AN ABSTRACT OF THE THESIS OF

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Title RADIOACTIVITY OF SEDIMENTS IN

THE COLUMBIA RIVER ESTUARY

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Sediment radioactivity was measured in the Columbia River by use of an *in situ* gamma-ray detector, calibrated to give activity in pc/cm^2. The probe has several advantages over methods which couple collection of sediments with laboratory radioanalysis. Most important is the rapidity with which field measurements can be made.

Sediments concentrate radioactivity and in the Columbia River the level of radiation is about an order of magnitude higher in the sediments than in the water above it. Benthic organisms which live in the sediments are constantly exposed to this higher radiation level.

Chromium-51 and Zn^{65} were the two man-made radionuclides present in greatest abundance in the Columbia River. The Cr^{51}/Zn^{65} ratio in the sediments varied, but Cr^{51} activity was about ten times as high as the Zn^{65} activity. Also identified in the Columbia River sediments, but with lower activities were K^{40}, Co^{60}, and Mn^{54}.

RADIOACTIVITY OF SEDIMENTS IN
THE COLUMBIA RIVER ESTUARY

by

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RADIOACTIVITY OF SEDIMENTS IN THE COLUMBIA RIVER ESTUARY

INTRODUCTION

Background

Radioactive materials introduced into the environment as fall-out from atomic weapons testing and as waste from the operation of reactors such as those used to produce plutonium at Hanford, Washington, are a potential hazard. The problem they present is basically different from that occurring with other industrial wastes. Radioactive wastes cannot be disposed of by chemical treatment but are eliminated from the environment only by radioactive decay. Their persistence in the environment can be very great in the case of a radionuclide with a long half-life.

Levels of radioactivity in the environment will grow in the future. Present predictions are that the nuclear power plant capacity in the United States will increase about fivefold every five years in the decades preceding 1980 (U. S. Atomic Energy Commission, 1957).

Anderson (1963) stated in his opening address to the symposium on the Studies of the Fate of Certain Radionuclides in Estuarine and Other Aquatic Environments in Savannah, Georgia,

> Before effective monitoring of radioactive wastes
> discharged to the marine environment can be

accomplished, the fate of these wastes and their pathways into man's marine food supply must be determined. To realize these objectives, an aquatic marine environment containing small amounts of man-made radioactive wastes should be investigated.

The Columbia River provides such an opportunity because of the reactors at Hanford, Washington.

Radioactivity wasted to a river is partitioned, according to its chemical properties. Plants and animals in the water may take up a portion, some may attach to the sediments, with the balance remaining in the water. A radioactivity budget for a particular radionuclide in a stream has been formulated by Sayre, Guy and Chamberlain (1963):

$$R + r_n = [r_w + r_s + r_o] e^{\lambda t} + r_d$$

in which

R   is the radioactivity in curies of waste discharged into the stream at time t = 0,

r_n is the background radioactivity in the stream due to natural causes and fallout at time t = 0,

r_w is the radioactivity carried in solution by the water at time t,

r_s is the radioactivity sorbed by stream sediments at time t,

r_o is the radioactivity taken up by living organisms and entering the ecological cycle, at time t,

r_d is the radioactivity discharged from the watershed, and

$e^{\lambda t}$ is a radioactive decay factor dependent upon time (t)

and the radioactive-decay constant (λ) which depends

on the nuclide.

While radioactivity in the stream is distributed among the biota,
water, and sediments, not all fractions can be sampled and analyzed
with equal ease. Biological sampling is often time consuming, but
analysis is no problem since the specific activity (counts per unit
volume)[1] of the samples can be increased by ashing. Likewise, wa-
ter sampling and analysis are readily accomplished, at least in the-
ory. For example, the specific activity can be increased by evapo-
ration and the salts distributed evenly in an agar agar gel. Once
concentrated, samples of the biota and water can be analyzed in the
laboratory by routine gamma-ray spectrometric techniques. Analy-
sis of sediment samples is not so straightforward, for the following
reasons.

Because artificial radioactivity has been present in quantity
only since 1944, man-made radionuclides are confined to the most
recently deposited marine sediments. This thin surface layer of

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[1] The term "specific activity" is used in several ways. In one
application specific activity is defined as the ratio of the quantity of
radioactivity of a particular isotope to the total amount of isotopes of
that element present (Revelle and Schaefer, 1957). Specific activity
can also refer to radioactivity per unit weight or volume of sample as
in this application (Chase and Rabinowitz, 1962; Wang and Willis,
1965).

sediment, though of greatest interest, is most difficult to remove in its undisturbed state in quantities sufficient for radioanalysis. Coring devices cause the least disturbance in the sample, but the proportion of surface sediments recovered is small. A coring device which collects cores of 15 cm diameter has been described (Nelson, Perkins and Nielsen, 1964) but it is effective only in fine sediments (Nelson, 1965). In the Columbia River estuary, most of the surface sediments are sand (Lockett, 1965) or are immediately underlaid by sand, making coring difficult.

Both dredged and grabbed samples contain relatively large proportions of surface sediments, but older, less radioactive sediments are often mixed into the desired sample and confound the analysis. Also some of the finer-grained sediments are likely to be washed out of samples as they are brought to the surface.

Use of an in situ probe eliminates the need to collect a sample and therefore avoids these problems.

In practice, the probe is lowered to the sediment where it remains for the duration of the radioactivity analysis (usually ten minutes in the Columbia River). Data are available immediately. If an area of particularly high radioactivity appears it can be noted and a further survey in that area pursued. This obviates the problem of returning to the laboratory only to discover that more samples in certain areas would have been helpful.

Since sampling and concentration of sediments is such a problem, the $\underline{in}$ $\underline{situ}$ probe described here should be useful in measuring the radioactivity sorbed by stream sediments, r_s, in the budget of Sayre, Guy and Chamberlain (1963).

It should be mentioned that all radioactivity levels discussed in this paper are very low and can only be detected by extremely sensitive instruments. When the terms "high radioactivity" or "low radioactivity" appear they are relative to other values found in this study. No radioactivity levels were found which were deemed to constitute a health hazard.

Purpose

The goals of this study were threefold:

(1) To develop a probe for making $\underline{in}$ $\underline{situ}$ measurements of the radioactivity in estuarine sediments.

(2) To determine areas of high and low radioactivity in the estuary.

(3) To consider physical, chemical, or biological processes which might cause variations in sediment radioactivity.

These goals, when fulfilled, would be of value in predicting the possible effects of future nuclear installations on natural waterways. The problem is by no means esoteric, since large fractions of certain radionuclides are removed from the water on sediments. The

water-sediment interface provides a residence for a large community of benthic animals. Background radioactivity of these animals could be appreciably increased by sediment sorbed radionuclides.

Several factors were considered essential in the instrument developed in this study. The probe should have some means of orienting itself so that geometry is reproducible and it must be sufficiently resistant to thermal and physical shock to reliably measure radioactivity in sediments. The casing material must not appreciably attenuate the 0. 32 MeV gamma-ray from Cr^{51}, a gamma emitter which is prevalent in the Columbia River. The instrument should be portable enough to be used on small (about 9 m) fishing boats and light enough to handle in the field (Figure 1). Once developed, the probe can be used to compile an inventory of sediment-sorbed radionuclides in the estuary. Levels of radioactivity, and ratios of gamma emitters can then be related to sediment type and size, and to water movements.
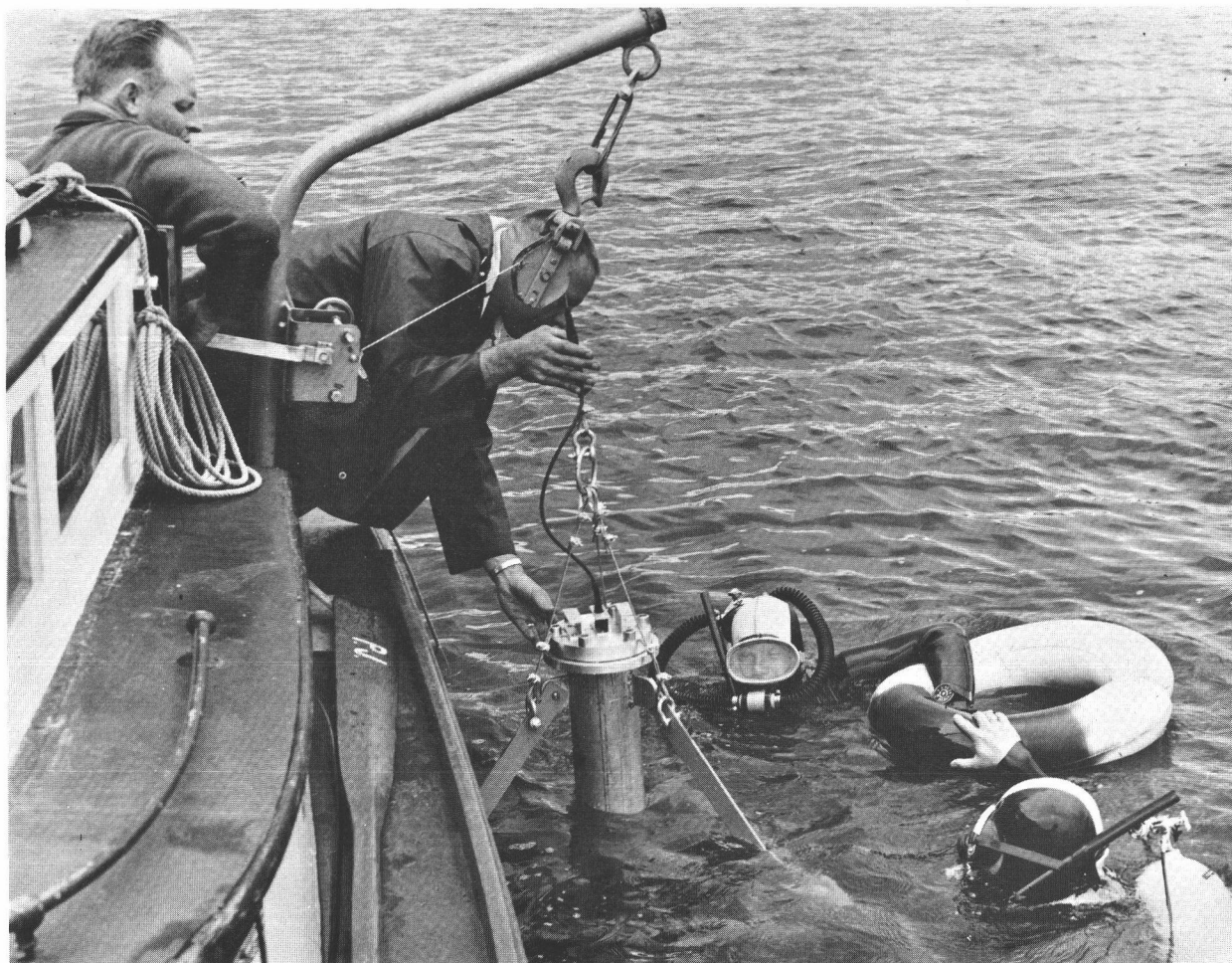
Figure 1.   Skin divers with the probe in its initial tests in the Columbia River. Observations were made to check the alignment of the probe on the bottom, and to determine if sediment was unduly disturbed by the probe.

PROBE DEVELOPMENT

History

Several detectors have been designed which measure gross gamma radioactivity in the field without distinguishing the particular radionuclides present.

Probably the first major effort to directly measure artificial radioactivity in the marine environment was made with a probe developed by Le Vine (Harley, n.d.). During May and June of 1954 this probe was towed behind the U. S. Coast Guard cutter TANEY in the vicinity of the Bikini atomic testing grounds, as a part of the Operation Troll survey. A plastic phosphor 7.6 cm in diameter and 76 cm long was enclosed in a 2.1 m long stainless steel casing. Operational difficulties of the fragile equipment and activity levels below the sensitivity of the instrument prevented significant measurements by this probe (Ibid).

A radiological sonde described at the Conference on the Peaceful Uses of Atomic Energy (Revelle et al., 1958) uses halogen-filled Geiger counters or gas discharge tubes as the detector. Ten or more of the counters are combined in parallel in a cylindrical protective shell. The sondes are roughly two meters long, with an outside diameter of 8 cm and a wall thickness of 7 mm. A

multiconductor cable returns the signal to the recording device aboard ship. These probes have been used for vertical radioactivity profiles and, by towing aft of a ship, for horizontal profiles of activity in the near surface layer. When provided with fins, the detector assemblies can be towed in groups of two or more behind ships. This technique is useful for continuous measurements at two or more depths down to about 400 meters.

The Russians (Khitrov and Kotlyarov, 1962) describe a radiometer in which the recording device is contained in a hermetically sealed pressure vessel. The 30×10 mm NaI crystal, associated electronics and recorder were enclosed in a piece of steel pipe of 20 cm diameter with a length of 140 cm and a wall thickness of 3 cm. The advantage of this instrument is its ability to withstand pressure of up to 1000 atm., thus it can be used at great depths.

Another detector measuring total γ-activity is described by the Japanese (Japan, n. d.). Like the Russian probe the recorder is enclosed in carbon steel pressure casing. It consists of a 3.8×3.8 cm NaI crystal and recording apparatus in a casing of 2.3 meters in overall length, 85 cm outside diameter and a 1.1 cm wall thickness. The wall thickness of the lower part is reduced to 0.5 cm to minimize attenuation of the gamma rays.

In California, Klingeman and Kaufman (1963) enclosed a 5×5 cm NaI(Tl) crystal in a 7.6 cm diameter $\times$ 38 cm long aluminum

cylinder of 1. 6 cm wall thickness and mounted the detector on an

underwater sled to survey fallout radionuclides in the bottom of

San Pablo Bay. This instrument was useful in determining cross-

distribution of total radioactivity across the channel. Higher radio-

activity values were found in quiet waters where finer sediments had

settled, with relatively lower values near the center of the channel

which is kept clean by current action.

Recently several probes have been developed which were

coupled with pulse height analyzers so that particular radionuclides

could be distinguished by energy analysis. One such detector was

tested in the Gulf Stream early in 1960 (Riel, Simons and Converse,

1964). This probe system was built around a 12. 7 × 15. 2 cm NaI(Tl)

detector. The crystal was loaded into a steel pressure vessel and

insulated with 2. 5 cm of styrofoam. Recent modifications in the

electronics of this system were described by Riel (1963). The probe

was used recently to measure the radioactivity of the Columbia River

effluent. The radionuclides identified in this study were neutron

activation products (Cr^{51} and Zn^{65}), naturally occurring K^{40}, and

fission products (Zr^{95} and Nb^{95}) (Gross, Barnes and Riel, 1965).

When the nuclear powered submarine the USS THRESHER sank in

1963 this in situ detector, known as the Naval Ordnance Laboratory's

DUNC, was used for a survey in the water where the submarine was

lost. A small NaI crystal (4. 5 × 5 cm) was placed in a container

capable of withstanding high pressures for a bottom survey in the same area. No fission products above usual atmospheric fallout were found either in the water or near the sediments (Riel, Simons and Converse, 1964).

In 1964 an in situ detector was built to be housed in the bathescaphe TRIESTE II to study more closely the possibility of a transfer of radioactive contamination to the ocean from the THRESHER reactor vessel (Dohne and Hoover, 1965). This instrument includes a 7.6 × 7.6 cm NaI crystal and associated circuitry in an aluminum pressure vessel. In this survey, the gamma energies identified can be assigned to one or more of the naturally occurring radioactive elements. No contamination from the THRESHER was observed in any of these tests.

Another probe for measurements of gamma activity in water was developed at the Boeing Company in 1962 (Proctor, Papudulpulos and Firminhac). It has a 12.7 × 10 cm NaI(Tl) crystal installed in a piece of 15 cm seamless carbon steel pressure tubing. A stainless steel dome 2 mm thick and lined with styrofoam protects the crystal. No artificial gamma emitters were identified in the reported tests in Puget Sound.

Mark I

The first probe constructed at the start of the author's project

in 1963 was made to detect radioactivity in surface water in the

Columbia River (Osterberg, Jennings and Cutshall, 1964). The

nuclide of particular interest was Cr^{51}. Styrene pipe (10 cm diam-

eter) was used to house the 7.6×7.6 cm NaI(Tl) crystal because of

its low absorption characteristics. The probe was equipped with two

12 m cables, one for high voltage to the photomultiplier tube and one

for signal return. Initial tests consisted of a 15 hour survey from a

dock in Astoria with the detector mounted on a float (Figure 2). The

crystal floated one meter below the water surface. Tidal variations

in Cr^{51} activity were very nearly a mirror image of the salinity.

This is due to dilution of the radioactive Columbia River water with

the less radioactive oceanic waters as the tide floods.

Mickey Mouse

The success of the initial survey resulted in mounting the probe

on a boat for a three-day cruise (Ibid.). This experiment began at

Hood River, 210 km above the mouth of the Columbia River. Thirteen

40-minute counts were taken at stations separated at about 16 km in-

tervals down to the mouth. Each night a 400-minute count was taken

while at anchor to see if other less abundant radionuclides would

appear in the spectrum. A 400-minute count taken on this cruise

(Figure 3) shows that Zn^{65} and Cr^{51} are the two principal gamma

emitters present in the water.

STYROFOAM INSULATION

3 x 3 INCH NaI (TI)
DETECTOR

LEAD WEIGHT
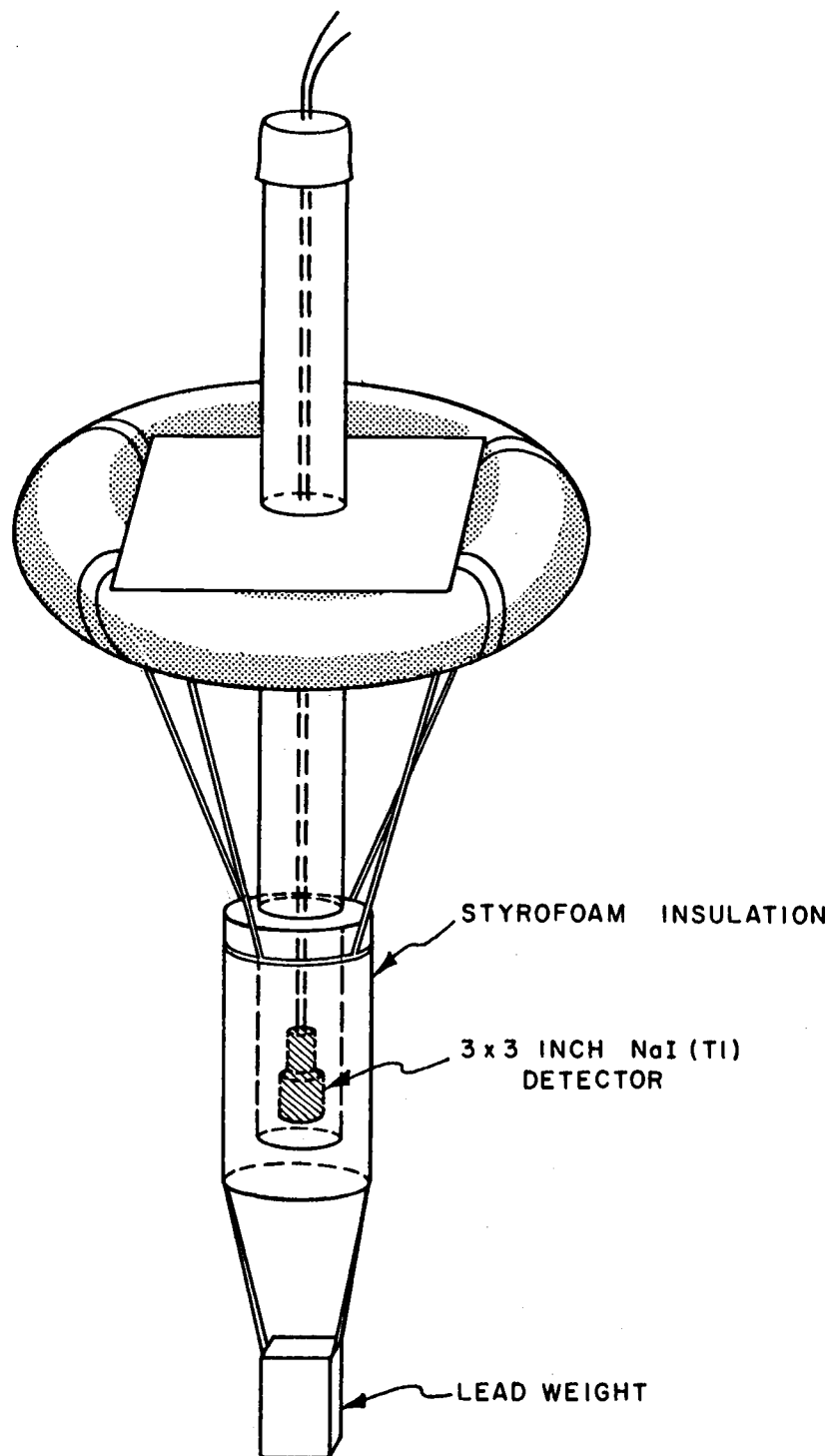
Figure 2. In situ gamma probe for measuring
radioactivity in surface water.
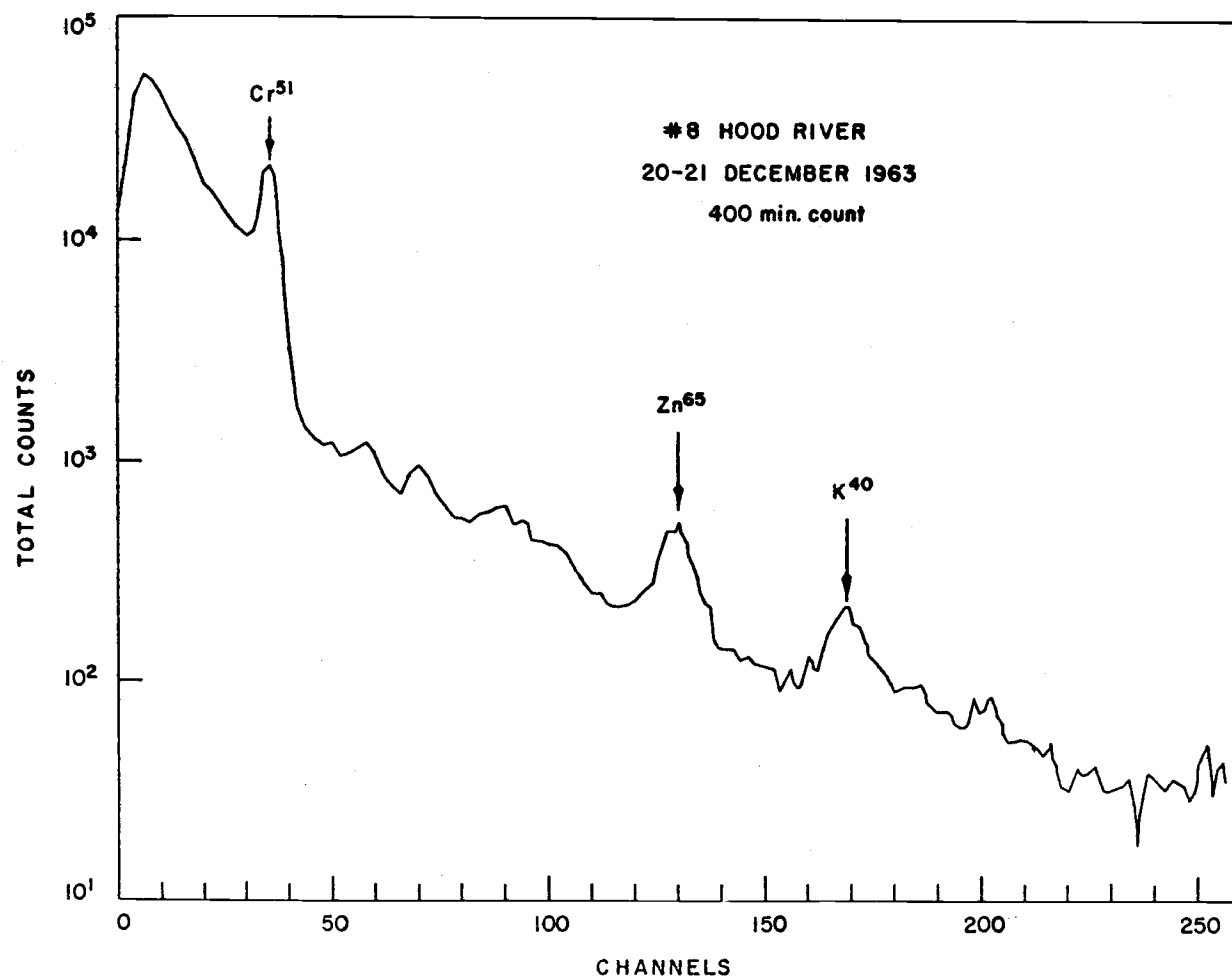
Figure 3. In situ spectrum of Columbia River water at Hood River, Oregon.

Sediment Probe

Few measurements have been made of sediment radioactivity. Since the sediment sorbed portion represents an important part of the total radioactivity budget, a probe was constructed for field measurements of sediment radioactivity (Jennings, Cutshall and Osterberg, 1965). Aluminum weighs less and shows less tendency to absorb photons than the commonly-used stainless steel, so it was chosen for the probe casing material. Samples of Cr^{51} counted in the laboratory with no absorber and with a 0.635 cm thickness of aluminum as an absorber show that the counting rate for Cr^{51} (0.32 MeV) is reduced only about 20% by the aluminum housing. There is, of course, even less attenuation of the higher-energy gamma-ray emitters such as Zn^{65} (1.12 MeV) and K^{40} (1.46 MeV).

The detector, a NaI(Tl) crystal (7.6 × 7.6 cm) with attached photomultiplier, plus a line-matching preamplifier and high voltage battery supply, is sealed in an aluminum pipe 14 cm in diameter, 63.5 cm long, and 0.635 cm thick. The probe is made hermetic by the installation of an O-ring on the outer flange of the cap and two Quad rings on the inside contact surface (see Figure 4). The detector is molded in Eccofoam to reduce thermal and physical shock with 2.5 cm of insulation between the crystal and the 0.635 cm aluminum bottom plate.

QUAD-RING

O-RING

PREAMP.

High
Voltage

Al CASING

PREAMP.
BATTERY

Dessi
cant

63.5 cm

ECCOFOAM
INSULATION

Photo-
tube

NaI(Tl)
Crystal
7.5

.635

14 cm
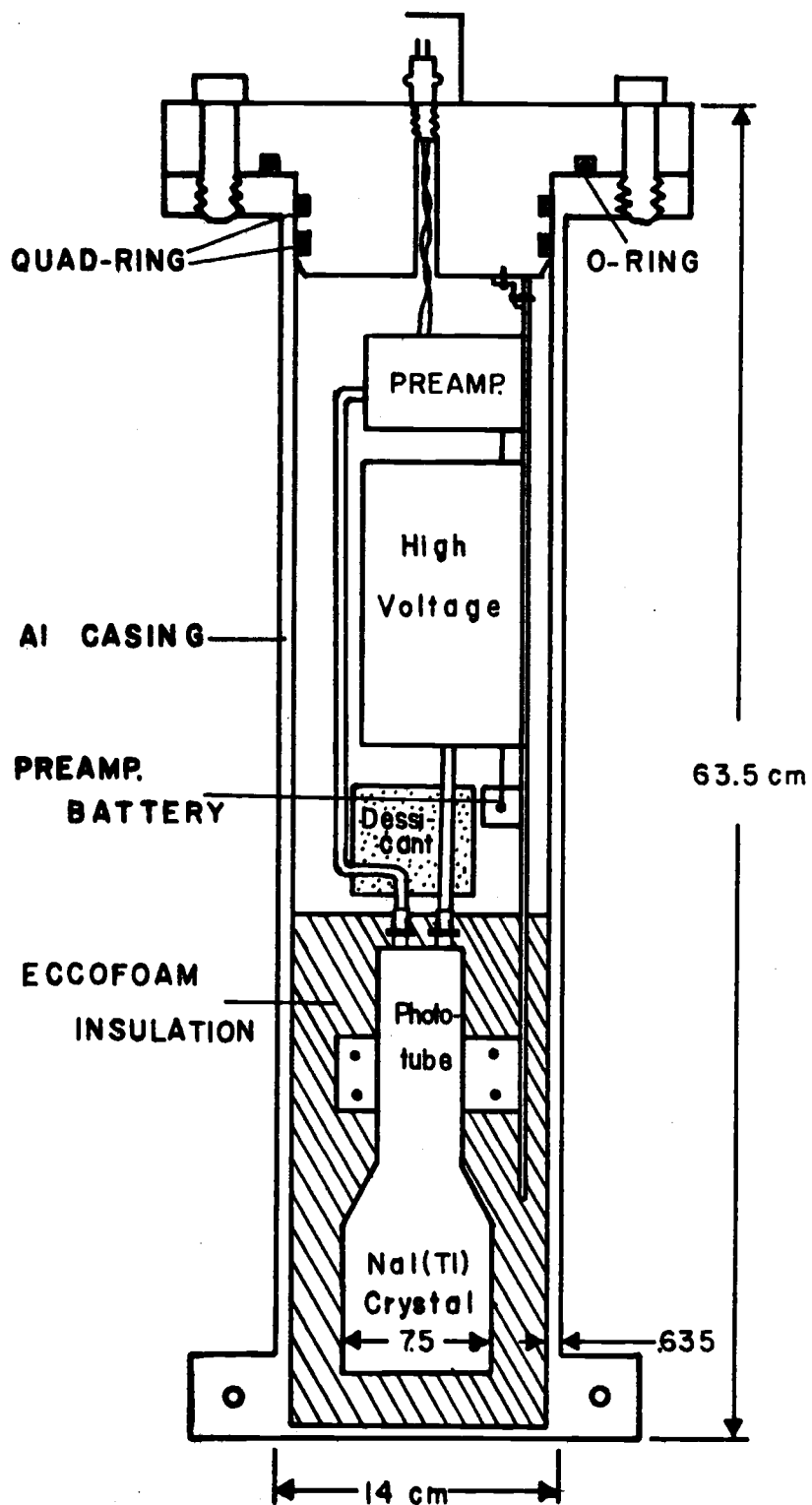
Figure 4. Cross section of sediment probe casing.

A tripod orients the detector on the bottom so that the positioning is generally reproducible (Figure 5). Scuba divers observing the device in operation in its initial tests reported that the probe showed no tendency to tip. However, a mercury switch shuts off the power to the photomultiplier tube in the event the probe is appreciably tilted. Thus, proper orientation of the probe while "counting" is insured, and the batteries can be conserved simply by tipping the probe on its side when not in use.

Signals from the probe are carried to an ND-130 AT 512 channel spectrometer in the boat through 0.318 cm steel-armored single-conductor cable (Amergraph No. 10H125, U. S. Steel). The cable has an insulated copper conductor covered with a polyvinyl chloride sheath with two layers of steel armor. Yield strength is 1500 lb, DC resistance of the conductor is 12.4 ohm/1000 ft., and armor resistance is 20.8 ohm/1000 ft. The characteristic impedance of the cable is 75 ohms. A 54 m length of the cable was donated by the U. S. Steel Co. for use in this experiment. A portable winch raises and lowers the probe by means of the single cable. The gamma-ray spectra are recorded on Tally punch-tape for laboratory analysis.

The high voltage battery supply consists of 36 30-volt batteries (Burgess Y20S) in series. Taps are provided so that voltage can be chosen at 60-volt intervals from 880 V to 1180 V. Figure 6 shows a schematic diagram of the line-matching preamplifier.
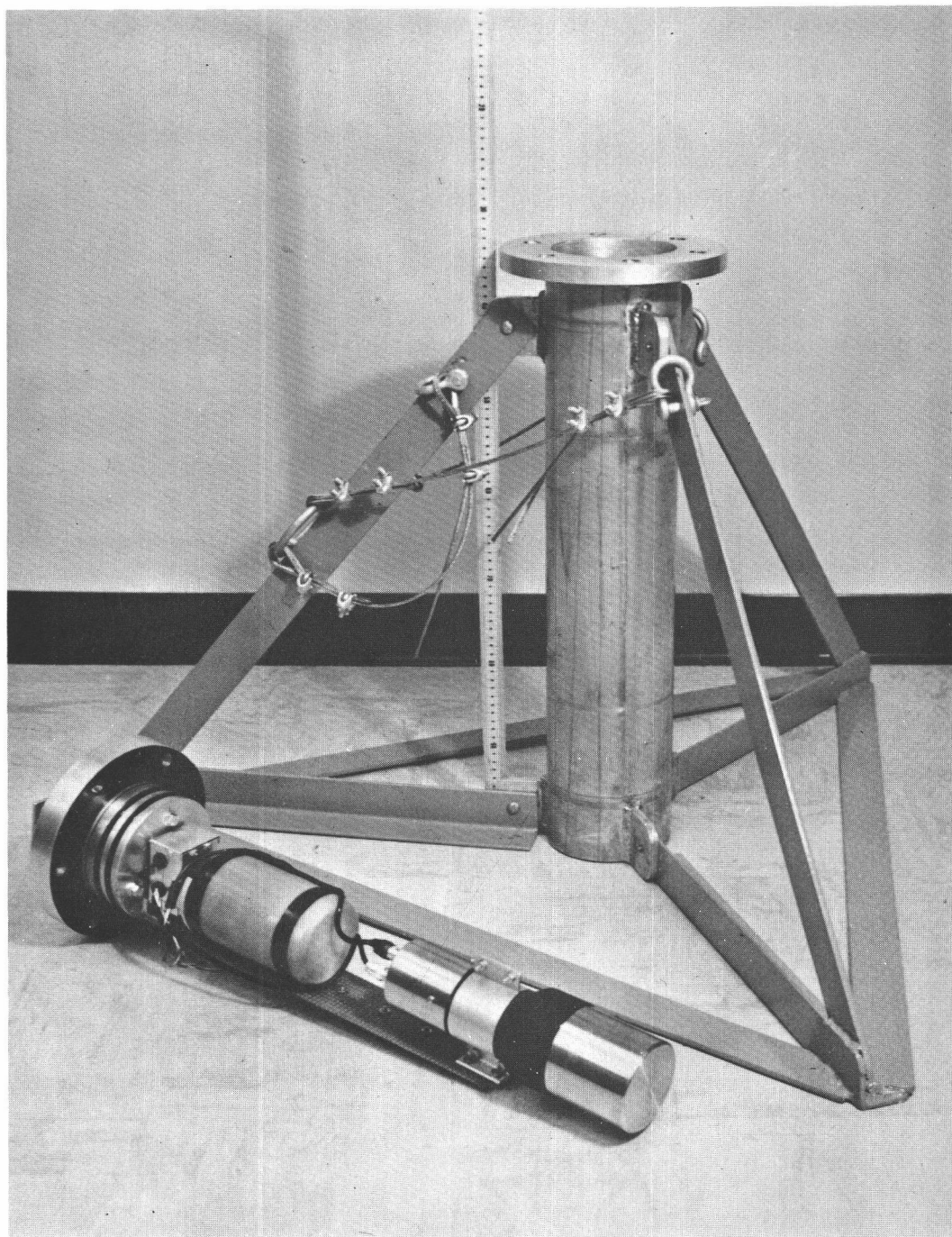
Figure 5.   Sediment probe with tripod attached.
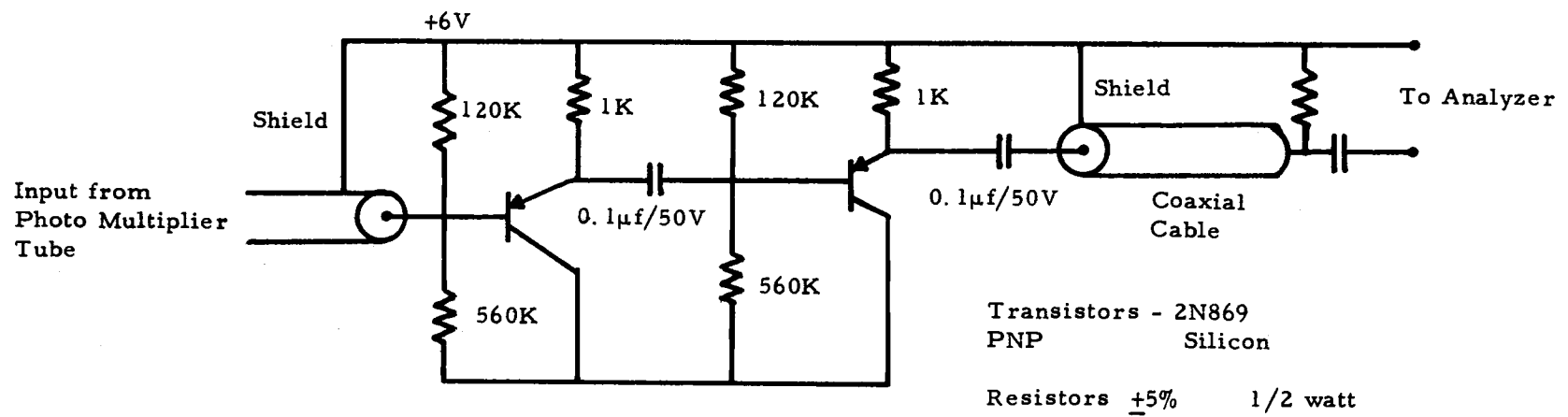
Figure 6. Schematic diagram of line-matching preamplifier designed by Roderick Mesecar.

USE OF THE PROBE

Cruise Preparation

A complete checkout of the electronic system before taking the probe into the field eliminates many operational problems. Of particular importance is a linearity check. This is accomplished by counting several gamma emitters with widely divergent photopeak energies and plotting energy vs. spectrometer channel number. These points must occur on a straight line to insure accurate identification of gamma emitters in the field samples. Figure 7 shows results of a linearity check for the in situ probe.

Two common causes of non-linearity were (1) a weak battery in the preamplifier and (2) poor pulse shape from the detector. The first difficulty is obviously solved by replacing the battery. The pulse shaping is rectified by matching the internal amplifier to the detector and input cable. A potentiometer for that purpose in the internal amplifier can be adjusted for optimum pulse shape (Nuclear Data, n. d.).

To test for contamination a count was made after each cruise with the probe in a lead cave in the laboratory. Figure 8 shows a spectrum of such a count with a typical sediment spectrum for comparison. The count rate is far below those seen on the sediments so that contamination presented no problem in this study.
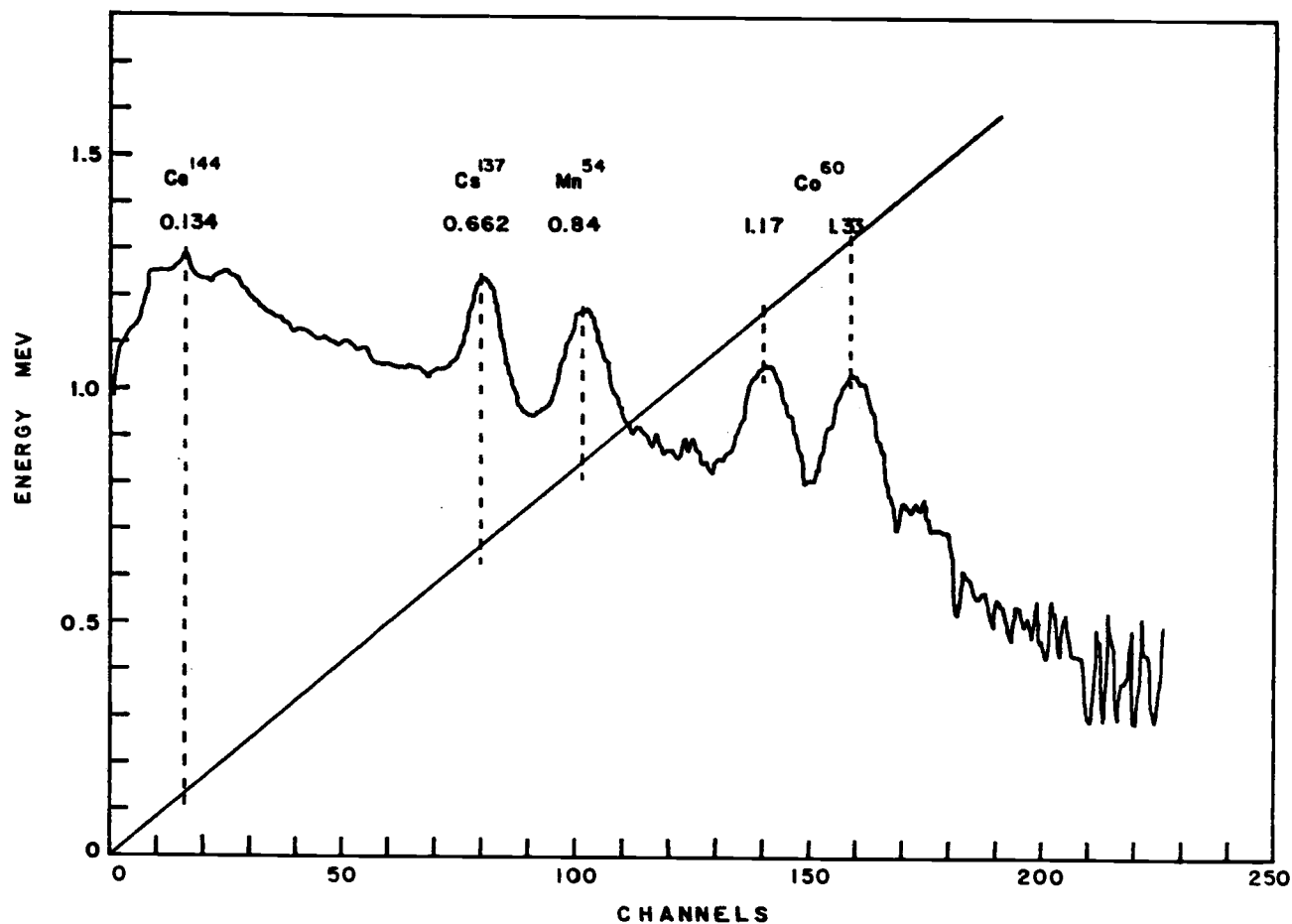
Figure 7. Linearity check of the sediment probe system. In order for gamma emitters to be identified by the spectrometer channel they fall in, the system must produce a linear relationship between γ-ray energy and spectrometer channel number of photopeak.
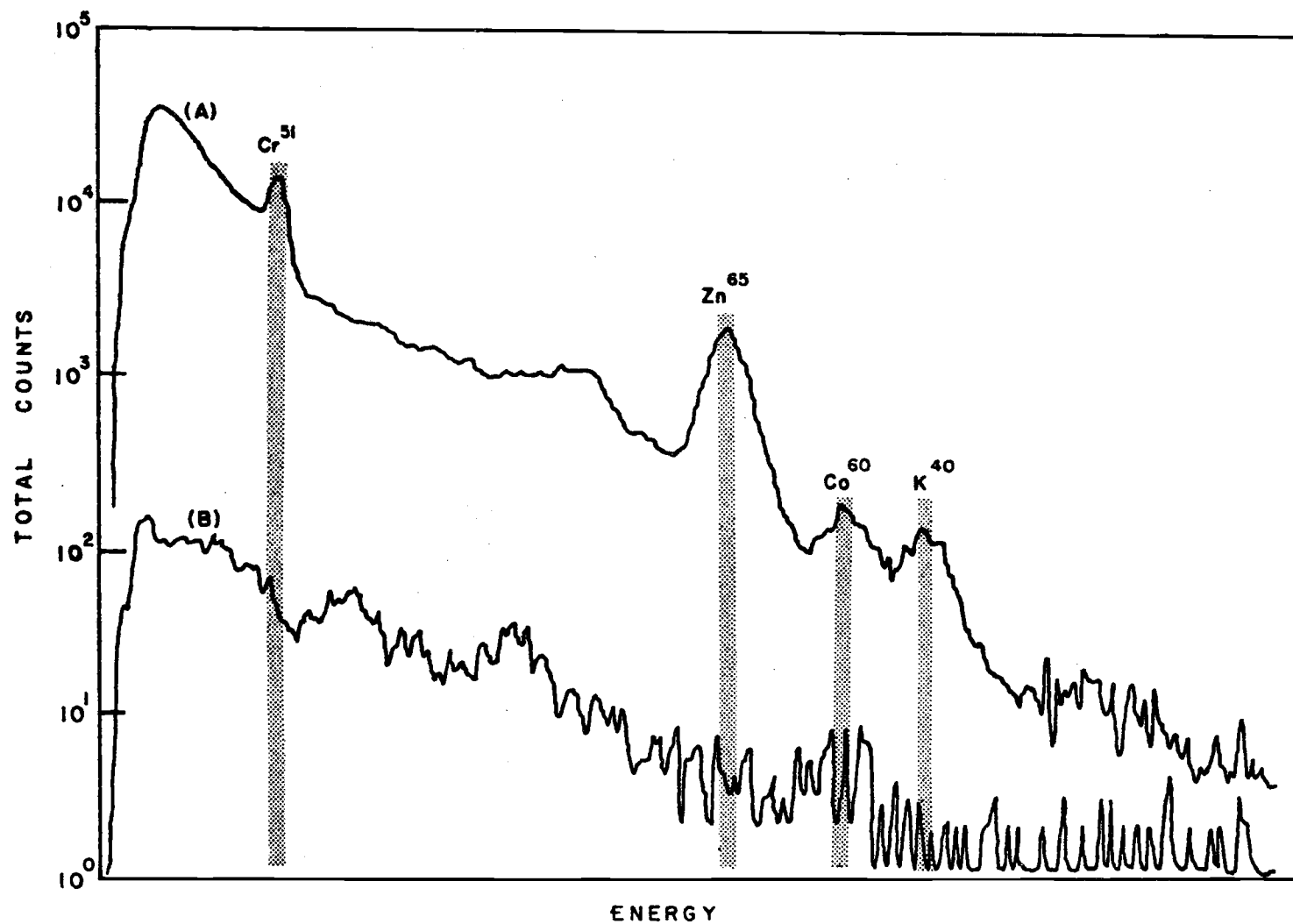
Figure 8. Spectrum of (A) surface sediment radioactivity in Youngs Bay and (B) background with probe in lead cave in the laboratory.

Field Work

The field sampling program primarily involved counting with the in situ sediment probe. Whenever possible core or grab samples of the sediment were also collected. Columbia River fishing boats provided a working platform for the sampling program. A portable 5 kw gasoline generator furnished the power to operate the analyzer.

Sampling sites in the Columbia River are seen in Figure 9, and Figure 10 shows the corresponding surface sediment radioactivity. Several factors entered into the choice of sample sites. (1) Finding the highest concentrations of radioactivity, which were felt to be of primary importance in an initial survey, required looking for areas of fine sediments. (2) A widespread survey was desired to give a general picture of the entire estuary, so the sampling sites were dispersed throughout the estuary. (3) The tidal variations in the estuary required that some thought be given to a cruise plan. Some areas of interest were inaccessible during low tide. Since running the boat against the tide wastes sampling time we chose stations that allowed us to move with the current as often as possible. (4) Finally, weather conditions and instrument problems occasionally made us opportunists. The onset of rough waters or fog sometimes required that the sampling plan be abbreviated or changed. Instrumental difficulties during the transportation and set up period often caused delays

WASHINGTON

OCEAN

PACIFIC

COLUMBIA

RIVER

ASTORIA

OREGON

0   5   10

MILES
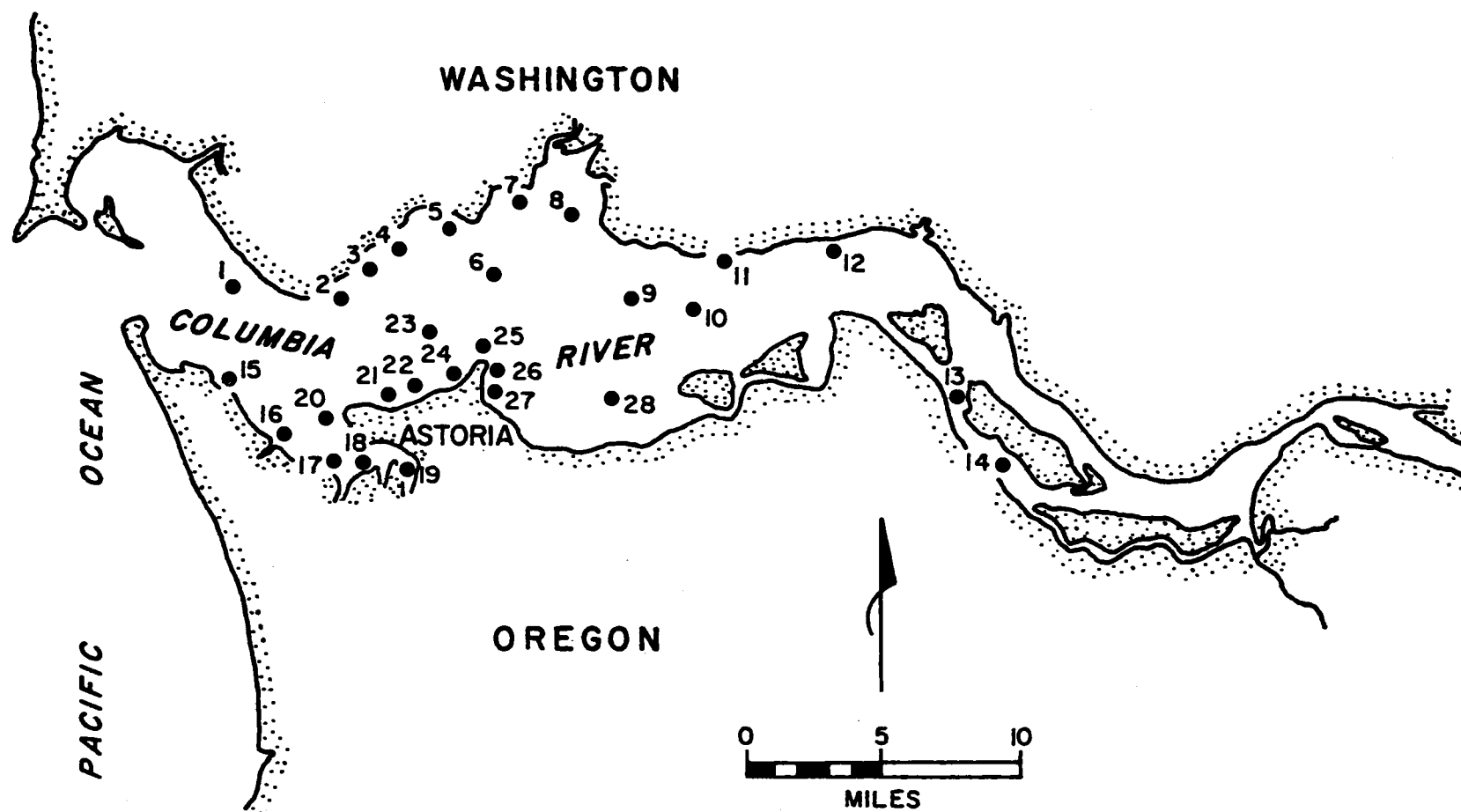
Figure 9. Sample site locations in the Columbia River. (See Figure
10 for surface sediment radioactivity.)

24
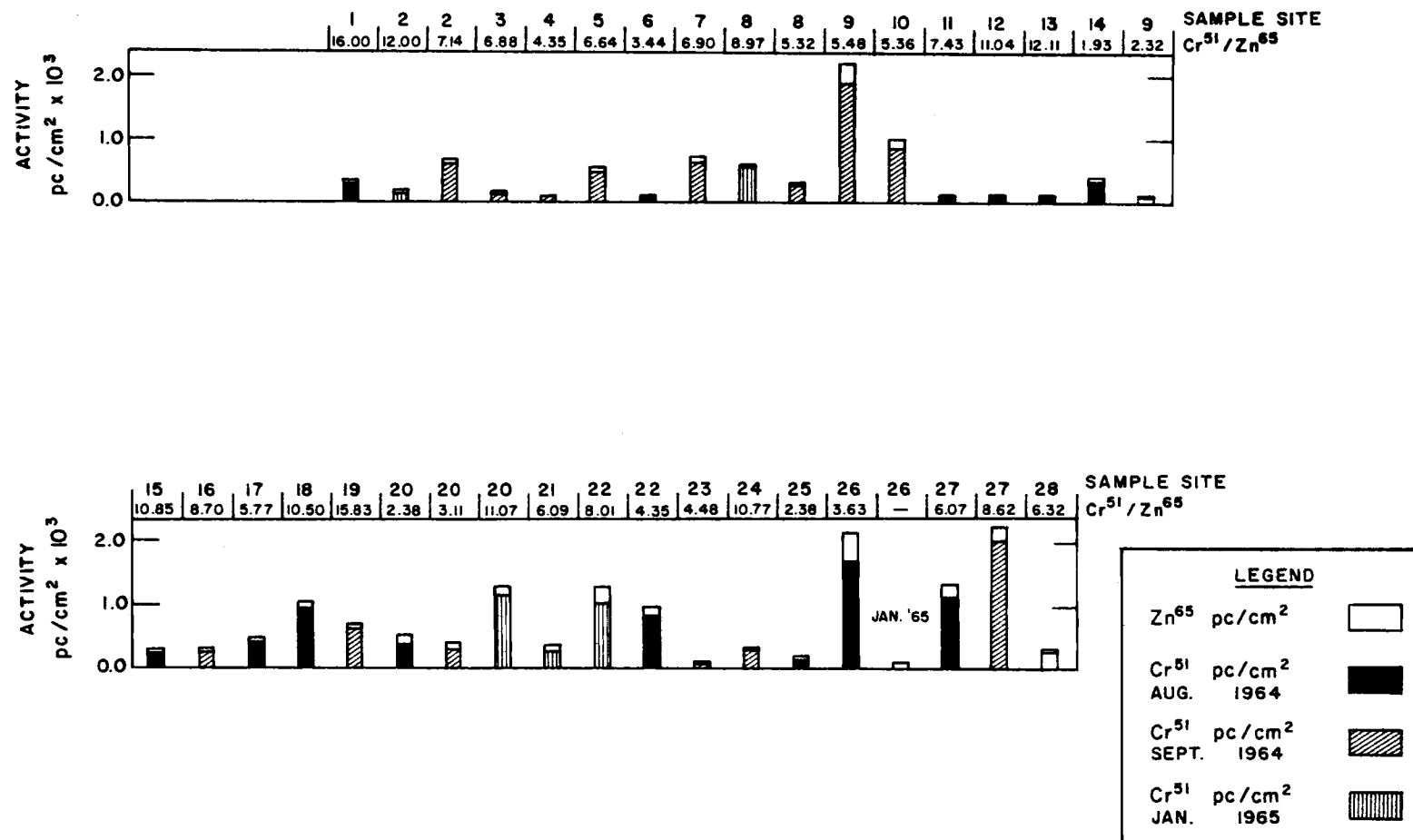
Figure 10. Surface sediment radioactivity in the Columbia River.
(See Figure 9 for sample site locations.)

which upset the schedule. Once in operation, however, the system was nearly trouble free.

At each station the boat was anchored before sampling began. The probe was lowered to the bottom and a few meters of slack were introduced into the cable to compensate for any movement of the boat. The winch was then locked and the multichannel analyzer connected at the jack on the winch. After a ten minute count the data were recorded on a tape punch and/or typewriter readout.

While the probe was counting an effort was made to obtain a core sample of the sediment for analysis in the laboratory. The coarse sands in the Columbia River estuary often made this task difficult. For this reason a grab sampler was sometimes substituted. It was not without problems either since sand grains would often hold the jaws open slightly and the sample would be washed out.

The top 1.5 cm of the cores collected were retained for the laboratory analysis. When a grab sample was taken a representative portion was saved for the laboratory work.

Following the count, the probe was brought aboard and tipped on its side to conserve batteries before proceeding to the next station.

Periodically a spectrum of a Co^{60} source was taken for future reference in identifying the radionuclides present in the sediments.

Calibration

The probe was calibrated so that sediment surface radioactivity measured in the field could be converted to picocuries per square centimeter and used in a radioactivity budget.

Duplication of the environment in which the probe was actually used is difficult. To do so, one would have to label a sediment sample with a known amount of activity and distribute it uniformly in a thick layer on the bottom of a tank of water. An uncertainty as to the depth of sediment to be used exists. Calculations (see Appendix) show that the half-thickness of a material with the density of sediment is about five centimeters for a 1.12 MeV gamma ray (e.g. Zn^{65}). Since use of large amounts of radioactive tracer was not feasible due to university restrictions, only the surface was coated with radioactivity. The surface layer of the sediments is expected to be the most active anyway since it contains the sediments which have been in most recent contact with Columbia River water.

Calibration was accomplished by distributing a known amount of radionuclide on the surface of a circular board and counting it with the sediment probe. A 30 cm radius was chosen because nearly 90% of the 1.12 MeV gamma-rays from Zn^{65} and over 95% of the 0.32 MeV gamma-rays from Cr^{51} are absorbed in 30 cm of water (see Appendix).

In addition to the absorption, geometrical spreading rapidly decreases the activity detected by the crystal as the source and the detector are separated. The conclusion is that the activity seen by the probe beyond this 30 cm radius is insignificant compared with the activity near the detector.

Chromium-51 (1.79 μc) and Zn^{65} (1.30 μc) were spread evenly over the circular board. Several coats of lacquer waterproofed the board so that a negligible amount of activity would be lost during the calibration. The board was placed in a tank of water 20 cm off the bottom and several counts were made with the sediment probe placed in the center of the board. Data reduction was identical to the method used for field observations (see Data Reduction). The areas under the Cr^{51} and Zn^{65} photopeaks of the river sediment spectra were compared with those of the calibration spectra to determine the activity of these two nuclides in pc/cm^2. Table 1 lists the factors obtained in this calibration.

In calibrating the probe, it was assumed that all radioactivity is confined to the sediment surface. This is not the case in the river, and some contribution to the spectra would be expected from gamma emitters in sub-surface layers. These would cause the surface layer to appear more radioactive than it actually is. Thus probe measurements give the maximum amount of radioactivity present in the surface layer. On the other hand, gamma rays from

TABLE 1 Photopeak areas for Cr^{51} and Zn^{65} calibration of in situ sediment probe.

Run number	Duration of count (min.)	Cr^{51}				Zn^{65}			
		Photopeak area	Planimeter factor	Corrected area	$\dfrac{\text{Area}}{\text{time}}$	Photopeak area	Planimeter factor	Corrected area	$\dfrac{\text{Area}}{\text{time}}$
1	2	0.185	0.5	0.092	0.0462	0.762	2.5	1.905	0.952
2	10	0.218	2	0.436	0.0436	0.980	10	9.80	0.980
3	10	0.220	2	0.440	0.0440	0.995	10	9.95	0.995
4	10	0.437	1	0.437	0.0437	0.943	10	9.43	0.943
AVERAGE					0.0444				0.968

radionuclides in sub-surface layers would be attenuated by overlying

sediments and would not be accurately evaluated by the probe. For

example, if a radioactive layer were covered with several centi-

meters of inert sediments, it would appear less radioactive to the

probe than if the layer were at the surface. Thus radioactivity of

the entire sediment column could be higher than probe data would

indicate. In summary, values for radioactivity with the probe de-

fine an upper limit for radioactivity in the surface layer, but more

nearly a lower limit for radioactivity distributed down through the

layers of sediment.

It should be pointed out that these errors are not particularly

objectionable, since they are essentially constant in all of the probe

measurements. They are minimized by the inherent property of

sediments which finds new (i. e. radioactive) sediments normally on

top of old sediments. Any radioactivity present in the old sediments

would be subjected to loss by decay, with Cr^{51} particularly vulner-

able. The radioactivity counted by the in situ probe corresponds

closely with that which benthic organisms experience. They are not

exposed to all the radioactivity of the sediment column either, be-

cause of attenuation of radiation from deeper sediments.

Data Reduction

In multichannel gamma ray analysis the amount of each

radionuclide present is directly proportional to the area under its
photopeak. If the gain is increased so that the photopeak is spread
out, the peak becomes shorter; if the gain is decreased, the photo-
peak becomes relatively more tall and narrow. However, the area
under the photopeak for a given sample remains the same with any
reasonable gain setting (Baird-Atomic, 1960). Although the in situ
probe was quite drift-free, a small amount of drift causing the gain
to change slightly does not affect the results of the count. This was
checked by comparing areas under the photopeak for a sample at
different gain settings. The results of this experiment are given in
Table 2.

TABLE 2   Photopeak areas for different gain settings of the
           internal amplifier.

Coarse gain	Fine gain	Planimeter areas
1	562	0. 157
1	962	0. 171
2	562	0. 178
2	962	0. 156
4	562	0. 139
4	962	0. 195

Amounts of Cr^{51} and Zn^{65} in surface sediments from each

station were determined graphically. That is, a linear display of the spectrum was made by plotting counts per channel vs. energy using digital data from the typed readout sheet.[2] Only the points which defined the respective Cr^{51} and Zn^{65} photopeaks were used. A smooth curve was fitted to the points and a baseline drawn to follow the background and Compton continuum underlying the photopeaks (Folsom, 1963).

The area between the photopeak and baseline is proportional to the amount of the radionuclide of interest. Areas were determined with a planimeter.

The activity of the sample was obtained by comparing the area of the photopeak to that of the calibration spectrum described previously. Photopeak areas obtained from the field data were then related to the pc/cm^2 value by the calibration factors listed in Table 3.

TABLE 3   Calibration factors for sediment probe.

Isotope	Amount of activity	Area of board	Photopeak area per minute counted	Calibration factor $\frac{pc/cm^2}{planimeter\ unit/min.}$
Cr^{51}	1.79 μc	2919 cm^2	0.0444	138×10^2
Zn^{65}	1.30 μc	2919 cm^2	0.968	4.5×10^2

---

[2]Owing to the large range of counting rates observed, several different vertical scales were used so that the greater part of a sheet of graph paper was used for each photopeak in order to minimize errors in planimetering. Each area was later multiplied by a proper factor.

RESULTS AND DISCUSSION

Gamma Emitters Present in Sediment

Columbia River water diverted through the Hanford reactors as a coolant contains more than 60 different trace radionuclides (Watson, Davis and Hanson, 1963). Most of these are produced by (n, γ) reactions on the parent materials in the river water. Fission products are normally present in much lower concentrations than activation products (Nelson, Perkins and Nielsen, 1964). The longer-lived radionuclides, Zn^{65} (245 days) and Cr^{51} (27.8 days) are the most prominent gamma emitters in estuarine water (Osterberg, Jennings and Cutshall, 1964) and sediments (Jennings, 1965). Also the neutron-induced radionuclides Sc^{46}, Co^{60}, Mn^{54} and the fission products Zr^{95}-Nb^{95}, $Ru^{103-106}$, Ce^{141}, and Cs^{137}, have been identified in the river water and on suspended sediments (Nelson, Perkins and Nielsen, 1964) and on the oceanic sediments near the Columbia River mouth (Osterberg, Kulm and Byrne, 1963), but are present only in very small amounts.

In spectra recorded with the _in situ_ probe from the Columbia River sediments Cr^{51}, Mn^{54}, Zn^{65}, Co^{60}, and K^{40} have been iden-tified. Cr^{51} and Zn^{65} were most abundant. The natural radioiso-tope of potassium, K^{40}, was readily identifiable at most stations and many samples showed distinguishable Co^{60} peaks. Mn^{54}

seemed to be present in several samples, but could never be iden-
tified with complete certainty from the field spectra. However,
samples of the cores counted in the well crystal in the laboratory
showed that Mn^{54} was definitely present.

When Mn^{54} (0. 84 MeV) is present in only small amounts it is
often masked by the high Compton edge resulting from the Zn^{65} (1. 12
MeV). Scattering and energy degradation are especially expected
when the source material is distributed through a dense medium such
as sediment. Proctor et al. (1962) report a high Compton continuum
when the source is distributed in water. Figure 11 compares the
spectrum of Zn^{65} distributed in water with that of Zn^{65} as a point
source. Spectra taken with the sediment probe resting on the sedi-
ment and suspended in the water one meter below the surface at sta-
tion 27 (Figure 12) show that more spreading occurs in the Cr^{51}
photopeak on the sediment than in the water. This is due to an even
greater energy degradation in the more dense sediments. Compton
continuum is present in both, however.

Figure 12 also presents an opportunity to compare the activity
levels of the water with those of the sediment. Both spectra are
graphed on the same scale and have equal counting times. The radio-
activity of the sediment is an order of magnitude higher than the wa-
ter with an enhancement of Zn^{65} with respect to Cr^{51}. Enhance-
ment is due to the greater tendency of Zn^{65} toward association with
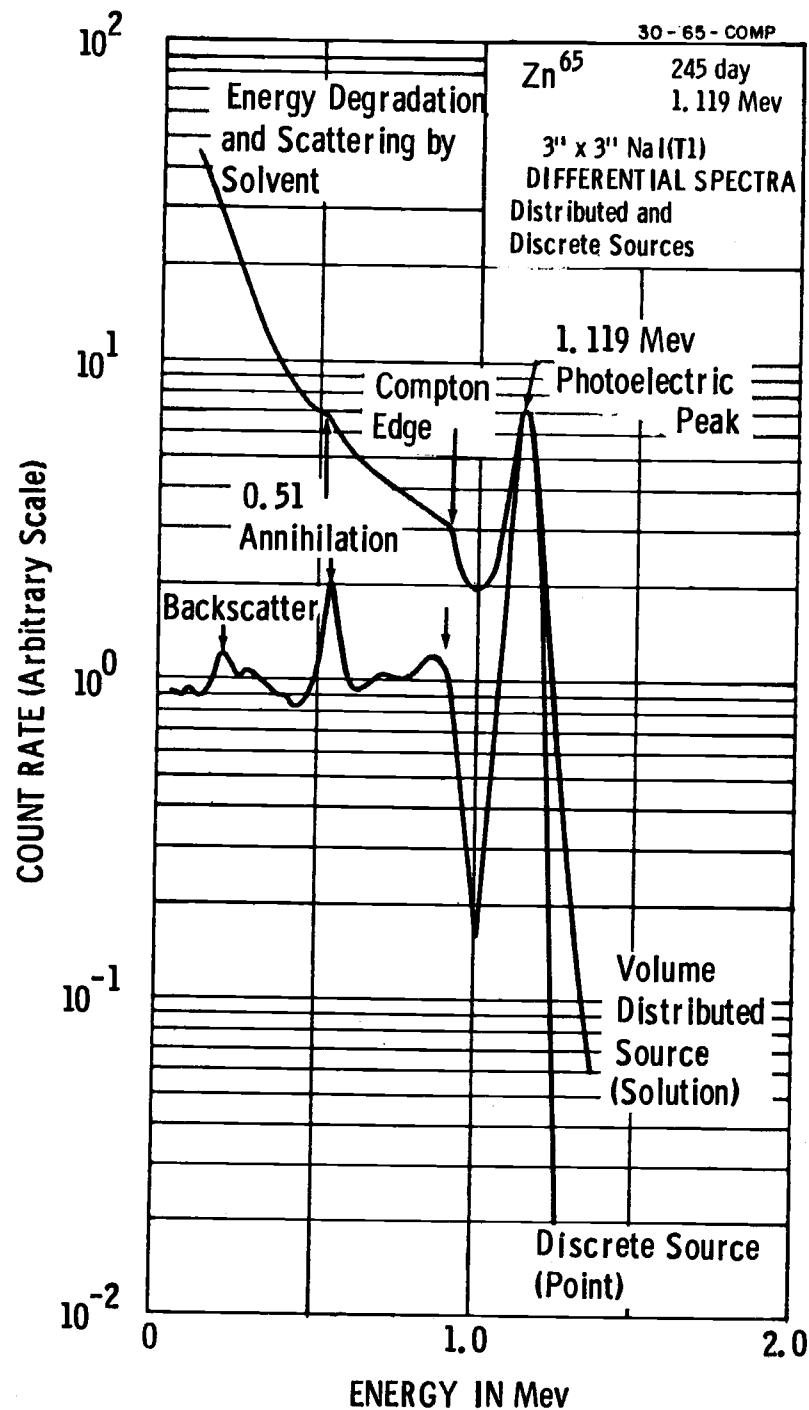
Figure 11. Comparison of the spectrum of a point source
of Zn[65] with the distributed source spectrum
from a detector immersed in 2,300 gal of a
solution of the nuclide. (Proctor et al., 1962,
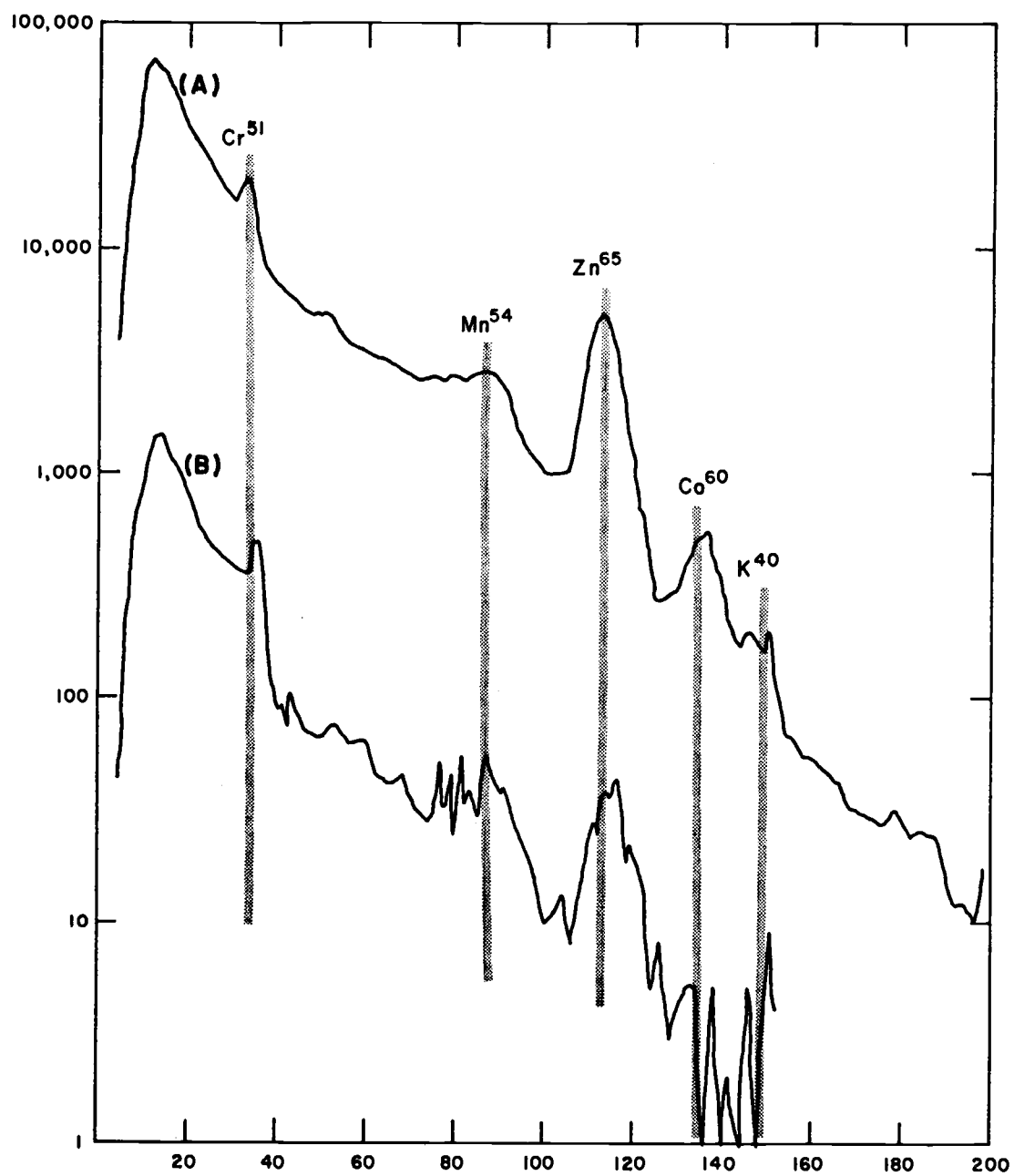reproduced by permission).

Figure 12.   Spectrum with (A) probe on sediment and
(B) probe at the water surface.   Counting
times were equal.

particulate material (Nelson, Perkins and Nielsen, 1964).

Values for Cr^{51} and Zn^{65} in the river sediments appear in Figure 10, with collecting sites shown in Figure 9. The areas highest in radioactivity are generally in protected regions of low river flow where sedimentation would be most likely to occur. Millers Sands, a small island near the river channel, protects station 9, where the activity was high. The area behind Tongue Point (Stations 26 and 27) appears to be a sediment trap, too. Station 22, in the quiet waters near the docks in Astoria, and Station 20 in Youngs Bay, are also shielded from the swift current, and were high in radioactivity. Not all stations in quiet water showed high levels of radioactivity, however. Stations 7, 8, and 28 are in protected areas but were not high in radioactivity. Variables other than current must affect the amount of activity in an area.

Current action and flocculation of colloidal particles by sea water are primary factors in the location of shoaling in the Savannah River estuary (Harris, 1963). These processes are likely important in the Columbia River as well. The area of sedimentation due to flocculation moves with the salt wedge. At stages of low tide, high river flow often limits maximum salinity intrusion to the first 9 km of the Columbia River estuary. At low river flow and high tide, the maximum salinity intrusion may extend over 28 km (Neal, 1965). The region of highest flocculation varies within these limits.

It has been determined that a salinity of about 5‰ is sufficient to completely flocculate Savannah River water within three to 12 hours (Harris, 1963). In the Savannah River (Ibid.) and in several Pacific coast streams (Waldichuk, 1961; Klingeman and Kaufman, 1963) rapid deposition of the bulk of the silt load appears to occur near the estuary. While this may also be true in the Columbia River, the fact that sands are the most prominent bottom material indicates that the swift tidal currents sweep away these finer sediments from the area of deposition.

Distribution of K^{40}, Co^{60}, and Mn^{54} in estuarine sediments is shown in Figure 13. The shadings indicate only whether the radionuclide was observed at that station but do not show the amount present. It is interesting to note that stations with high total radioactivity usually had all three of these minor radionuclides present. Areas of low activity often contained only Co^{60} and K^{40}, or K^{40} alone.

Although Columbia River sediments do not constitute a health hazard now, the fact that they concentrate radionuclides demands attention. Sayre, Guy and Chamberlain (1963) expressed their concern when they wrote:

> In a natural stream environment, however, uncontrolled sorption of radioactive substances by sediments must be considered a potential hazard. Radioactivity could build up over long periods of time in sections of a stream where the rate of accumulation of radioactivity induced by sediment deposition exceeds the rate of natural radioactive decay.
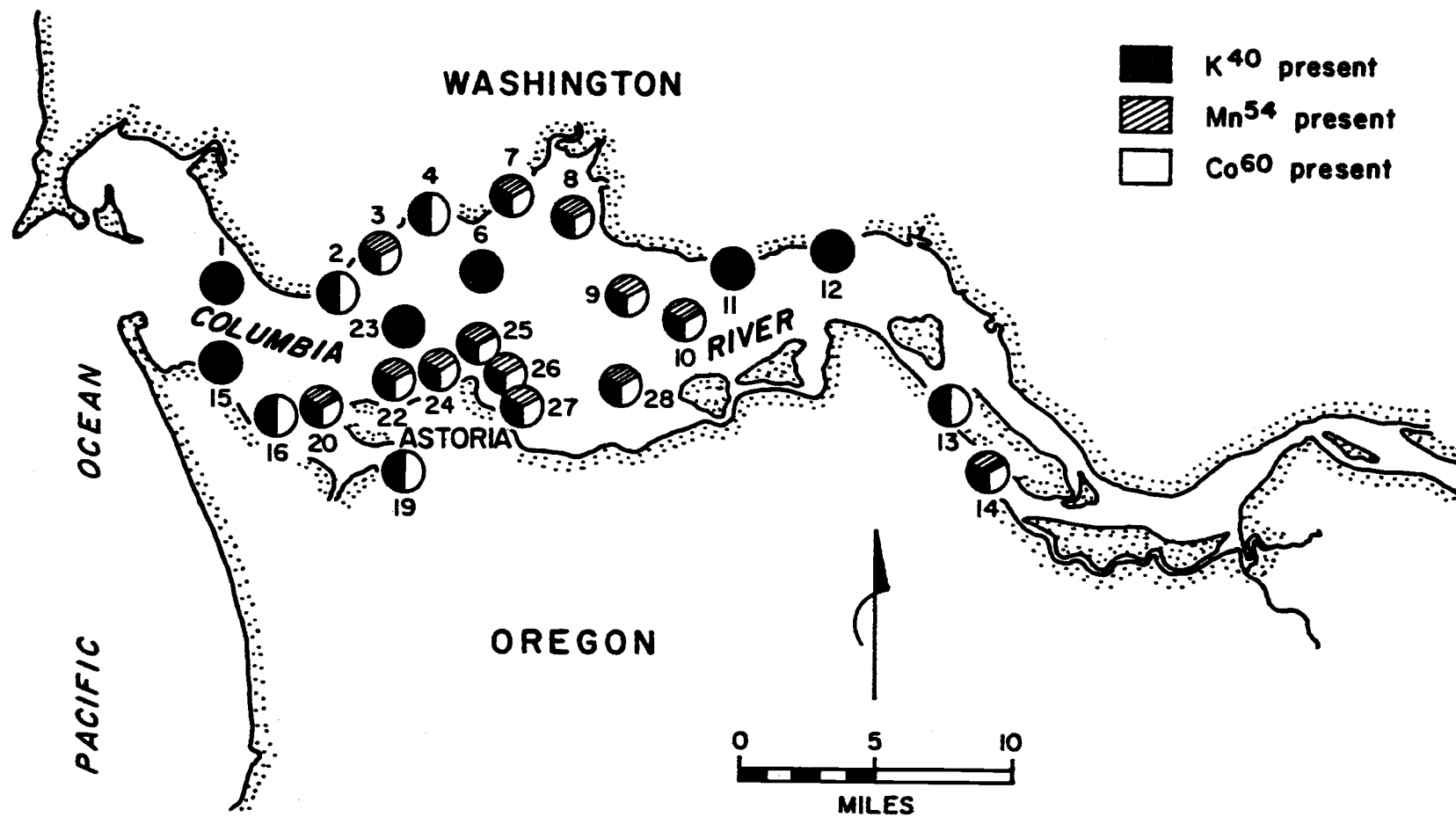
Figure 13. Distribution of K^{40}, Co^{60}, and Mn^{54} in Columbia River estuarine sediments.

Scouring of Sediments During High Water

The flood during the Christmas week of 1964 afforded an excellent opportunity to see the results of high river flow on the sediment radioactivity. Fishermen in the area reported that during the high flood waters the direction of river flow did not change with the ebb and flow of the tide. During flood tide the velocity of outflow would decrease but tidal reversal, which occurs under normal conditions, was not observed.

Surface sediment radioactivity at several stations three months before the flood and about two weeks after the flood are plotted in Figures 14 and 15, respectively. This survey was made as soon as flood conditions allowed, while the river was still high and the water was extremely muddy from suspended material.

Before the flood, stations 9 and 26 had high surface radioactivity. The sediments in these areas were very fine sands and muds. After the flood the activity was much lower, and the bottom was now sandy. It seems plain that high waters had scoured these protected areas which previously had been regions of sedimentation. At station 9 the Cr^{51}/Zn^{65} ratio before the flood was 5.48; after the flood it was 2.32 (Table 4). Chromium-51 has a shorter half-life than Zn^{65}, so that the lower ratio after the flood indicates that these sediments were older than those sampled before the flood. A
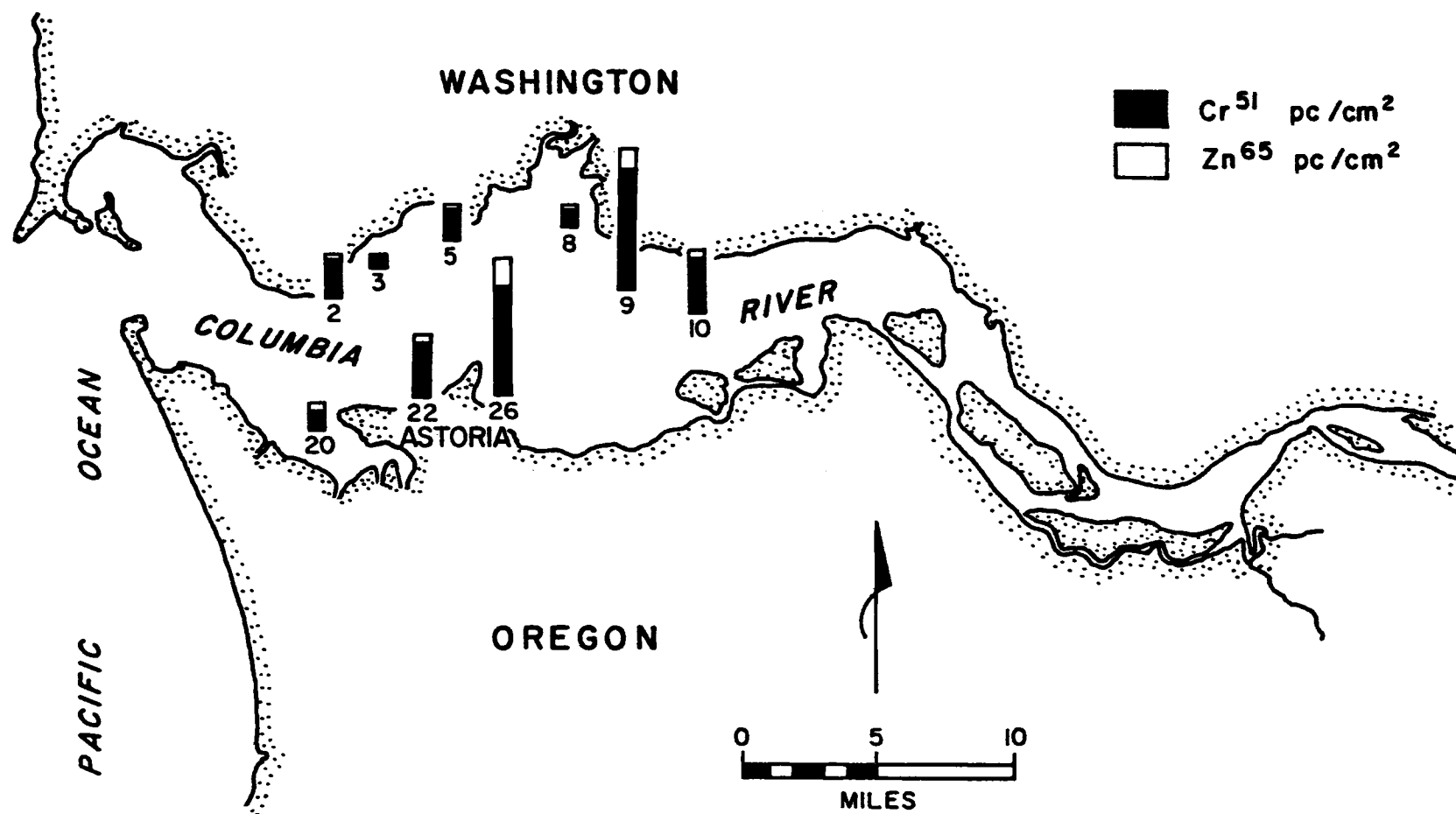
Figure 14. Sediment radioactivity before the flood of Christmas week 1964.
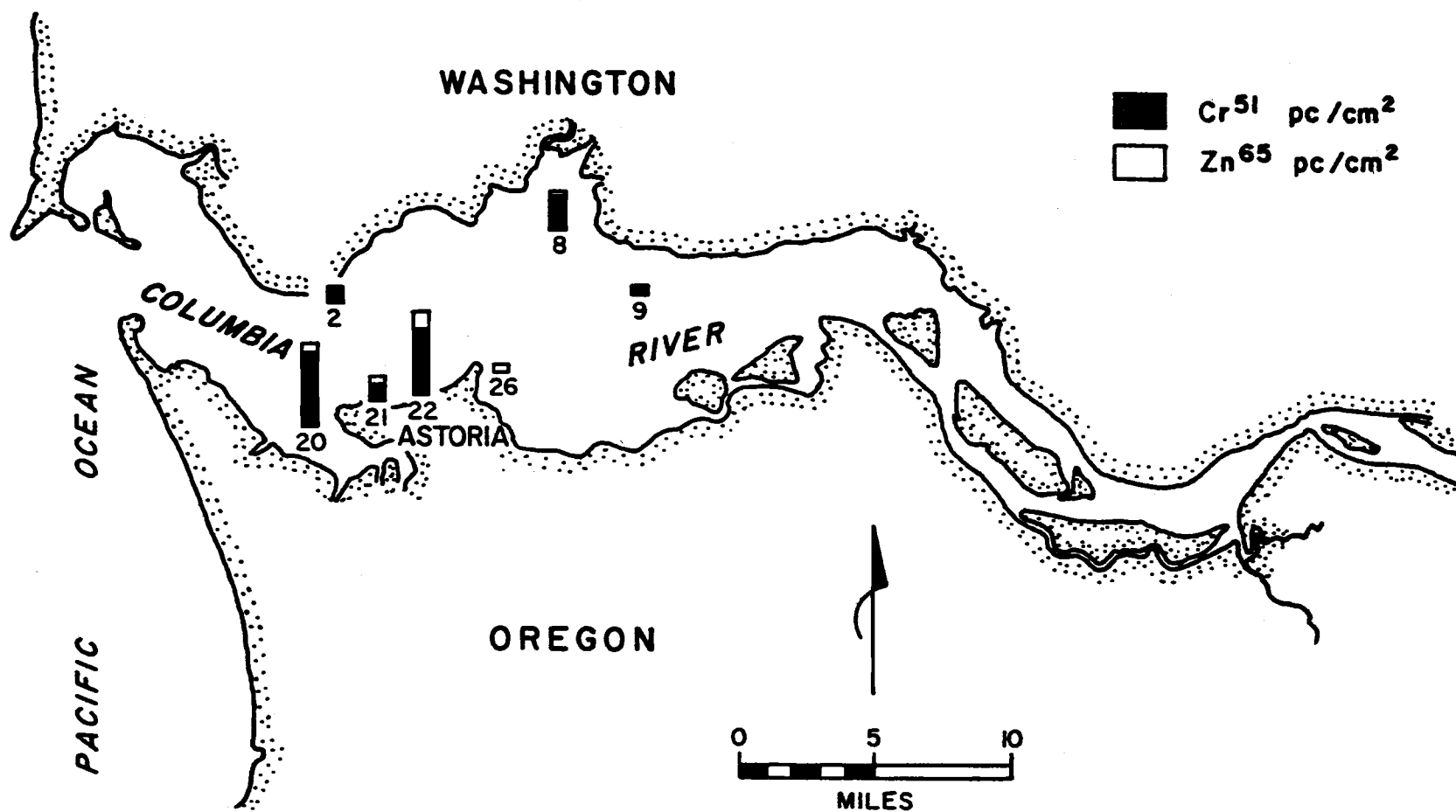
Figure 15. Sediment radioactivity after the flood of Christmas week 1964.

similar situation occurs at station 26 where the Cr^{51}/Zn^{65} ratio

before the flood was 3.63. After the flood the Cr^{51} was too low to

measure, indicating that only old sediments remained.

On the other hand, stations 8, 20, and 22 have higher surface

radioactivity values after the flood (Figure 14). These are presum-

ably areas of redeposition of sediments scoured from points up river.

The Cr^{51}/Zn^{65} ratios at these stations before and after the flood

show higher ratios after the flood in each case (Table 4), thus sub-

stantiating the conclusion that these are areas of recent deposition.

TABLE 4 Cr^{51}/Zn^{65} before and after the flood of Christmas
week 1964.

Station	Cr^{51}/Zn^{65} before flood	Cr^{51}/Zn^{65} after flood
2	7.14	12.00
9	5.48	2.32
26	3.63	Very low
8	5.32	8.97
20	2.38	11.07
22	4.35	8.01

The data from Station 2 appear anomalous because of the lower

surface activity and a higher Cr^{51}/Zn^{65} ratio after the flood. This

station, near the ferry dock at Megler, Washington, is close to the

river channel and may be kept free of sedimentation by the current at all times.

Scouring of sediments during spring high waters has also been reported in the upper Columbia River (Nelson, Perkins and Nielsen, 1964). This was determined indirectly by water radioactivity measurements. A comparison of radionuclides in the river water at Pasco, Washington, Hood River, Oregon, and Vancouver, Washington, showed that those radionuclides associated with particulate material are depleted from the river water by a factor of five to ten during periods of low flow rate. During the spring freshet, when the river flow rate increases, the transport of these radionuclides at Vancouver may equal or exceed the input at Pasco. This situation is due to scouring action which re-suspends deposited sediments (Perkins, Nelson and Hauschild, n. d.).

<div align="center">Particle Size Distribution</div>

Whenever possible a core sample was collected with a Phleger coring device at each station. This worked best in fine sediments, but not very well in coarse sands. The core samples were dried and sieved through Tyler standard screens, separating the sediments into six size fractions with screens of 0. 991 mm, 0. 495 mm, 0. 256 mm, 0. 124 mm, and 0. 061 mm. These portions were weighed and the Φ-mean diameter of each sample calculated by an IBM 1410 computer

by the equations of Inman (1952).

The finer sediments (larger Φ values) seem to contain more of the radioactive material than do the coarser sediments, but the relationship was not linear. Perhaps radioactive decay and local variations in the sediments tend to mask the effect.

The mean diameters calculated imply that most Columbia River sediments fall into the very fine sand category of 3.0 to 4.0 Φ-units (1/8 mm to 1/16 mm) (Shepard, 1963). This conclusion is biased by the fact that only fine sediments could be collected. Lockett (1965) shows that much of the sediment falls in the medium sand range of 1.0 to 2.0 Φ-units ($\frac{1}{2}$ mm to $\frac{1}{4}$ mm). A more representative cross section would be required to establish the definite relationship between mean particle size and radioactivity. However, a trend toward higher activity in fine sediments was reported by Nelson, Perkins and Nielsen (1965).

Youngs River

A sidelight to the Columbia River study occurred when the probe was taken into Youngs River. The sediment in Youngs Bay just below Astoria is primarily mud; also Youngs River has a bottom composed mostly of mud with some rocky places. Flow rate of the river is low so that at high tide considerable penetration of

Columbia River water occurs at certain times of the year.[3] This

exposes the Youngs River sediments to the Hanford-induced radio-

nuclides.

On two cruises measurements were made to determine how

far up Youngs River artificial radioactivity could be detected.

Sampling sites are shown in Figure 16 and the sediment radioactiv-

ity for each station is shown in Figure 17. The trip in September

1964, was halted about 1. 5 km above the confluence of the Youngs

River with the Klatskanine River at station 35 (Figure 16). At this

point Cr^{51} and Zn^{65} could still be detected although the level was

very low.

In January 1965, following the Christmas week flood, the

measurements were repeated, and activities were much lower.

The explanation must be that the sediments containing the higher

activity were washed out by the flood waters.

An interesting relationship appears when the Cr^{51}/Zn^{65} ratios

for the September cruise are plotted against distance from the mouth

of the Youngs River (Figure 18). The Cr^{51}/Zn^{65} ratios increase to

a point 3 km up the river and then decrease to the end of our sampling

stations.

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[3] The average flow rate for Youngs River in 1956 was 218 cfs.
During the month of September the flow rate was below 7. 0 cfs on
some days (Wells, 1959).
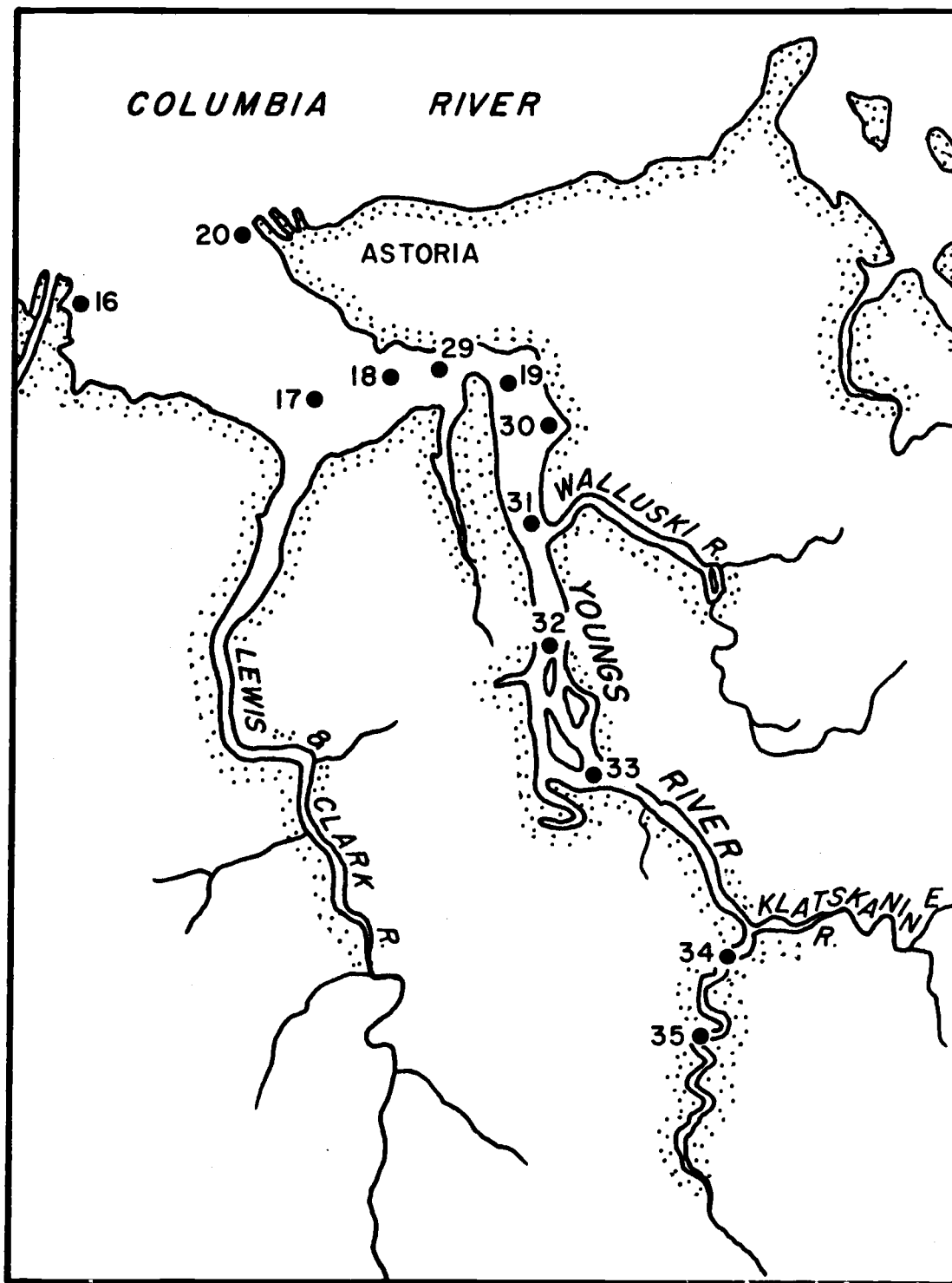
Figure 16. Sample site locations in Youngs River. (See Figure 17 for surface sediment radioactivity.)

	16	17	17	18	18	18	29	29	19	19	30	31	32	32	33	33	34	34	35	SAMPLE SITE
	8.70	5.23	5.77	10.50	11.70	7.00	2.02	4.97	15.83	12.15	2.92	11.64	7.41	5.42	5.71	—	6.58	—	—	Cr^{51}/Zn^{65}

LEGEND

Zn^{65} pc/cm^2

Cr^{51} pc/cm^2
AUG. 1964

Cr^{51} pc/cm^2
SEPT. 1964

Cr^{51} pc/cm^2
JAN. 1965

Figure 17. Surface sediment radioactivity in Youngs River. (See Figure 16 for sample site locations.)
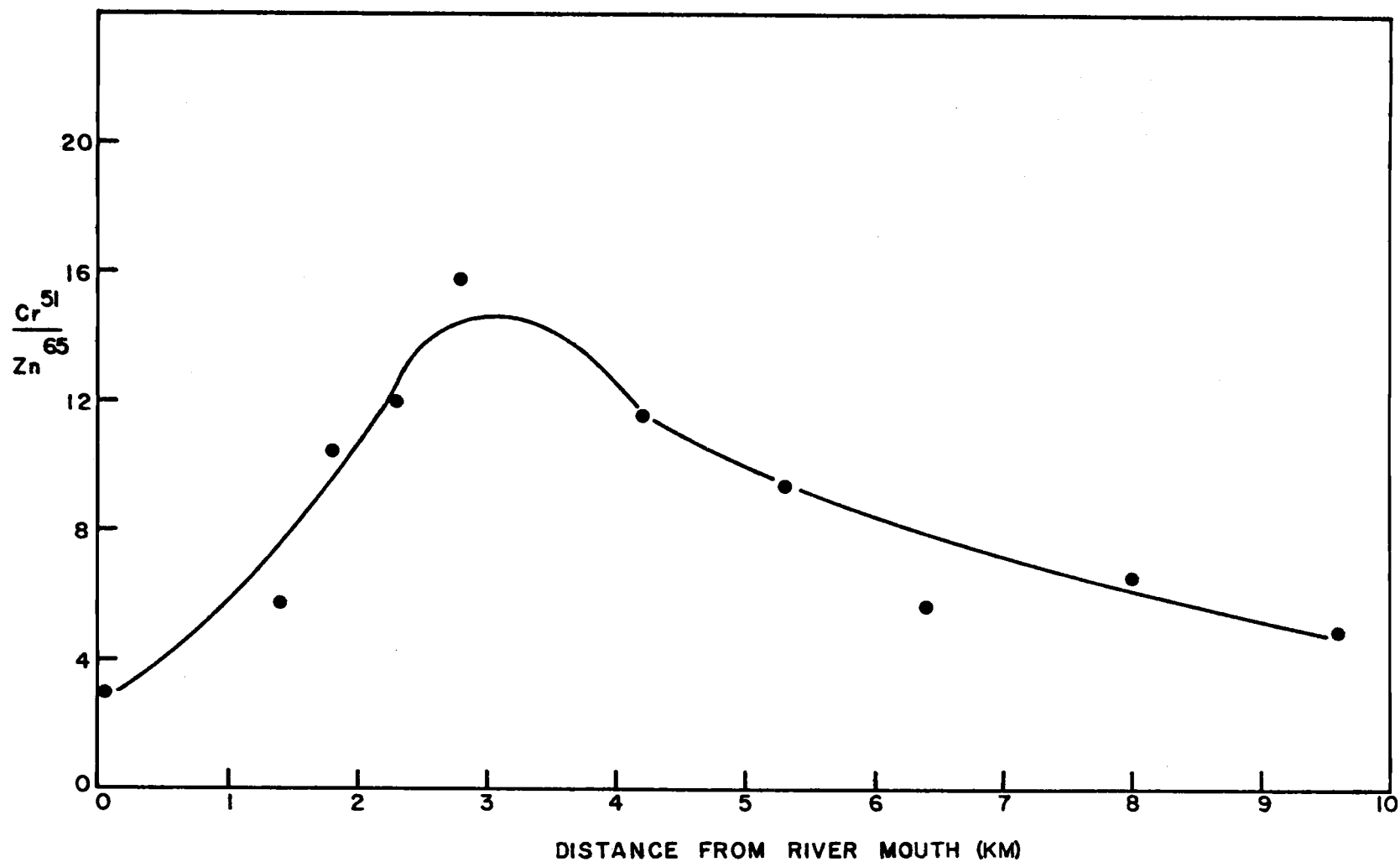
Figure 18. Cr^{51}/Zn^{65} activity ratio vs. distance up Youngs River from Pier 3 in Youngs Bay.

Up the river from station 19 the decrease in the ratio of Cr^{51} to Zn^{65} can be explained by the fact that these sediments are not in contact with Columbia River water as often as those downstream. Station 35 would be expected to contact Columbia River water only at times of high tide and low flow from Youngs River. The downstream stations would see the radioactive water more often. In terms of contact with the Columbia water the sediments appear to be older and older at points upstream from station 19.

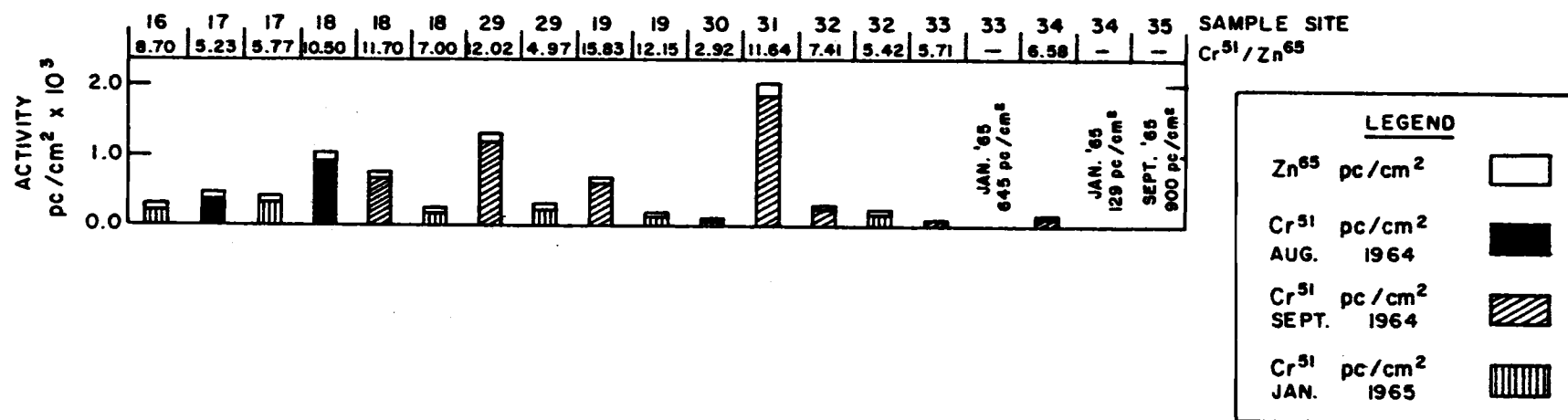Owing to the low flow of the Youngs River and the relatively high tides which occur in the Columbia River, station 19 probably contacts water from the radioactive Columbia on almost every tidal cycle. Cr^{51} seems to be increasingly sorbed by the sediments from the mouth of Youngs River to station 19.

This phenomenon may be due to the chemical behavior of chromium. Present data indicate (Nelson, Perkins and Nielsen, 1964; 1965) that Cr^{51} in the river water remains almost completely in the form of its hexavalent anion, $Cr_2O_7^{-2}$ or CrO_4^{-2}. Experiments by Nelson and Cutshall (1965) show that Cr^{51} in the form of the trivalent cation is rapidly taken up by the sediments (see Figure 19). Chromium-III is taken up completely and rapidly whether or not the sediment has been oxidized with H_2O_2. Chromium-VI is taken up on normal sediment slowly compared to Cr-III. If, however, the sediments are previously oxidized with H_2O_2, there is no change in the
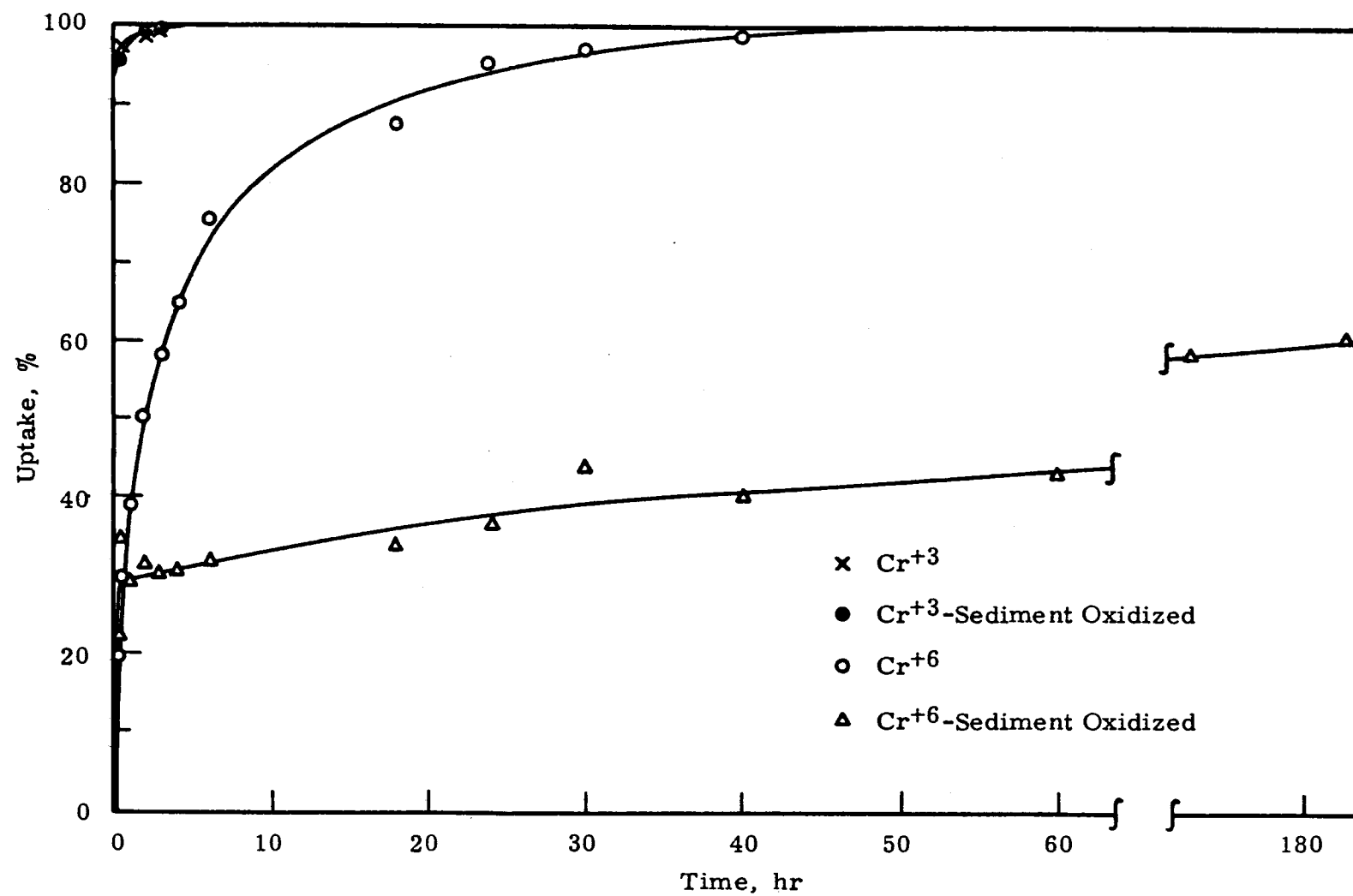
Figure 19. Rate of uptake of Cr^{51} on Columbia River sediment. (Nelson and Cutshall, 1965, reproduced by permission).

uptake of Cr-III but uptake of Cr-VI occurs very slowly. This indi-
cates that if Cr-VI is reduced to Cr-III it will be taken up by the sedi-
ments. Thus, reducing environment would enhance the sorption of
Cr^{51} on sediments. The region of Youngs Bay from its mouth to
station 19 appears to have the proper environment to allow Cr^{51} to
associate with the sediments. Further experiments are indicated
to establish this hypothesis.

Ocean Sediments

In addition to the Columbia River and Youngs River analyses
a few sediment counts were made in shallow coastal waters of the
Pacific Ocean. Figure 20(a) shows a spectrum counted 4.8 km off
the mouth of the Columbia River. Figure 20(b) gives a similar spec-
trum beneath 30 m of water, 1.6 km off Newport, Oregon. Counting
time was 40 minutes but the spectrum was normalized to ten minutes
(all counts multiplied by 0.25) for comparison with Figure 20(a).
There is less scatter in the high-energy portion of this spectrum as
a result of better statistics from the longer count time and an actual
higher counting rate from the emitting substance whose peak falls at
the far right of the spectrum. Cr^{51}, Zn^{65}, and Co^{60} are principally
from Hanford, Washington, and K^{40} and Bi^{214} are naturally occur-
ring. The high-energy peak (lower right) has been identified as
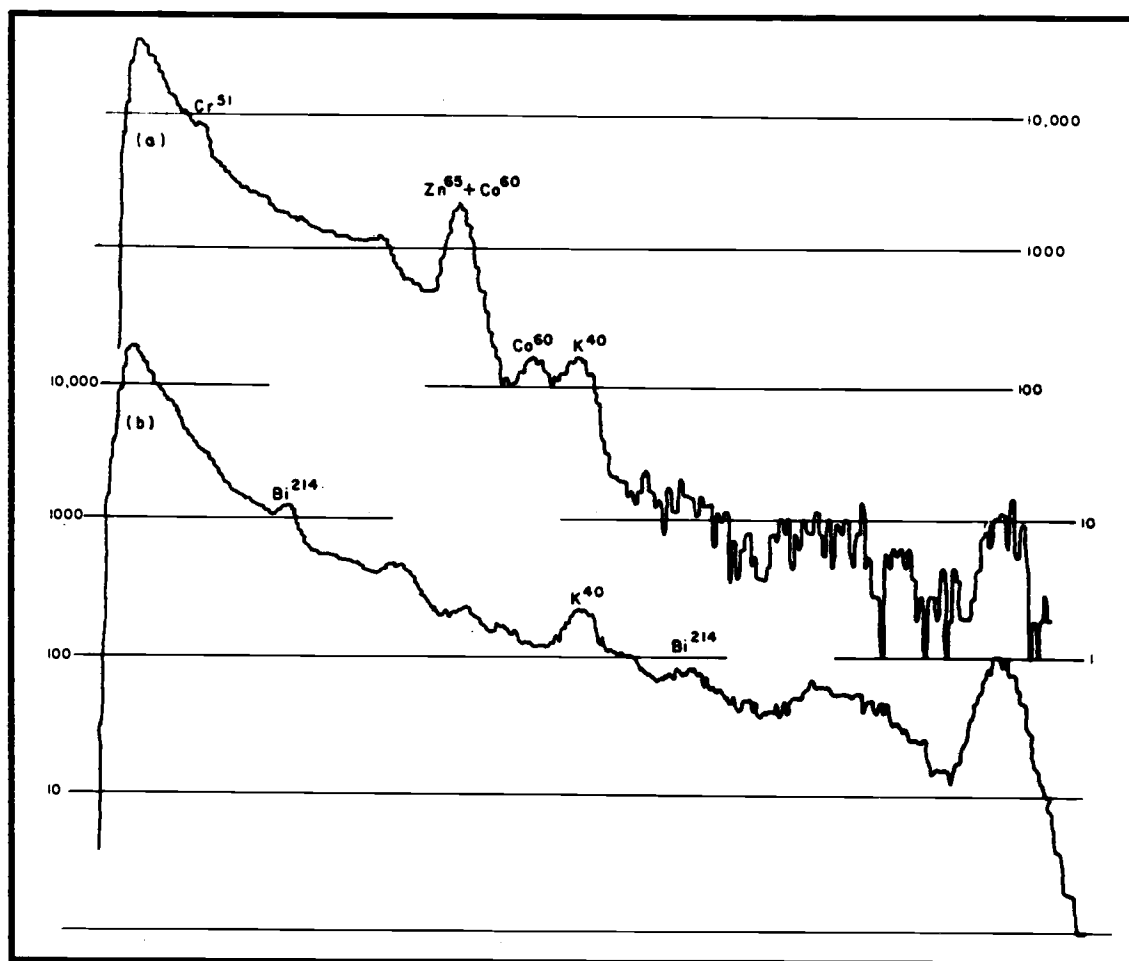Tl^{208}.

Figure 20.   Spectrum of (a) fine sand beneath 21 m of water, 4. 8 km off the mouth of the
Columbia River and (b) coarse sand and gravel beneath 30 m of water, 1. 6 km
off Newport, Oregon.   (Jennings, Cutshall and Osterberg, 1965).

SUMMARY AND CONCLUSIONS

Determination of the sediment sorbed radioactivity is greatly facilitated by the use of an in situ probe. The regions of high and low radioactivity can be determined with less expenditure of time than a sampling program which couples field collection with laboratory radioanalysis. A primary advantage of the probe is that it can sample any kind of sediment while collection of sediments is difficult in some areas. Data are available immediately so that the sampling plan can be altered if the values so dictate.

The primary disadvantage of the probe is its failure to produce a sample to be studied in detail in the laboratory. Collection of a sample in the same general area is helpful, but not completely comparable owing to some striking local variations.

The inherent complexity of the Columbia River physical-chemical-biological system makes it difficult to relate the radioactivity distribution to any one variable. However, certain processes have been suggested which affect the sediment radioactivity distribution.

Flocculation of colloidal material and variations in tidal currents are important factors in sedimentation. Physiography of the estuary also enters into the sedimentation pattern. Islands and river bends cause a decrease in velocity allowing finer particles to settle out.

The chemical state of a radionuclide also modifies its occurrence in the sediment. For example, chromium as trivalent cations seems to have a much greater affinity for the sediments than in the form of its hexavalent anion. Thus an environment in the Columbia River capable of reducing $Cr-VI$ to $Cr-III$ would likely have more Cr^{51} in the sediment.

Biological organisms have been shown to concentrate radionuclides from the Hanford reactors (Osterberg, 1962; 1963; Watson, Davis and Hanson, 1961). As these organisms die they could settle out of the water, increasing the radioactivity of the sediments in a particular region. Acceleration of sinking rates of fallout radionuclides in the ocean has been attributed to faecal pellets of herbivorous zooplankton (Osterberg, Carey and Pearcy, 1963). Although Zn^{65} was not found to move with fallout, this mechanism is important for some radionuclides.

In this study it has been demonstrated that an _in situ_ probe coupled with a spectrometer gives valuable information about the distribution of radioactivity in the sediments. A survey characterizing the surface radioactivity more completely by measuring transects at several places along the estuary should be valuable for future work. Also a nearly untapped source of knowledge lies in a study of sediment radioactivity in the ocean.

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APPENDIX

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Calculation of absorption of Cr^{51} and Zn^{65} gamma-rays in water and sediment.

The equation for attenuation of gamma-rays in matter is

$$I = I_o e^{-\mu x} \qquad \text{Eq. 1}$$

where $\quad$ I_o = original radiation exposure rate,

$\quad$ I = attenuated radiation exposure rate,

$\quad$ μ = linear absorption coefficient (cm^{-1})

$\quad$ $= 0.693/_{x_{\frac{1}{2}}}$, and,

$\quad$ x = absorber thickness (cm) (U. S. Department of Health, Education, and Welfare, 1960).

Solving for the penetration distance gives,

$$x = \frac{2.303 \log I_o/I}{\mu} \qquad \text{Eq. 2}$$

The distance where 90% of the γ-rays from a Zn^{65}(1.12MeV) source are absorbed in water occurs when $I/I_o = 0.1$ and $\mu = 0.068$ cm^{-1} (Ibid.). Equation 2 gives $x_{0.9} = 33.88$ cm for a 1.12 MeV γ-ray in water.

The distance required for 95% of the γ-rays from a Cr^{51}(0.32 MeV) source to be absorbed in water occurs when I/I_o 0.05 and

$\mu = 0.115$ cm^{-1} (Ibid.). Equation 2 gives $x_{0.95} = 26.03$ cm for a

0.32 MeV γ-ray in water.

To calculate the half-thickness of the γ-rays in sediment it is assumed that the linear absorption coefficient for concrete can be used.

For the 1.12 MeV γ-ray, $\mu = 0.143$ cm^{-1} giving $x_{\frac{1}{2}} = 4.86$ cm of sediment for the γ-ray from Zn^{65}. For the 0.32 MeV γ-ray, $\mu = 0.251$ cm^{-1} giving $x_{\frac{1}{2}} = 2.76$ cm of sediment for the γ-ray from Cr^{51}.