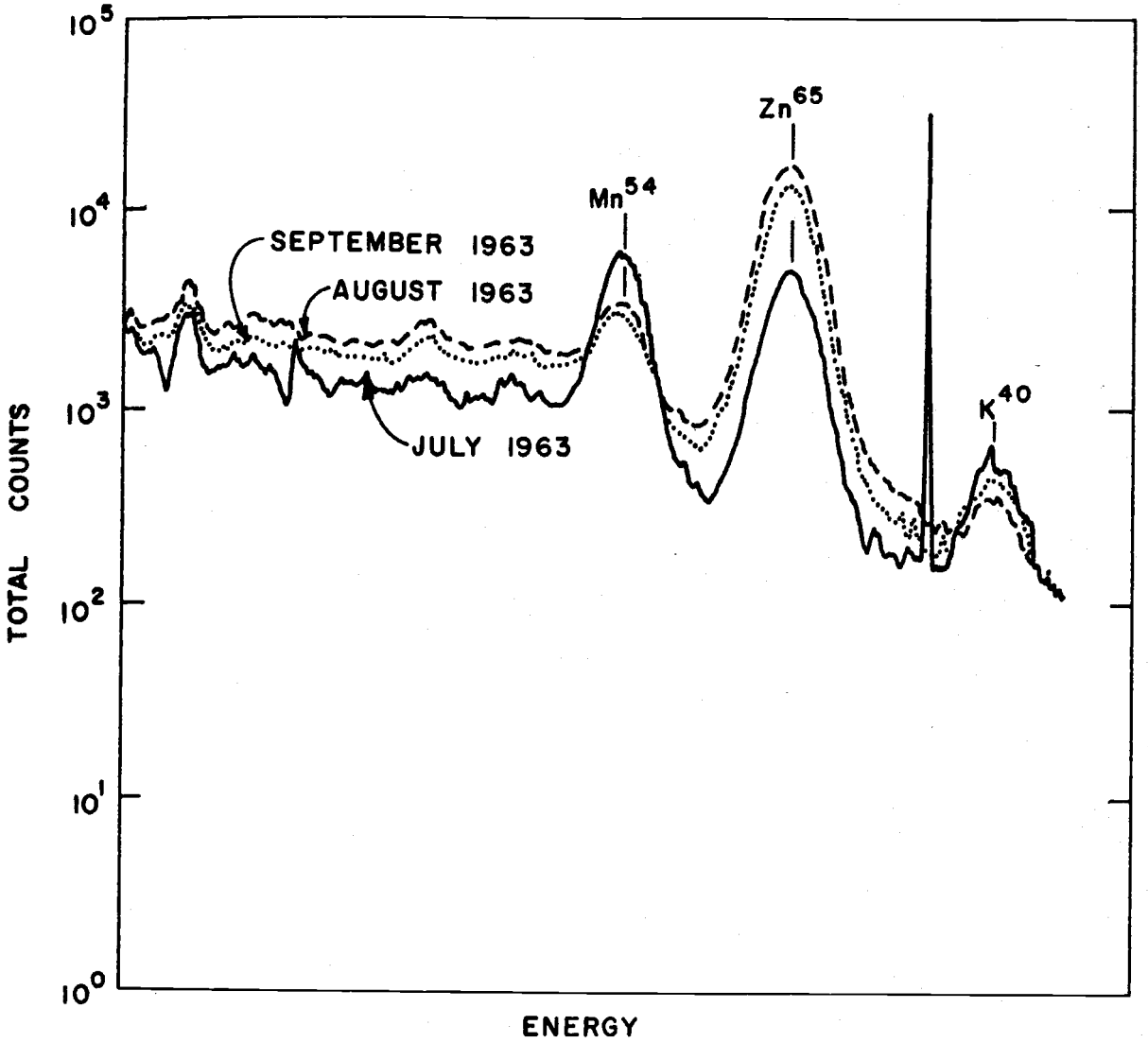

Figure 17. Average spectra of tuna livers for months of July, August and September 1963. Six samples of tuna liver from July were averaged, eight from August, and four from September. All spectra were adjusted for equal dry weights. Albacore tuna were from off Oregon.

potassium, should vary directly with the salinity of the water. While the presence of the freshwater plume of the Columbia River makes Oregon coastal waters less saline during this season, the decrease appears too small to account for this difference.

The plume of the Columbia River does explain the rapid increase in Zn^{65} noted. Macroplankton and micronekton from off central Oregon generally show the highest levels of Zn^{65} in the summertime. Analyses of stomach contents of albacore show that most of the organisms therein concentrate Zn^{65}.

Further studies will be necessary to assess the implications of this year's data.

G Variation in Radioactivity in Sea Cucumbers From the Same Sample

Charles Osterberg and Lois Haertel

A large number of sea cucumbers (Laetmophasma fecumdum Ludwig) were collected from 600 m 34 miles off Newport on 27 April 1963. These were freeze-dried, ground with mortar and pestle, packed into ten 12 cc portions, and each portion analyzed to check for variations in activity from Z^{95}-Nb^{95}.

The impetus from this study came from two sources: (1) the rather surprising evidence that Zr^{95}-Nb^{95} were present in sea cucumbers from 2800 m (see Osterberg, Carey and Curl, 1963), and (2) discovery of relatively large "hot particles" in Columbia River sediments (Cutshall and Osterberg, in press). These observations suggested that perhaps large, "hot" fallout particles settled quickly to the bottom where they were picked up by benthic feeders.

Earlier work (Osterberg, Pearcy, and Curl, 1964) showed that euphausiids in surface waters of the ocean from nine successive tows over a period of eight hours had only small variations in Zr^{95}-Nb^{95} activity (i.e., 13.6 pc/g $\pm$ 1.2)(8.8 percent). This indicated that most of the fallout particles in surface waters were low in activity and that a single particle contributed only a small fraction of the total radioactivity of a sample. The present experiment, therefore, was designed to see if particles in animals from 600 m were appreciably larger (hotter) than those from surface waters.

Results of counts of the ten portions of our sample are shown in Table 8.

Variations in the Zr^{95}-Nb^{95} activity of the ten portions are quite small. We must therefore conclude that fallout radioactivity at 600 m is not caused by a few larger, hotter than average particles. Instead,

Table 8. Sea Cucumbers (600 m)

	Weight (grams)	Net counts (100 min) K^{40}	Net counts (100 min) Zr^{95}-Nb^{95}	Counts/g K^{40}	Counts/g Zr^{95}-Nb^{95}
1.	8.73	2,113	41,013	242	4,698
2.	8.54	2,117	43,228	248	5,062
3.	8.55	2,194	43,686	257	5,109
4.	9.84	2,324	48,154	236	4,894
5.	9.64	2,251	42,964	234	4,457
6.	9.33	2,116	44,144	227	4,731
7.	9.71	2,321	48,449	239	4,990
8.	10.05	2,127	47,612	212	4,738
9.	9.94	2,179	49,830	219	5,013
10.	9.91	2,128	46,085	215	4,650
				x = 233	= 4,834
				σ = 13.8 (5.9%)	σ = 205 (4.2%)

the particles are not appreciably different from those in euphausiids from surface waters. This seems to add credence to the hypothesis (Osterberg, Carey and Curl, 1963) that herbivorous zooplankton in surface waters are responsible for the fallout radioactivity present in deep benthic animals. Herbivorous zooplankton pick up the radionuclides while feeding and release them as fecal pellets which rapidly sink to the bottom.

H An Observation of the Effect of Upwelling on the Zn^{65} Content on Mussels

Charles Osterberg

The mussel Mytilus californianus is found on rocky shores that are open to direct assault from the ocean. Unlike the oyster, it does not grow in sheltered bays or in areas of low salinity. For this reason, Folsom considers mussels to be excellent monitors of radioactivity in oceanic waters.

We collected M. californianus for neutron-activation studies in the fall of 1963 at the stations indicated on the map (Fig. 18). We were curious whether levels of stable zinc, cobalt, etc., would be the same in mussels from all four locations. The samples were therefore taken off the rocks, placed in plastic freezer bags, and frozen with dry ice. After return to the laboratory, they were thawed and then dissected

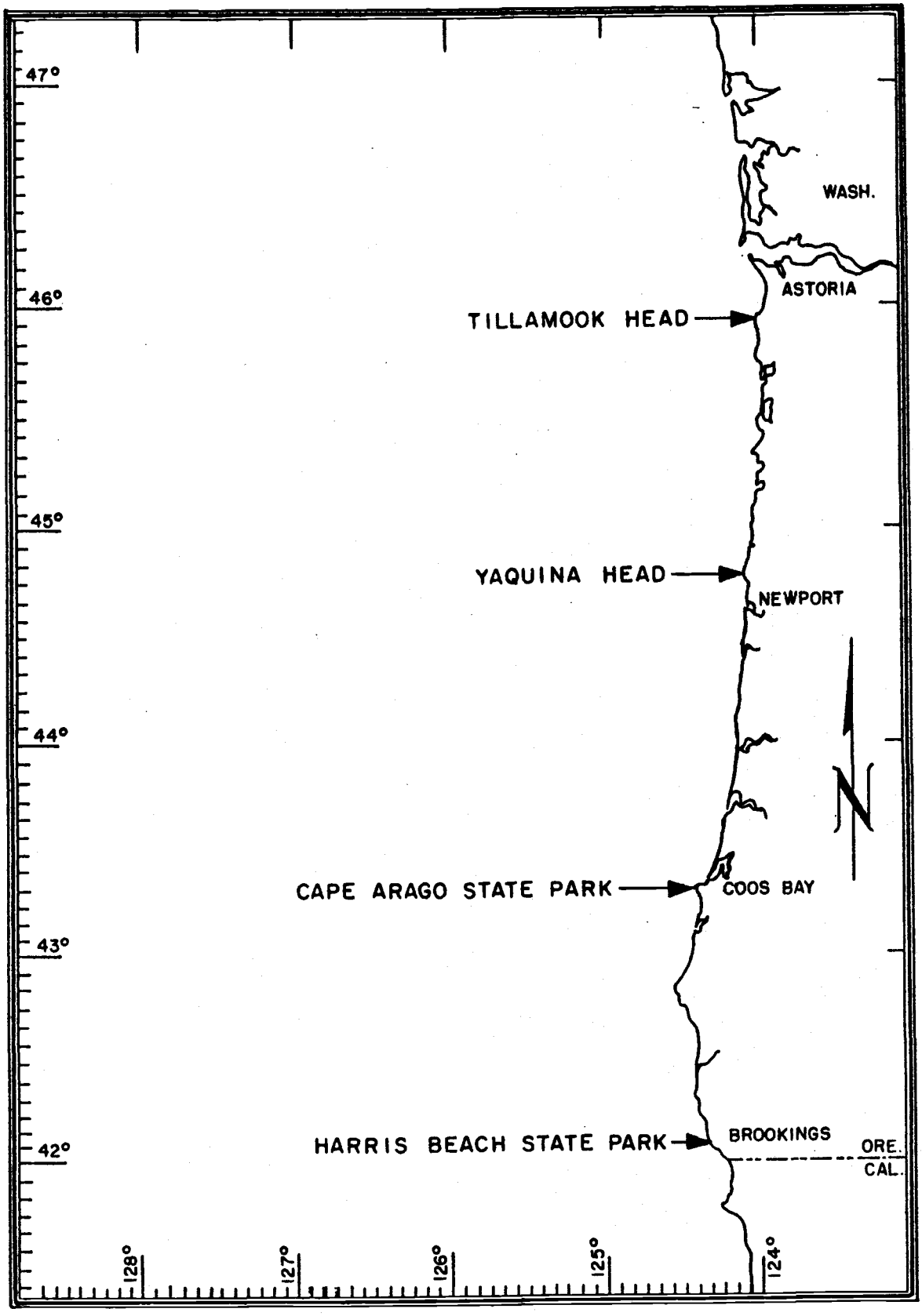

Figure 18. Location of mussel sampling sites. September 1963.

with a plastic knife. Processing was in accordance with the techniques listed under Methods, Neutron-activation.

When the initial spectra (before neutron-activation) were made, it was readily apparent (Fig. 19) that Zn^{65} was greatest at Tillamook Head, nearest the Columbia River, but fell off gradually with distance. There was a sharp drop in Zn^{65} content off Brookings, Oregon, despite the fact that the increased distance (compared with Coos Bay) from the mouth of the Columbia was relatively small.

Physical measurements of salinities at sea demonstrated that the plume of the Columbia River was held well offshore from Brookings in late summer. Zinc-65 associated with the plume was evidently likewise unable to reach shore, as reflected in the Zn^{65} content of the mussels.

Although the plume of the river normally stays offshore (due to prevailing winds and Coriolis effects (see Figs. 2-6 in Osterberg, Pattullo, Pearcy 1964), late summer upwelling off southern Oregon helps isolate it from coastal waters. Salinity profiles for 1961 (Fig. 2, ibid) and 1962 (Fig. 6, ibid) show that upwelling is a seasonal phenomenon. Zinc-65 data (Fig. 6, ibid) likewise show that euphausiids in the upwelled water contained much less radioactivity than those from further offshore in the plume .

Our mussel data therefore confirm physical measurements of the plume and illustrate the role of upwelling in isolating the coastal region from effects of plume waters. The spectrum of the mussels from off Tillamook Head also indicates the presence of Cr^{51}. This represents, more or less, the greatest distance from the Columbia River that we can measure Cr^{51} in marine organisms.

I Trace Element Analysis

Charles Osterberg and Jerry Wiese

Most of the radionuclides present in the Columbia River as a result of the reactors at Hanford Laboratories are neutron-induced. The two reactions of importance to our studies are:

$$(1)\quad {}_{30}Zn^{64} + {}_{0}n^{1} \longrightarrow {}_{30}Zn^{65} + \gamma$$

$$(2)\quad {}_{24}Cr^{50} + {}_{0}n^{1} \longrightarrow {}_{24}Cr^{51} + \gamma$$

Stable Zn^{64} has a neutron cross section of 0.5 barns, and makes up 48.89 percent of the zinc found in nature. Zinc is ubiquitous in the environment, and presumably, much of the zinc which is activated at Hanford enters the reactors in coolant water. The balance might come from corrosion products, i.e., from metallic parts of the reactor.

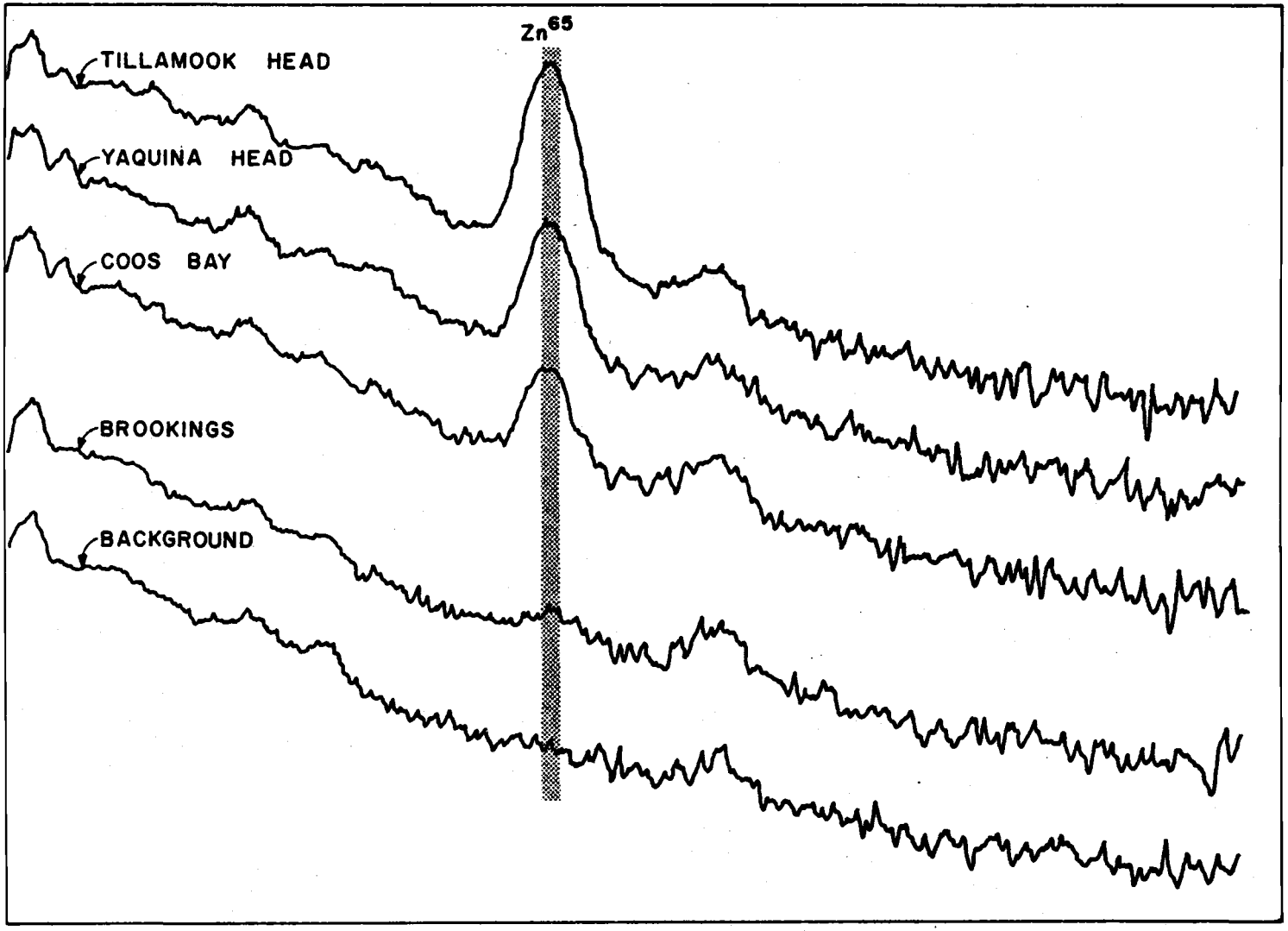

Figure 19. Comparison of radioactivity in mussel samples from Tillamook Head, Yaquina Head, Coos Bay, and Brookings, Oregon. Note gradual decrease in Zn^{65} peak with distance from the Columbia River. Sudden drop at Brookings is attributed to upwelling, which forces Columbia River water well offshore.

Chromium-51 has a thermal neutron cross section of 16.0 barns but makes up only 4.31 percent of natural stable chromium. Most of the chromium activated originates from sodium dichromate, added to the reactor coolant water to inhibit corrosion.

The half-lives of Zn^{65} and Cr^{51} (245 d and 27.8 d, respectively) favor their persistence, and they are readily measurable in certain phases of the environment at the mouth of the river. Their loss with passage down the river seems more related to their different chemical behavior than to their half-lives (see section D).

Since the Zn^{65} and Cr^{51} that we measure in organisms near the mouth of the river originate from neutron-activation, it was decided to duplicate the process in our laboratory. The reasoning was that organisms with an affinity for Zn^{65} would have a similar affinity for stable zinc. Therefore, if the organisms were irradiated in a reactor, a great enhancement in the Zn^{65} peak should occur. The increase in the area under the photopeak would be a function of neutron flux, and under controlled conditions could be used to measure stable zinc.

A very large number of radioisotopes, due to neutron activation, appear in the reactor effluent. Many have short half-lives and disappear from the river quite rapidly. We therefore delayed analyzing our irradiated plankton sample until several months had passed to also eliminate such complicating short half-lived gamma emitters from the spectrum.

Problems are bound to occur since the relative abundance of trace elements in marine plankton is different from that of coolant water in the Hanford reactors. Since some trace elements have tremendous nuclear cross sections, the possibility of obtaining a complex spectrum is real. Not all nuclear reactions are simple n, γ reactions, so that some chemical separations may be required to interpret results. Nevertheless, the technique seems ideal for our study since our objectives are limited. That is to say: (1) Our primary interest is in radioisotopes which originate from their stable counterparts by neutron-activation. (2) We are only interested in those with sufficient half-lives to survive the lengthy trip to sea. (3) Errors in measurement are inherently small. The neutron-activation data on stable zinc, for example, are obtained from the same sample using the same gamma-ray spectrometer and identical conditions to those used to obtain data for Zn^{65}. Therefore, since specific activity is expressed as a ratio, most errors are eliminated (e.g., those errors that appear as a factor in both numerator and denominator).

Methods

Samples of marine organisms, fish, clams, sediments, etc., are:

(1) collected with extreme care to prevent contact with metals,

(2) frozen in sealed polyethylene freezer bags, *

(3) dried (or freeze-dried) in glass trays,

(4) ground with mortar and pestle,

(5) packed into preweighed 5-dram polyethylene tubes (Olympic plastics),

(6) reweighed,

(7) sealed with hot soldering iron and immersed in hot water to check for leaks,

(8) placed in well of 5x5-inch NaI(Tl) crystal and counted for 100 minutes using ND130-AT gamma-ray spectrometer,

(9) washed in acetone,

(10) 14 samples are placed in a plastic baby bottle (Evenflo),

(11) baby bottle with enclosed samples is irradiated for 3-4 hours (swimming pool reactor, Washington State University),

(12) samples are stored at reactor facility allowing several months for nuclear decay,

(13) step 8 is repeated for the irradiated sample, and

(14) the two spectra are compared for differences due to neutron activation.

Results from a sample of marine plankton (Euphausia pacifica) are seen in Figure 20. The bottom trace (Fig. 20) is background, which has not been subtracted from either spectrum. Before irradiation (Fig. 20), a peak of Zn^{65} was apparent. After irradiation (Several months decay period elapsed before analysis), the peak due to Zn^{65} was greatly enhanced. There are also "sum" peaks of Sc^{46} and Co^{60} (Removing the sample from the well and recounting it outside the crystal verifies that both of these peaks are sum peaks.)

Four other peaks appear at the low energy end of the spectrum. By following these for decay, it was apparent that three of the peaks were the 136, 280, and 405 kev peaks of Se^{75}. A plot of the decay of the largest peak (Fig. 21) yields approximately the correct half-life, i.e., 127 d.

*Some of our duplicate samples have been preserved in isopropyl alcohol for comparison. There is indication that fish preserved in alcohol lose both Zn^{65} and Cr^{51} to the solution. This is subject of further investigation.

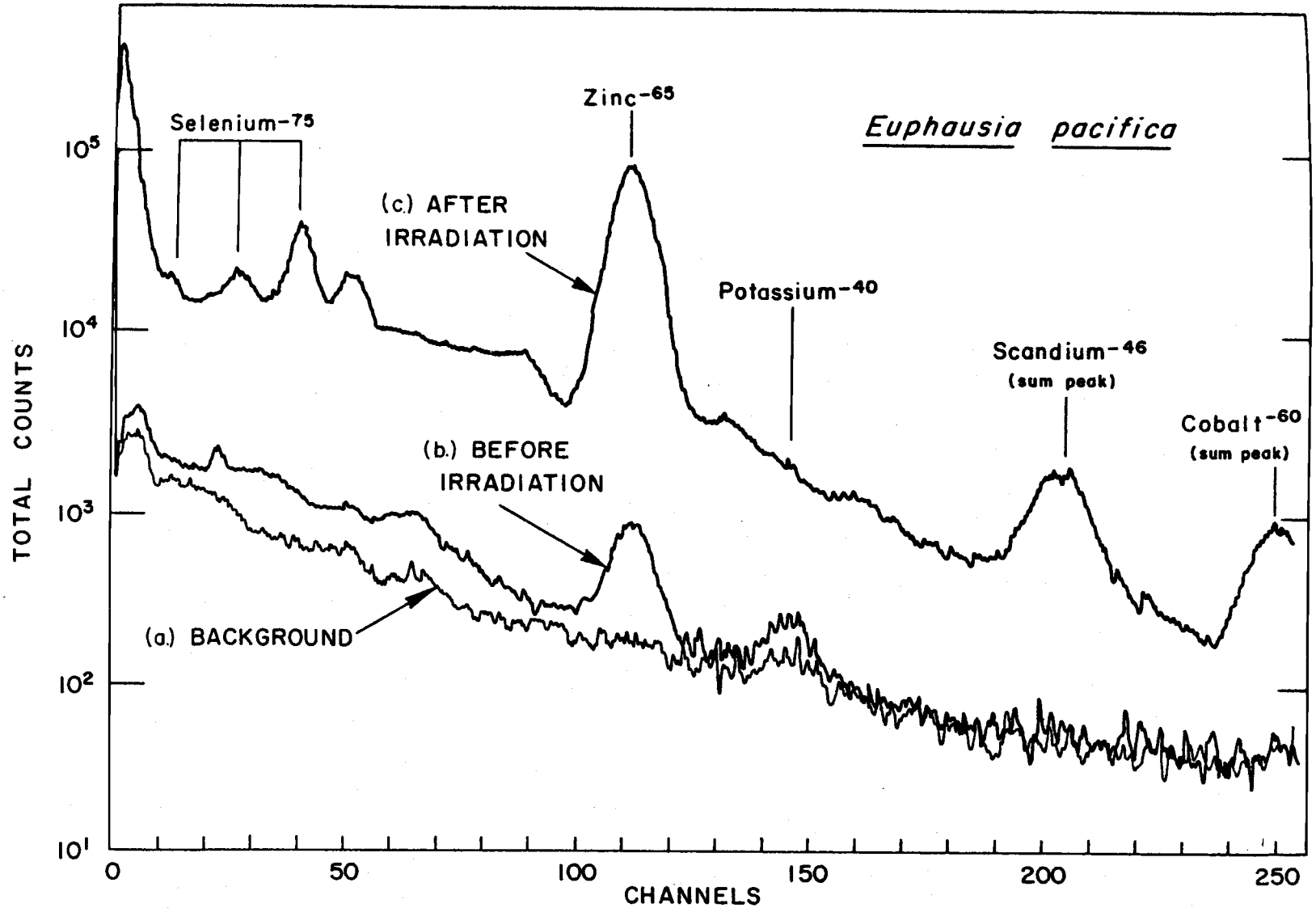

Figure 20. Comparison of plankton sample (Euphausia pacifica) before and after neutron activation. 100 minutes.

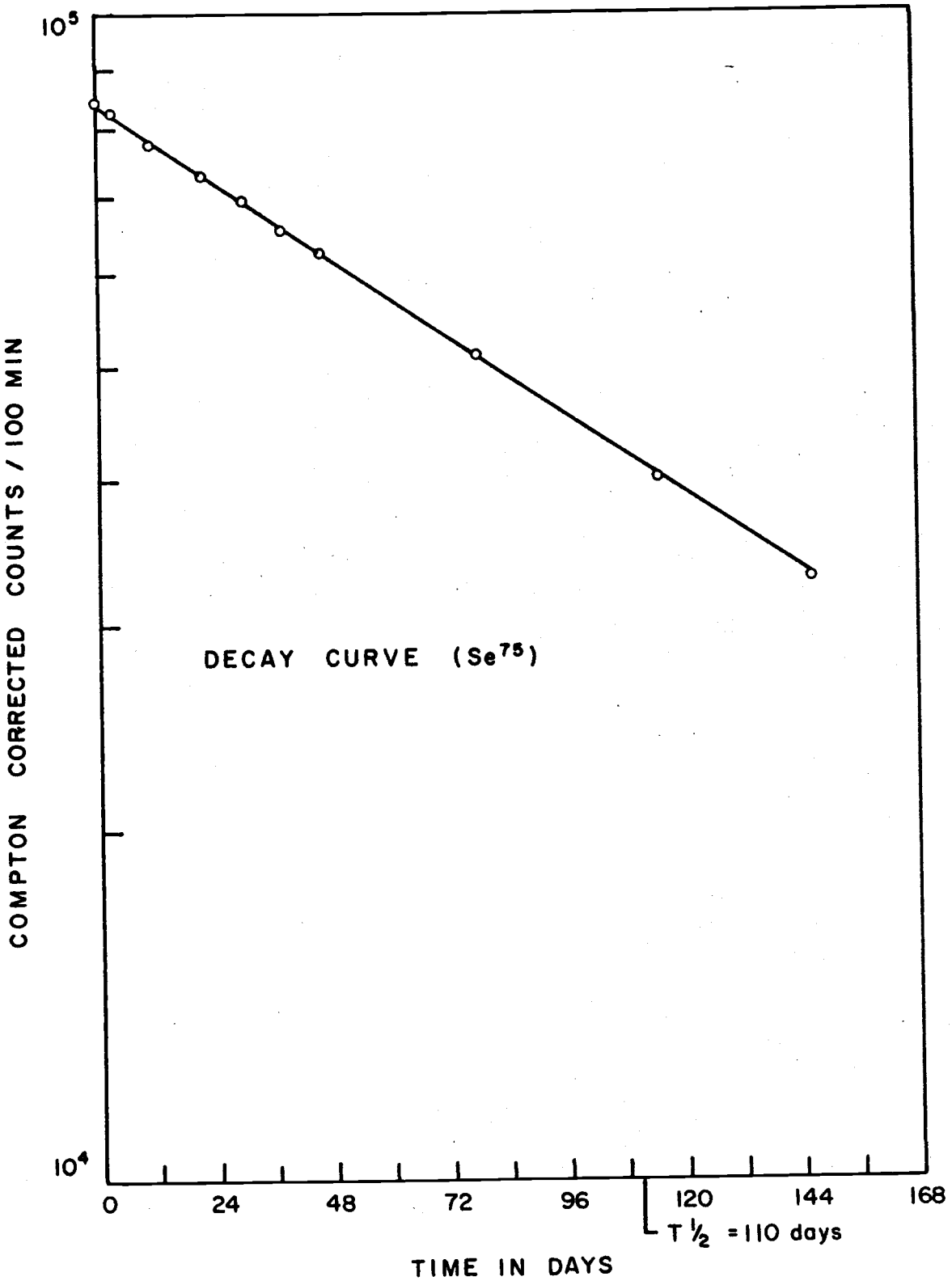

Figure 21. Decay curve of low energy peak in Figure 20. Both decay curve and energies of gamma rays indicate selenium-75.

A source of concern is the origin of Sc^{46}. Yield should be very good by the following reaction since 100 percent of stable scandium is Sc^{45}, and the cross section for thermal neutrons is 12 barns.

$$Sc^{45}\ (n, \gamma) \qquad Sc^{46}$$

however the following reaction is also possible:

$$Ti^{46}\ (n, p) \qquad Sc^{46}$$

Titanium is much more abundant in sea water than scandium, and has been reported in marine organisms, but less information is available on scandium.

Titanium-46 makes up only 7.95 percent of stable titanium, and the yield from the n, p reaction is very low. Nevertheless, since this apparently represents the first observation of scandium and selenium in euphausiids, * we intend to carry out a number of chemical steps to eliminate possible contaminants. Very little faith can be placed on a single observation using a new technique.

Our "wet chemistry" has been limited to date by a lack of facilities. When the radiochemical laboratory in the new Oceanography Building is completed, capabilities will be greatly increased.

The neutron-activation program is, however, seriously limited by the amount of spectrometer time required to identify various gamma emitters. This is particularly true if the gamma-emmitters turn out to be from previously unreported trace elements, such as scandium and selenium. Spectrometer time required is actually not excessive, but our single spectrometer is already operated 24 hours per day on radioanalysis of various marine samples, under contracted AEC research.

A large number (56) of various samples were irradiated at the Washington State reactor facility on 15 November 1963. Total dose level at 12 inches on 18 November 1963 was 100 mr/hr. These samples (Table 9) will be shipped to us after sufficient cooling time has elapsed to eliminate the need for excessively heavy lead shipping containers.

At the time we first contacted WSU with regard to irradiating our samples, we also negotiated with Hanford Laboratories. The much greater heat generated by the higher neutron flux in the Hanford reactors demanded special protection for the sample sent there for irradiation. Therefore, they were ashed and sealed in quartz prior to irradiation. No information regarding exposure time, neutron flux, etc., was available from Hanford because of security restrictions.

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*According to Vinogradov, selenium has been reported in Globigerina and Radiolaria silts (p. 175).

Table 9. Activated Samples Awaiting Sufficient Decay for Radioanalysis.

Sample	Name	Part	Date Collected	Location	Process
MY 1	Mytilus californianus	muscle	9/20/63	Cape Arago (Oreg.)	freeze dried
MY 2	Mytilus californianus	soft part	9/20/63	Cape Arago (Oreg.)	freeze dried
MY 3	Mytilus californianus	soft part	9/20/63	Cape Arago (Oreg.)	freeze dried
MY 4	Mytilus californianus	soft part	9/20/63	Cape Arago (Oreg.)	freeze dried
MY 5	Mytilus californianus	soft part	9/19/63	Tillamook Head (Oreg.)	freeze dried
MY 6	Mytilus californianus	soft part	9/19/63	Tillamook Head (Oreg.)	freeze dried
MY 7	Mytilus californianus	soft part	9/19/63	Tillamook Head (Oreg.)	freeze dried
MY 8	Mytilus californianus	muscle	9/19/63	Tillamook Head (Oreg.)	freeze dried
MY 9	Mytilus californianus	soft part	9/20/63	Brookings (Oreg.)	freeze dried
MY 10	Mytilus californianus	soft part	9/20/63	Brookings (Oreg.)	freeze dried
MY 11	Mytilus californianus	soft part	9/20/63	Brookings (Oreg.)	freeze dried
MY 12	Mytilus californianus	muscle	9/20/63	Brookings (Oreg.)	freeze dried
MY 13	Mytilus californianus	soft part	9/18/63	Yaquina Head (Oreg.)	freeze dried
MY 14	Mytilus californianus	soft part	9/18/63	Yaquina Head (Oreg.)	freeze dried
MY 15	Mytilus californianus	soft part	9/18/63	Yaquina Head (Oreg.)	freeze dried
MY 16	Mytilus californianus	muscle	9/18/63	Yaquina Head (Oreg.)	freeze dried
NH 1(NA)	Cardium nuttalli	whole minus shell	9/7/63	Yaquina Bay (Oreg.)	freeze dried
NH 2(NA)	Schizothaerus nuttalli	whole minus shell	9/7/63	Yaquina Bay (Oreg.)	freeze dried
L 28	Usnea florida	whole	8/14/63	Corvallis (Oreg.)	dried
2NH1	Euphausia pacifica	whole	10/2/63	NH-25 (Newport Harbor)	freeze dried
2NH2	Euphausia pacifica	whole	10/2/63	NH-25 (Newport Harbor)	freeze dried
ES 121	Cottus asper	cleaned fish	10/7/63	Harrington Point (Columbia River)	freeze dried ashed
ES 122	Spirinchus dilatus	cleaned fish	10/7/63	Astoria (Columbia River)	freeze dried ashed
ES 123	Spirinchus dilatus	gills and organs	10/7/63	Astoria	freeze dried ashed
ES 124	Cymatogaster aggregata	whole fish	10/7/63	Harrington Point	freeze dried ashed
ES 125	Cottus asper	gills and organs	10/7/63	Harrington Point	freeze dried ashed
ES 126	Leptocottus armatus	gills and organs	10/7/63	Harrington Point	freeze dried ashed
ES 127	Leptocottus armatus	cleaned fish	10/7/63	Harrington Point	freeze dried ashed
ES 128	Mylocheilus caurinus	whole fish	10/7/63	Harrington Point	freeze dried ashed
ES 129	Catostomus macrocheilus	gills and organs	10/7/63	Harrington Point	freeze dried ashed
ES 130	Catostomus macrocheilus	cleaned fish	10/7/63	Harrington Point	freeze dried ashed

Table 9. Activated Samples Awaiting Sufficient Decay for Radioanalysis (Continued)

ES 132	Platichthys stellatus	whole fish	10/7/63	Harrington Point	freeze dried ashed
ES 133	Platichthys stellatus	gills and organs	10/7/63	Chinook Point (Columbia River)	freeze dried ashed
ES 134	Platichthys stellatus	cleaned fish	10/7/63	Chinook Point	freeze dried ashed
ES 135	Psettichthys melanostictus	whole fish	10/7/63	Chinook Point	freeze dried ashed
ES 136	Leptocottus armatus	gills and organs	10/7/63	Astoria	freeze dried ashed
ES 137	Leptocottus armatus	cleaned fish	10/7/63	Astoria	freeze dried ashed
ES 138	Crangon franciscorum	whole	10/7/63	Harrington Point	freeze dried ashed
ES 140	Leptocottus armatus	fin	10/7/63	Astoria	freeze dried ashed
ES 141	Microgadus proximus	cleaned fish	10/7/63	Astoria	freeze dried ashed
ES 142	Microgadus proximus	gills and organs	10/7/63	Astoria	freeze dried ashed
ES 143	Cancer magister	gills and organs	10/7/63	Chinook Point	freeze dried ashed
ES 144	Crangon franciscorum	whole	10/7/63	Astoria	freeze dried shed
ES 145	Cancer magister	muscle and shell	10/7/63	Chinook Point	freeze dried ashed
ES 146	Cancer magister	shell	10/7/63	Chinook Point	dried
ES 147	Lumpenus sagitta	cleaned fish	10/7/63	Astoria	dried
ES 148	Lumpenus sagitta	gills and organs	10/7/63	Astoria	dried
ES 149	Platichthys stellatus	cleaned fish	10/7/63	Astoria	dried
ES 150	Platichthys stellatus	gills and organs	10/7/63	Astoria	dried ashed
ES 151	Platichthys stellatus	cleaned fish isopropyl	10/7/63	Astoria	dried ashed

The total flux was apparently great, and our two small plankton samples were sent to us in a 250 pound shipping container and were much too "hot" to count even after six months. A one-minute count of the smaller sample (0.045g), still encapsulated in quartz, shows the presence of zinc, cobalt, etc. (Fig.22). Ashing would be expected to volatilize the selenium (the chemical behavior of selenium should be similar to that of sulphur), and this element was not observed in the spectrum of the Hanford sample. Sc^{46} was still present; however, scandium is known to be a ubiquitous contaminant.

J. Lichens as Collectors of Fallout Radioactivity

Lauren Larsen and Charles Osterberg

The ability of lichens to absorb large quantities of moisture from the atmosphere enables them to obtain vital nutrients needed for growth. Moisture, in the form of dew, mist, fog, and principally rainfall, contains dissolved nutrients. When the elements have been absorbed and concentrated within the thallus, lichens have no means of excreting them (1). Radioactive debris from nuclear testing may also be included with the nutrients. Condensation of water vapor into droplets within the atmosphere is facilitated by the presence of dust, combustion particles, salt, nuclear debris, and other impurities. As the raindrops fall, they may accumulate impurities or coalesce with other small droplets. In this manner scavenging takes place, cleansing the atmosphere of such impurities. Libby (2) estimated that the lower 10,000 feet of air is washed clean by precipitation on an average of every three days. Since fallout is deposited largely by precipitation, its geographical distribution is related to rainfall (3).

An initial analysis was made using a gamma-ray spectrometer on a sample of lichen collected 12 June 1963 near Otis Junction, Oregon (Fig. 23). The following radioactive nuclides were present: $Ce^{141-144}$, $Ru^{103-106}$, Zr-Nb^{95}, Cs^{137}, and Mn^{54}. Samples were counted in the well of a 5x5-inch NaI(Tl) crystal. Counting time was 100 minutes.

Ratios of the peak values for Zr-$Nb^{95}/Ce^{141-144}$ for lichens from the Oregon coast plotted against time, yield a line with negative slope (Fig. 24), indicating that the fallout has a minimum age greater than 100 days (Radiological Health Handbook, p. 96). An age dating curve constructed from the apparent half-life of a $Ce^{141-144}$ ratio with Zr^{95}-Nb^{95} (constructed by Norman Cutshall) estimates the fallout to have a minimum age around 180 days. Information obtained from "Radiological Health Data" lists nuclear detonations from previous months, both foreign and domestic. During the months between January

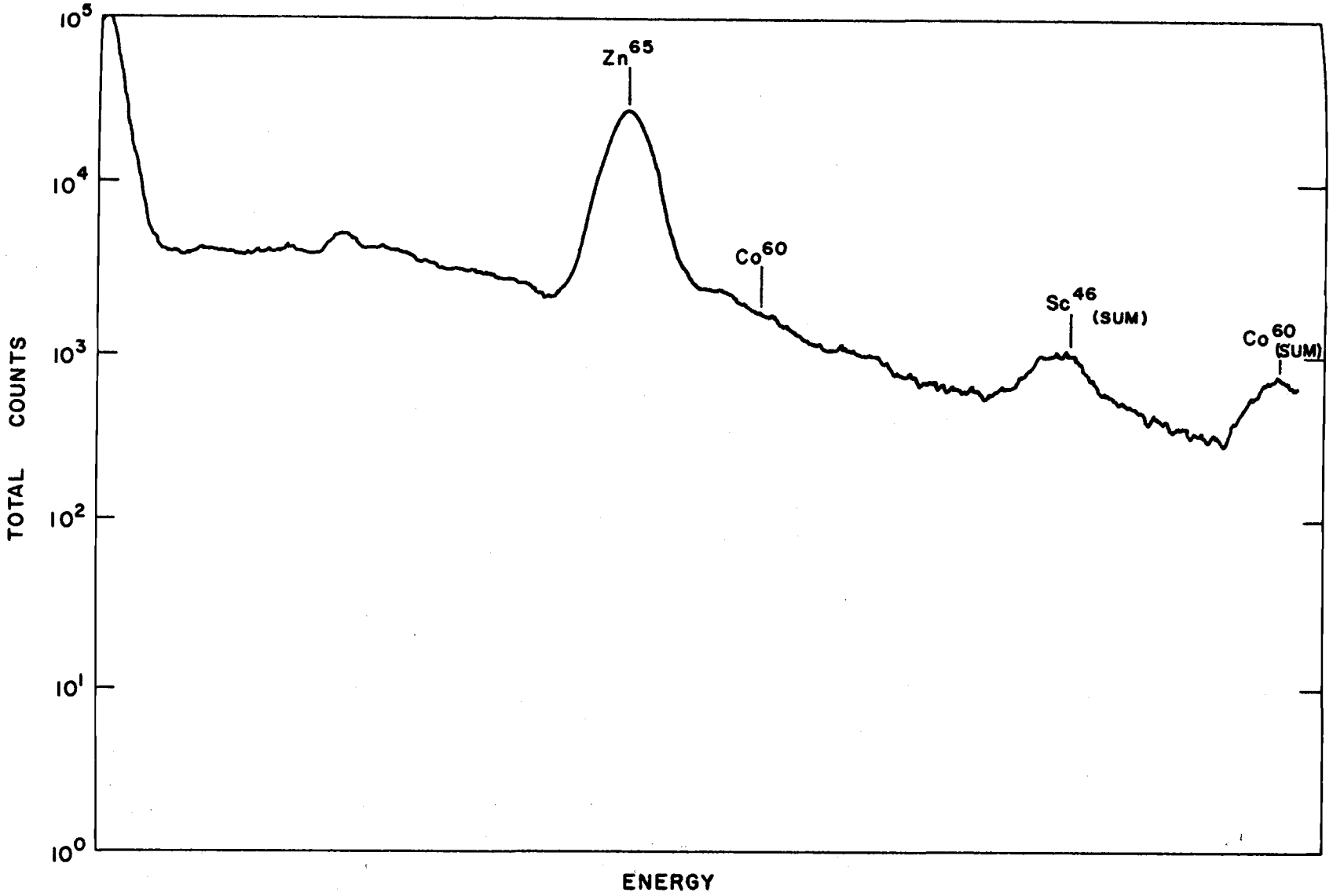

Figure 22. Ashed plankton sample (Euphausia pacifica) after irradiation. 1 minute count. This sample is too active for our equipment.

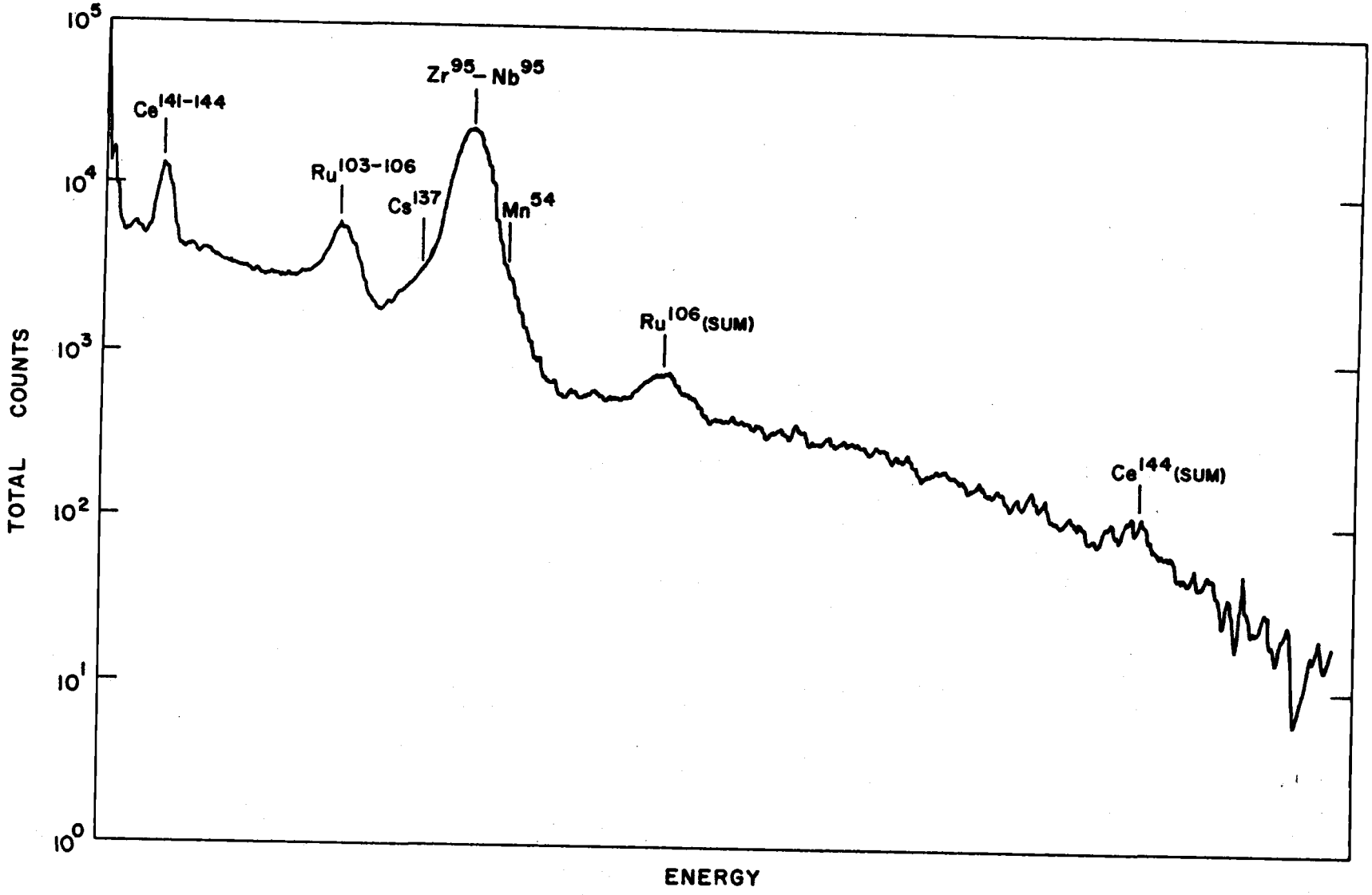

Figure 23. Spectrum of lichen, showing typical fallout peaks. 100 minute count. Later counts (and also chemical separations) verify the presence of Mn^{54} and Cs^{137}.

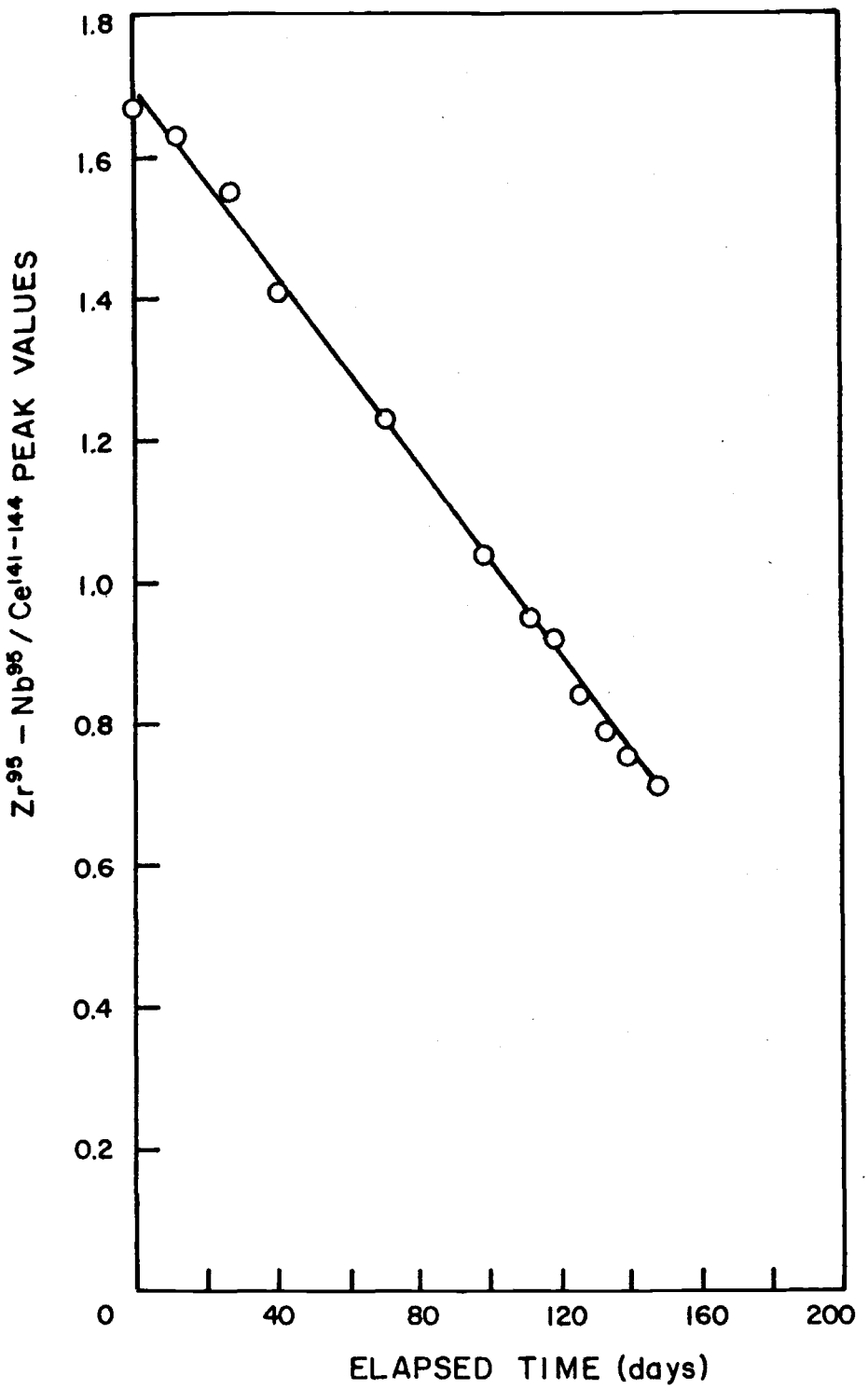

Figure 24. Ratios of Zr^{95}-Nb^{95}:$C^{141-144}$ versus time, to determine age of fallout.

and September 1963, there were approximately 15 Nevada underground tests. The latest confirmed atmospheric tests reported were those conducted by the U. S. S. R. in December, 1962. (The Sahara French test in March 17 was not listed in "Radiological Health Data.") A total of seven such detonations were reported. Atmospheric testing is the prime contributor to nuclear debris in the atmosphere; date of the latest atmospheric test generally agrees with the minimum age of the fallout in the lichen samples.

Fallout, which is related to rainfall, may be high in areas of high precipitation. Lichens, which absorb large quantities of moisture, may therefore show greater radioactivity in areas with greater rainfall. Davis, et. al. (4) show that, although a positive correlation exists between precipitation and deposition of certain fallout radionuclides, this relationship may be altered by other ecological factors. Data on pine needles indicated that radiocesium in the needles generally increased with additional rainfall. However, the increase of Cs^{137} was not in direct proportion to the increase in precipitation. This was attributed in part to washing away of radioactive materials from the surface of the vegetation by heavy rains. Lichens however, may not be as easily washed as pine needles. The thallus of a lichen can "sop" up anywhere from 3 to 35 times its own weight in water (1). Because of this high absorption of water and the matt or net-like growth pattern of lichens, washing may not be accomplished as readily as in other types of foliage, such as pine needles.

The northwestern part of Oregon was divided into 3 general areas of study. These included: 1) the coastal area, 2) the upper Willamette Valley, and 3) the Northern Cascades (Fig. 25). This breakdown was approximately that given by the U. S. Weather Bureau (6), so that rainfall data might also be compared with amounts of radioactivity.

Results are given only for $Ce^{141\text{-}144}$, $Ru^{103\text{-}106}$, and $Zr\text{-}Nb^{95}$, and Compton corrections were made using these nuclides. Amounts given are only approximate because Cs^{137} and Mn^{54} were also present, but IBM data processing was not carried out for these particular nuclides.

Sites of sample collection, species, and data on radioactivity are summarized in Table 10.

Ratios from Table 11, which compare U. florida (from the coast): U. florida (Willamette Valley), show the mean of the coastal lichen to be 2.1 times more radioactive than that from the Willamette Valley. Rainfall (Table 12) for the same areas is 1.4 times greater for the coastal area. Comparing means from the coast and Northern Cascades for U. plicata, we find the coastal mean to be approximately 1.7 times greater. In a similar comparison, rainfall is 1.1 times greater.

Our results, though preliminary, show that high amounts of radioactive fallout are associated with lichens, and that the lichens are good, easily collected monitors of fallout. The correlation of radioactivity with rainfall is poorer than the literature had led us to believe.

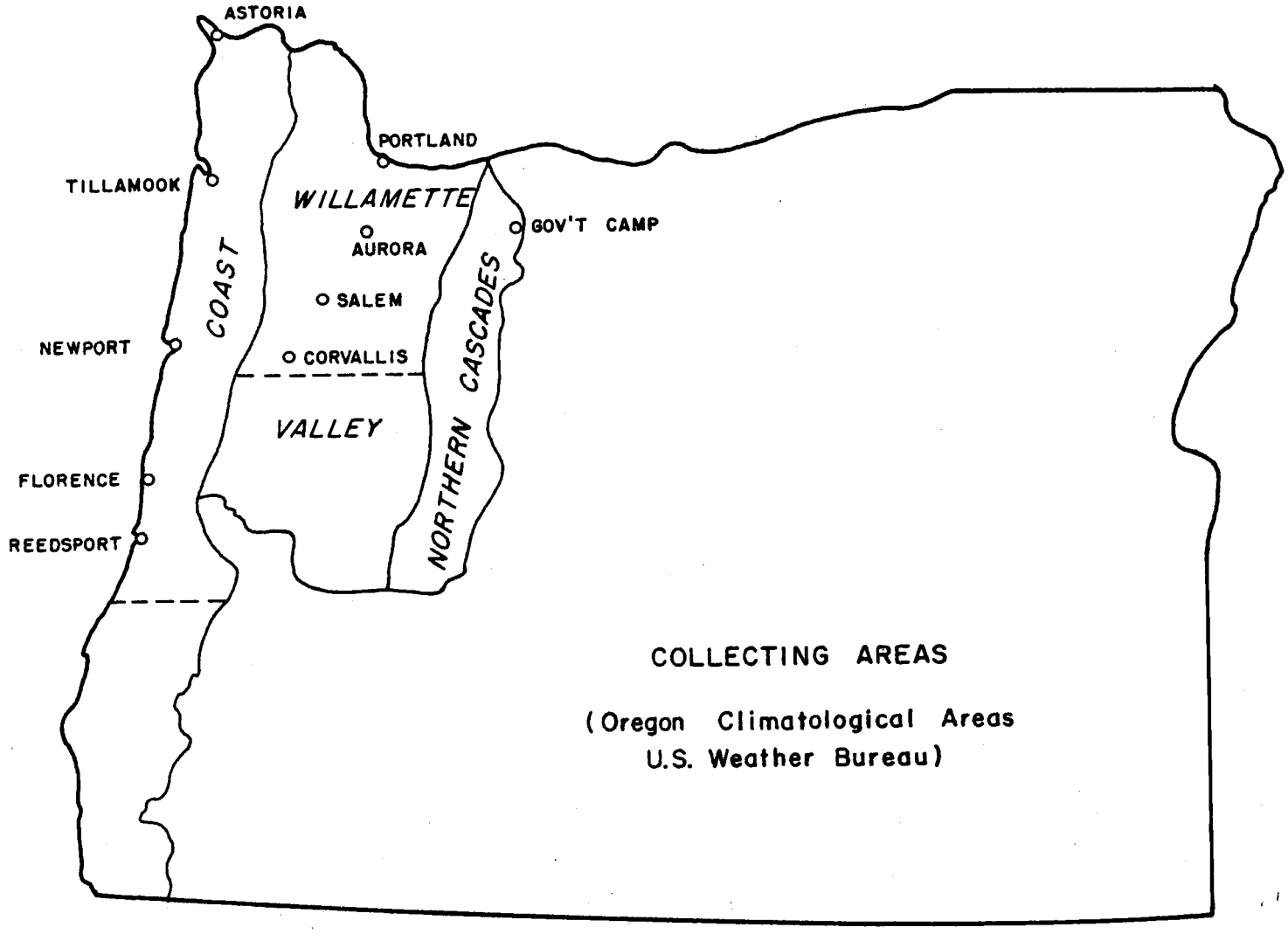

Figure 25. Map showing three climatological regions of Oregon, and areas from which lichen were collected.

Table 10. Fallout Radioactivity in Lichen

	Approximate picocuries per gram dry weight		
	Ce	Ru	Zr-Nb
COASTAL			
Usnea florida			
West Port	109	120	320
Svensen	104	97	540
Fort Clatsop	117	139	359
Garibaldi	223	243	616
Tillamook Park	148	160	399
Otter Crest	112	118	231
Heceta Head	171	182	392
Talkenitch Lake	194	167	394
Umpqua Head	145	108	252
Remote	250	190	329
Mean	157	152	383
Standard Dev.	48.5	42.7	112.7
Usnea plicata			
Fort Stevens	73	86	228
Mary's Peak	88	86	149
Beach Side Park	64	94	299
Mean	75	89	225
Standard Dev.	9.9	3.8	61.3

	Approximate picocuries per gram dry weight		
	Ce	Ru	Zr-Nb
WILLAMETTE VALLEY			
Usnea florida			
Silverton	45	55	116
Aurora	61	76	188
Rainier	50	68	150
Helmick Park	40	52	127
Hwy. 99W & 233	76	92	245
Estacadia	111	116	259
Mean	64	77	181
Standard Dev.	24.1	22.1	55.3
Alectoria oregana			
	330	368	1122
Usnea plicata			
	143	157	281

	Approximate picocuries per gram dry weight		
	Ce	Ru	Zr-Nb
NORTHERN CASCADES			
Usnea plicata			
Breitenbush	69	51	88
Timberline Lodge	20	31	57
Wemme	100	96	185
Santiam Pass	36	52	74
Larch Mountain	70	73	147
Mt. Jefferson Park	35	77	82
Mean	55	63	106
Standard Dev.	27.2	21.1	45.2
Breitenbush:			
A. oregana	83	108	262
U. plicata	69	51	88
Timberline Lodge:			
A. oregana	54	86	203
U. plicata	20	31	57
Mt. Jefferson Park:			
A. oregana	29	86	174
U. plicata	35	77	82

Table 11. Comparison of Means Taken From Table 10.

		Ce	Ru	Zr-Nb	Sum of Mean
Usnea florida	Coast / W. Valley	$\frac{157}{64} = 2.4$	$\frac{152}{77} = 2.0$	$\frac{383}{181} = 2.1$	$\frac{692}{322} = 2.1$
Usnea plicata	Coast / N. Cascades	$\frac{75}{55} = 1.4$	$\frac{89}{63} = 1.4$	$\frac{225}{106} = 2.1$	$\frac{389}{241} = 1.7$

Table 12. Monthly Precipitation (inches) for Given Locality. Data From U.S. Weather Bureau Climatological Data by Sections, Oregon

	Coast	Willamette Valley	Northern Cascades
January	4.35	2.57	3.09
February	8.95	6.02	11.15
March	8.91	7.11	7.88
April	10.00	5.79	6.74
May	5.85	4.24	5.27
June	2.25	2.24	3.68
TOTAL	40.31	27.97	37.81

Ratio for given areas

$\frac{\text{Coast}}{\text{W. Valley}} = \frac{40.3}{28.0} = 1.4$

$\frac{\text{Coast}}{\text{N. Cascades}} = \frac{40.3}{37.8} = 1.1$

We thank Dr. Theodore F. Folsom, Scripps Institution of Oceanography, who first pointed out to us the unique properties of lichens. We acknowledge the assistance of members of the Botany Department, Oregon State University, in identifying our samples.

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2. Libby, W. F. Radioactive Fallout Particularly from the Russian October Series. Proceedings of the National Academy of Science. 45(7): 959-976 (1959).

3. Cited by J. J. Davis, the Effects of Environmental Factors upon the Accumulation of Radioisotopes by Ecological Systems. Proceedings of the Second Annual Texas Conference on the Utilization of Atomic Energy. November 12-13, 1959.

4. Davis, J. J., Hanson, W. C., and Watson, D. G. Some Effects of Environmental Factors upon Accumulation of Worldwide Fallout in Natural Populations. Radioecology, Edited by Schultz, Vincent, and Klement, Alfred W. Jr., Division of Biology and Medicine, U.S. Atomic Energy Commission. Reinhold Publishing Corporation and the American Institute of Biological Science. 1963.

5. Radiological Health Data, Dec, 1962 to Sept. 1963.

6. U. S. Dept. of Commerce, Weather Bureau, Climatological Data, Oregon. Volumn 69, 1963.

7. Radiological Health Handbook. U.S. Department of Health, Education, and Welfare. Public Health Service. Bureau of State Services. Division of Radiological Health, Washington 25, D.C. September, 1960.

K. Geochemical Studies

Vern Johnson, Norm Cutshall, and Charles Osterberg

Abstract

Water, filter and sediment samples were collected at five sites along the Columbia River ranging from 60 to 303 miles from Hanford.

Water filter samples indicate that Zn^{65} is predominantly associated with particulate material throughout the river while Cr^{51} is in solution.

A particle size versus activity study shows that Zn^{65}, Cr^{51}, Co^{60}, and Sc^{46} show a general increase in activity with a decrease in particle size while Zr^{95} exhibits a random distribution.

Leaching experiments reveal that approximately 20-25 percent of the Zn^{65} present on sediments examined is in a readily exchangeable form, with lesser amounts of Mn^{54} and Co^{60}. Cr^{51} and Sc^{46} appear to be in an entirely fixed form.

There is good evidence that exchangeable gamma emitters associated with suspended material are displaced by sea water as the suspensoids contact saline waters of the estuary.

Introduction and Purpose

From the work of Osterberg et. al. (1963) on gamma emitters in marine sediments near the mouth of the Columbia River, interest arose as to how the radionuclides might be transported to the site of marine deposition and how they were associated with the sediment. This led us to begin an investigation of the mechanisms by which radionuclides are removed and held by sediment and suspended material in the Columbia River.

Experimental Work

Water, filter, and sediment samples were collected from July 22 to July 26, 1963, at five sample sites ranging from 60 to 303 miles from the Hanford activities (Fig. 26).

Radioanalysis of the water and filter samples from the five stations showed that Zn^{65} was predominantly associated with particulate material while Cr^{51} was not.

Results from a study of the distribution of the various gamma emitters in sediments (Fig. 27) indicate a general increase in activity with a decrease in particle size for Zn^{65}, Cr^{51}, Co^{60}, and Sc^{46}, but Zr^{95} exhibits a random distribution.

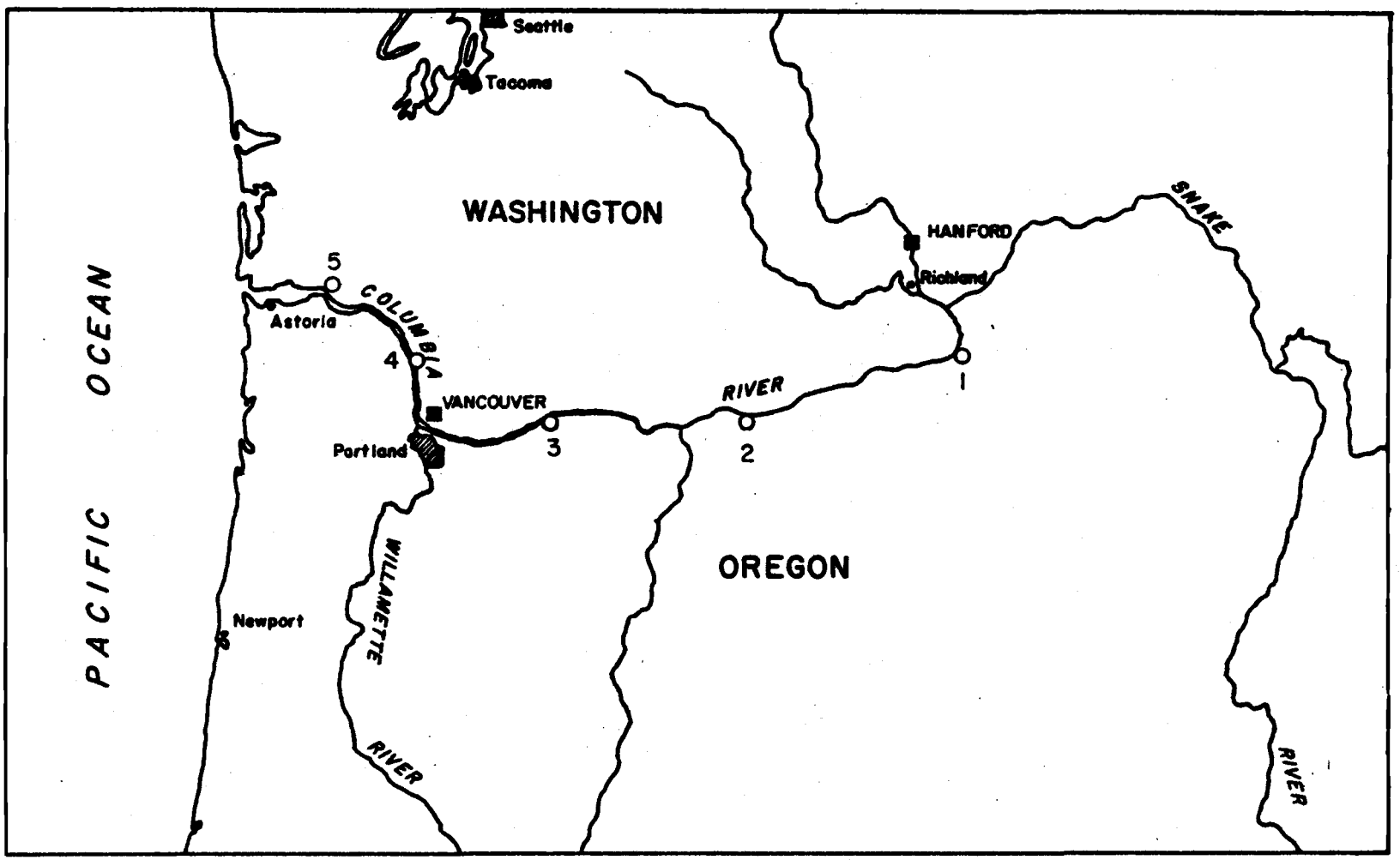

Figure 26. Columbia River sediment collecting sites, July 1963.

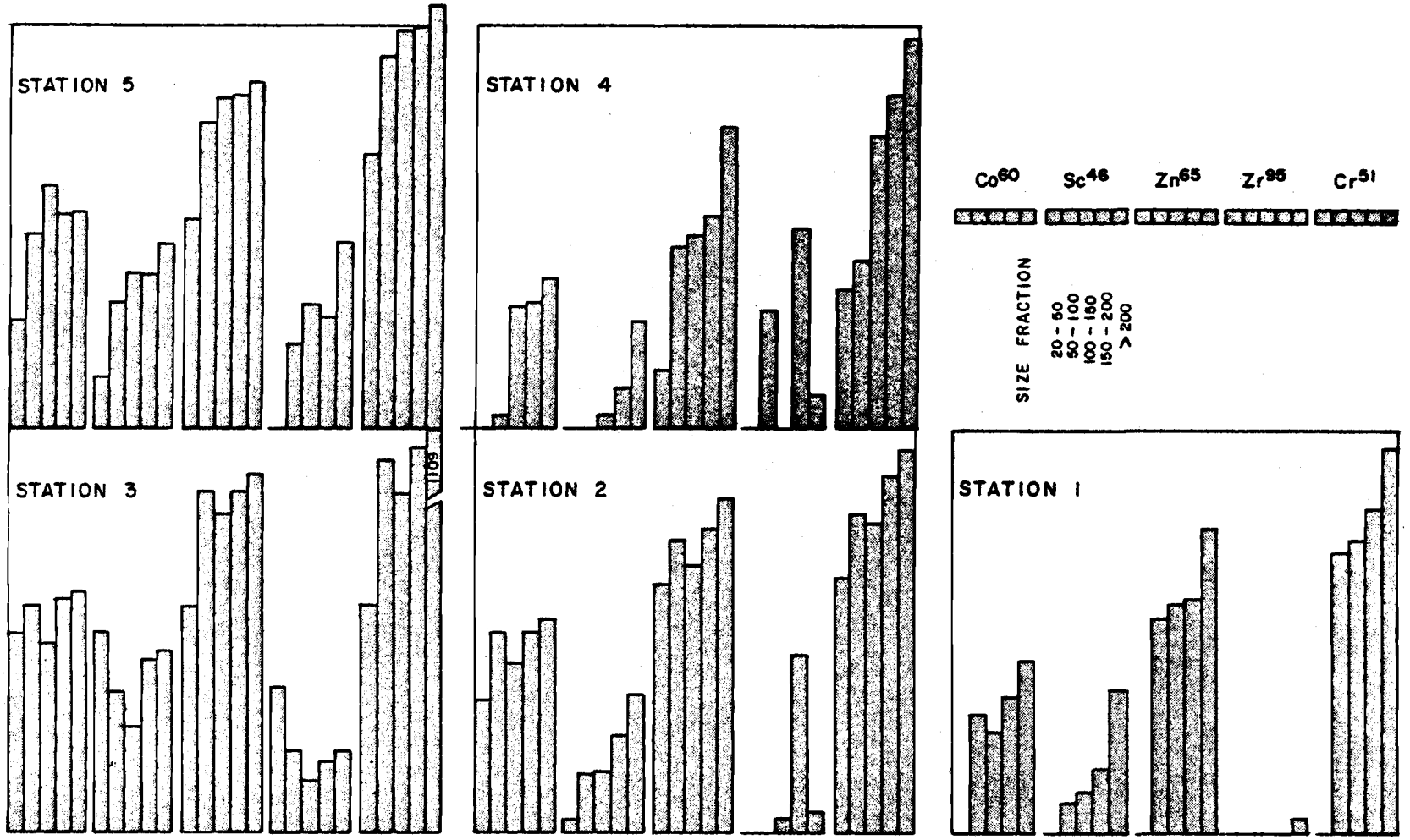

Figure 27. Sediment radioactivity versus particle size distribution. Key in upper right hand corner.

Leaching experiments were begun to determine directly the exchangeable gamma emitters present on sediment. A vacuum system (Fig. 28), which allows a continuous flow of leachate for radioanalysis, was used for this purpose. (A combination of three-way stop cocks and a common ballast tank permit one receiving tube to be emptied while the other fills.) This arrangement made it possible to estimate relative rates of removal of various radionuclides, in addition to total amounts that could be displaced with leaching solutions.

Four leaching solutions were used (Table 13). Gamma-ray spectra for acetic acid, ammonium acetate, and standard sea water leachates indicated that some Mn^{54}, Zn^{65}, and Co^{60} were present in exchangeable form. Figure 29 illustrates a typical gamma-ray spectrum for these leaching solutions. The EDTA leachate revealed, in addition to the above radionuclides, a small peak due to Sc^{46} (Fig. 30).

Table 13. Percent Zn^{65} Removed with Various Leaching Solutions

Solution	Volume	Sediment sample	Percent displaced
Neutral normal amonium acetate	250 ml	10 g 200-, Puget Is. (Sta. 5)	15.8
.5 N acetic acid	250	" "	24.6
Standard sea water	250	" "	31.3
5 percent EDTA @ pH 7	180	" "	33.1
.5 N HAc	250	10 g 200-, Blalock (Sta. 2)	28.4

Leaching curves for acetic acid, standard sea water, and EDTA (Fig. 31-34) represent, in each case, the activity removed from 10 g of air-dried 200 minus Puget Island sediment, with successive aliquots of leaching solution.

Leaching curves for EDTA (Fig. 31) indicate that displacement is extremely rapid, with the greatest portion of the displacement being completed in the first 40 seconds, with the first 5 ml of solution.

In contrast, acetic acid leaching curves (Fig. 32) illustrate that displacement with this reagent is a much slower process.

Standard sea water also demonstrates rather rapid displacement relative to acetic acid. This can be explained in terms of the concentration of the respective displacing cations, and the law of mass action (Sea water is over two orders of magnitude greater in concentration of Na^+ than the concentration of H_3O^+ in .5 acetic acid (3.1×10^{-3}M).

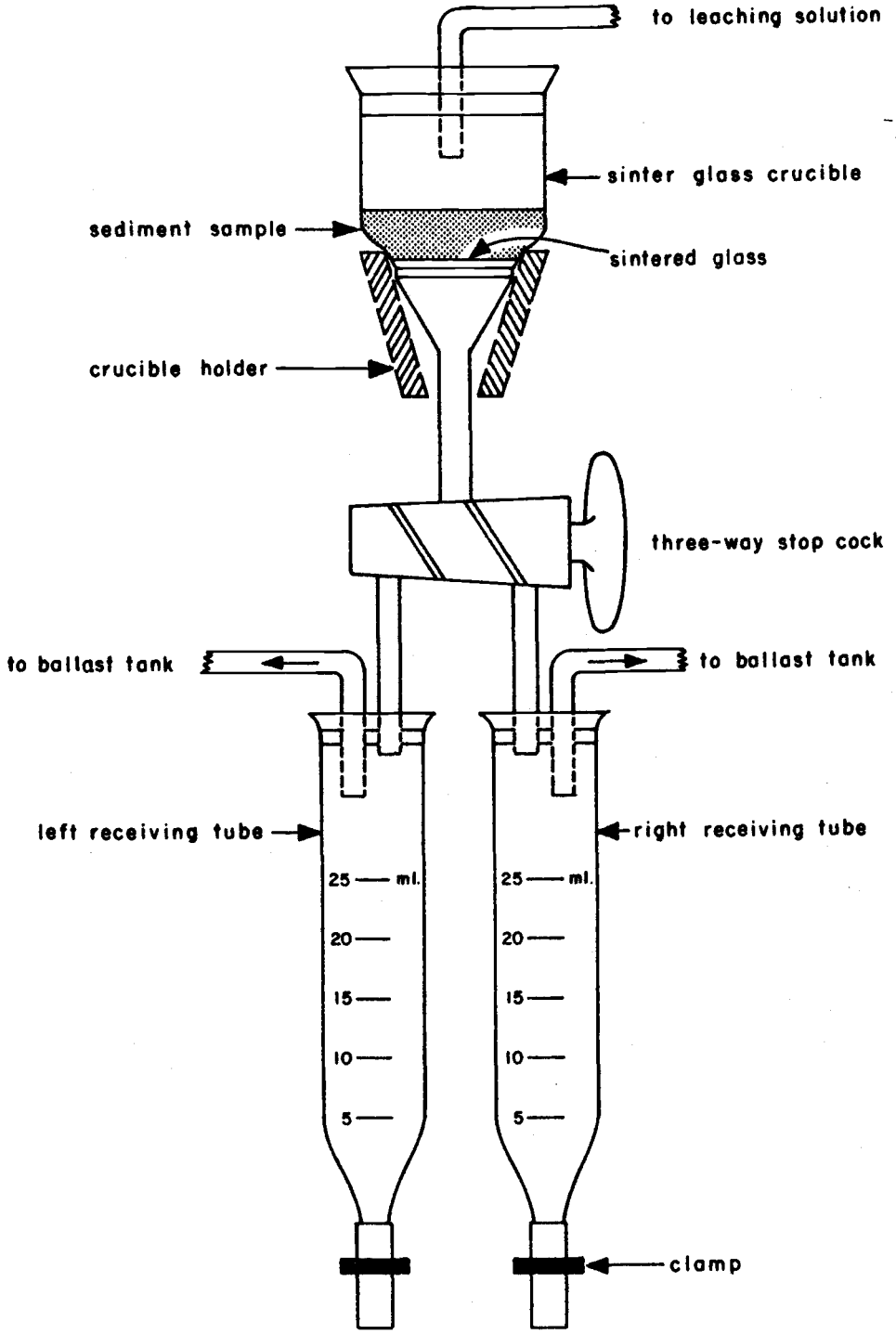

Figure 28. Device used to trap successive portions of leaching solutions after solutions had passed through sediments. (This device was designed and built by Johnson and Cutshall).

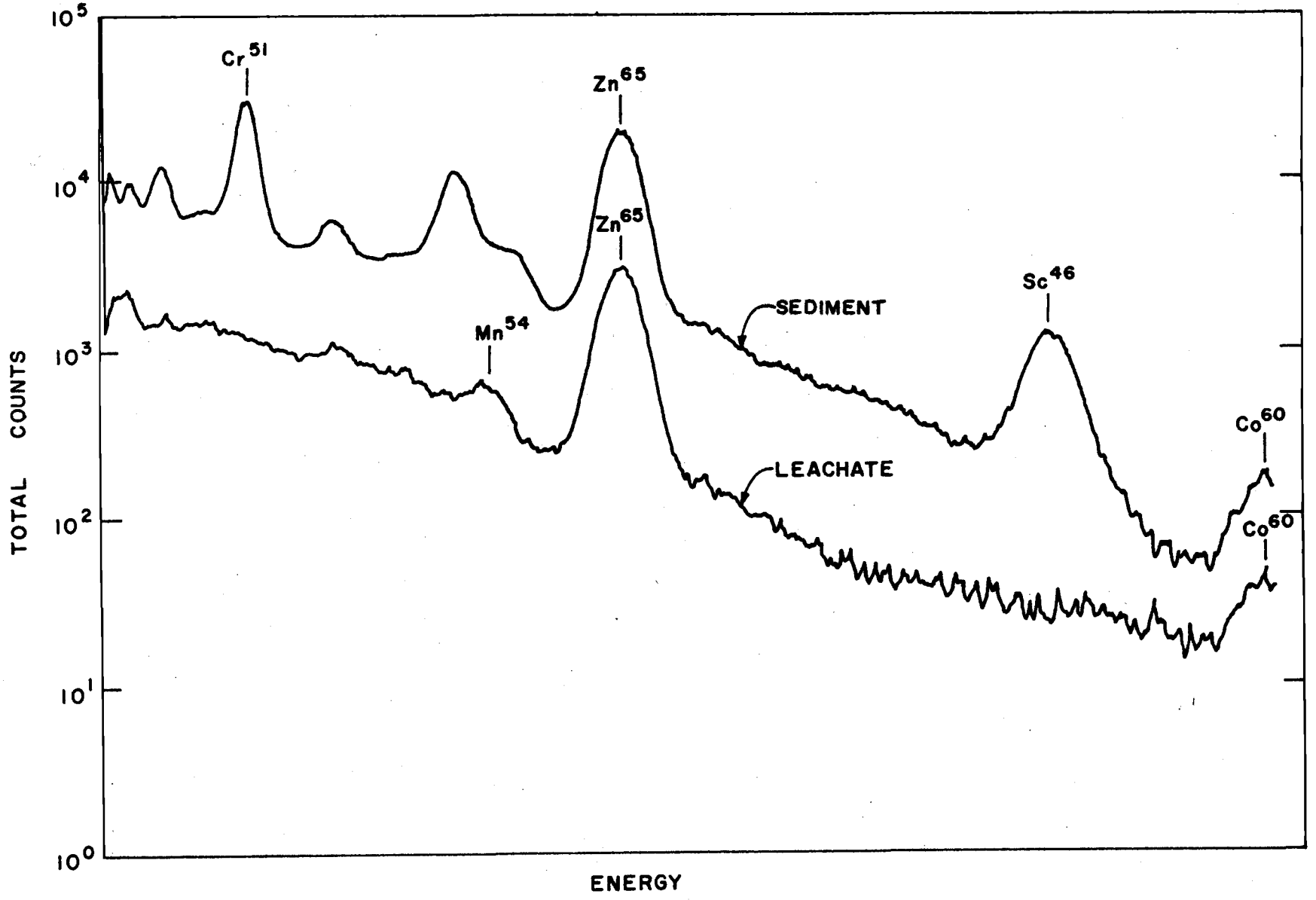

Figure 29. Gamma-ray spectra of Columbia River sediment and

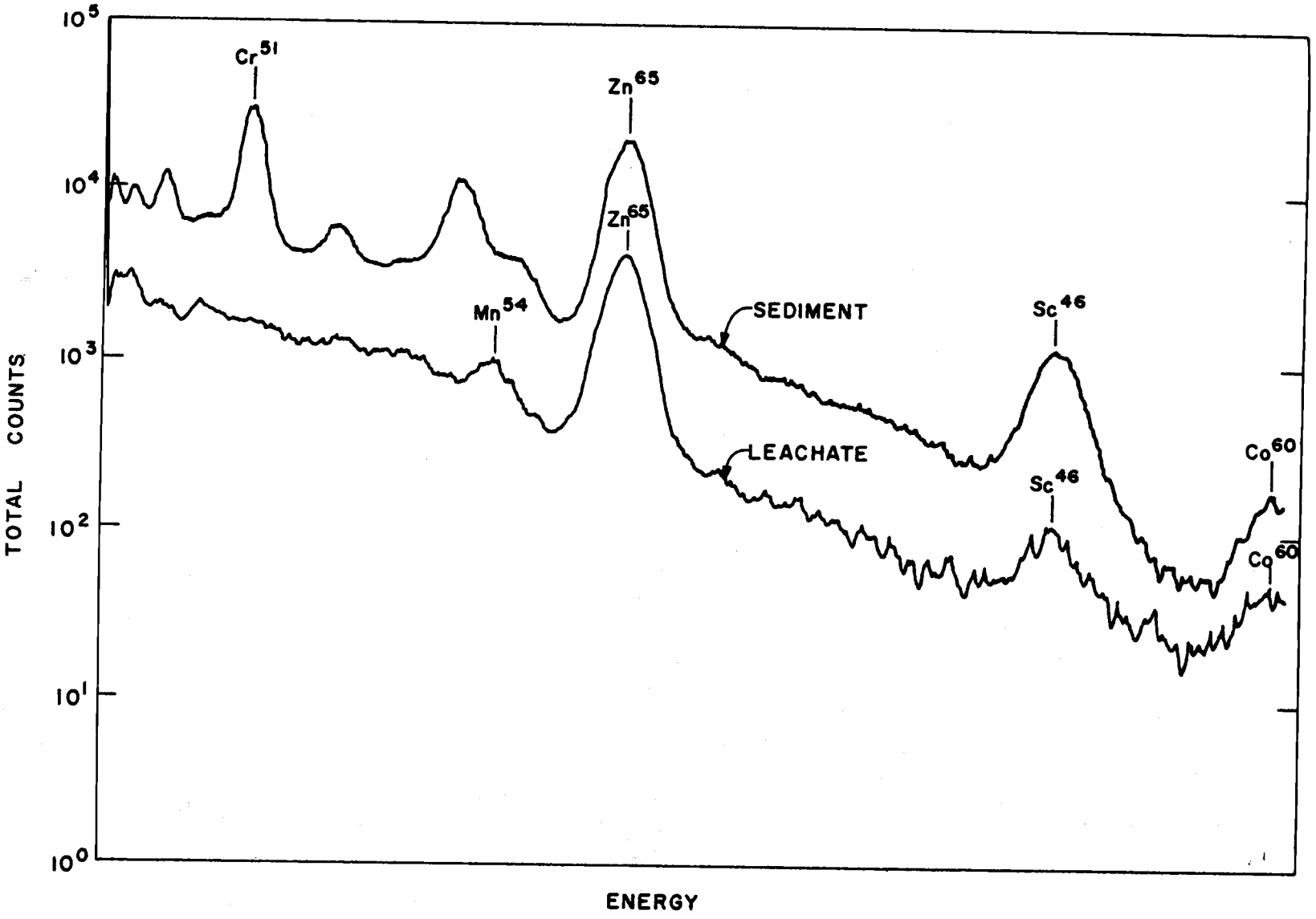

Figure 30. Gamma-ray spectra of Columbia River sediment and EDTA leachate. Some Zn^{65}, Co^{60}, and Mn^{54} are removed. Note also that some Sc^{46} appears in the leachate.

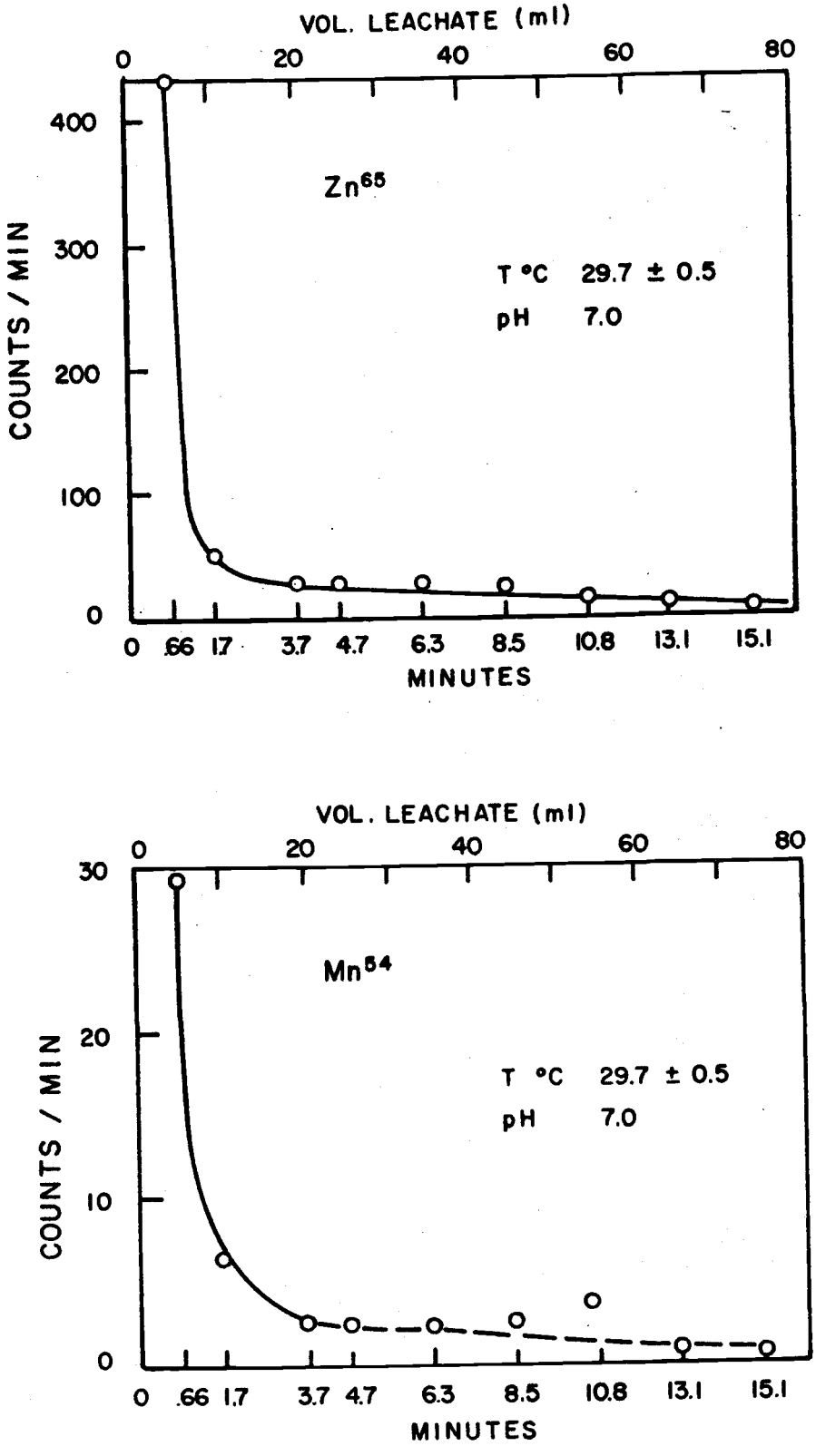

Figure 31. Counts due to Zn^{65} and Mn^{54} in EDTA leachate. Note that most of the radioactivity is quickly removed by the first 10 ml of leachate.

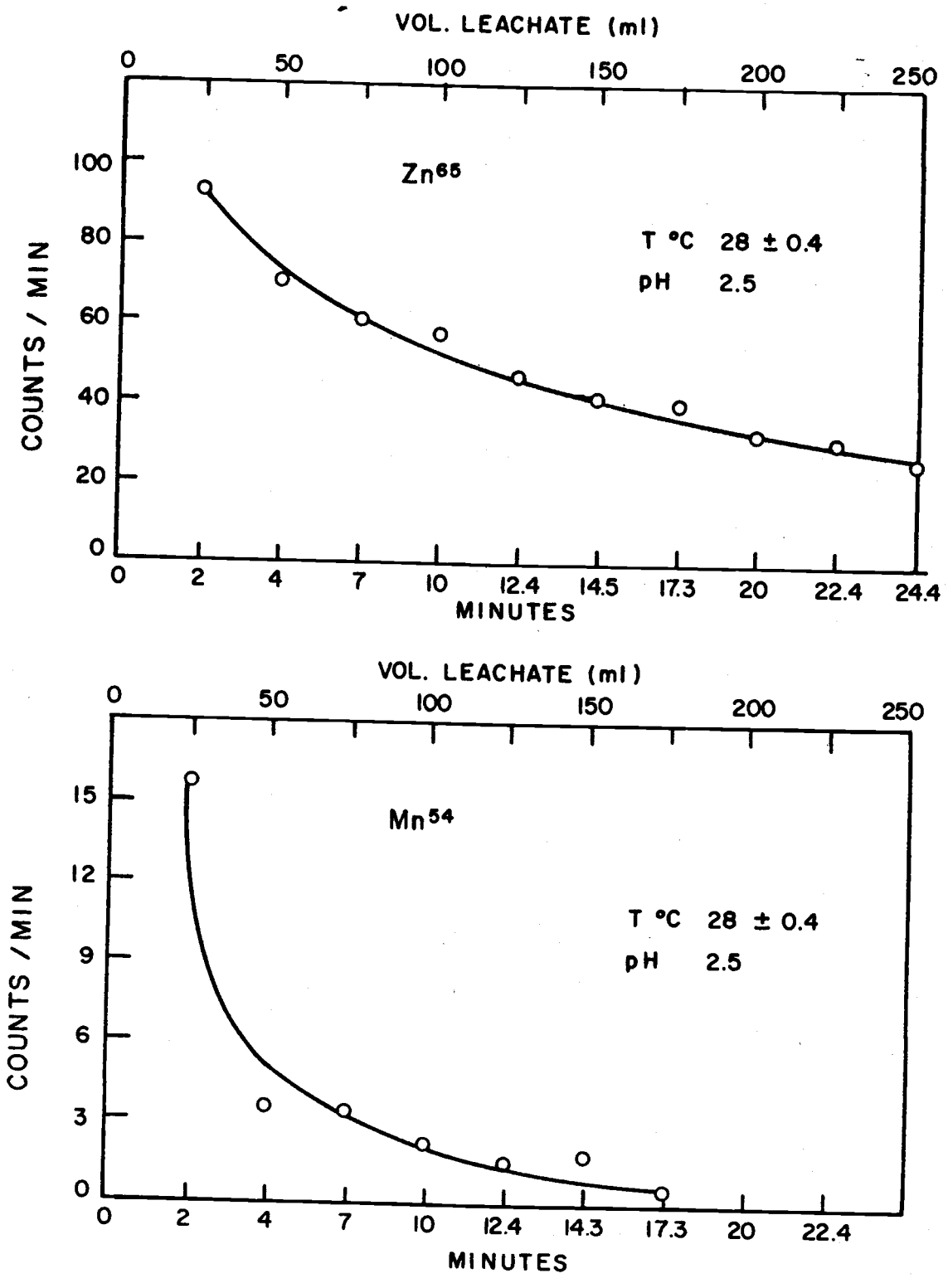

Figure 32. Counts due to Zn^{65} and Mn^{54} in 0.5N acetic acid leachate. Note gradual removal rate of Zn^{65}, but rapid removal for Mn^{54}. Station 2.

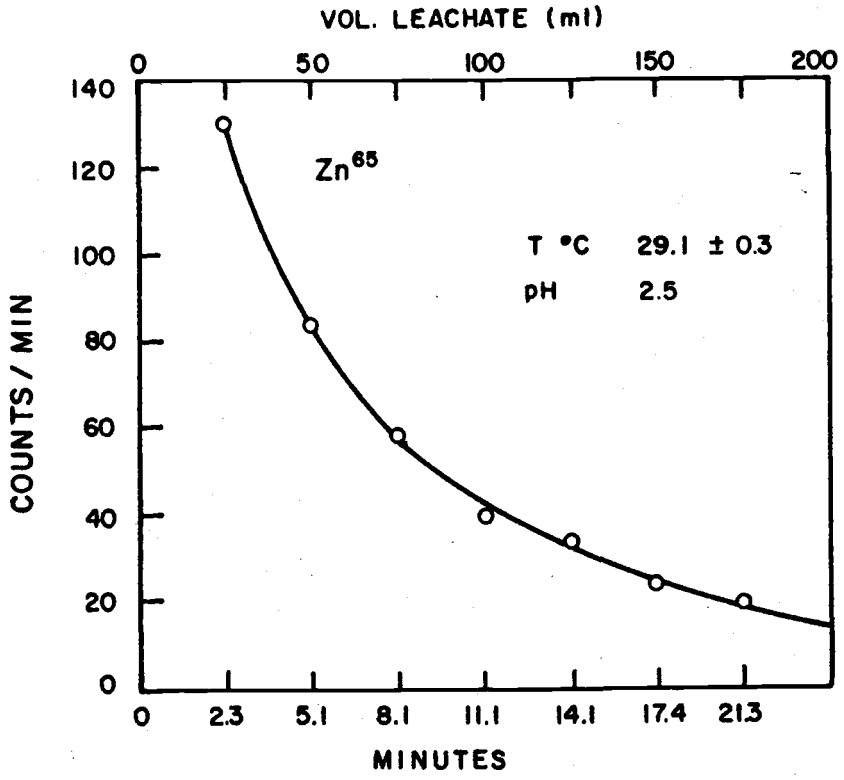

Figure 33. Counts due to Zn^{65} in 0.5N acetic acid leachate. Station 5.

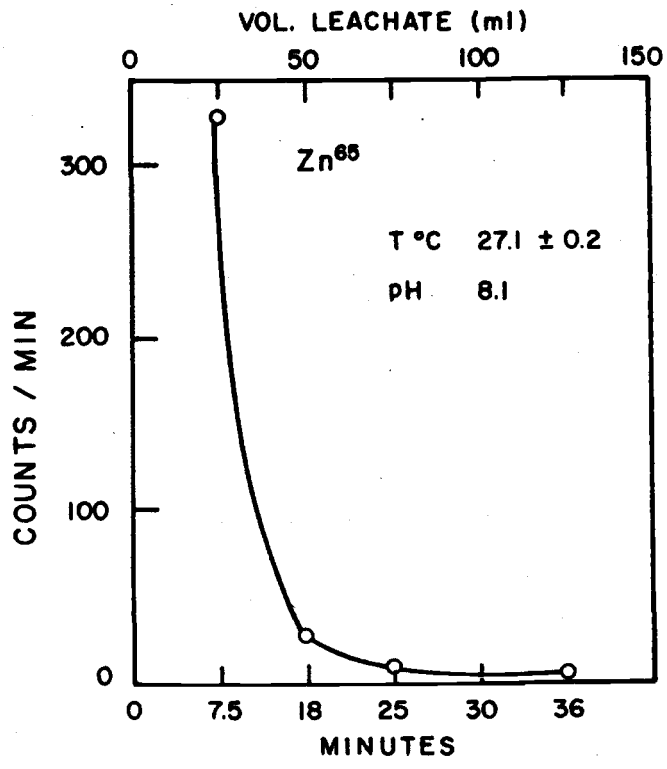

Figure 34. Counts due to Zn^{65} in standard sea water leachate. Note rapid removal rate, compared with 0.5N acetic acid.

Discussion and Summary

(1) Although the particle size versus activity study shows a general increase in activity with decrease in particle size, there are several anomalies (Fig. 27). These deviations suggest a change in mineral composition, and corresponding exchange capacities, from one size fraction to another (at least in some cases). Plans are underway to examine the exchange capacities of various fractions to determine if anomalies can be related to cation exchange capacity (c. e. c.), and changes in mineral composition.

(2) The fact that only 20-25 percent of the Zn^{65} appeared to be in exchangeable form suggests an irreversible mechanism of removal is involved, besides ordinary cation exchange. For example, it has long been known that appreciable amounts of zinc and cobalt can be irreversibly removed from aqueous solution by clay minerals and become fixed in a non-exchangeable form (Tiller et. al. 1961). More recent work, employing the use of acetic acid and EDTA leaching solutions (combined with special sorption conditions), has revealed that at least two different specific sorption sites are present in certain clay soil systems, in addition to the regular cation exchange positions. These sites can lose cations to acetic acid or EDTA but not to strong common salt solutions (i. e. NaCl, NaAc, BaAc, etc.) (Hodgson 1960; Tiller et. al. 1963).

Whether or not the observed difference between the amount of Zn^{65} removed by sea water and that removed by EDTA shown in Table 12 can be attributed to the "specifically bound" form cannot be concluded at the present time. However, as more data are collected, the apparent "fixed" Zn^{65} present in Columbia River sediments should become better defined as a result of this type of work.

(3) On the basis of our work and that of others, it must be concluded that ammonium acetate is not reliable for determination of exchangeable cations in natural systems. Since the amount of displaceable with NH_4Ac was consistently lower than with sea water (Table 12), and since Sawney et. al. (1959) found that vermiculite exhibits a c. e. c. (using N NaAc) just double that using neutral normal ammonium acetate, the amount of Zn^{65} removed with sea water is probably a representative value for exchangeable Zn^{65} (Sea water is approximately .5 M in NaCl and .1 M $MgCl_2$, and should be comparable to the 1 M NaAc used by Sawney).

(4) Since sea water is capable of rapidly displacing the exchangeable gamma emitters from Columbia River sediments as in Figures 29 and 34, it seems likely that exchangeable radionuclides associated with suspended material will be rapidly exchanged for Na^+ and Mg^{++} as the suspensoids contact saline waters in the estuary. Further evidence for a mechanism of this type was demonstrated by Cr^{51}/Zn^{65} ratios on filter samples taken from Hood River to the mouth of the Columbia (See Section E). This process is shown schematically

(Fig. 35). Although ion exchange has been commonly listed as a mechanism which tends to concentrate radioactive materials in an estuary (Rice 1963), the proposed mechanism illustrated (Fig. 35) should instead act to disperse radionuclides.

Further work will involve sea water leaching of both river and marine sediments, and filter samples as well. Marine sediments should show little if any exchangeable gamma emitters compared to river sediments.

It should also be mentioned that future leachings will be carried out on site to avoid any biologically or physically induced chemical changes that could occur when samples are collected in the field and then brought back to the lab for processing.

In summary, our preliminary work shows the Columbia River (and its radio-tagged trace elements) to be a powerful tool that can help solve some of the geochemical problems concerning the distribution of various elements in the terrestrial and marine environment.

L. Biological Study of the Columbia River Estuary

Lois Haertel and Gerald Wiese

Introduction

The Columbia River estuary is an area of great economic importance, serving as a source of water for industry, and avenue for shipping, a resource for both commercial and sport fisheries, and a disposal for dumping of industrial and radioactive wastes. For this reason it is desirable to understand its natural processes as well as possible. However, other than some isolated studies on pollution and game fish, little information is available on the ecology of the estuary.

The nuclear reactors at Hanford release radioisotopes into the river waters in amounts sufficient to make possible their use as ecological tracers in the estuary. Because of this, radioanalysis of organisms collected in the estuary can give useful information about the circulation and concentration of isotopes throughout the trophic levels of the estuary ecosystem. However, such radioanalysis should be coupled with a general ecological study so that results of radioanalysis may be accurately interpreted and at the same time contribute to the understanding of the general ecology of the estuary.

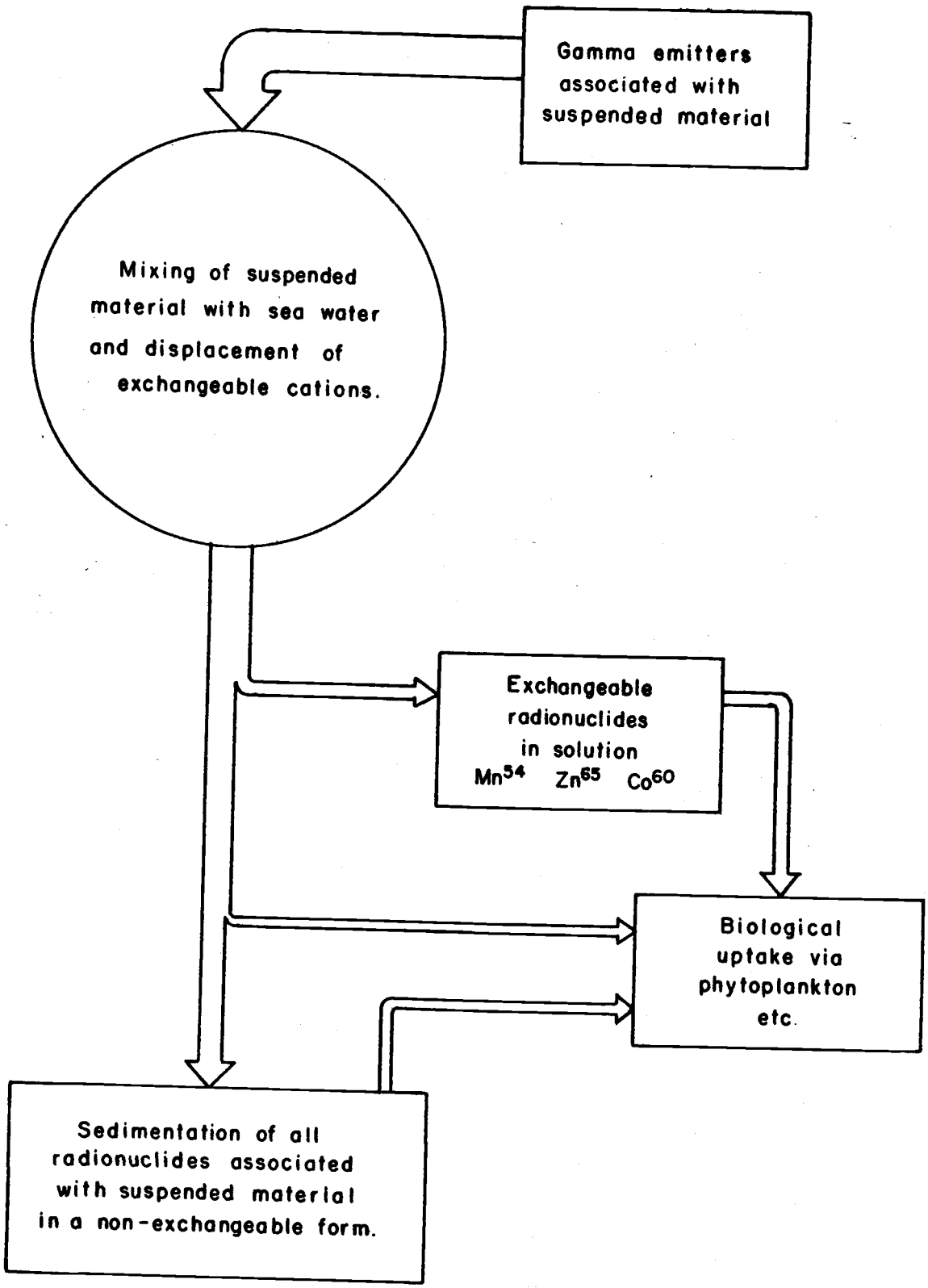

Figure 35. Schematic diagram shows that exchangeable gamma emitters are released from suspended materials by salt water.

Methods

Samples were collected October 7, November 5, and December 5, 1963, and January 8, 1964. Three sampling sites were chosen in order to sample fresh, brackish and nearly saline conditions. Sampling points were located about seven miles apart at Harrington Point (buoy 3), Astoria (buoy 39) and Chinook Point (Fig. 36), making it possible to sample all three stations in one day. On all dates but January 8, samples were collected upriver at Harrington Point near low tide in order to obtain as nearly fresh conditions as possible, and downriver at Chinook Point near high tide in order to obtain as nearly saline conditions as possible. Samples were collected at midtide at Astoria on all four dates. This sampling program made it necessary to travel against the tide going from point to point, thus avoiding the possibility of sampling the same water twice.

At each sampling point surface and bottom temperatures and salinities were taken by means of an induction salinometer. Fish and crab samples were collected by towing a 22-foot otter trawl along the bottom for about 20 minutes at each location. On October 7 plankton samples were collected at Harrington and Astoria by towing a #1 mesh meter net at various depths for 20 minutes. Plankton samples were collected at Harrington and Chinook on December 5 and at all three stations on January 8 by towing a #6 mesh half-meter net for 10 minutes in each location.

All otter trawl samples were analyzed as to the number of each species present in the trawl. November, December, and January samples were also analyzed as to wet weight of each species. The length of each individual was measured and length-frequency distribution plotted in order to determine the age composition of each species. Fish lengths taken were standard lengths and crabs were measured across the widest part of the carapace. Fish otoliths were examined in several cases to supplement length-frequency data. Qualitative stomach-content analysis was done on January samples.

Plankton samples were analyzed by diluting the settled volume up to five times and extracting one or more cubic centimeter aliquots with a stempel pipette. The aliquot was then placed on a gridded petri dish and all zooplankton counted. Numerical values for zooplankton given in the charts were obtained by multiplying the counted aliquot(s) by a factor to give the values for the total number of cubic centimeters of diluted sample.

Results and Conclusions

Temperature and salinity values are shown in Table 14 and graphed in Figures 37 and 38. Bottom waters at Chinook Point appear to have the highest salinity. Surface waters at Chinook and bottom waters

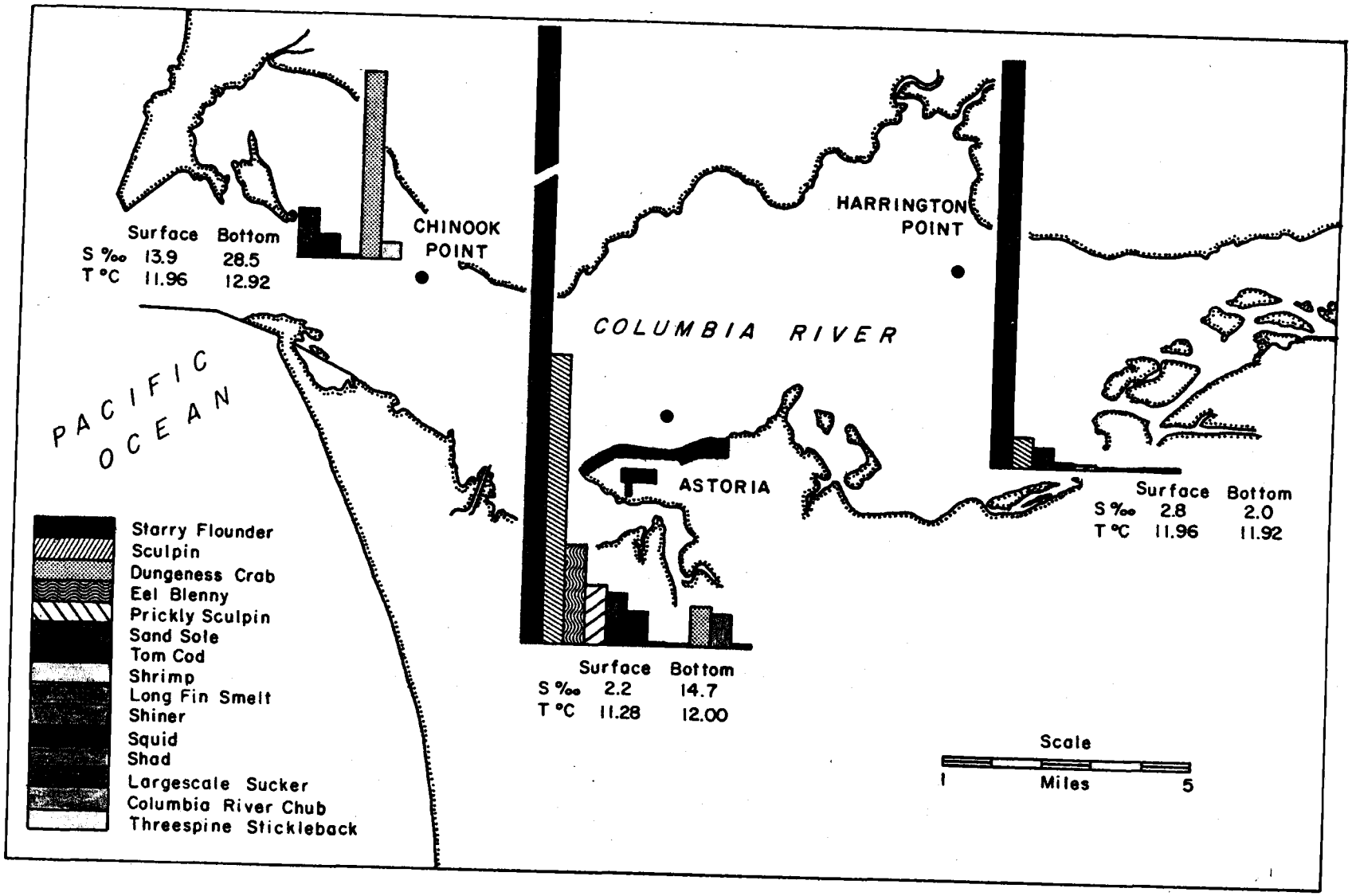

Figure 36. Biomass (grams) of organisms taken in 20 minutes with a 22-foot otter trawl, at three locations in the Columbia River estuary. 5 November 1963.

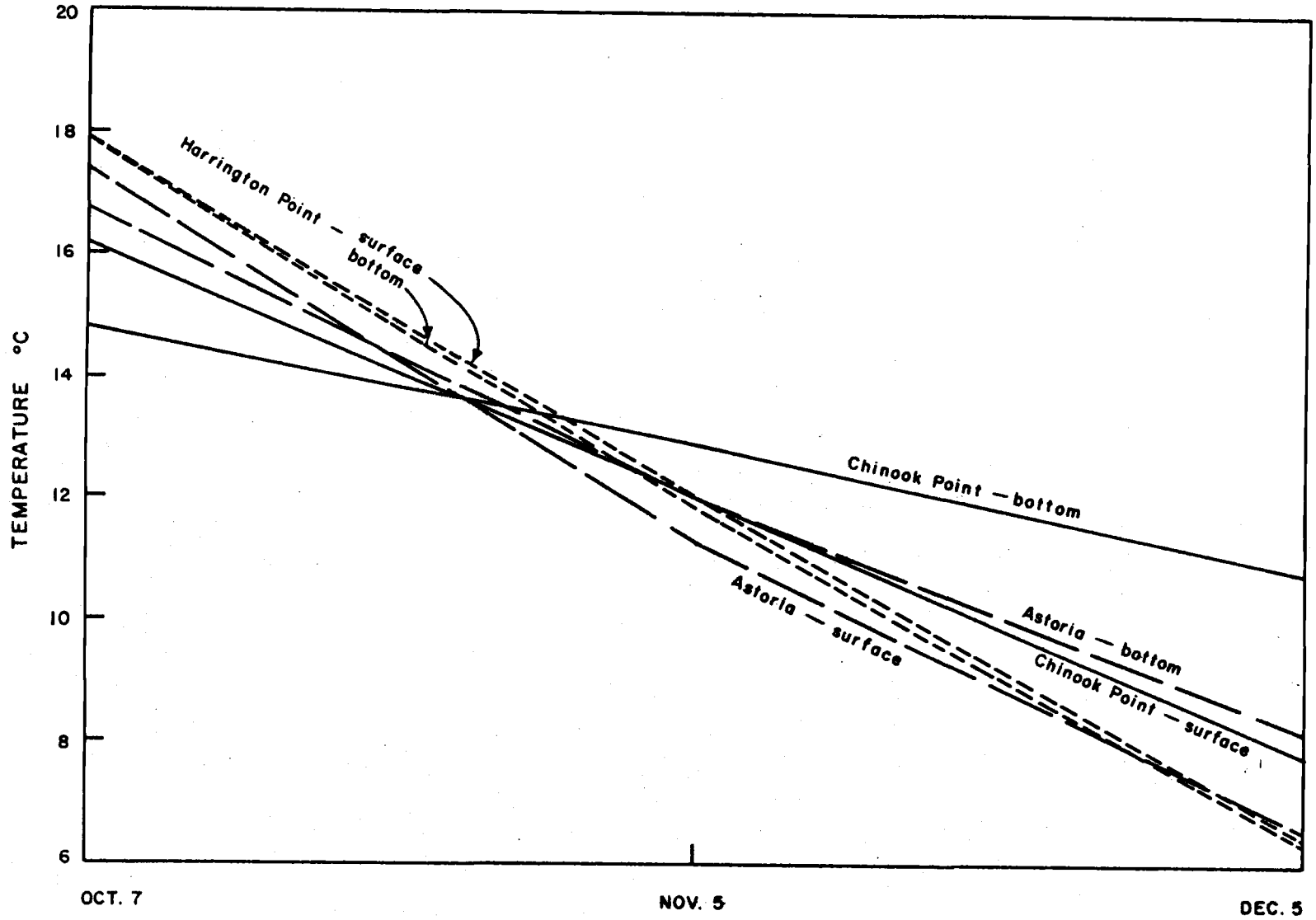

Figure 37.. Surface and bottom temperatures at three sampling stations in the Columbia River estuary, for 7 October, 5 November, and 5 December 1963.

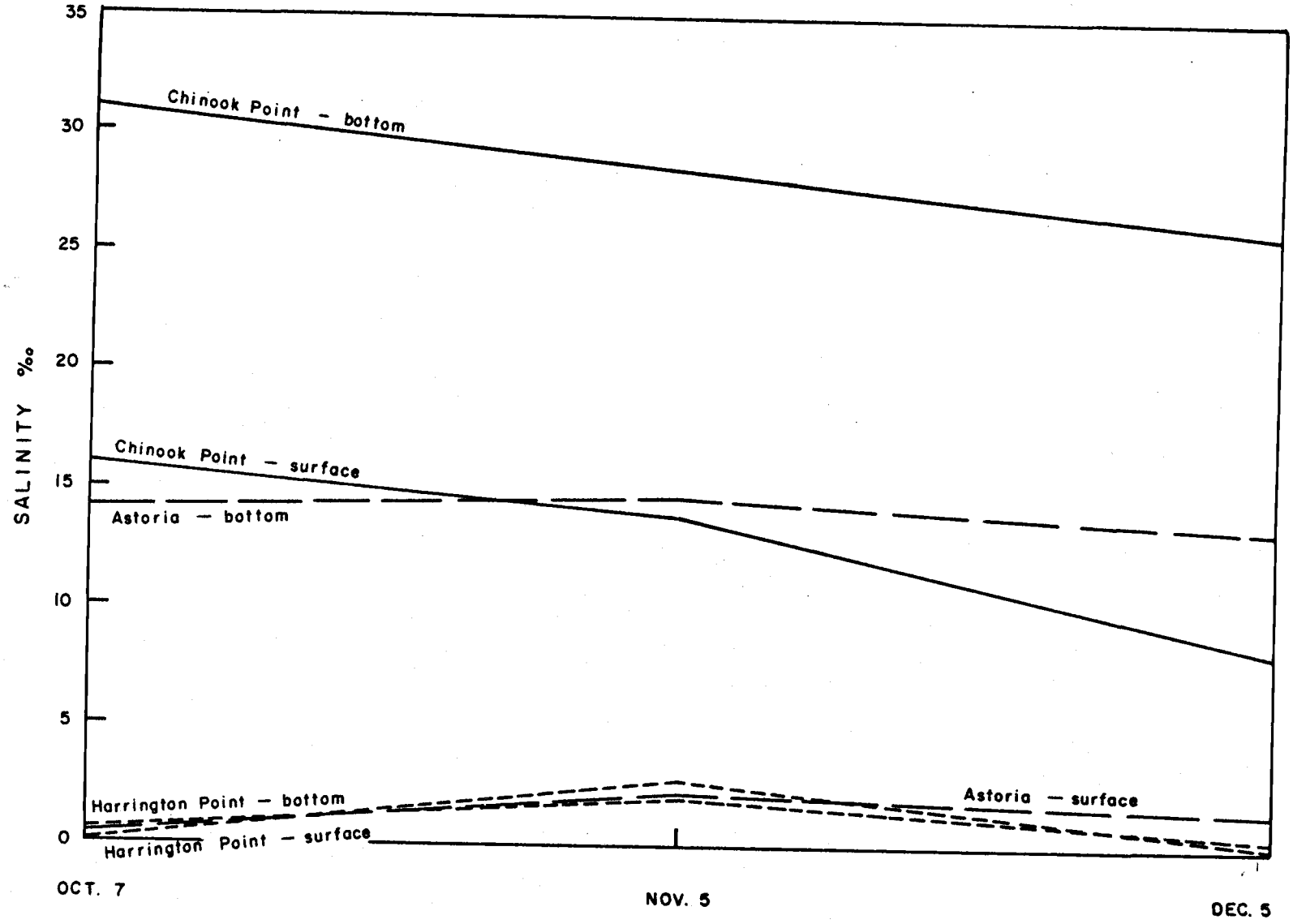

Figure 38. Surface and bottom salinities at three sampling stations in the Columbia River estuary, for 7 October, 5 November, and 5 December 1963.

Table 14. Comparison of Salinity and Temperature Values Obtained at all Stations on all Dates of Sampling.

Location	10/7/63		11/5/63		12/5/63	
	Sal.	Temp.	Sal.	Temp.	Sal.	Temp.
Harrington Point:						
Surface	0.2	17.9	2.8	12.0	0.2	6.5
Bottom	0.5	17.9	2.0	11.9	0.4	6.4
Astoria:						
Surface	0.4	17.4	2.2	11.3	1.5	6.6
Bottom	14.0	16.8	14.7	12.0	13.4	8.2

at Astoria appear similar in salinity and temperature. Surface waters at Astoria appear to resemble waters at Harrington Point in salinity and temperature and are relatively fresh. Overall salinity appears to show a decrease between the months of October and December. The temperature graph shows that an inversion of the temperatures of the three stations has occurred between October and December, undoubtedly because of the great seasonal change in temperature of the river water.

Table 15 shows the organisms which have been collected to date and the frequency with which they have been collected at each station. The organisms present indicate that Chinook Point may be a primarily marine environment and Harrington Point a primarily fresh-water environment. Presence of many fresh-water planktonic forms at Chinook can be explained by their being carried there by the current of the river. Quite a few fish and several invertebrates appear to be euryhaline as they are found frequently at all three stations.

Table 16 compares the organisms caught at different stations on December 5, 1963, and shows that the largest number of otter trawl species was taken at Chinook Point on this date. In October and November the largest number of species was taken at Astoria (Novembe results are shown in Figure 36. The change in the location of the largest number of species may possibly be correlated with the decreasi salinity in the estuary at this time or the generally warmer temperature afforded at Chinook in December or both. Table 17 also shows the increase of otter trawl species taken at Chinook at the later dates. Number of species taken at the other stations did not appreciably change during the period of sampling.

Stomach analyses of January samples indicate that larger fishes of all species ate primarily smaller fish and Crangon franciscorum. Smaller sand sole (Psettichthys melanostictus), probably representing the 0 and 1-year classes, were found to have ingested primarily Mysidaceae, and 0 and 1-year classes of starry flounder (Platichthys stellatus) were found to have eaten primarily Coriophorum. Most

Table 15. Frequency of Collection of Different Organisms at Different Stations.

Habitat*	Scientific Name	Common Name	Number of times collected		
			Harrington Pt.	Astoria	Chinook Point
	Otter trawl invertebrates:				
Fr. W.	Cottus gulosus	Riffle sculpin	2		
Fr. W.	Catostomus macrocheilus	Largescale sucker	3		
Fr. W.	Mylocheilus caurinus	Peamouth	3		
Both	Gasterosteus aculeatus	Threespine stickleback	3		
Both	Cottus asper	Prickly sculpin	4	4	
Both	Leptocottus armatus	Pac. staghorn sculpin	4	4	2
Both	Platichthys stellatus	Starry flounder	4	4	3
Both	Alosa sapidissima	American shad	3	1	2
Both	Spirinchus dilatus	Longfin smelt	3	4	2
Both	Cymatogaster aggregatus	Shiner perch		2	1
Marine	Lumpenus sagitta	Pacific snakeblenny		2	
Marine	Microgadus proximus	Pacific tomcod		4	4
Marine	Psettichythys melanostictus	Sand sole		3	4
Marine	Trichodon trichodon	Pacific sandfish			1
Marine	Osmerus dentex	Arctic smelt			1
Marine	Ammodytes hexapterus	Pacific sand lance			2
Marine	Allosmerus elongatus	Whitebait smelt			2
Marine	Xeneretmus latifrons	Blacktip poacher			1
	Otter trawl invertebrates:				
Marine	Crangon franciscorum	Sand shrimp	3	4	4
Marine	Loligo opalescens	Squid		1	
Marine	Cancer magister	Dungeness crab		1	4
	Plankton net:			**	***
Fr. W.	Unid. Oligochaeta	Aquatic earthworm	1		
Fr. W.	Unid. Insecta	Insect	1		
Fr. W.	Unid. Cladocera	Cladoceran	3	2	1
Fr. W.	Daphnia pulex	Cladoceran	3	1	1
Fr. W.	Diaptomus sp.	Copepod	2	1	1
Fr. W.	Bosmina sp.	Cladoceran	3	1	3
Fr. W.	Daphnia longispina	Cladoceran	3	3	3
Fr. W.	Cyclops sp.	Copepod	3	3	3
Marine	Coriophorum sp.	Amphipod	1	1	
Marine	Eurytemora hirundoides	Copepod	1	3	3
Marine	Acartia tonsa	Copepod		2	3
Marine	Cirripedia nauplius	Barnacle nauplius		2	2
Marine	Corycaeus affinis	Copepod			3
Marine	Pseudocalanus minutus	Copepod			2
Marine	Calanus finmarchicus	Copepod			2
Marine	Unid. Chaetognatha	Arrow-worm			1
	Unid. Mysidaceae	Mysid shrimp	1	2	2
	Unid. Gammaridae	Amphipod	1	3	1
	Unid. Polychaeta larvae	Polychaete worm larvae		2	2
	Unid. Pelecepoda larvae	Clam larvae		2	
	Unid. Fish larvae	Fish larvae	1		

* Habitat for fish taken from American Fisheries Society 1960, for invertebrates taken from Edmondson, 1959, and Light 1961.

** October value multiplied by 2 as no December sample available.

*** December value multiplied by 2 as no October sample available.

Table 16. Comparison of Organisms Collected at Three Stations, 5 December 1963.

Organism	Harrington Pt. No.	%	Wt.*	%	Astoria No.	%	Wt.	%	Chinook Pt. No.	%	Wt.	%
Cottus gulosus	4	1	22	4								
Catostomus marcocheilus	2	4	21	4								
Cottus asper	79	12	1513	18	5	3	77	2				
Leptocottus armatus	7	1	171	2	11	6	168	4	1	1	19	1
Platichthys stellatus	498	73	5970	71	129	69	2220	49	24	33	667	41
Alosa sapidissima	16	2	54	1					6	8	32	2
Spirinchus dilatus	79	12	637	8	14	8	126	3	8	11	55	3
Cymotogaster aggregata									2	3	12	1
Microgadus proximus					26	14	1850	41	8	11	119	7
Psettichthys melanostictus					1	1	98	2	19	26	518	32
Trichodon trichodon									1	1	159	10
Osmerus dentex									1	1	32	2
Ammodytes hexapterus									1	1	8	4
Allosmerus elongatus									1	1	1	4
Crangon franciscorum	5				67				194			
Cancer magister									28			
Daphnia pulex	167	4										
Bosmina sp.	333	9							11	4		
Daphnia longispina	1167	30							77	3		
Cyclops sp.	1667	44							77	3		
Eurytemora hirundoides	333	9							2288	40		
Acartia tonsa									66	2		
Cirripedia nauplius									11	4		
Corycaeus affinis									154	5		
Pseudocalanus minutus									77	3		
Calanus finmarchicus									33	1		
Polychaeta larvae									11	4		

* Weights given in grams

Table 17. Comparison of Organisms Taken at Chinook Point on Four Different Dates.

Organism	10/7/63		11/5/63				12/5/63				1/8/64			
	No.	%	No.	%	Wt.*	%	No.	%	Wt.	%	No.	%	Wt.	%
Leptocottus armatus							1	1	19	1	8	5	418	15
Platichthys stellatus	8	73	11	58	186	65	24	33	667	41				
Alosa sapidissima							6	8	32	2	2	1	13	1
Spirinchus dilatus							8	11	56	3	9	6	39	1
Cymatogaster aggregata							2	3	12	1				
Microgadus proximus	1	9	1	5	4	2	8	11	119	7	67	44	1413	49
Psettichthys melanostictus	2	18	7	37	94	33	19	26	518	32	55	36	958	33
Trichodon trichodon							1	1	159	10				
Osmerus dentex							1	1	32	2				
Ammodytes hexapterus							1	1	8	1	1	1	1	1
Allosmerus elongatus							1	1	1	1	10	7	15	1
Xeneretmus latifrons											1	1	5	1
Crangon franciscorum	10		45				194				278			
Cancer magister	4		3				28				13			
Unid. Cladocera														
Daphnia pulex											750	6		
Diaptomus sp											90	1		
Bosmina sp.											300	3		
Daphnia longispina							11	1			1110	9		
Cyclops sp.							77	3			2280	19		
Eurytemora hirundoides							77	3			2460	21		
Acartia tonsa							2288	40			5340	45		
Cirripedia nauplius							66	2			60	1		
Corycaeus affinis							11	1						
Pseudocalanus minutus							154	5			60	1		
Calanus finmarchicus							77	3						
Unid. Chaetognatha	+						33	1						
Unid. Mysidacea	+													
Unid. Gammaridae											+			
Unid. Polychaeta larvae											+			
							11	1						

* Weight given in grams

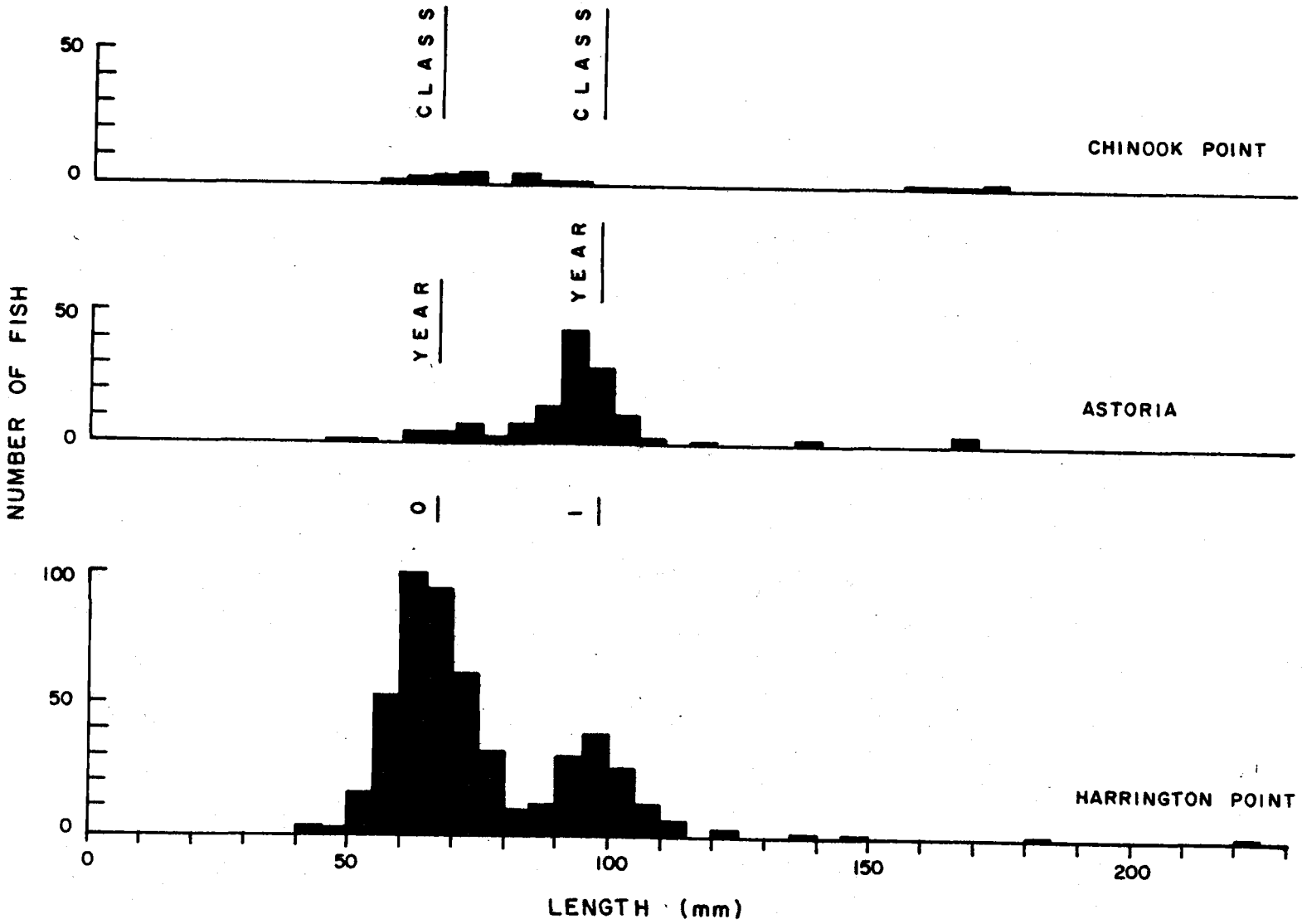

Figure 39. Size distribution of starry flounder at three stations in the Columbia River estuary, 5 December 1963. Note that most of the 0-year class fish are in fresh water (Harrington Point), while most of the fish from off Astoria are 1-year class. Very few young fish are found in salt water (Chinook Point).

other fish analyzed were found to have a variety of species in their stomachs. One species of fish (Lampetra tridentata, the Pacific lamprey) that had not been collected in the otter trawl, was found in the stomachs of at least four prickly sculpins (Cottus asper). Much more work is necessary to get an accurate picture of the food habits of species present.

A comparison of length-frequency distribution of starry flounder at all three stations (Fig. 39) on December 5 indicates that the bulk of the very young (0-year class) fish are found upriver in fresh water. Length-frequency data for January indicate that these young fish may be beginning to move down the estuary; however, data from the coming months will be necessary to confirm this.

Sampling to date has indicated that quite a few species of fish may be removed regularly in sufficient quantity for radioanalysis. The several species of both fish and invertebrates which are sufficiently euryhaline to be found frequently in saline, brackish and fresh water provide a basis of radiological comparison between the three stations. Continued sampling on a year-round basis will probably give a more complete picture of food-chain relationships and seasonal distribution of organisms within the estuary. Weather conditions permit sampling at all seasons of the year.

The data presented herein are not complete, but represent an example of the study being conducted. A full report will be prepared after observations have been carried out over a complete year.

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M. Radiological Study of the Columbia River Estuary

Gerald Wiese and Charles Osterberg

Since October, 1963, a monthly radiological study of the biota in the Columbia River estuary has been underway. The primary purpose of this study is to discover the behavior of radionuclides in the estuary and its biota. Seasonal variations, concentrations, and metabolic activities are all parts of this behavior. This radiological study is meant to contribute to an understanding of the ecological processes of the estuary.

Samples were processed to a suitable form for radioanalysis and then analyzed in a gamma-ray spectrometer. In processing, some organisms were dissected into different parts, dried, and ashed. Some samples that did not provide enough bulk were simply dried. Duplicate samples of the same organism were counted and results were compared. Dissecting consisted of cutting the animal into two or three specific parts. At first, fishes were divided into two parts, the cleaned body, and gills and internal organs. Later it was felt that more could be learned from dissecting the organism into three parts; the cleaned body, internal organs, and gills. Duplicate samples of the same organism at the same station were usually done with whole animals.

Methods of collection of these samples are presented in the previous portion of this report (see Haertel and Wiese). Locations of sampling stations bottom salinities and bottom temperatures are also contained in the biological section. Data reduction was by IBM 1620. Reduced data were recorded as picocuries per gram dry weight.

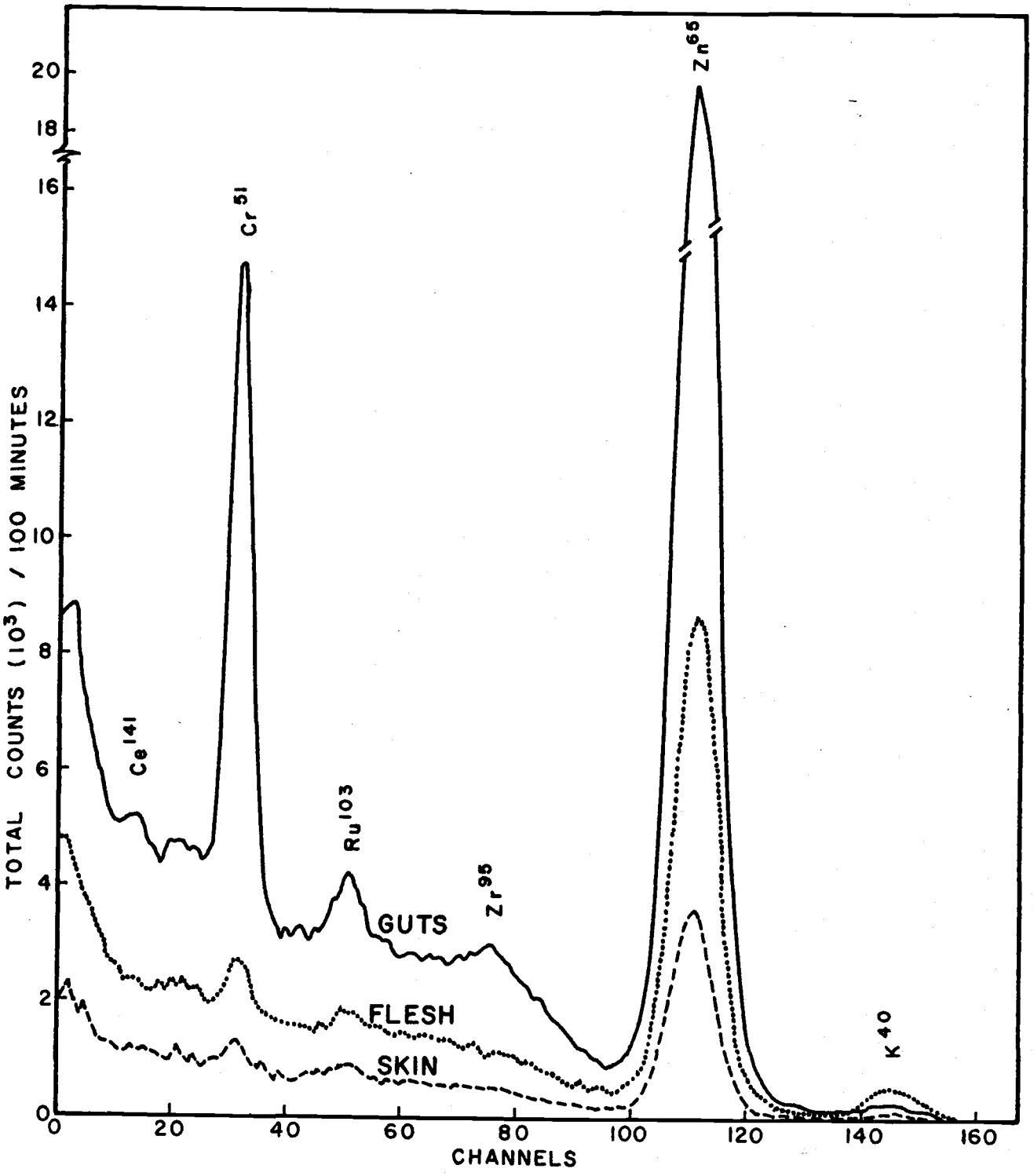
Cr^{51}, Zn^{65}, and K^{40} were found in most of the samples analyzed. Figure 40 shows a typical spectrum.

Average values of Cr^{51} for organisms were higher at buoy 3 (Fig. 41). Values for Cr^{51} at buoy 39 appear to be higher than those for Chinook Point, but large differences are not present (Fig. 41). Relationships shown for Cr^{51} are similar for every month studied (Fig. 42). Similar changes in Zn^{65} are not apparent (Fig. 43).

The gut of Platichthys stellatus seems to be consistently high in Cr^{51} at most stations compared to other samples from the same station (Table 18). At buoy 3, the gut of P. stellatus and whole Crangon franciscorum appear high in Cr^{51} (Table 17). C. franciscorum was high in Cr^{51} at buoy 39 both months it was taken. Other animals show similar tendencies for or against certain radionuclides.

No radiological relationship between levels for separate months has yet been established; insufficient time has elapsed to expect seasonal cycles.

Although this study has been of an introductory nature with little financial backing and low priority, certain conclusions are apparent:



SCULPIN

Leptocottus armatus

Figure 40. Typical spectrum of a fish from the Columbia River estuary.

C.L. OSTERBERG, 1963

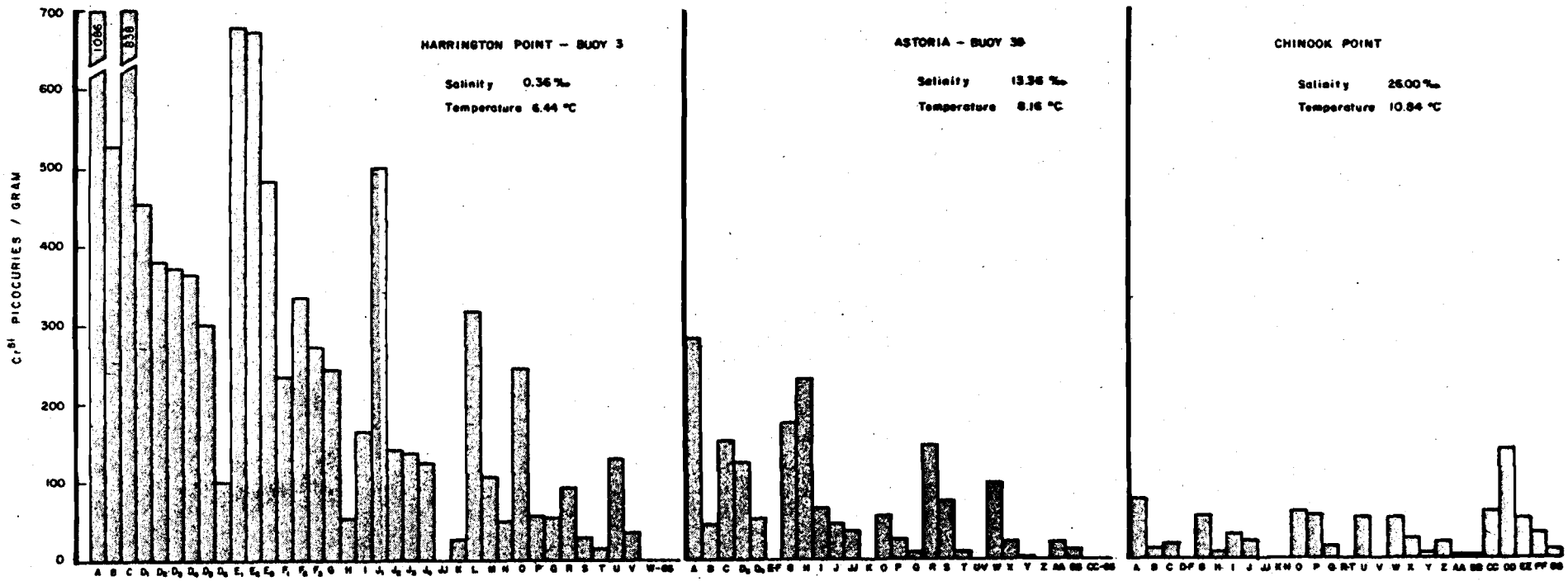


Figure 41. Chromium-51 levels in a number of organisms, from three stations in the Columbia River estuary, collected 12-5-63. Note higher levels of Cr^{51} are found in fresh water. (See list of organisms, page 81.)

LEGEND FOR FIGURES 41 AND 43.

A	Platichthys stellatus	starry flounder	gut and internal organs
B	Platichthys stellatus	starry flounder	gill
C	Platichthys stellatus	starry flounder	cleaned
D1	" "	" "	whole
D2	" "	" "	whole
D3	" "	" "	whole 1 year
D4	" "	" "	whole
D5	" "	" "	whole
D6	" "	" "	whole
E1	" "	" "	skin 1 year
E2	" "	" "	skin
E3	" "	" "	skin 0 year
F1	" "	" "	gill, guts, internal organs, and skinless body 1 year
F2	" "	" "	gills, guts, internal organs, and skinless body
F3	" "	" "	gills, guts, internal organs and skinless body 0 year
G	Crangon franciscorum	shrimp	whole
H	Spirinchus dilatus	longfin smelt	gut and internal organs
I	" "	" " "	gills
J1	" "	" " "	whole
J2	" "	" " "	whole
J3	" "	" " "	whole
J4	" "	" " "	whole
J5	" "	" " "	whole
JJ	" "	" " "	cleaned
K	" "	" " "	eggs
L	Cottus gulosus	riffle sculpin	gut and internal organs
M	" "	" "	gills
N	" "	" "	cleaned
O	Leptocottus armatus	staghorn sculpin	gut and internal organs
P	" "	" " "	gills
Q	" "	" " "	cleaned
R	Cottus asper	prickly sculpin	gut and internal organs
S	" "	" "	gill
T	" "	" "	cleaned
U	Alosa sapidissima	American shad	whole
V	Catostomus macrocheilus	largescale sucker	whole
W	Microgadus proximus	Pacific tomcod	gut and internal organs
X	" "	" "	gill
Y	" "	" "	cleaned
Z	Psettichthys melanostictus	sand sole	gut and internal organs
AA	" "	" "	gills
BB	" "	" "	cleaned
CC	Cancer magister	dungeness crab	gut and new shell
DD	" "	" "	gills
EE	" "	" "	shell
FF	" "	" "	whole
GG	Cymatogaster aggregata	shiner perch	whole

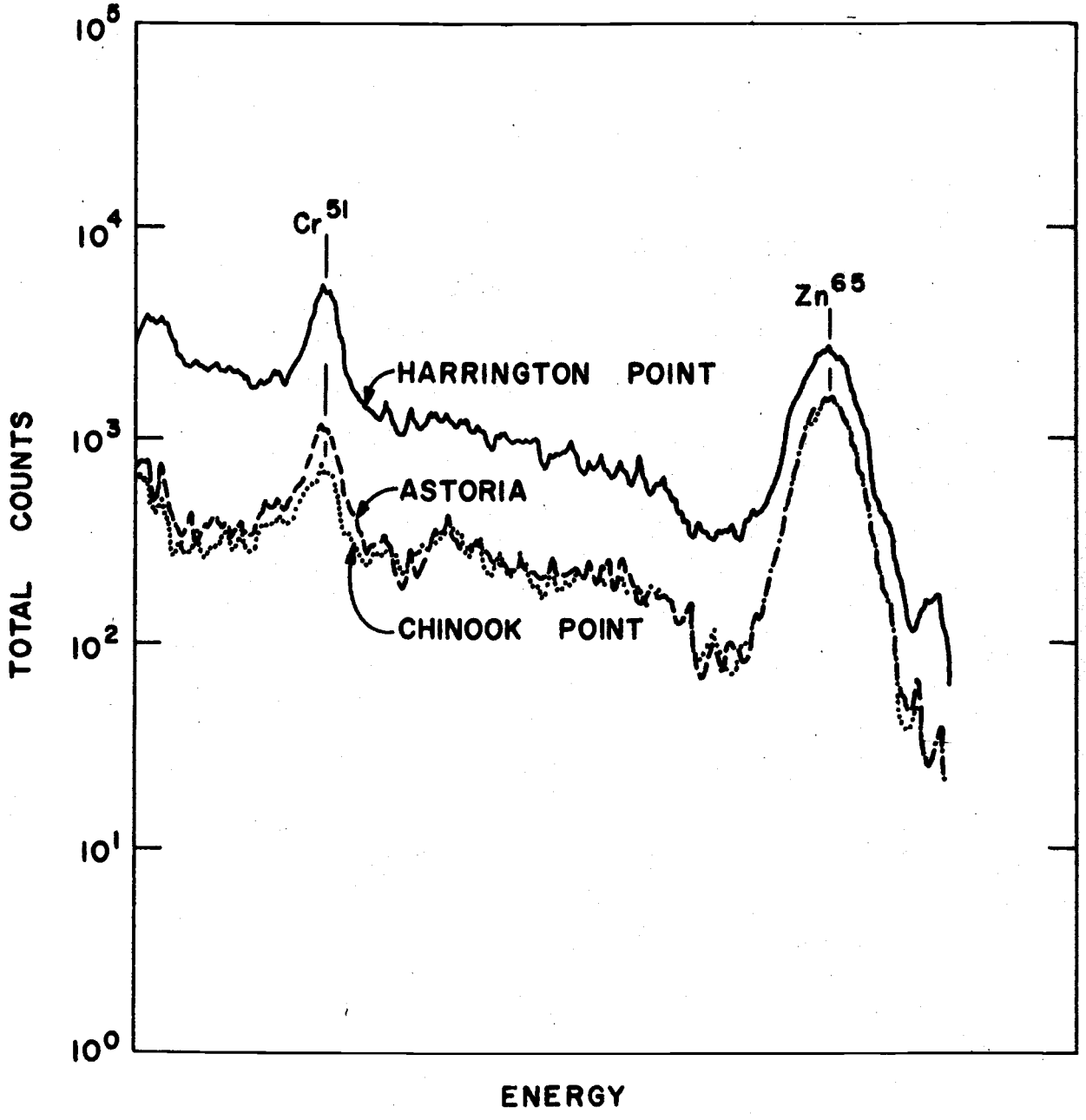


Figure 42. Comparison of spectra of sand shrimp (Crangon franciscorum) from three stations in the Columbia River estuary.

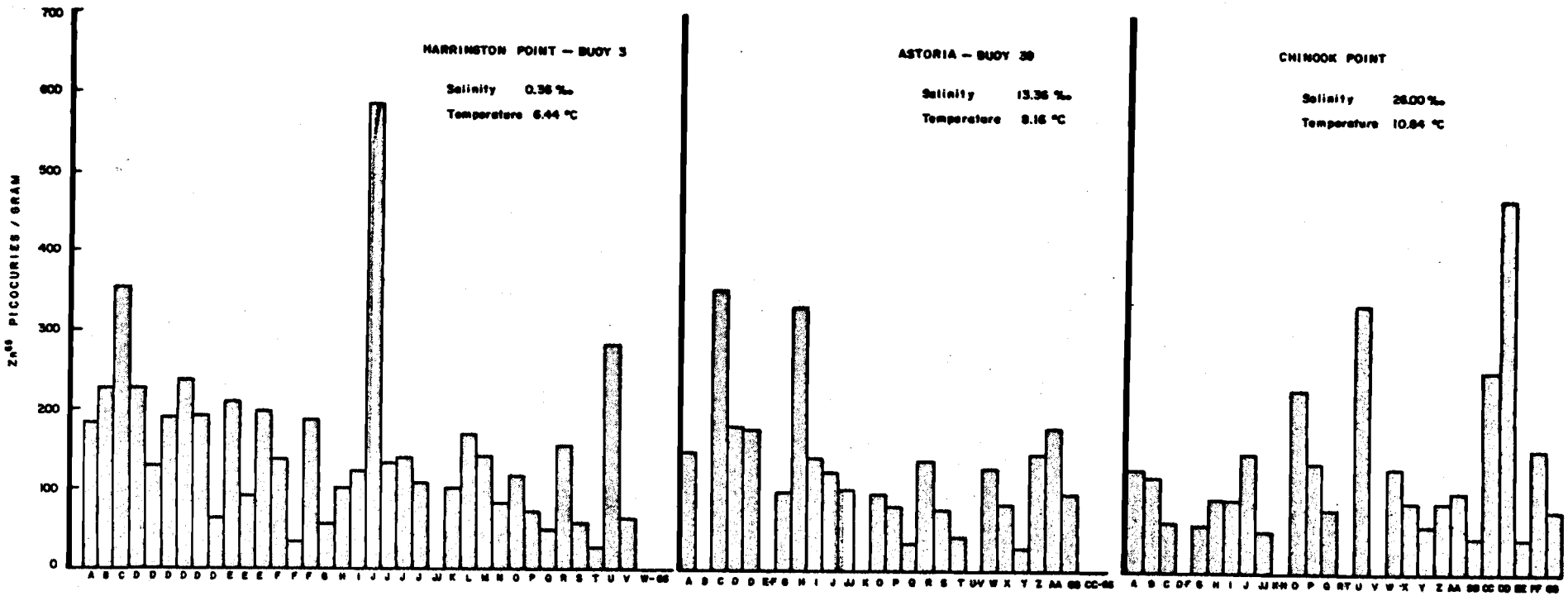


Figure 43. Zinc-65 levels in a number of organisms, from three stations in the Columbia River estuary, collected 12-5-63. No great changes in Zn^{65} levels are apparent. (See list of organisms, page 81.)

1. Radionuclides from Hanford appear in the water, sediments, and biota of the estuary.

2. Isotope concentrations vary markedly between different samples, proving that different mechanisms are at work.

3. Levels of radioactivity are sufficiently high to make radioecological studies practical with short counting times.

4. A large number of resident and migratory organisms are readily available throughout the year.

5. Weather conditions in the estuary usually are not severe enough to eliminate field trips.

6. Boat charters are reasonable.

7. Salt water and fresh water environments lie adjacent to each other.

8. All factors tend to make the Columbia River and its estuary an ideal natural laboratory for the study of radioecology.

Table 18. Organisms Highest in Cr^{51} (picocuries/gram dry weight).

BUOY 3 HARRINGTON POINT			BUOY 39 ASTORIA			CHINOOK POINT		
Species	Part	Pc/gr	Species	Part	Pc/gr	Species	Part	Pc/gr
10/7/63								
Crangon franciscorum	whole	491.44	Lumpenus sagitta	guts, gills, internal organs	503.73	Cancer magister	shell	57.17
Platichthys stellatus	whole	290.58	Spirinchus dilatus	guts, gills, internal organs	418.38	P. stellatus	guts, gills, organs	18.12
Leptocottus armatus	guts, gills, organs	246.19	C. franciscorum	whole	134.50	C. magister	muscle, shell	12.80
Cottus asper	guts, gills, organs	221.14	Microgadus proximus	guts, gills, organs	75.24	P. stellatus	cleaned*	4.29
P. stellatus	guts, gills, organs	176.23	Cymatogaster aggregata	whole	63.99	Psettichthys melanostictus	cleaned	1.98
11/5/63								
P. stellatus	guts, organs	647.42	L. sagitta	guts, organs	475.34	C. magister	gills	501.33
L. armatus	guts, organs	632.05	C. franciscorum	whole	139.33	C. magister	shell	117.93
C. franciscorum	whole	510.84	M. proximus	guts, organs	475.34	C. magister	guts, new shell	110.08
P. stellatus	cleaned	135.20	L. sagitta	gills	72.25	P. stellatus	guts, organs	77.51
Alosa sapidissima	whole	132.35	C. asper	gills	49.16	C. franciscorum	whole	71.48
12/5/63								
P. stellatus	guts, organs	1086.48	P. stellatus	guts, organs	285.96	C. magister	gills	140.48
P. stellatus	cleaned	837.69	S. dilatus	guts, organs	233.31	P. stellatus	guts, organs	75.96
P. stellatus	cleaned	837.69	C. franciscorum	whole	177.75	L. armatus	guts, organs	62.72
P. stellatus	gill	527.48	P. stellatus	cleaned	154.02	C. magister	guts, new shell	60.19
S. dilatus	whole	502.40	C. asper	guts, organs	148.89	L. armatus	gills	58.08

*Cleaned whole fish minus gills, guts, and internal organs.

IV. PAPERS PRESENTED AND MEETINGS ATTENDED

A panel discussion on "Atomic Waste Disposal Monitoring and Predictions -- Fact or Fancy" was sponsored by the Pacific Fisheries Bioligists at a meeting 20-22 March 1963, at Gearhart, Oregon. Dr. Richard Foster of General Electric was moderator. Panel members were Dr. Allyn Seymour, University of Washington; Mr. Harold Bissell, California Department of Fish and Game; and Dr. Charles Osterberg, Oregon State University.

Mr. Lauren Larsen presented a paper entitled "Gamma-ray Analysis on Suspended Material from the Willamette River" at a meeting of the Oregon Student Science Conference held at Reed College, Portland, on 27 April 1963. At this same meeting Mr. Norman Cutshall presented a paper entitled "Chromium-51 as a Tracer in the Hydrosphere."

Dr. Osterberg, as co-author with Dr. Herbert Curl, Jr., presented a paper at the meeting of the American Society of Limnology and Oceanography at Stanford University, 17-21 June 1963. Title of the paper was "Uptake of Chromium-51 by particles in Seawater." While in California, Dr. Osterberg went aboard a vessel in San Pablo Bay to observe in situ gamma analysis of sediments by means of the "Kaufmann sled." Two selected mud samples were carried to Oregon for detailed gamma analysis, and the results were made available to Mr. Peter Klingeman for comparison with his in situ results.

Dr. Osterberg attended the AEC sponsored symposium on "The Natural Radiation Environment" at Rice University, Houston, Texas, 11-13 April 1963, and a meeting of the Society for Applied Spectroscopy at San Diego on 14-18 October 1963. The latter meeting offered an opportunity to confer with Dr. Theodore Folsom at Scripps. Three samples of Oregon mussels and three of gooseneck barnacles were given to Dr. Folsom at this time. These samples were furnished to help Dr. Folsom substantiate earlier findings made with California organisms.

Dr. Osterberg represented Oregon State University at meetings of the Columbia River Working Committee 25 June 1963, in Portland and 5 February 1964, in Richland, Washington.

V PAPERS PUBLISHED, BUT REPRINTS NOT YET AVAILABLE

ACCELERATION OF SINKING RATES OF RADIONUCLIDES IN THE OCEAN

by

Drs. Charles Osterberg, Andrew G. Carey, Jr.,
and Herbert Curl, Jr.

Zirconium-95 - niobium-95 and cerium-141-144 were detected by gamma-ray spectrometry in two samples of sea cucumbers collected off the Oregon coast west of Newport in April 1963. Sea cucumbers of the genus Paelopatides sp. were found at 2800 m, 65 miles offshore; Stichopus californicus were taken in 200 m of water 25 miles off Newport. The gamma-ray spectra of these two benthic sediment feeders were remarkably similar, except for the lower zinc-65 peak in the spectrum of Paelopatides sp. (Fig. 1). Since these animals are bottom feeders, the presence of short half-lived fission products in the deeper sample suggests a more rapid sinking rate for fallout particles in the sea than would be expected from purely physical processes.

All benthic animals were collected with a 22-foot shrimp-type otter trawl. Samples, preserved in formalin and freeze-dried, were ashed at 500° C overnight. Twelve cc of ash were counted in the well of a 5 x 5-inch NaI(Tl) crystal, the signal from which was resolved with a 256-channel pulse height analyzer.

Recounts of both samples over a 114-day period indicated a 66-day half-life[1] for Zr^{95}-Nb^{95}. The calculated half-life of the Ce^{141}-^{144} mixture gradually lengthened due to the decreasing percentage of 33-day Ce^{141} compared to 282-day Ce^{144}. Potassium-40, the naturally radioactive isotope of potassium, has a very long half-life and showed no significant changes in activity over the period of observation.

Particles of fallout radionuclides exist in a wide range of sizes, but Sisefsky[2] found particles larger than 4.5μ from worldwide fallout to be very rare. These particles would not be likely to sink in sea water at a rate faster than that determined from Stokes' Law. According to the calculations of Carritt and Harley[3] based on Stokes' Law a 10μ spherical granite particle would require almost a year to sink to 2800 m and a 1μ particle would require 100 years, in the absence of turbulence.

There are few measurements of the settling rate of fallout particles in the ocean. Near Eniwetok immediately after a nuclear test, Lowman[4] observed that radioactivity moved downward in the surface-mixed layer (about 75 m thick) at 2.5 m per hour. Below the thermocline, it increased to 10 m per hour. At these high rates, a particle would sink to 2800 m in approximately 12 days. Such high sinking rates are probably due to the predominately large particles present in local fallout,[5] most of which have a diameter greater than 25μ.

Sugihara and Bowen[6] reported that cerium-144 in the Atlantic Ocean, away from local fallout, is removed from surface waters on particles sinking at about 100 m per month, or slower. Fallout traveling at this rate would require 28 months to reach 2800 m. The half-life of Zr^{95} is only 65 days, and the peak due to this isotope in the spectrum of our deep sample is a prominent one. Clearly the Zr^{95} in our collecting area, also far from any nuclear test site, must have descended at a rate much faster than 100 m per month.

The similarity of the fallout peaks (Zr^{95}-Nb^{95} and $Ce^{141-144}$) for the two samples (Fig. 1) suggests a very short transit time downward for these radionuclides. Unfortunately, however, absolute values for settling times require many assumptions. For example, we may assume that both deep and shallow sea cucumbers should have the same K^{40}:Zr^{95}-Nb^{95} ratios, and attribute the small observed differences to radioactive decay. The larger ratio for the sea cucumbers from 2800 m would result from a decay time (i.e., transit time) of seven days.

We might also assume that both groups of animals should have equal amounts of Zr^{95}-Nb^{95} (per unit weight) if they were feeding together at the same depth. However, the deeper sample was observed to have less Zr^{95}-Nb^{95} than the shallow sample. The observed difference in activity, if attributed to radioactive decay while the fallout settled to the greater depth, yields a transit time of about 12 days. Although neither assumption is known to be valid, the small difference in the spectra points to the need for a process which is capable of carrying fallout down to the ocean depths before appreciable radioactive decay can occur.

We suggest that biological processes are responsible for the relative abundance of fission products in deep bottom feeders. Particulate fallout radionuclides are known to be concentrated by herbivorous copepods, euphausiids, salps, and pteropods[7,8,9]. These zooplankton, which make up the bulk of second trophic level marine organisms, ingest fallout particles attached to microscopic plants in the surface waters of the ocean. Unassimilated radionuclides are released in faecal pellets, which, because of their greater size, sink rapidly. The biological turn-over time for these radionuclides is quite short. Chipman[10], in a laboratory experiment, observed that the copepod Tigropus californicus excreted 97% of ingested Ce^{144} in one day. He concluded that fission products in general were poorly absorbed from the digestive tracts of zooplankton. Apparently most herbivorous plankton are rapid "processors" of particulate fallout radionuclides, continually converting small particles into larger ones.

Preliminary measurements of the sinking rates of faecal pellets have been made in our laboratory by Curl (in prep.). Euphausia pacifica, a very abundant euphausiid, was fed diatoms (Skeletonema costatum). The average sinking rate for the resulting faecal pellets (100μ by 1000μ to 500μ by 3000μ) was 43 m per day. Under similar conditions (still water, temperature 10°C, salinity 33‰) these pellets would require about 65 days to reach 2800 m. Of course, there are many factors in the ocean which could alter the sinking rate. Nevertheless radionuclides incorporated into faecal pellets would reach bottom much faster than "unprocessed" fallout particles.

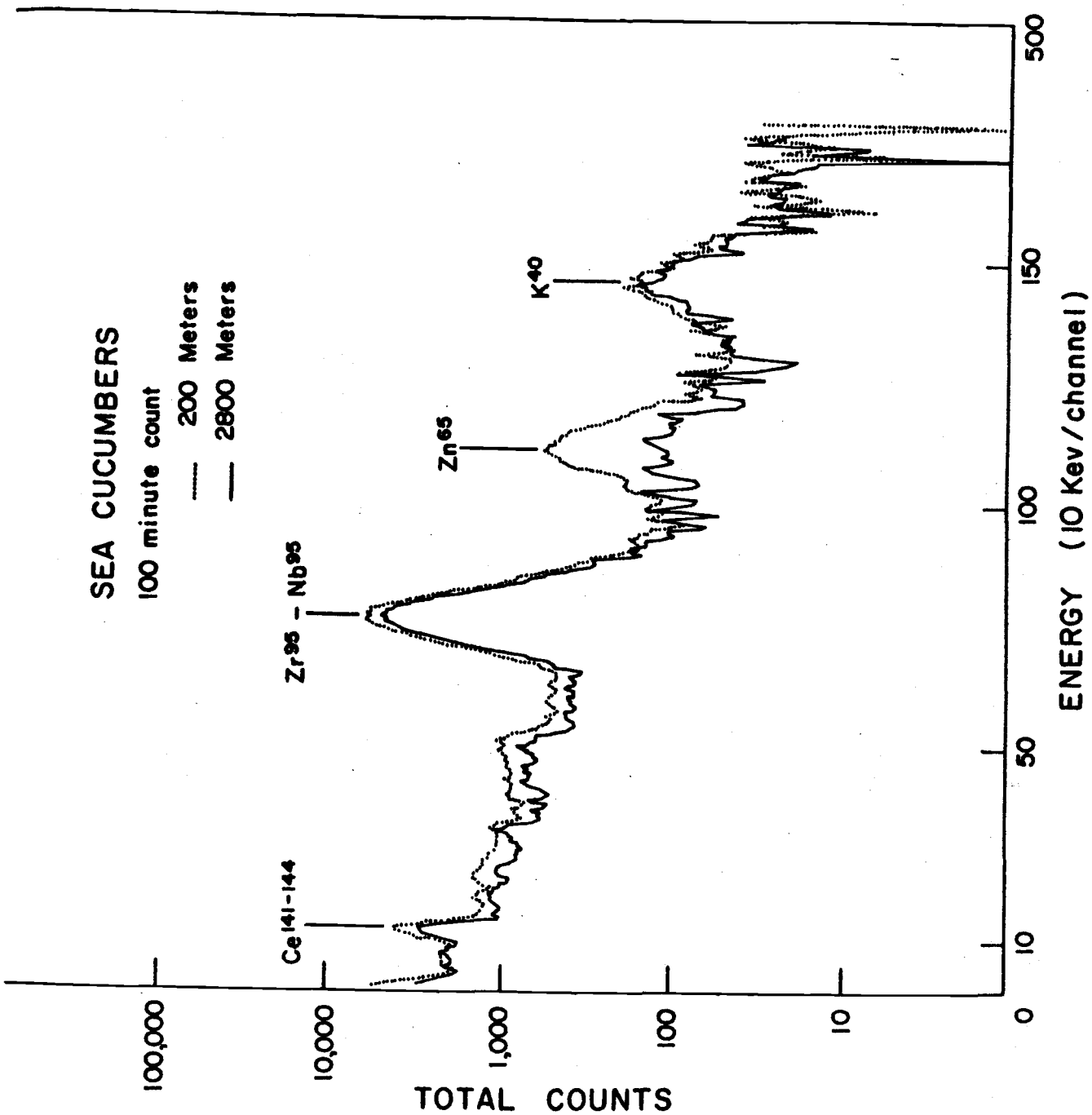


Figure 1. Gamma-ray spectra of the sea cucumbers Paelopatides sp. and Stichopus californicus. To avoid the confusion of overlap, the spectrum of the sea cucumber from 2800 m has been displaced downward slightly. Zr^{95}-Nb^{95} and $Ce^{141-144}$ are from fallout; Zn^{65} is principally from the Hanford, Washington, reactors; and K^{40} is the naturally radioactive isotope of potassium.

Predators which feed on herbivorous zooplankton carry fission products into higher trophic levels. These predators are in effect concentrating fallout radionuclides, since most of the Zr^{95}-Nb^{95} and $Ce^{141-144}$ are not assimilated[9]. It is conceivable that the excretory products of higher trophic level organisms are in such a form as to further accelerate settling rates of these radionuclides.

The ratios of Zr^{95}-Nb^{95}: $Ce^{141-144}$, which were higher in the deep sample throughout the observed decay period, indicate that more $Ce^{141-144}$ than Zr^{95}-Nb^{95} was lost during the longer transit time to 2800 m. Preferential loss could result from a slightly greater solubility of $Ce^{141-144}$, either in sea water or in the digestive tract.

Zinc-65, from atomic reactors (Hanford Laboratories, Washington) on the Columbia River, did not behave in the same manner as the fission products. It was abundant in sea cucumbers from 200 m, but much less was found in animals from 2800 m (Fig. 1), despite a longer half-life (245 days) than Zr^{95}-Nb^{95}. Unlike zirconium, niobium, and cerium, zinc is present in a number of enzymes. Apparently the biological turnover time for zinc is quite long, and little radiozinc reaches the bottom. Either it is not readily excreted, or, if excreted, most Zn^{65} is lost from faecal pellets before they reach 2800 m.

The possibility that our samples were contaminated with fallout either at sea or in the laboratory was carefully checked. Of the 25 sea cucumbers taken in the deep sample, only 16 were ashed to obtain the spectrum in Fig. 1. Six others were dissected, and their digestive tracts (which would not be subject to surface contamination) were analyzed separately. The same radionuclides were present. The spectrum of seven unidentified anthozoans (see ananomes) from the same trawl showed comparable levels of K^{40} and Zn^{65}, but the Zr^{95}-Nb^{95} peak was only one-eighth as high. This difference would not be expected if the sample were contaminated, but is typical of the discrimination against Zr^{95}-Nb^{95} which has been observed in some oceanic animals[9]. As a further check, three and one-half months later a second collection was made from 2600 m, 145 miles offshore. The levels of radioactivity if Paelopatides sp. were lower, but evidence of fallout was clearly present. Benthic worms, which are considered to be deposit feeders, were also collected in this trawl. They too contained Zr^{95}-Nb^{95}.

These observations lead us to conclude that fallout radionuclides can reach the ocean bottom quite rapidly under certain conditions. We propose that the great numbers of herbivorous zooplankton in the upper layers of the ocean are largely responsible for this phenomenon.

We acknowledge the assistance of Danil Hancock, Lauren Larsen, and the captain and crew of the R/V ACONA. We thank Elizabeth Deichmann and J. McCauley for identification of our sea cucumbers. This work was supported by contracts AT(45-1)1750 and AT(45-1)1758 with the Atomic Energy Commission, and NONR 1286(02) with the Office of Naval Research.

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VI PAPERS IN PRESS

RADIOACTIVITY AND ITS RELATIONSHIP TO OCEANIC FOOD CHAINS

by

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

ABSTRACT

Gamma-ray spectra of some primary producers (single-celled plants), filter-feeding herbivores, and carnivores, assigned to trophic levels I, II, III, 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, compared with spectra of herbivores.

INTRODUCTION

The oceans receive a substantial share of radioactive fallout resulting from nuclear testing because of their large areas 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 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, ed., 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 for us 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 by references from the literature.

METHODS

Phytoplankton and detritus were collected by passing surface sea water through a 5-inch membrane filter (0.65 microns), plus glass fiber

pre-filter (Gelman Instrument Company). The filters were ignited in an open crucible to destroy the membrane filter, the residue 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 6-foot Isaacs-Kidd midwater trawl towed for 30 minutes from 200 m depth to the surface. Plankton and nekton samples were freeze-dried after formaldehyde preservation. 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). Tubes containing the prepared samples were placed in the well of the 5" x 5" NaI(TI) primary crystal of the Hanford, Washington, total absorption anticoincidence spectrometer (Perkins, et al., 1960) for analysis. Counting time was 30 minutes, with 30-minute background subtracted.

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).

RESULTS

Trophic Level Relationships

The first trophic level in the pelagic environment consists of single-celled 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 sea water was passed. Most animals were removed by pre-filtering through a #6 mesh net.

Trophic Level II

Euphausia pacifica. Euphausiids may feed on phytoplankton, small crustacea such as copepods, or detritus (Ponomareva, 1954; MacDonald, 1927; Marshall, 1954). Although Ponomareva (1954) noted that *E. pacifica* occasionally fed on crustacea, it is primarily a filter-feeding herbivore. Setae of the thoracic legs of our adult *E. pacifica* are about 20-40 μ apart; thus, the filtration apparatus is equipped to collect most marine diatoms.

Calanus cristatus, *Salpa* spp., and *Clio pyramidata* are pelagic zooplankton which feed on suspended particles, principally phytoplankton. *Calanus* 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 which were apparently dismembered and masticated before ingestion. Chitinous remains of crustacea 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 them 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 lantern fish, 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 stomach contents by volume was due to the presence of several lantern fish. T. macropus appears to be intermediate between trophic level III and IV.

Trophic Level III-V

Thunnus alalunga. Several hundred migratory 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 were empty or less than one-quarter full. Cephalopods composed about 75 percent of the bulk of the stomach contents, fish about 18 percent, and crustacea about 5 percent.

Radioanalyses

Gamma-ray spectra of organisms from several trophic levels taken at the same time and location are shown in Figure 1. Trophic level I is represented by a membrane filter through which surface sea water 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 (probably as clay particles) was present. The K^{40} level on this filter was nevertheless higher than in filter samples from further offshore. Cerium-141, ruthenium-103, and zirconium-95 - niobium-95,[2/] which are particulate in sea water (Greendale and Ballou, 1954), were trapped by the filter, whereas zinc-65, which is ionic in sea water, was low even

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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}.

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 sea water 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 are 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 while feeding or they may adsorb to larger particles, which are eaten. Concentration of these radionuclides does not appear to be due to adsorption on the surface of the zooplankton, however (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, although in some cases small amounts of Cr^{51} are found in euphausiids.

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

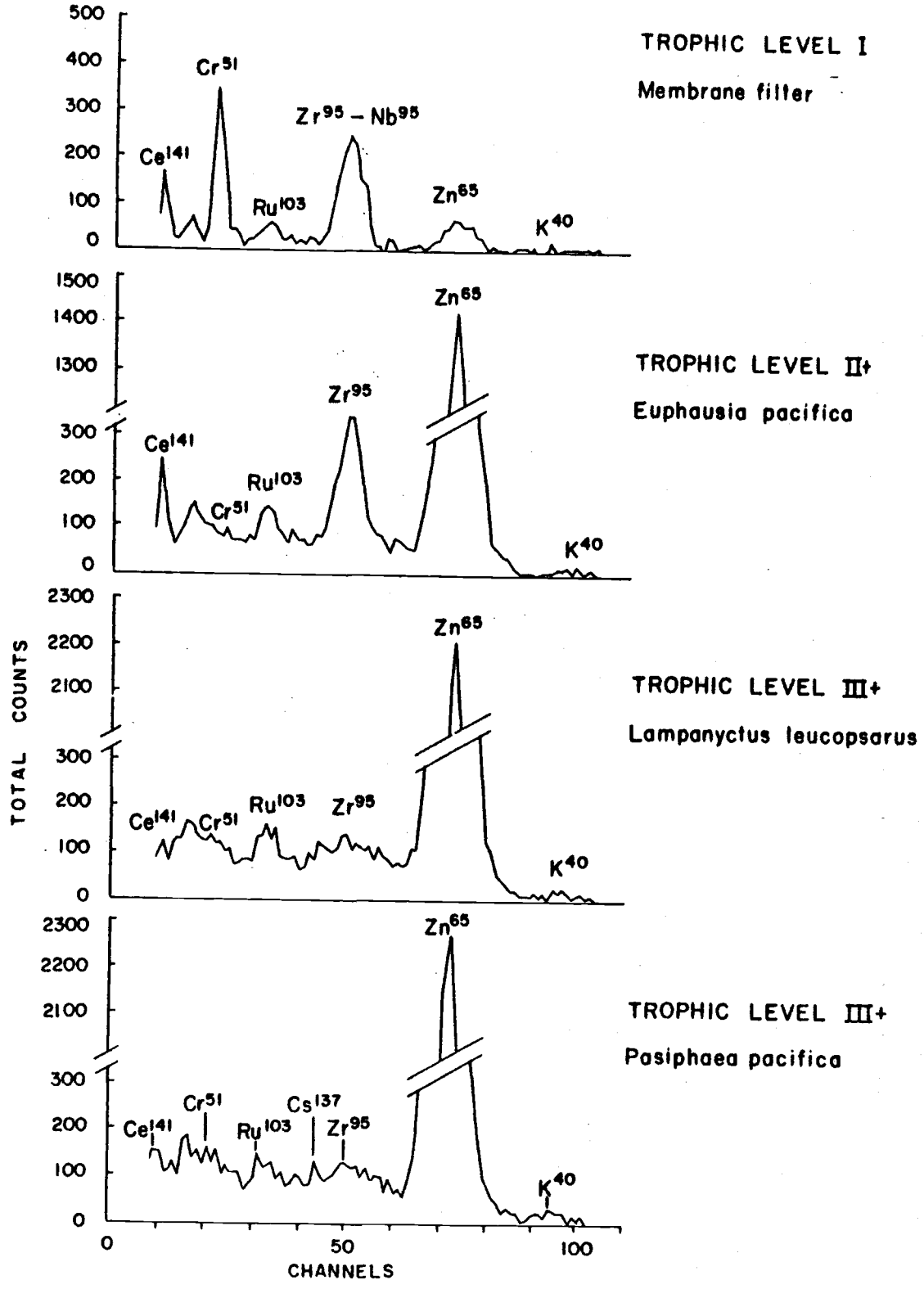

Figure 1. Comparison of gamma emitters from several trophic levels. Organisms for all four spectra were collected 15 miles off Astoria on 5-6 April 1962. All trophic level II and III animals were from the same trawl sample.

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

E. pacifica often contributes the greatest biomass in our trawl collections. 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

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3/ Short period variations in fission product levels in euphausiids from a single location are small. Nine consecutive tows, made over a period of 8 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/gram and Ce^{141}, 17.5 ± 2.8 pc/g, dry weight.

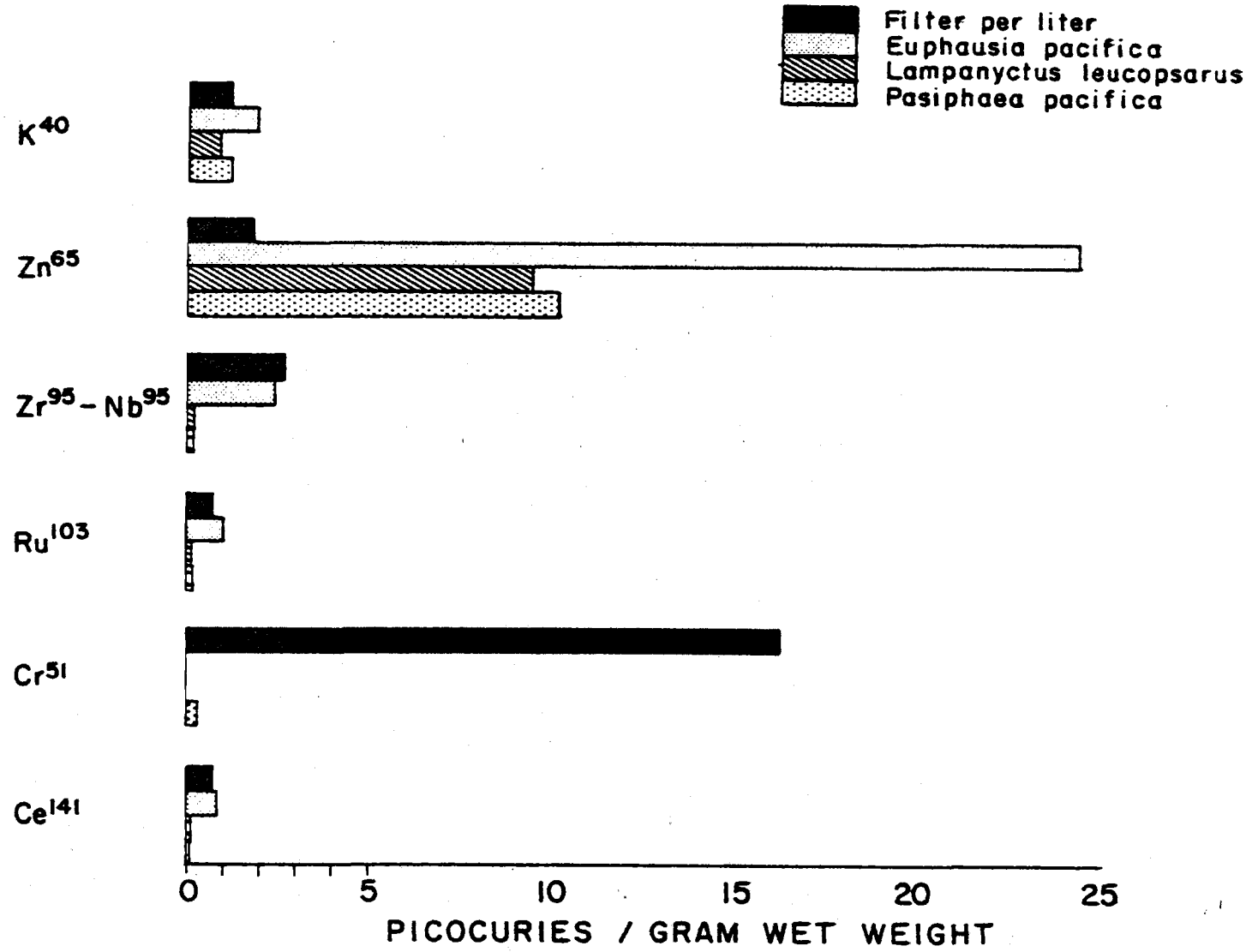

Figure 2. Concentrations of gamma emitters from several trophic levels. All organisms were collected 15 miles off Astoria on 5-6 April 1962. Note different units for filter sample.

the same time and place. Our comparisons were usually 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 Figure 3. These spectra are somewhat typical of our oceanic herbivores. That 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 Figure 1 form a general pattern. That is, 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 comparisons from the same sample were possible, and 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 a 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 non-cationic species regularly found in trophic levels III-V. It was present in the tuna liver, and has been reported in the liver of a cotton rat (Sigmodon hispidus) (Kaye and Dunaway, 1962), although Chipman (1960) observed little uptake of Ru^{106} from digestive tracts of

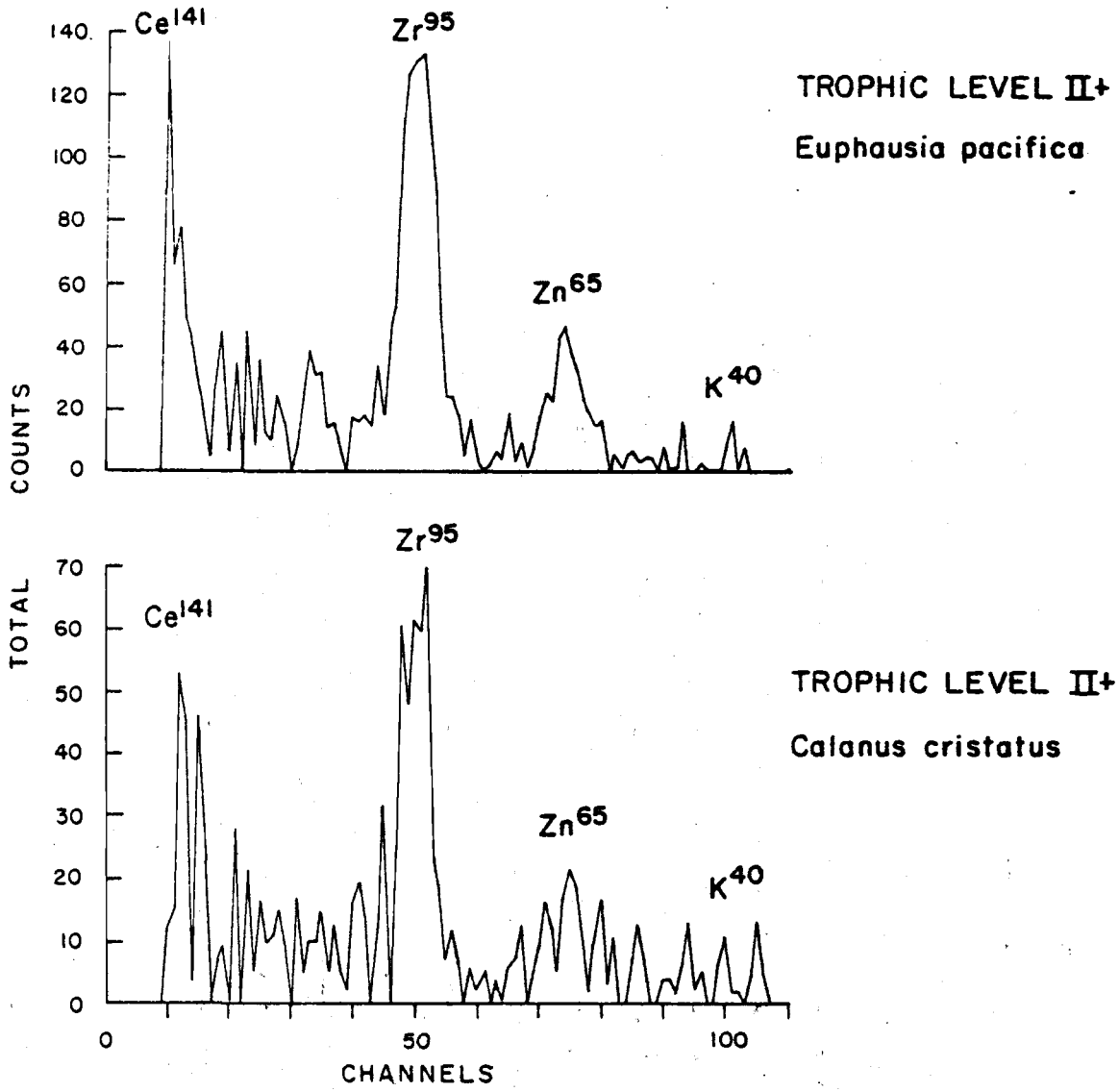

Figure 3. Comparison of spectra of euphausiids and copepods from the same sample, collected 105 miles off Astoria on 6 June 1962. The scatter in the lower spectrum is due to small sample size.

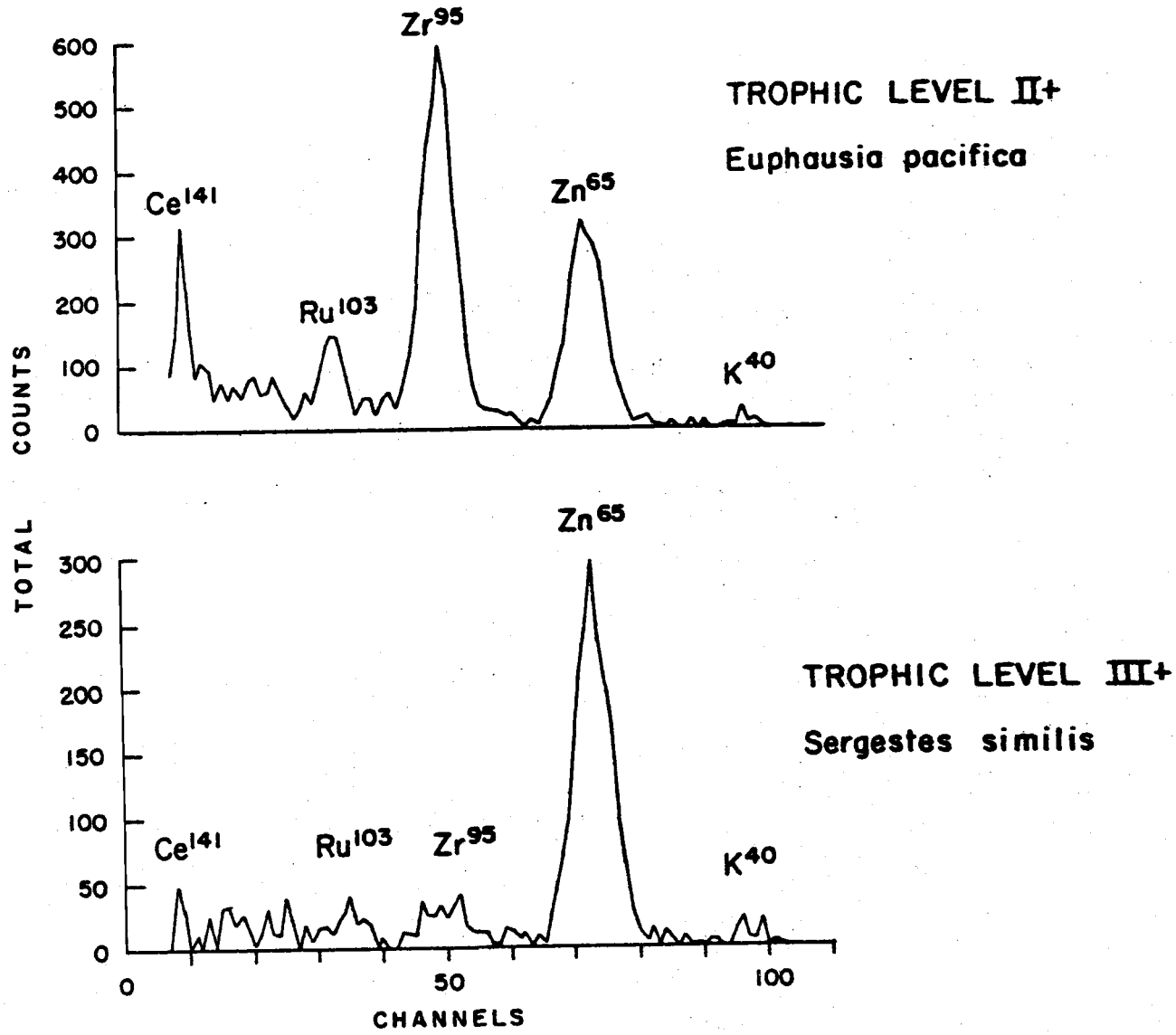

Figure 4. Comparison of spectra of euphausiids and sergestids from the same sample, collected 25 miles off Newport on 6 November 1961.

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) show 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 being reversed on land. Zn^{65} is a prominent isotope in fresh-water fish from the Columbia River (Davis, 1958). Joyner (1962) found that carnivorous, marine, lagoon fish concentrate Zn^{65} to a greater extent than do herbivores. However, the levels of Zn^{65} reported in oysters (Watson, et al., 1961) near the Oregon-Washington coast were much higher than 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 presence of the Hanford reactors on the Columbia River makes Zn^{65} the most common gamma emitter in marine organisms from the 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 to the same isotopes in marine herbivorous animals collected simultaneously. The highest trophic levels in the ocean, as evidenced by our tuna sample, almost completely discriminate against particulate fission products, but 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 intercomparing organisms from the same trawl sample.

We conclude that particulate radioactive fallout will be concentrated at the second trophic level by filter-feeding plankton, but very little of this radioactivity will be 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 is one of the more innocuous radioisotopes, since it decays principally by electron capture and few ionizing particles are emitted. 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 fallout.

ACKNOWLEDGMENTS

We thank R. W. Perkins of General Electric's Hanford Laboratories for making his equipment available to us, and for his assistance with certain technical aspects of the gamma-ray spectrometry.

This research was carried out under grants AT(45-1)1750 and AT(45-1) 1726 with the Atomic Energy Commission, Nonr 1286(02) with the Office of Naval Research, and Grant G 23103 with the National Science Foundation.

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ZINC-65 IN EUPHAUSIIDS AS RELATED TO COLUMBIA RIVER WATER OFF THE OREGON COAST

Charles Osterberg, June Pattullo and William Pearcy

Abstract

Most of the zinc-65 in the northeast Pacific Ocean originates in the Columbia River as a result of the Hanford, Washington, nuclear reactor operations. The Columbia River is also the principal source of fresh water in the region, so that at certain seasons the plume of the river is detectable far at sea because of its low salinity. In an effort to determine the fate of the river water at sea, euphausiids from the ocean off Oregon were used as biological monitors. The Zn^{65} content of the euphausiids was measured by gamma-ray spectrometry, and the relationship between Zn^{65} content and the salinity distribution over the area was examined. Some correlations were apparent, although seasonal changes of Zn^{65} content in euphausiids were not great. Zn^{65} levels remained fairly high off Oregon even when Columbia River water was not evident as a low salinity plume. This indicates that water from the Columbia River, which accumulates as a plume off Oregon in the summer, influences the Zn^{65} content in animals in the area throughout the year. The long half-life of Zn^{65} in the mixed layer, the diurnal migrations of euphausiids, and the seasonal reversal of currents all may contribute to the maintenance of this reservoir of Zn^{65} in the ocean off the Oregon coast.

INTRODUCTION

The outflow of the Columbia River dilutes the salinity of the surface water in certain sections of the northeast Pacific Ocean, forming a shallow lens of low salinity water which over-rides the more dense oceanic water. This lens of water is known as the plume of the Columbia River, and, by definition, consists of water having a salinity of 32.5 °/oo or less. In summer, prevailing winds and currents carry this less saline water southward, sometimes forming a pool approximately 100,000 square miles in area off the Oregon-California coast. In winter, surface waters are mixed to a greater depth, and Columbia outflow is diminished, while coastal rivers attain maximum flow. At this time the plume becomes smaller and less distinct and appears to be driven northward by the prevailing winds (Barnes et al. 1957; Anderson et al. 1961, 1962; Budinger et al. 1963).

Water from the Columbia River is used to cool the nuclear reactors at Hanford, Washington. The intense neutron flux in the reactors activates certain trace elements in the coolant water. Stable zinc-64, for example, captures a neutron to become radioactive zinc-65. An estimated 38 curies per day of Zn^{65} passes Vancouver, Washington, en route to the sea. This rate of replenishment and the 245-day half-life of Zn^{65} lead to an equilibrium value of 14,000 curies of this isotope in the Pacific ocean as a result of Hanford activities (Nelson ed. 1961).

Zinc-65 has been found in sessile organisms along the Oregon-Washington coasts (Watson et al. 1961), in marine plankton at some distance from shore (Osterberg 1962, 1963), and in marine sediments near the Columbia River (Osterberg et al. 1963b). Zinc-65 in sea water is more difficult to detect because of the low concentrations that exist. It is the affinity of marine

organisms for zinc that makes them useful monitors of Zn^{65}.

The constant low-level radioactivity in the Columbia River and the inherent sensitivity of radiotracer techniques invite an attempt to trace river water at sea by using marine animals which concentrate Zn^{65} to readily measurable levels. If Zn^{65} is a characteristic unique to Columbia River water, and if marine organisms accurately reflect the radioactivity of their environment, then Zn^{65} in animals should be a good indicator of the presence of Columbia River water. This hypothesis is explored, using Euphausia pacifica as a monitor of Zn^{65}. Zinc-65 concentration in this abundant macroplankter is correlated with salinity distribution in an effort to delineate the seasonal distribution of Columbia River water in the ocean.

METHODS

Euphausiids were collected by using a six-foot Isaacs-Kidd midwater trawl at three east-west series of stations extending 5 to 165 nautical miles offshore from the Columbia River (Astoria), Newport, and Coos Bay. In one case, an additional series was made off Brookings, Oregon. Oblique tows from 200 meters to the surface, averaging 30 minutes duration, were made at night.

The bulk of the samples usually consisted of euphausiids, with Euphausia pacifica predominating. Of the 194 midwater trawl collections throughout the period of study, 132 contained sufficient E. pacifica for radioanalysis (i.e., at least 40 g. wet weight).

E. pacifica were separated from the preserved samples, freeze dried, and ground with mortar and pestle. Approximately 13 cc of the sample were

packed into 15 cc plastic spectrometer counting tubes, corked and sealed with paraffin. Samples were counted using General Electric's total absorption anticoincidence spectrometer (see Perkins et al. 1960) at Hanford Laboratories, Washington. Counting time was 30 minutes. Data reduction and statistical analyses were made using an IBM 1620 computer.

Filter samples from rivers were obtained with 5-inch diameter membrane filters (0.65 micron) and glass prefilters (Gelman Instrument Co.). Two filter units from each site were packed into each 15 cc counting tube. Samples (500 ml) of the cation exchange resins, which were used with river water, were counted in the well of a 9-inch crystal (Perkins 1961).

Variability of Zn^{65} in Euphausiids

Euphausiids concentrate Zn^{65} (Osterberg 1962). However, before comparing seasonal or geographic variations of Zn^{65}, some knowledge of the short-term variability of Zn^{65} in E. pacifica is desirable. Euphausiids were taken in a series of nine repeated tows over a 16-hour period at a station 50 miles off Newport, 11-12 April 1962. The mean radiozinc content was 11.0 picocuries/gram dry weight, with a standard deviation of 1.5 pc/g. This indicates a fairly consistent concentration of Zn^{65}, and variations which are small compared to seasonal or geographic trends.

The Columbia River as a Source of Zn^{65}

Before the Zn^{65} content of E. pacifica can be considered indicative of Columbia River water, it must be shown that fall-out Zn^{65} occurs in comparatively insignificant amounts in this region. Although Zn^{65} is not a fission product, it is associated with nuclear testing (Lowman 1960).

To check for Zn^{65} in euphausiids from areas relatively free from the effects of the Columbia River water, euphausiids found off Alaska and off San Diego and Los Angeles, California, were analyzed.[1/] Results show that a low-level background of Zn^{65} from fall-out does appear to exist. The existence of a widespread background of Zn^{65} has been further confirmed by its presence in a variety of foods in the eastern United States (Murthy et al. 1959), in fish and prawns in India (Chhabra and Hukkoo 1962), and in polar cod (Boreogadus saida) from Arctic waters (Osterberg and McCauley lab notes). However, the quantity of Zn^{65} normally occurring off Oregon far exceeds that found in other areas of the world, except near nuclear testing sites.

Additional evidence on the relative lack of fall-out Zn^{65} is furnished by comparisons of the radioactivity in the Columbia and Willamette Rivers, which join near Portland. Fission products were present in the particulate matter filtered from the waters of both rivers, but Zn^{65} was readily detectable only in the Columbia River (Fig. 1). There is a possibility that fall-out Zn^{65} might be in a form chemically different from the radiozinc induced in the Columbia River. Therefore, filtrates from both rivers were passed through cation exchange columns (Dowex-50) to remove Zn^{++} in solution in the water. This experiment also showed that the Zn^{65} peak was prominent only in the spectrum of Columbia River water. Since the Willamette River (and all of the other Oregon rivers that we have sampled[2/]) is relatively free from Zn^{65}, we conclude that most of this isotope found in the Oregon coastal region was produced at Hanford. This situation is

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1/ We thank Frank Hebard, U.S. Fish and Wildlife Service, for the Alaska sample, and Edward Brinton, Scripps Institution of Oceanography, for those collected off California.

2/ Nehalem, Nestucca, Siletz, Suislaw, Umpqua, Marys, N. Santiam, S. Santiam, Yamhill, Clackamas, Deschutes, Malheur, Owyhee, Burnt, Snake, John Day, Metolius, Rogue, Coquille, Alsea, and McKenzie Rivers.

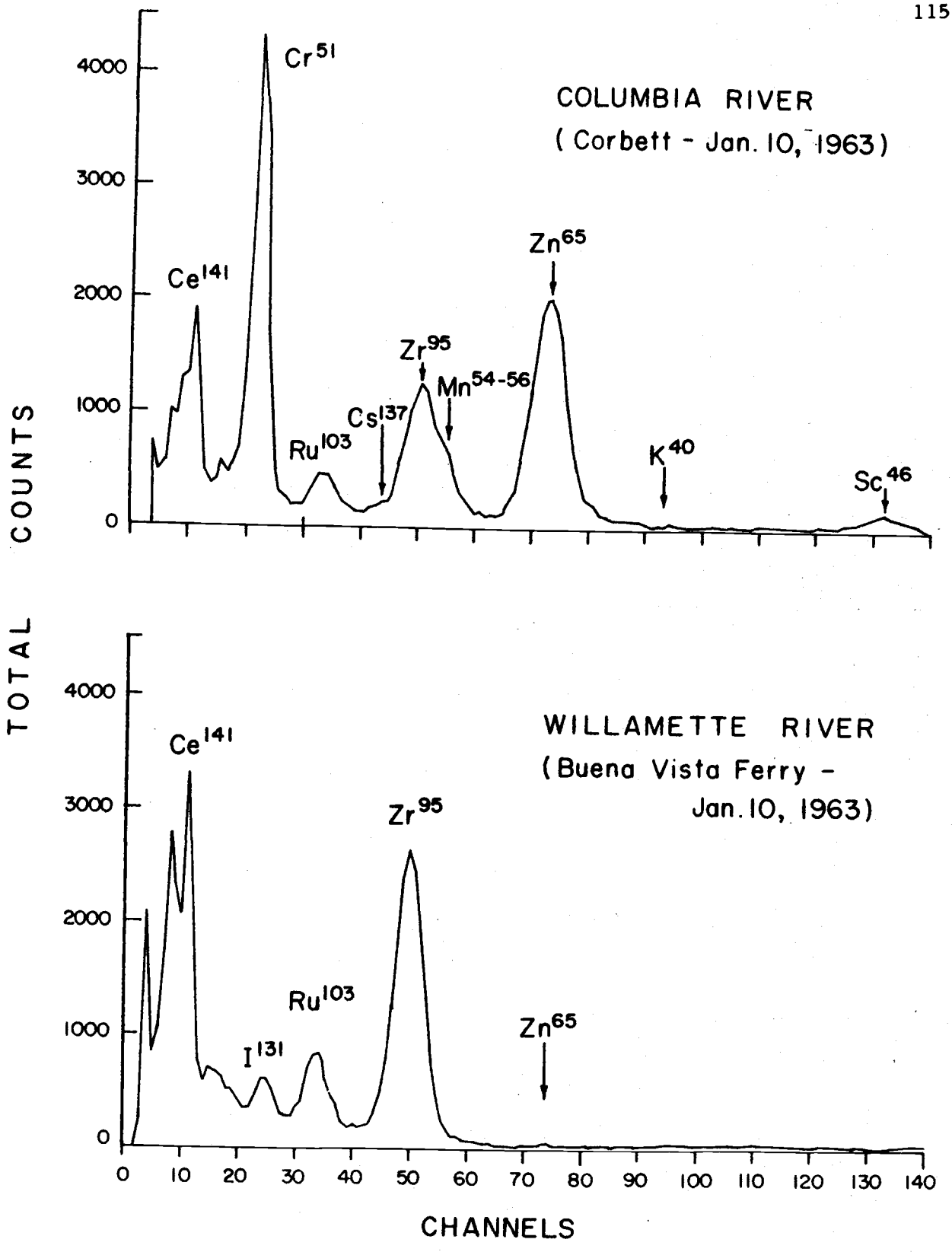

Figure 1. Gamma-ray spectra of particulate material in two Oregon rivers. Note chronium-51 and zinc-65 peaks in the Columbia River spectrum.

subject to change, however, since each nuclear detonation is a possible new source of fallout Zn^{65}.

RESULTS

Zn^{65} in Euphausiids and Related Salinity Distributions

If Zn^{65} in euphausiids reflects the distribution of Columbia River water at sea, then a negative correlation between Zn^{65} and surface salinity is expected. Figures 2-6 show amounts of Zn^{65} in euphausiids and salinity distributions at approximately the time when the euphausiids were collected.

During July-August 1961 (Fig. 2), Zn^{65} was more abundant in euphausiids from Newport than in the sample from Astoria, at the mouth of the Columbia River. Lower values, but relatively uniform with distance from shore, were found in euphausiids taken off Coos Bay. The salinity data, taken a month earlier, indicate a tongue of relatively fresh surface water extending at least as far south as the California border (salinity 32.5 $^{o}/_{oo}$). This shallow lens of water is the plume of the Columbia River (Anderson et al. 1962, Budinger et al. 1963).

During November 1961 (Fig. 3), low Zn^{65} values were found everywhere except at the station nearest the mouth of the Columbia River. Seaward from this station the Zn^{65} content of euphausiids decreased markedly. In addition, close to shore there was a gradual decrease in radioactivity to the south. Again, our salinity data, as well as those of Anderson et al. (1962), indicate that the Columbia River plume extended south of Coos Bay.

Figure 4 shows that in January Zn^{65} was still present in appreciable quantities in euphausiids taken off Newport, but values at stations off Coos Bay, particularly near shore, were lower than in November. The low

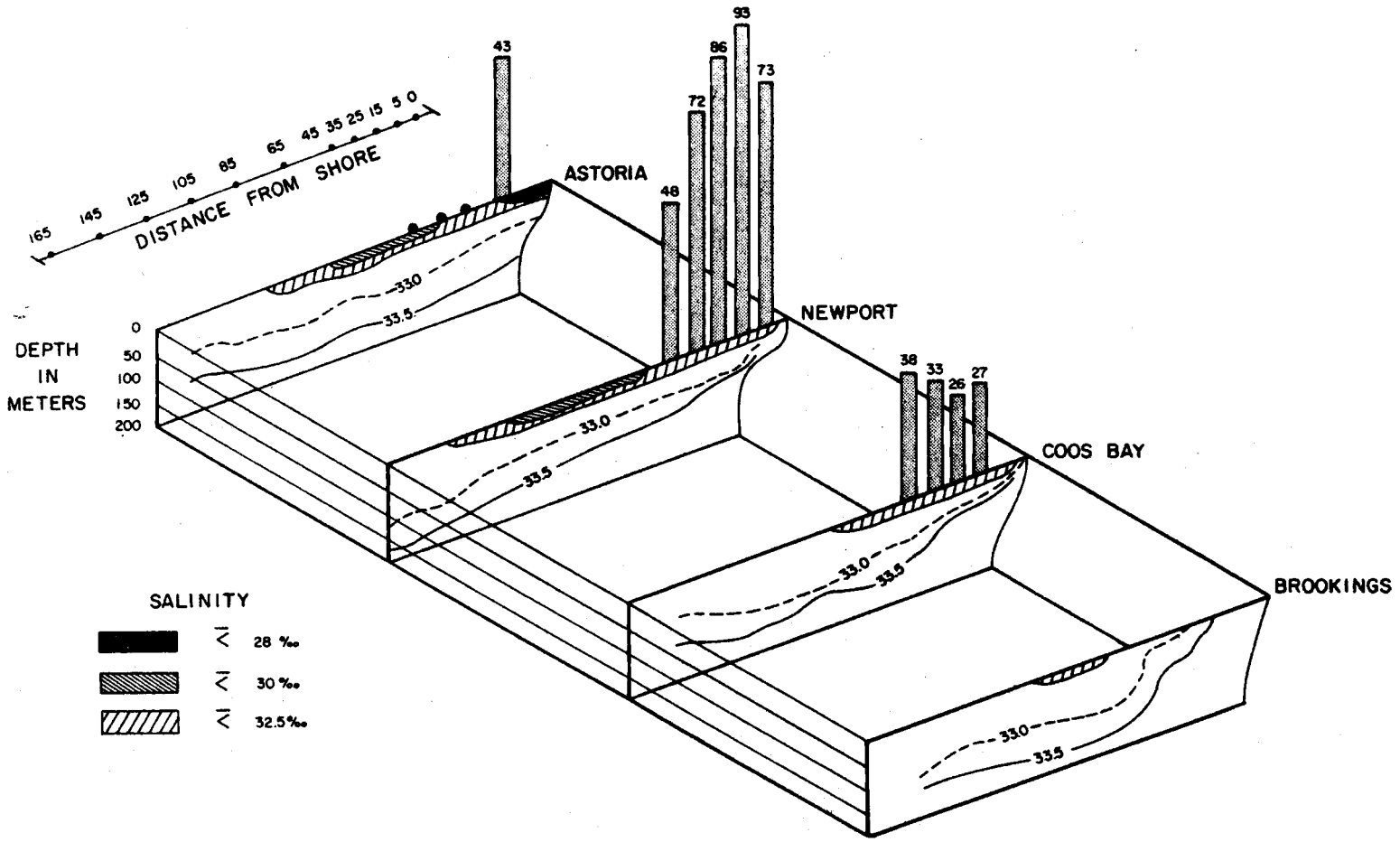

Figure 2. Zinc-65 in euphausiids (bars) and salinity distribution (contours) during summer, 1961 (see text for dates). Numbers above bars indicate picocuries per gram dry weight of euphausiids. All stations where euphausiids were collected are indicated by bars or dots. Dots show stations where euphausiids were insufficient for radioanalysis. In no case where analysis was possible was Zn^{65} absent.

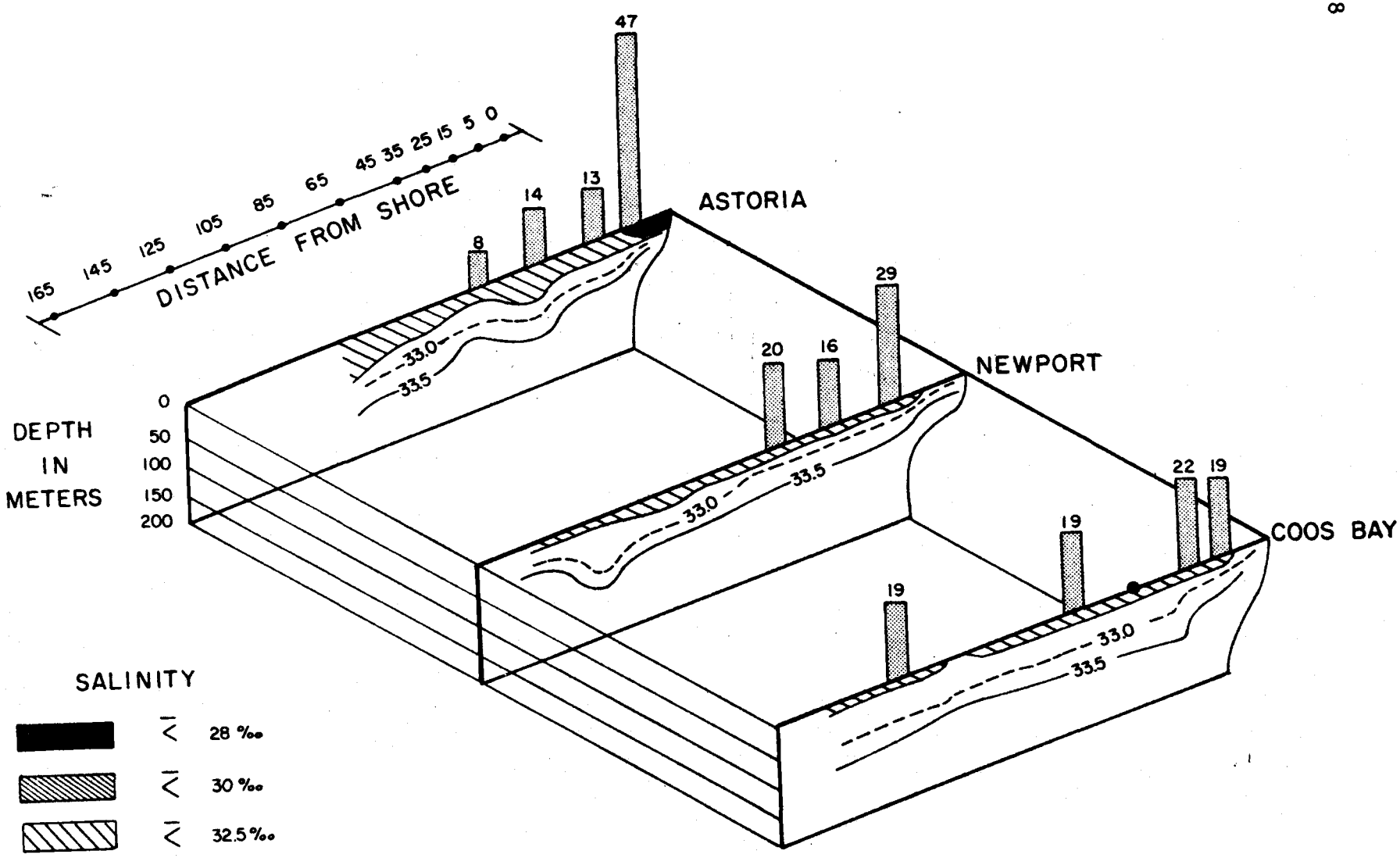

Figure 3. Same as Figure 2 for November 1961.

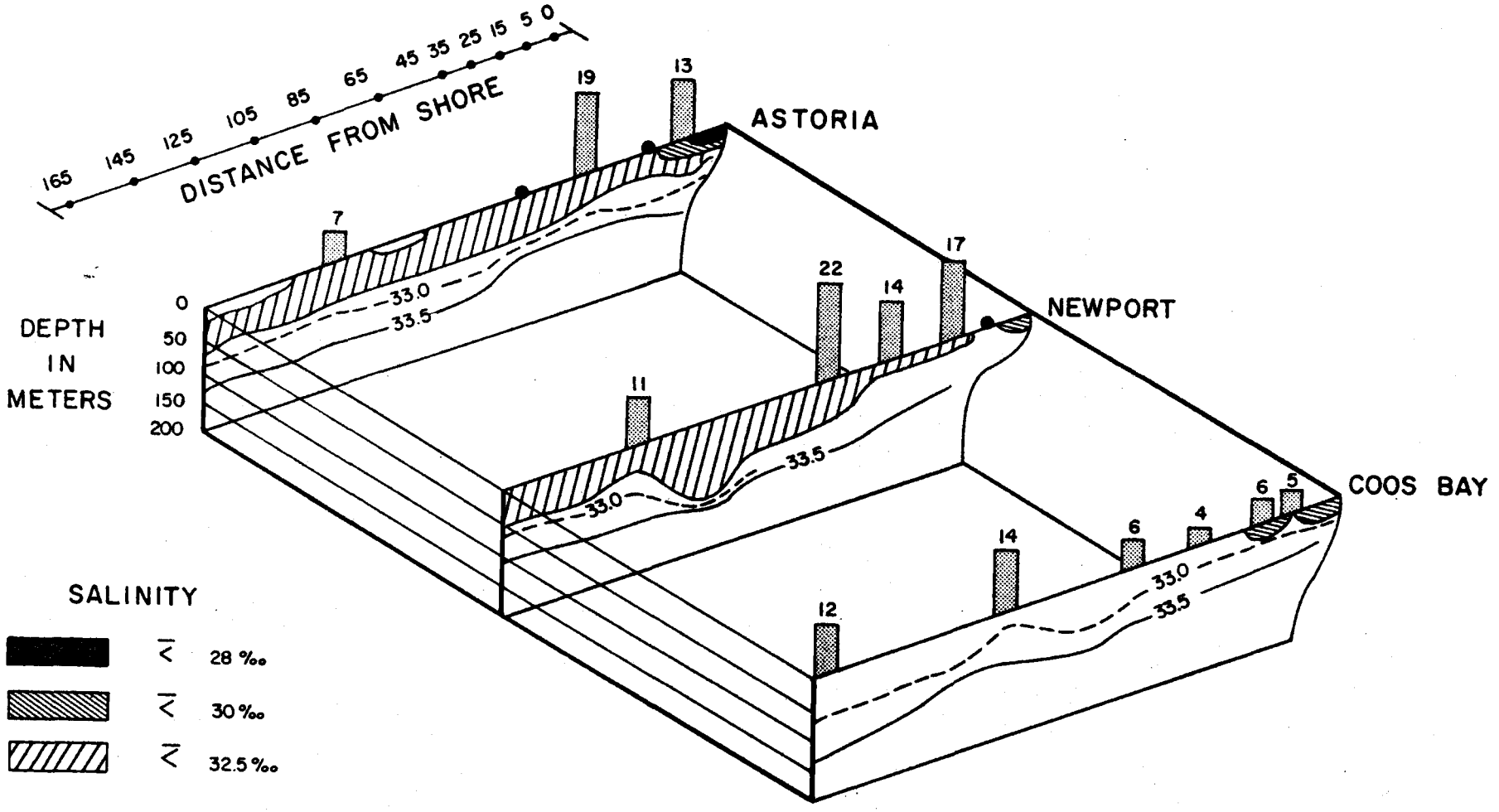

Figure 4. Same as Figure 2 for January 1962.

salinity of this inshore water was undoubtedly due to increased winter runoff from the nearby Umpqua and Coos Rivers, rather than to the Columbia plume. Zinc-65 values at the four stations nearest shore off Coos Bay may also have been reduced by admixture of water from the south (Davidson Current) which occurs frequently in this area at this season, whereas, water further offshore continues to flow from the north (H. O. Publ. 570 1947; Sverdrup et al. 1942; Anderson et al. 1962).

Anderson et al. (1962) point out that prevailing winds in winter probably carry the Columbia plume northward. However, low salinity and relatively high Zn^{65} values off Newport during January indicated the continued influence of plume water in this area. On the other hand, euphausiids from 125 miles off Astoria contained little Zn^{65}, although low salinity water was present there. We believe that this low salinity water off Astoria was not from the Columbia River, but instead was part of the general low-salinity belt (modified Sub-Arctic Water) extending to the Gulf of Alaska (Anderson et al. 1961). This conclusion is based both on the low Zn^{65} value and on the fact that winds that would drive the plume westward are absent.

Exceptionally high values of Zn^{65} were found during March-April 1962 very near the mouth of the Columbia River (Fig. 5). These high Zn^{65} values occurred at a time when the plume was "pooled" close to the mouth of the river by several months of strong winds from the south-west (Anderson et al. 1962). High radioactivity off Newport seems to be associated with water fresher than 32.5 $^{o}/_{oo}$ in spite of the fact that the Columbia River plume is not thought to extend this far south during the spring (Anderson et al. 1962).

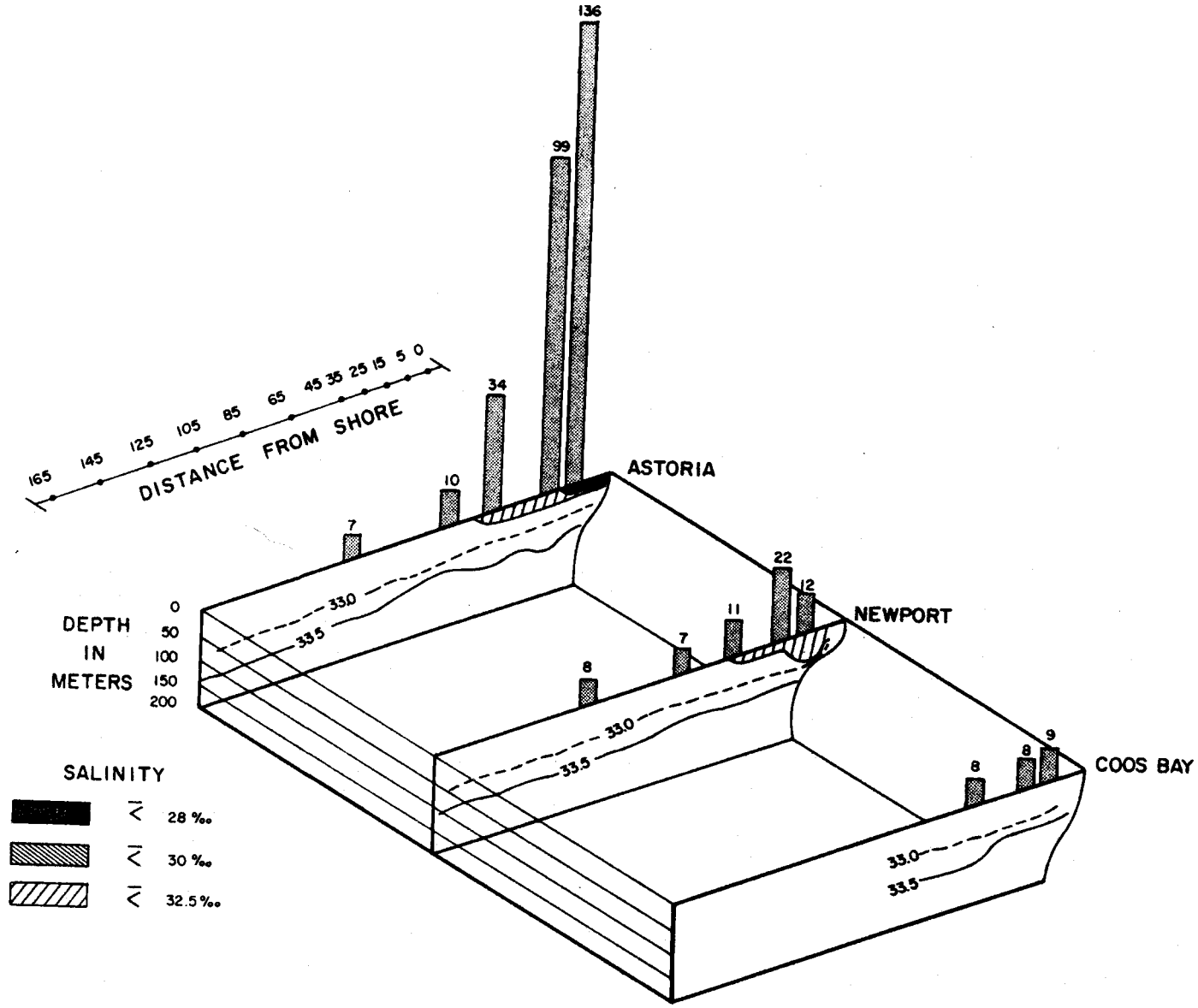

Figure 5. Same as Figure 2 for March-April 1962.

Figure 6 (July-August 1962) represents the same season as Figure 2. Zn^{65} was again lower off Astoria than off Newport or, during this year, even Coos Bay. Relatively high values of Zn^{65} in euphausiids were found off Brookings in the tongue of fresher water offshore, which again seems to be part of the Columbia River plume. Much lower Zn^{65} values were found in the four stations nearest the coast where the salinity was almost 34 $^{o}/_{oo}$ (undoubtedly due to upwelling).

In summary, the maximum value of Zn^{65} in E. pacifica, was found off the mouth of the Columbia River in spring, 1962, (Fig. 5). However, large amounts were found off the central coast of Oregon throughout the year. In summer, these high values were from euphausiids taken in the area of the Columbia River plume, as indicated by salinity measurements. It is surprising that values of Zn^{65} were sometimes higher off central Oregon than off the mouth of the Columbia River (Fig. 2, 4, and 6). Moreover, relatively high concentrations were found off southern Oregon as much as 250 miles from the Columbia (Fig. 6). It was particularly unexpected to find that seasonal variations in the Zn^{65} in euphausiids were not great, and that comparatively high values were found off Oregon in the winter, when the plume presumably extends northward and is absent off the Oregon Coast.

Zn^{65} as an Indicator of Columbia River Water

In the absence of fallout Zn^{65}, the Zn^{65} which we measured in euphausiids must be derived from Hanford activities, i.e., the Columbia River water. In interpreting the distributions of euphausiids containing Zn^{65}, and the disparity between this and the location of the Columbia River

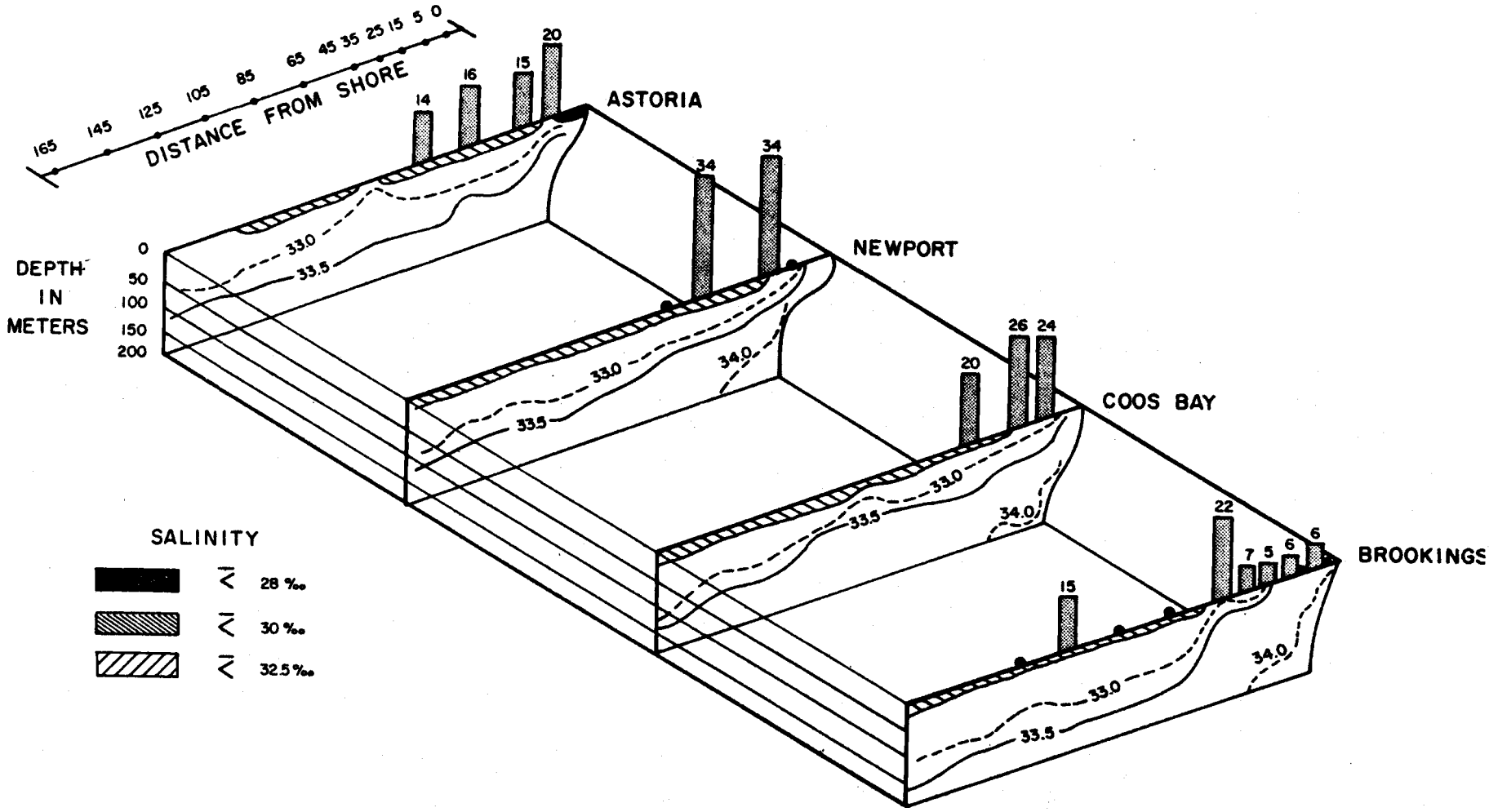

Figure 6. Same as Figure 2 for July-August 1962.

plume, we consider the following physical and biological factors: (1) variations in the amount of Zn^{65} introduced into the Columbia at Hanford, (2) amounts of Zn^{65} reaching the ocean, (3) losses at sea due to nuclear decay, (4) currents affecting the movement of the plume, (5) effect of currents and vertical migrations on transport of Zn^{65}, (6) half-life of Zn^{65} in the mixed layer, and (7) uptake and retention of Zn^{65} by euphausiids.

(1) Variations in the Amount of Zn^{65} Introduced into the Columbia at Hanford

The quantity of Zn^{65} entering the Pacific Ocean undoubtedly varies, but data are not available on the variations in the amount of Zn^{65} released by the reactors. Recent reports indicate that improved techniques have reduced Zn^{65} output by about 25 percent (Schneller 1962), but the date of this change was not given. However, equilibrium values of Zn^{65} in the Pacific Ocean for 1961 (Nelson, 1962) are unchanged from the previous year (Nelson, 1961), which indicates that this reduction occurred after 1961. Perhaps the lower values off Newport in the summer of 1962 (Fig. 6), compared with the summer of 1961 (Fig. 2), reflect this reduction.

(2) Amounts of Zn^{65} Reaching the Ocean

Because of the transitory nature of Zn^{65}, the amount which reaches the ocean will be affected by the time it takes water to flow from Hanford to the sea. Dams on the Columbia, by increasing this time, allow more decay and thus tend to reduce the output of Zn^{65} into the ocean. Another factor is uptake by the biota of the river (Davis and Foster, no date). We have no quantitative estimates of these complex factors, but believe their bearing on the problem of tracing the Columbia River is negligible

compared with other uncertainties.

(3) Losses at Sea Due to Nuclear Decay

If only physical decay is considered, half of the Zn^{65} is lost in 245 days. However, steady state conditions for Zn^{65} exist in the ocean off Oregon in that decay is approximately balanced by replenishment of Zn^{65} from the Hanford operations (Nelson 1961). But as the plume of the river flows southward in the ocean (as it appears to do in the summer), decay will result in a gradual decrease in radioactivity with time and distance. If this were the only loss, the transit time of the water could be computed by measuring the change in Zn^{65} concentration in the water.

Let us assume for the moment that measurement of Zn^{65} in euphausiids is equivalent to measurement of Zn^{65} in water. The spatial and temporal changes in Zn^{65} concentration in euphausiids off Astoria are very large. Therefore, interpretation of this series of samples would be difficult. However, Zn^{65} values for the Newport and Coos Bay series are less variable and more confidence can be placed in averages. The average of the Coos Bay series was always lower than that of the Newport series. If the lower amounts of Zn^{65} at Coos Bay are attributed entirely to nuclear decay as the water flows from north to south, the travel time ranged from 34 days in November 1961 to 309 days in summer 1961.

Anderson et al. (1961) state that "the current drift (of the southerly-setting California Current) as computed from the dynamic topography of the sea surface is small, with average values of the order of 5 cm/sec or less and maxima up to 20 cm/sec (10 miles per day) in the spring and

summer." Zn^{65} carried at these rates by the southward drift would cover the distance between Newport and Coos Bay in about 8 to 40 days; this is considerably less than the times computed from the rate of physical decay of Zn^{65}.

This leads us to conclude that, although radioactive decay is undoubtedly a factor effecting a decrease in Zn^{65} concentration as the water flows from the latitude of Newport to that of Coos Bay, it cannot be the only or even the most important factor.

(4) Currents Affecting the Movement of the Plume

The plume itself is a very shallow phenomenon and may be thought of as a thin anticyclonic gyre embedded in the prevailing or "steering" surface currents. Currents within 100 miles of the Oregon coast are generally southward in summer, and, less consistently, northward in winter (H. O. Publ. 570 1947). The outer edge of the plume generally lies in the prevailing southward drift. Most of these currents are weak with speeds of less than half a knot. In addition to the general coastal drift, offshore motion of the upper layers is present during summer when upwelling occurs along the coast (Pattullo et al. in prep.).

Consider the behavior of a hypothetical plume that leaves the mouth of the Columbia as a discrete discharge during the month of June. It would drift southward (at about 5 cm/sec at the core), and seaward, undoubtedly spreading and becoming saltier with time. Zinc-65 concentration in the plume would be diluted by mixing with salt water, but, on the whole, would tend to remain near the surface. Eventually, the plume would disappear as a result of turbulent mixing.

Although grossly oversimplified, this general pattern agrees with the behavior of the plume as described by Anderson et al. (1961,-1962). The principal difference between the simplified "model" and the real river plume is that the flow is not a single burst in June but has some persistence during the summer months. Therefore, the plume body does not simply detach itself from the river as a fixed quantity of fresh water, but increases in freshwater content, as well as in area, during the summer. Winter mixing is such, however, that the plume completely loses identity each year and can be considered an annually recurring event. Occasionally a secondary maximum in Columbia River flow is observed during winter. This does not reinforce the old plume body off the Oregon coast, normally, but is driven northward along the Washington coast instead.

Surface drift is directly influenced, apparently, by winds. During summer the North Pacific high pressure area is well developed. The Oregon coast lies under the influence of the northwesterly winds at the northeastern edge of the high. During winter, this high retreats to the southwest and the southeastern sector of the Aleutian low frequently dominates the area; this leads to southerly winds. However, at any particular time the meteorological conditions may not conform to this generalized pattern, and deviations from prevailing flow can be expected to result. During winter southwesterly winds result in a "pooling" of river water along the shore adjacent to the mouth of the Columbia River (Anderson et al. 1962). At this season, river discharge is low and so is the area of the "pool" compared with the summertime pattern. However, the Zn^{65} content is high because of the high concentration of river water in a restricted area.

(5) Effect of Currents and Vertical Migrations on the Transport of Zn^{65}

If all the Zn^{65} released by the Columbia River were retained entirely within the plume, its study would involve only surface flow and transport. Since euphausiids and other vertical migrants are found to concentrate Zn^{65}, however, we must consider the motions of the euphausiids and the currents that affect them, as well as the currents which affect only the plume.

Euphausia pacifica is known to make daily vertical migrations (Esterly 1914, Tucker 1951, Barham 1956, Brinton 1962). Off Oregon, E. pacifica is abundant in catches from the upper 200 m at night. It is uncommon during the day, presumably descending into deeper water. This migration takes euphausiids well below the Columbia River plume water, and, in fact below the temperature-salinity gradients that separate the low density waters of the surface mixed layer from denser waters below. Thus, biological transport of zinc across the pycnocline by such animals as E. pacifica is undoubtedly more important than transport through this layer by physical processes (see Ketchum and Bowen 1958).

Direct measurements of currents indicate that the pycnocline (at 100 to 200 m) is a region of velocity shear. Recent data show that throughout the year surface waters have had southward velocity relative to the deeper waters (Pattullo et al. in prep.). Therefore, for vertically migrating animals such as euphausiids, which reside in deep waters during the day, the net drift to the south would be less than that of the plume. Hence an atom of Zn^{65} in a euphausiid would move less rapidly out of the local area than one floating freely in the plume, and concentration of Zn^{65} in euphausiids could be expected to lag behind the plume body.

Seasonal variations in subsurface currents also may affect euphausiid distribution. During wintertime, both subsurface (at least to 200 m) and surface flow are probably slightly northward (Pattullo et al. in prep.). This would tend to return to Oregon coastal areas euphausiids and Zn^{65} that had drifted southward during the summer. Consequently, the patterns of currents and vertical distributions of E. pacifica, both on a daily and seasonal basis, appear to reduce the dispersion of Zn^{65} from the local area off Oregon and may contribute to the absence of large seasonal variations of this radionuclide in euphausiids. The effects of vertical migrations on the horizontal distributions of Antarctic euphausiids have been described by Hardy and Gunther (1935) and Mackintosh (1937). They also show that both daily and seasonal vertical migrations may affect the retention of a population within a geographic area.

(6) Half-life of Zn^{65} in the Mixed Layer

Budinger et al. (unpublished) report that mixing of the plume with salt water occurs in such a way that radionuclides in the plume tend to remain in the surface layers. That is, salt water mixes vertically upward into the overlying plume, while little fresh water is lost through the resulting halocline. Diffusion occurs along the outer edges of the plume, and often "cells" of fresher water become separated from the plume (Anderson et al. 1962). Thus, radioactivity per unit volume of surface water in the plume decreases with distance from the mouth of the river even though little physical loss occurs vertically. Thus most of the Zn^{65} from the Columbia River is retained in the euphotic zone where it readily enters into food chains.

Dilution of river water at sea makes measurement of Zn^{65} extremely difficult, except in marine organisms, which concentrate the zinc. Most of our non-biological measurements have, therefore, been made in or near the Columbia estuary, where levels are much higher. There, Zn^{65} is found in several conditions: (1) as ionic zinc in the filtrate (concentrated on cation and chelating resins), (2) attached to particles (collected by filtering surface water through membrane filters), and (3) in organisms.

The concentration of ionic zinc is low and diminishes rapidly seaward, so that it is not generally detectable by our techniques beyond 25 miles from the river's mouth (Osterberg and Cutshall lab notes). Zn^{65} in or on particulate material can be detected farther at sea. A small but distinct Zn^{65} peak appeared in the spectrum of a filter through which had been passed 109 liters of surface sea water from 45 miles off Astoria. Zn^{65} in marine organisms remains easily detectable over the entire area of our observations. This indicates that much of the Zn^{65} is associated with organisms. Major loss from the mixed layer therefore may be caused by excretion and predation below the thermocline by animals undertaking diurnal migrations.

(7) Uptake and Retention of Zinc-65 by Euphausiids

The biological half-life of Zn^{65} in E. pacifica is not known. If it is long, as it is in some marine fish (Chipman et al. 1958), radio-zinc may be accumulated and retained for several months. This would help explain the small difference in Zn^{65} content between winter (when the plume is not evident) and summer (when the plume is obviously present).

Zinc is a constituent of several enzyme systems (Vallee 1957), and marine organisms probably have a physiological need for it (Joyner 1962). Metabolic demand for this element, which was the most common artificial gamma emitter in marine animals off the Oregon coast (Osterberg et al. in press), may be greater during certain seasons of the year, or for certain stages in the life history of euphausiids than at other times. Davis and Foster (no date) show that fluctuations in the radioactivity of freshwater minnows (Richardsonius balteatus), for example, were strongly temperature-dependent. They attribute the 75-fold increase in concentration of radioisotopes in the fish between winter and late summer to different metabolic requirements, due largely to temperature changes.

High Zn^{65} values near the mouth of the Columbia River in spring and somewhat increased values in the fall correspond roughly to spring and fall plankton "blooms." E. pacifica grazes on phytoplankton (Ponamereva 1954), from which it appears to obtain Zn^{65} (Osterberg et al. 1963a). Thus, a relationship is suggested between phytoplankton abundance and increased Zn^{65} content in euphausiids. This relationship may be due either to increased Zn^{65} content per unit of phytoplankton during rapid growth of the population, or to increased grazing rates by the euphausiids.

Foster and Davis (1955) state that freshwater diatoms reach equilibrium with reactor effluent water in about an hour. Marine diatoms also appear to concentrate radionuclides rapidly, since maximum Zn^{65} values are found in filter samples from nearest the mouth of the Columbia River (Osterberg and Curl lab notes). Nevertheless, the highest Zn^{65} content in euphausiids was generally found off Newport, well to the south. This suggests that radioactivity of euphausiids is dependent on the time that

the animal has spent in water containing Zn^{65}. That is, a long period is required for maximum concentration to be reached in the euphausiid.

In contrast, near the mouth of the Columbia River, Zn^{65} content of mussels (Mytilus spp.) was found to be generally higher than that of euphausiids, but a similar comparison at Coos Bay showed less in mussels (Watson et al. 1961). The smaller gradient of Zn^{65} concentration in euphausiids with distance is probably typical of plankton which drift with the moving water. On the other hand, the water surrounding sessile organisms is continually renewed with changes of wind and tide.

CONCLUSIONS

Surface salinity patterns off the Oregon coast change in response to the position of the Columbia River plume, which in turn is influenced by a seasonal change in prevailing winds. While Zn^{65} is shown to be associated with the Columbia River, seasonal variations of Zn^{65} in euphausiids are not marked, and the amount of Zn^{65} does not diminish greatly in the absence of the plume in winter. Vertical stability in the ocean in this area (which generally precludes mixing to great depths), seasonal current reversal and the migratory habits of euphausiids appear to maintain high levels of Zn^{65} in euphausiids throughout the year. A long biological half-life of Zn^{65} would also help maintain high Zn^{65} levels in the winter.

Maximum concentration of Zn^{65} in euphausiids occurs close to the mouth of the river in spring when the river's effluent is driven inshore and "pooled" by winds from the southwest. Otherwise levels are generally higher off Newport, to the south, perhaps due to a longer residence time

in plume waters and time lag in Zn^{65} incorporation. Levels are lower off Coos Bay due to dilution and spreading of the plume and to decay of Zn^{65} Two prominent local phenomena, the inshore wintertime current from the south (Davidson Current) and summertime upwelling off Brookings, are both reflected by lower Zn^{65} concentrations in euphausiids from those areas.

The great affinity of marine organisms for zinc and sensitivity of modern gamma-ray spectrographic techniques make Zn^{65} in euphausiids easy to measure. Unfortunately, however, use of biological organisms as monitors introduces many uncertainties. The most important is that we do not know how accurately the radioactivity of the euphausiid reflects the radioactivity of his immediate environment. This difficulty is compounded if variations in stable zinc occur; local variations seem likely because of the affinity of marine organisms for zinc. However, no comparable data exist for stable zinc.

Another approach to the problem might be through the study of a radioisotope that is not utilized extensively by marine organisms and thus would be a more conservative indicator of plume waters. Chromium-51 meets this requirement and furthermore is even more abundant in Columbia River water than is Zn^{65} (Fig. 1) (Nelson 1961). However, this lack of biological concentration of Cr^{51} makes it especially difficult to measure, except near the mouth of the river before much dilution occurs. Nevertheless, we are striving to improve our techniques so that radioactive elements in the Columbia River can become a more useful tool in physical and biological studies of the plume and its effects in the Oregon coastal region. It is this low-level radioactivity which distinguishes the plume from other sources of fresh water in the area, and seems to offer the most promise as a tracer of the river water at sea.

ACKNOWLEDGMENTS

We thank R. W. Perkins of General Electric's Hanford Laboratories for allowing us to use his equipment, and for his assistance with certain technical aspects of the gamma-ray spectrometry. We are indebted to Harriet Van Arsdale, I. Lauren Larsen, Norman Cutshall, and the captain and crew of the R/V ACONA for their work in the laboratory and at sea. We gratefully acknowledge the use of salinity data made available to us by Bruce Wyatt.

This research was carried out under grants AT(45-1)1750 and AT(45-1)1726 with the Atomic Energy Commission, Nonr 1286(02) with the Office of Naval Research, and grant GP-622 with the National Science Foundation.

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VERTICAL DISTRIBUTION OF ZINC-65 AND ZIRCONIUM-95--NIOBIUM-95 FROM OCEANIC ANIMALS[1]

William G. Pearcy and Charles L. Osterberg

ABSTRACT

Zn^{65} per gram wet weight was approximately the same in small oceanic animals collected at depths of 0-150, 150-500, 500-1000 m, but Zr^{95}-Nb^{95} was about five times more concentrated in animals above 150 m than in deeper water. Total amounts of Zn^{65} and Zr^{95}-Nb^{95} in animals filtered from 1000 m^3 water were highest near the surface during the night; during the day Zn^{65} was highest at 150-500 m.

Variations in concentrations of radionuclides indicate that much of the measured radioactivity was incorporated in animals that undertake daily vertical migrations across the region of the halocline at 150 m.

More radioactivity was found in animals from a column of water 1000 m deep in night samples than in day samples, which means that both changes in availability of animals and vertical migrations are important considerations in interpreting the vertical distribution of radionuclides at sea.

INTRODUCTION

The ocean off Oregon contains two classes of radionuclides; fallout isotopes such as Zr^{95}-Nb^{95} which are fission products, and neutron induced isotopes such as Zn^{65} and Cr^{51} which are produced principally by nuclear reactors on the Columbia River. Although both types of radionuclides initially enter surface waters of the ocean either by atmospheric fallout or in the fresh water effluent of the Columbia River, variations in concentration of isotopes with depth have not been studied. Some marine organisms off Oregon, including animals that migrate vertically (Pearcy 1964 and in preparation), accumulate Zn^{65} and Zr^{95}-Nb^{95} (Osterberg et al. 1964). Daily vertical migrations provide an obvious mechanism whereby radionuclides may be transported across density gradients at rates faster than physical processes alone would permit (Ketchum and Bowen 1958).

This examination of differences in concentrations of Zn^{65} and Zr^{95}-Nb^{95} in small oceanic animals collected from various depths and during day and night periods is preliminary. Since signing of the nuclear test-ban, Zr^{95}-Nb^{95}, because of their short half-lives, are no longer present in reliably measurable quantities in pelagic animals, so repetition of this study is unlikely.

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[1]This research was supported by the Atomic Energy Commission Contracts AT(45-1)1726 and AT(45-1)1750. Ship operations were sponsored by grants from the National Science Foundation and the Office of Naval Research.

METHODS

Animals were collected with an Isaacs-Kidd midwater trawl with a Multiple Plankton Sampler as an opening-closing cod-end unit. This provided samples from three depth intervals: 0-150 m, 150-500 m, 500-1000 m (Pearcy in press). Volume of water filtered was estimated from the cross-sectional area of the mouth of the trawl and distance towed at each depth interval as determined from geographic fixes (Pearcy in preparation). A single mesh size (12 mm stretched measure) was used as a liner throughout the body of the trawl. Two tows during the day and two during the night provided material for radioanalysis. Tows were made 50 miles off the central Oregon coast (in the vicinity of 44° 39'N, 125° 15'W) during 7-8 January, 1963.

Samples from each depth interval, consisting of mixed-species collections (mostly mesopelagic fishes, crustaceans, and chaetognaths), were drained to remove excess fluids, weighed (wet-preserved weight), ashed at 450°C, and packed in 15 cc plastic counting tubes, reweighed and sealed. Samples were analyzed in the well of a 5x5-inch Harshaw NaI(Tl) crystal used with a gamma-ray spectrometer (512 channel Nuclear Data Model 130 A) according to the procedures of Perkins (1958). Counting time was either 100 or 400 minutes, depending on sample activity. Counts were converted to pico-curies per gram wet weight, per gram ash weight, and per 1000 m^3 water filtered.

RESULTS

Data on the activity of Zn^{65} and Zr^{95}-Nb^{95} per gram of ash weight and wet weight are given in Table 1. Total variations in the concentration of Zn^{65} in the oceanic animals ranged from 4.7 to 19.4 pc/g ash and 0.2 to 1.0 pc/g wet weight. Concentrations of Zr^{95}-Nb^{95}, on the other hand, showed a wider total variation, 3.2 to 121.2 pc/g ash and 0.2 to 3.2 pc/g wet weight.

Differences at various depths are apparently in the activity of these radionuclides. Zr^{95}-Nb^{95} per gram of animal was always highest in the 0-150 m sample. Average concentration in samples from three depth intervals (Table 2) shows that animals collected below 150 m contained only one-fifth to one-ninth the amount of Zr^{95}-Nb^{95} per gram as surface animals. Zn^{65}, however, demonstrated surprisingly little variation with depth in the upper 1000 m. Deep-water (500-1000 m) animals contained almost as much Zn^{65} per gram as animals collected in surface waters. Undoubtedly some differences between wet weight and ash weight ratios for a depth interval are due to types of animals collected. For example, surface catches during daylight periods often contain a higher percentage of gelatinous or watery forms (e.g., medusae, salps) than those collected after dark. The latter contained more animals with relatively high ash weight (e.g., fishes, euphausiids). Hence, ash weight/wet weight ratios for day samples may be lower than those for night samples.

Table 1. Picocuries of (A) Zn^{65} and (B) Zr^{95}-Nb^{95} per g of animals, and (C) weight of animals per 1000 m^3 water filtered at three depth intervals, 500 miles off Newport, Oregon, 7-8 January 1963

	Depth Interval (m)	DAY				NIGHT			
		Sample 1 0753-1102 hr PST		Sample 2 1249-1612 hr PST		Sample 3 1822-2134 hr PST		Sample 4 2358-0309 hr PST	
(A)									
Zn^{65}/g wet wt.	0-150	0.4		0.2		0.9		0.8	
	150-500	0.8		0.8		0.4		0.7	
	500-1000	0.6		0.3		0.3		1.0	
Zn^{65}/g ash wt. (Std. dev.)	0-150	19.4	(8.5)	4.7	(2.6)	18.0	(0.9)	16.4	(0.4)
	150-500	19.1	(0.4)	24.2	(0.6)	19.9	(0.5)	12.8	(0.4)
	500-1000	9.0	(0.6)	5.4	(0.6)	6.7	(0.5)	15.4	(0.6)
(B)									
Zr^{95}-Nb^{95}/g wet wt.	0-150	2.8		2.3		3.2		1.3	
	150-500	0.2		0.4		0.2		0.4	
	500-1000	0.2		0.2		0.3		1.0	
Zr^{95}-Nb^{95}/g ash wt. (Std. ev.)	0-150	121.2	(17.3)	58.0	(3.2)	62.5	(2.3)	25.1	(0.4)
	150-500	5.3	(0.3)	13.7	(0.5)	8.6	(0.4)	7.7	(0.3)
	500-1000	3.2	(0.6)	3.8	(1.3)	7.0	(0.6)	15.7	(0.6)
(C)									
g wet wt/1000 m^3	0-150	1.1		1.2		6.7		14.0	
	150-500	6.9		8.0		8.6		7.3	
	500-1000	1.7		3.3		2.6		2.3	

Table 2. The relative content of Zn^{65} and Zr^{95}-Nb^{95} from each depth interval with respect to the content in animals from 500-1000 m.

	Zn^{65}		Zr^{95}-Nb^{95}	
Depth (m)	Ash Wt.	Wet Wt.	Ash Wt.	Wet Wt.
0-150 m	1.6	1.1	9.0	5.4
150-500 m	2.1	1.2	1.1	0.7
500-1000 m	1.0	1.0	1.0	1.0

Vertical distribution of Zn^{65} and Zr^{95}-Nb^{95} in animals filtered per 1000 m^3 of water, a function of the biomass of animals and their radioactivity per gram, is shown as histograms in Figure 1. Concentration of radionuclides were calculated as follows:

$$\text{pc/1000 m}^3 = \frac{\text{(pc/g ash) (total g ash)}}{\text{1000 m}^3 \text{ water filtered}}$$

The striking feature of the histograms is the marked day-night difference between concentrations in surface waters. The amount of both Zn^{65} and Zr^{95}-Nb^{95} in animals collected after dark is highest at the surface and is progressively lower at mid-depths and in deep-water. During daylight periods, concentrations of both radionuclides are lower at the surface than at night. During daylight, Zn^{65} was highest at mid-depths and lower at the surface than any other depth. Similar enhancement of Zr^{95}-Nb^{95} at mid-depths during the day is not evident.

Differences between vertical concentrations of these radionuclides may result from variations of radioactivity in the animals or from variations of radioactivity in the animals or from variations of biomass, or both. The fact that biomass is substantially larger in 0-150 m night collections (Table 1) explains most of the nocturnal increase in radioactivity in this layer. Zn^{65} content per gram wet weight in 0-150 m night samples is about twice that of day samples and contributes to the order of magnitude increase in radiozinc. Increased Zn^{65} activity at mid-depths during the day, however, is not explained by increased biomass, but rather by higher activity per gram of animals.

DISCUSSION

Diel or diurnal vertical migration of animals, a well-known phenomenon for oceanic animals, largely explains the observed day-night differences in depth variations of Zn^{65} and Zr^{95}-Nb^{95}. Such migrations by mesopelagic animals off Oregon have been evidenced by increased catches in surface water at night and concomitant decreased catches at mid-depths (Pearcy 1964, in preparation). Occurrence of animals with highest Zn^{65} activity at mid-depths during the day but at surface during the night may be interpreted as evidence for such migrations. Thus the histograms indicate that a considerable portion of the measured radioactivity is incorporated in animals that migrate into the upper 150 m at night As the depth of the halocline is about 150 m, animals cause a daily flux of these radionuclides across this density discontinuity from surface waters into waters below the halocline.

To compare total amounts of these two radionuclides in animals collected during day and night periods, estimates of Zn^{65} and Zr^{95}-Nb^{95} were calculated per m^2 in a column of water 1000 m deep. The amount of Zn^{65} was 2.6 and 2.8 pc/m^2 in organisms collected during the day and 2.7 and 4.6 pc/m^2 in those collected during the night. Zr^{95}-Nb^{95} was 1.2 and 2.1 pc/m^2 during the day but 4.3 and 3.3 pc/m^2 during the night. Greater quantities of radionuclides in night collections are related to larger biomass in night samples (Table 1). Apparently some animals more effectively avoid or escape the net during daylight hours than at night (Pearcy, in preparation).

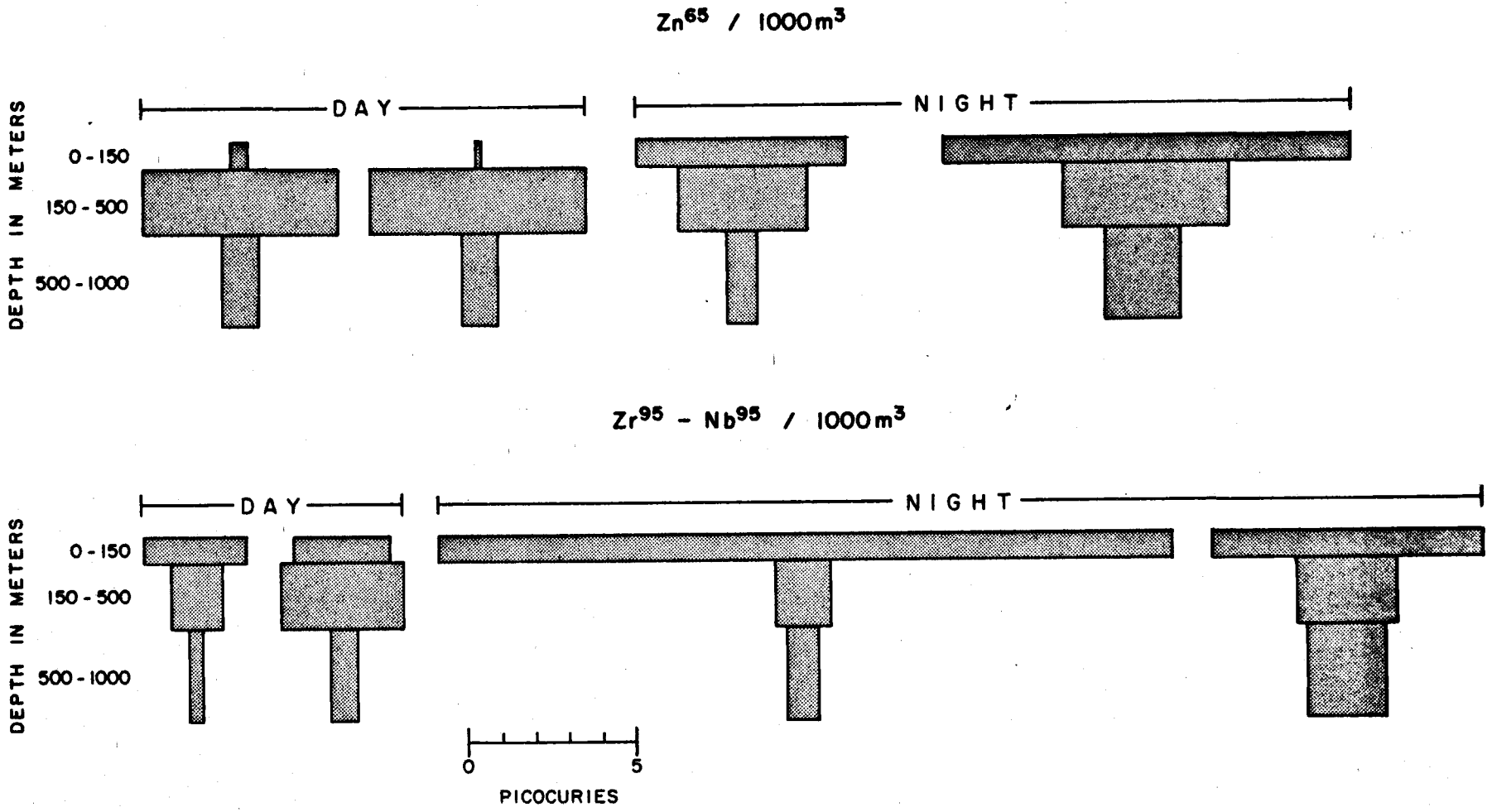

Figure 1. Zn^{65} and Zr^{95}-Nb^{95}. Activity of oceanic animals at three depth intervals for the day and night periods, 7-8 January 1963, 50 miles off Newport, Oregon.

Absence of an increase in Zr^{95}-Nb^{95} at mid-depths during the day (Fig. 1) may be due to day-night differences in catches of herbivores, which have been found to accumulate particulate fallout more than carnivores (Osterberg et al. 1964). However, since Chipman (1958) found that another fission product, Ce^{141}, was rapidly excreted by copepods, there is the added possibility that some of the day-night variations of Zr^{95}-Nb^{95} may be caused by actual changes in amounts of these radionuclides in the animals. Zr^{95}-Nb^{95}, which probably passes directly through the digestive tract, may occur in relatively high amounts in animals that actively graze in upper waters at night. Lower amounts may remain during the day after some fallout has been excreted.

Osterberg et al. (1964) found that Zn^{65}, which is believed to be metabolically active with a long turnover time, was accumulated by all trophic levels examined; on the other hand Zr^{95}-Nb^{95}, which is presumably absorbed to phytoplankton but not absorbed by the animals, was accumulated mainly by herbivores and not carnivores. The relatively high ration of Zn^{65}/Zr^{95}-Nb^{95} per gram found in deep-water organisms were carnivores.

Loss of both Zn^{65} and Zr^{95}-Nb^{95} downward is accelerated by vertical migrations of animals. Migrators may excrete radionuclides or be preyed upon below the pycnocline. Sinking particles, such as dead animals and fecal pellets, may also be important agents in the transport of radionuclides to deep water or sediments (Osterberg et al. in press).

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"HOT PARTICLE" IN COLUMBIA RIVER SEDIMENTS

by

Norman Cutshall and Charles Osterberg

ABSTRACT

A "hot particle" containing only fission product radioactivity was isolated from Columbia River sediments. A number of characteristics make the particle unusual, and its origin uncertain.

* * * *

A single particle with fission product radioactivity in the millimicrocurie range has been isolated. High activity, large size and apparent lack of fractionation make it quite unlike other reported individual fallout particles.[1,2,3] The particle was isolated from Columbia River sediments taken 23 July 1963 near Blalock, Oregon, about 130 miles below nuclear reactors at Hanford, Washington.

Surface sediments were collected from beneath about 2 m of fast flowing water; then air-dried and passed through a 200 mesh screen. Seven 5-gram portions of sediment (less than 74μ in diameter) were counted in the wall of a 5x5-inch NaI(Tl) crystal. The signal from the detector was resolved with a 256-channel pulse-height analyzer.

Unusually high activity was noted in one of the samples (#5, Table I). The single particle was isolated by repeated divisions of the highly active portion. Fission product radioactivity of the particle was equivalent to that found in about 200 grams of average sediment from the same location.

When examined under a microscope, the 11 by 25μ particle appeared colorless and isotropic except from several small, poorly defined anisotropic inclusions. One large inclusion was brown in color. The particle itself was highly angular and had at least 3 cleavage planes. Adams, et al.,[4] describe similar particles and inclusions associated with surface shots at the Nevada test site.

Neutron-induced radioactivity in sediments results from trace elements in river water used to cool nuclear reactors at Hanford, but very few fission products arise from this source.[5] Neutron-induced radionuclides appear to be more evenly distributed throughout sediments than are fission products. Scandium -46, for example, showed only small variations between portions of the same sample (Table I). This would seem to indicate that fission products are associated with relatively fewer sediment particles than are neutron-induced isotopes.

The gamma spectrum of portion #5 (Figure 1a) contains peaks due to neutron-induced radionuclides such as Zn^{65}, Co^{60} and Sc^{46}. When the particle was isolated from the sediment and recounted, its spectrum (Figure 1b) showed peaks only for the fission products Zr^{95}-Nb^{95}, Ru^{103}, and $Ce^{141-144}$. The slight increase in fission product activity after separation is due to changes in geometry and self absorption.[6] Equations of Rock et al.,[7] for changes with time in the apparent half-life of the Zr^{95}-Nb^{95}, were used to set early April 1963 as the approximate date for the formation of the particle. No Russian tests were reported from December 1962 to June 1963, however, underground tests were conducted at the Nevada site during this period.[8] A test was reportedly conducted in the Sahara Desert by the French government on March 18.[9]

A number of collections were made at four other stations on the river between Hanford and the ocean, but no similar "hot particle" was found. Measurements of fallout radionuclides in marine plankton and of filter samples from Oregon rivers and from rain water have failed to reveal anomalous particles; that is, dividing our samples generally resulted in a corresponding division of radioactivity. For example, Osterberg et al.,[10] report that average Zr^{95}-Nb^{95} activity in marine organisms (euphausiids) from nine consecutive tows was 13.6 pc/g, with a standard deviation of only 1.2 pc/g. This indicates that generally individual fallout particles do not contribute appreciably to the total fallout radioactivity present in the diet of these animals. The small fraction of fallout particles large enough to drop out of suspension in the river, and the large fraction of very small "suspendable" particles in the ocean, would lead one to expect a more even distribution of fission products in plankton than in sediments. Clearly the "hot particle" was in a class of its own; it contained 7300 pc of Zr^{95}-Nb^{95}, 3600 pc of Ru^{103} and 1400 pc of Ce^{141} on the date collected.

Table 1. Fission product activity and neutron-induced activity in 5 gram portions of Columbia River sediment.

Portion Number	Fission Product Zr^{95}-Nb^{95} picocuries/gram	Neutron Induced Sc-46 picocuries/gram
1	33	44
2	22	40
3	82	40
4	11	34
5	760	34
6	22	47
7	14	38
Mean	31 #5 not	40
Standard Deviation	+24 included	± 4.5

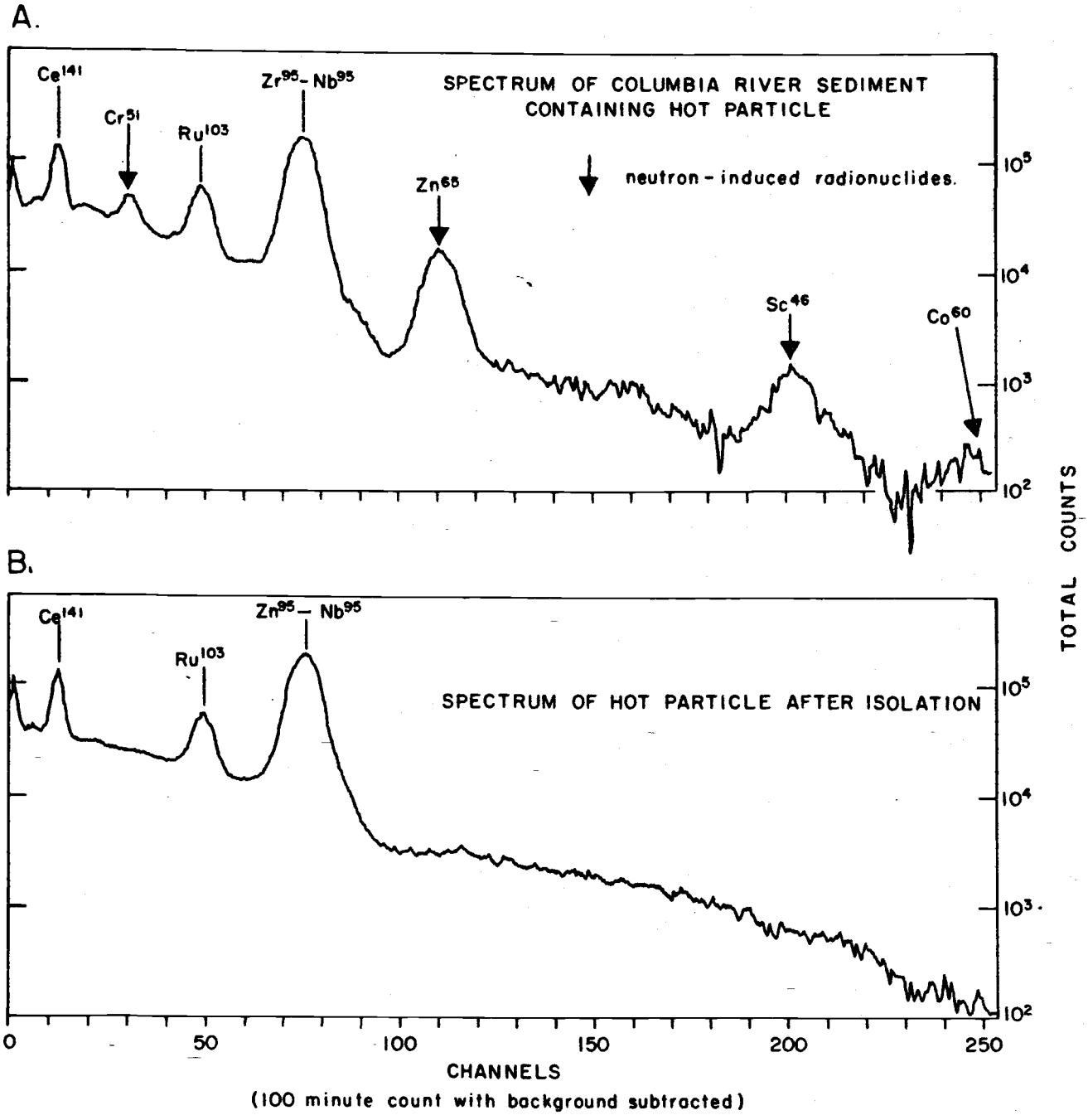

Figure 1a. Spectrum of 5 g sediment sample. Peaks indicated for Sc^{46} and Co^{60} are "sum peaks."

1b. Spectrum of hot particle after isolation. (1 channel = 10 kev)

Origin of the particle is not obvious. Its large size makes it unlikely, but not impossible, that air currents carried it 500 miles from the test site in Nevada (25μ is considered to be the division between local and worldwide fallout,[11] with most of the latter smaller than 4.5μ[12]). It is also unlikely that a particle this large could be carried from the Sahara Desert eastward to the Northwestern U.S. On the other hand, a particle of this size released into the river at Hanford might be expected to settle out in the quiet waters of Lake Wallula (McNary pool), which lies between Hanford and the collection site.

We conclude that the particle is unusual because of its high radioactivity, large size, and lack of fractionation, and also because its location in the river makes its origin an enigma.[13]

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13. This work was supported by contract AT(45-1) 1750 with the Atomic Energy Commission. We appreciate the help with petrographic examination given by Neil Maloney and Vern Kulm, and assistance in sample collection and preparation by Vern Johnson.

GAMMA EMITTERS IN A FIN WHALE

Charles Osterberg, William Pearcy, and Norman Kujala

Abstract

Qualitative and quantitative variations in gamma radioactivity were observed in tissues of a baleen whale captured off Oregon. Some contained concentrations of radionuclides appreciably different from those found in zooplankton removed from the whale's stomach. Zinc-65 levels in this large migratory mammal were comparable to those in resident animals of the area.

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Baleen whales are prodigious collectors of marine zooplankton. Zooplankton in turn concentrate radionuclides. Both fission products from fallout, and neutron-induced radionuclides (mostly Zn^{65}) from Hanford Laboratories, Washington, have been reported in marine zooplankton off Oregon[1,2]. An opportunity to measure the same radionuclides in various tissues of a plankton-feeding mammal occurred when a Fin Whale, Balaenoptera physalus Linnaeus, was captured west of Depoe Bay, Oregon, on 19 September 1963.

Tissue samples of a 55-foot (16.8 m) male Fin Whale were removed from a fresh animal at the Bioproducts, Inc. facility near Astoria, Oregon. Most samples were preserved by freezing, but stomach contents, liver, eyeball, and intestine were fixed in formalin. Samples were weighed, freeze-dried, and ashed at about 500°C. Oily tissues were burned in an open flame before ashing. Residual ash was analyzed in the well of a 5x5-inch (12.7 cm) NaI(Tl) crystal, using a 256 channel pulse height analyzer, with 100- to 200-minute counting times. The eye was an exception. It was counted whole, lens downward, on top of the crystal. The external location of the eyeball and its shape complicated the counting geometry and its value is subject to this additional error.

Zinc-65 was the most prominent peak in spectra of all whale tissues analyzed (Fig. 1), and the large variations among tissues are probably related to differences in stable zinc and in individual turnover rates.[3] Potassium-40, the naturally radioactive isotope of potassium, was next in prominence, followed by Mn^{54}, Cs^{137}, Zr^{95}, Nb^{95}, and $Ce^{141-144}$ in some tissues. Unlabeled peaks at 0.5 Mev arise from both $Ru^{103-106}$ and Zn^{65}

The spectrum of muscle had a small peak due to Cs^{137}, in addition to Zn^{65} and K^{40}, and evidence of $Ce^{141-144}$ and Zr^{95}-Nb^{95} (Fig. 1a). Cesium is chemically similar to potassium and is probably taken up in the same way. The abundance of potassium in the sea reduces uptake of Cs^{137} by marine animals, compared with terrestrial and freshwater animals.

The tongue contained much less Zn^{65} than did other tissues in Figure 1. Fission products (Zr^{95}-Nb^{95}, and $Ce^{141-144}$) were present, with some indication of Cs^{137}, even though the amount of K^{40} was quite low (Fig. 1b). While the Zr^{95}-Nb^{95} peak may result from surface adsorption, neither the spectrum of the skin (both dermis and epidermis) nor lungs (Fig. 1c) show comparable evidence of this airborne fallout radionuclide. The spectrum of the lungs is most nearly typical of tissues whose spectra are not individually illustrated, e.g., testes, intestine, skin, and blubber.

The liver contained Zn^{65}, K^{40}, traces of Cs^{137}, and $Ce^{141-144}$ and a distinct peak of Mn^{54} (Fig. 1d). Folsom et al.[4] reported Mn^{54} both in seawater and in marine organisms from off southern California. Osterberg et al.[5] found much larger Mn^{54} peaks in the liver of albacore, Thunnus alalunga, from off Oregon, and freshwater clams, Anodonta wahlamatenses, from the Columbia River. Although Mn^{54} is present in fallout[4], it may be even more abundant in the vicinity of the Columbia River, but no direct comparisons have been made.

The stomach of the whale contained tremendous quantities of zooplankton ("krill"). Only four liters (primarily Euphausia pacifica) were removed, and a portion analyzed. The resulting spectrum (Fig. 1e) contained mostly Zn^{65} and K^{40}, but $Ce^{141-144}$ and Mn^{54}, and possibly a trace of Zr^{95}-Nb^{95} were present.

In general, spectra of tissues from the Fin Whale resembled those of small mesopelagic fishes from the same trophic level. Since fallout is normally concentrated by herbivorous zooplankton and is discriminated against at higher trophic levels[2], the presence of Zr^{95}-Nb^{95} in the whale tongue seems anomalous. Little, if any, appeared in zooplankton taken from the whale's stomach. Radioanalysis of euphausiids collected off Newport in October 1963, verified the virtual absence of fallout radionuclides in euphausiids at this time. Levels of fallout in plankton from surface waters off Oregon are much reduced compared with those observed during the past two years.[5] Therefore, the tongue may have been contaminated from terrestrial sources after capture of the whale, or by fallout retained from a previous exposure. Since Fin Whales are highly mobile and our knowledge of the fallout pattern at sea is limited, neither alternative can be readily discounted.

The activity of Zn^{65} per gram wet weight of the various tissues is given in Table 1. Liver, lungs, testes and intestine contained the greatest amounts of this radionuclide, while the eyeball and blubber contained the least. Maximum values for various small mesopelagic fishes, typical of levels of Zn^{65} usually found in marine plankton and nekton in this area, are comparable with those in the whale. Since the Columbia River is the principal source of this radionuclide in the northeast Pacific Ocean[6], whales apparently accumulate Zn^{65} rapidly during their sojourn in Oregon waters. Levels are extremely low and no health hazard exists. However, if whales retain this radioactive souvenir of the Columbia River for a sufficiently long period, the Zn^{65} should provide a useful tracer for migration studies of these ocean wanderers.

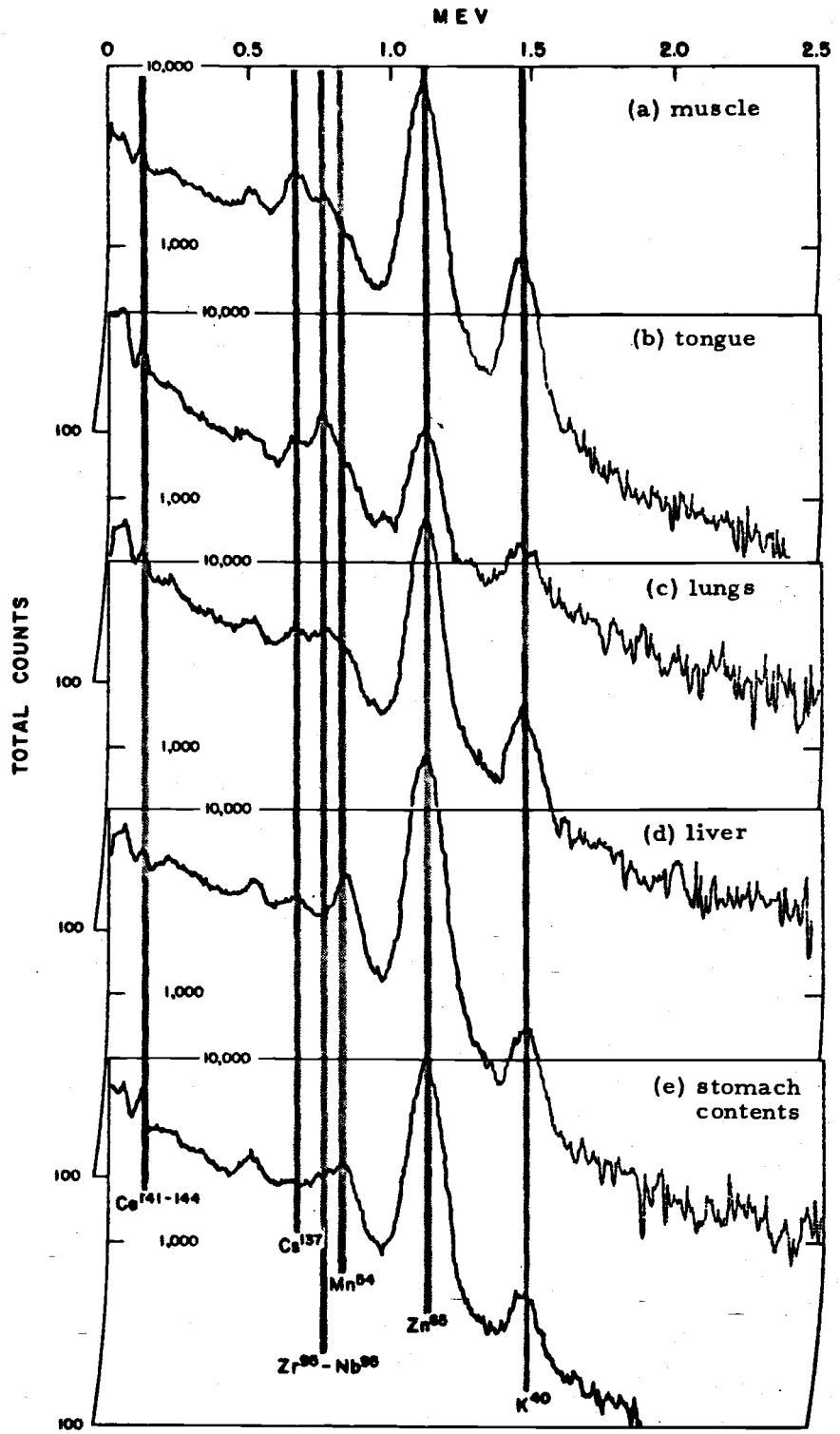

Figure 1. Gamma-ray spectra of whale tissues and stomach contents all adjusted to the same wet weight (949 g). Gray bands show location of gamma emitters permitting direct comparison of relative amounts of radionuclides in each tissue. Dotted tracings of spectra indicate overlap. Counting times were 100 minutes with no background subtracted.

These preliminary results warrant further study, particularly since the great size of the whale provides samples of almost all tissues in quantities suitable for radioanalysis. Unfortunately, the small 90-foot (27.4 m) whaling boat was damaged when it prematurely tied to an 80-foot (24.4 m) whale.[7] This required a delay for repairs, and the subsequent onset of winter storms ended the whaling season.[8]

Table 1. Zn^{65} in tissues of a whale (picocuries/gram wet weight). The fish, used as a comparison, were various lantern fish from near the same area in which the whale was captured.

Sample	Zn^{65} pc/g Wet Weight
Liver	7.4
Lung	5.2
Stomach contents	4.0
Testes	3.6
Intestine	3.1
Muscle	2.7
Skin	2.3
Tongue	0.7
Eye (whole)	0.4
Blubber	0.2
Mesopelagic fishes	1.7-7.3

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