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Technical Report No. 84

PRELIMINARY REPORT ON THE SEDIMENTS AND RADIOACTIVITY
IN THE VICINITY OF THE COLUMBIA RIVER EFFLUENT

by

M. Grant Gross, Dean A. McManus, and
Joe S. Creager

Brown Bear Cruise 291
29 July to 12 August 1961
and
Brown Bear Cruise 311
23 July to 14 August 1962

U.S. Atomic Energy Commission
Contract AT-45-1-1725
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Contract Nonr-477(10)
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January 1963

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ABSTRACT

The sediments in the area affected by the runoff from the Columbia River are being studied to determine the distribution of sediment types and the presence and amount of radioactivity in the sediments. The study of the dispersal processes and distributional patterns of the sediment and its radioactivity is in the initial phase of a three-phase program.

Various sedimentological analyses of 178 samples have provided preliminary data on sediment distribution. The inner part of the continental shelf is covered with sand which grades into silt along the outer part of the shelf. The sediment of the abyssal plains is principally a silty clay, although local coarsening of the sediment is produced by the accumulation of tests of microorganisms. The presence of quartz sand in samples from the abyssal plain appears to be restricted to those samples collected from deep-sea channels.

Analyses of the gamma-ray spectra of sediments collected near the Columbia River mouth in 1961 indicate that radionuclides, in addition to the naturally occurring potassium-40, are present in the sediments. The sandy sediments from Grays Harbor and the beaches adjacent to the Columbia River had very low activities although chromium-51, zinc-65, cobalt-57 and cobalt-60 were detected. The fine-grained sediments in and near the river mouth had the highest concentrations of chromium-51 and zinc-65 which decrease by a factor of 10 within one hundred miles of the river mouth. Detectable amounts of chromium-51 and zinc-65 were present in sediments 180 miles south and west from the river mouth. The activity in the silt and clay fractions was five to ten times greater than the activity of the sand fraction.

INTRODUCTION

During 1961 and 1962 the initial phase of an investigation of the marine sediments adjacent to the Oregon-Washington coast was begun as part of a larger study of the Columbia River outflow and its effect on the Northeast Pacific Ocean. The results of the biological, chemical and physical research will be discussed in separate reports. The ultimate objective of the present study is to delineate the areal distribution of Columbia River sediment and to investigate the processes involved in the distribution and deposition of the sediments, as well as the chemical changes that affect the sediments, their radioactivity, and their trace element composition.

As presently envisioned, the study will consist of three phases:

Phase 1. A survey of the sediments in the area affected by the runoff from the Columbia River to determine the distribution of different types of sediment and to detect the presence and amount of radioactivity in the sediments.

Phase 2. A study of the physical, chemical, and mineralogical properties of the sediments for the purpose of determining the sources of the sediments and the sources of their associated radioactivity.

Phase 3. Intensive study and experimentation on basic problems of marine sedimentation and marine geochemistry using the information obtained from the preliminary surveys.

Much of the time in 1961 and 1962 was spent in completing the first phase of the study, i.e. a survey of the sediments and their radioactivity. The data reported here are only the preliminary results of this study. Analyses of the sediments are continuing, and the results of these analyses will be reported in the near future. Appendix 1 shows the present status of the collection and analyses of sediments from the coastal and offshore areas.

SURVEY OF OFFSHORE SEDIMENTS

Two cruises were made in 1961 and 1962 for the purpose of collecting bottom samples in the area of the Columbia River effluent. The first of these cruises, (Brown Bear Cruise 291), had as its objective a reconnaissance of the deposits on the continental terrace, and the eastern edge of the Cascadia Abyssal Plain (Fig. 1) between California and Vancouver Island.

Samples from this cruise indicated the regional distribution of sediments on and adjacent to the continental terrace. However, no data were available to serve as a basis for interpreting the seaward extent of Columbia River sediments and sedimentation. For this reason, a second cruise, (Brown Bear Cruise 311) was made to sample the sediments of the Cascadia Plain. Samples have now been collected, therefore, from the continental terrace, the Cascadia Abyssal Plain, and from the Seamount regions forming the western perimeter of the plain. The 133 samples collected provide only one bottom sample for each 1360 nautical miles. The physiographic and bathymetric complexity of the area (Figs. 1 and 2) limit the usefulness of these samples for regional interpretation. Three physiographic and sedimentary provinces are recognized in the study area: the continental terrace, Cascadia Abyssal Plain, and the Seamount Province, each of which consists of several distinct subdivisions. Although careful selection of sampling localities has provided samples which are thought to be representative of the sediment distribution in the study area, the physiographic complexity and the large size of the area limit description and interpretations of the general sediment distribution to approximately the same level of detail as that reported by Shepard (1939) and Hurley (1960).

The survey of the sediment distribution includes the investigation of at least five individual problems, each of which will be discussed in turn:

- 1) Determination of the magnitude of variations in the composition of bottom sediments in the three sedimentary provinces.
- 2) Estimation of the amount of sediment deposited in each of the sedimentary provinces from terrigenous, pelagic, and seamount sources.
- 3) Examination of the channels across Cascadia Abyssal Plain as significant dispersal routes for sediment from the Columbia River.
- 4) Evaluation of the role of Astoria Canyon in transporting sediments from the Columbia River.
- 5) Selection of samples to be used for determining the sediment depositional rate.

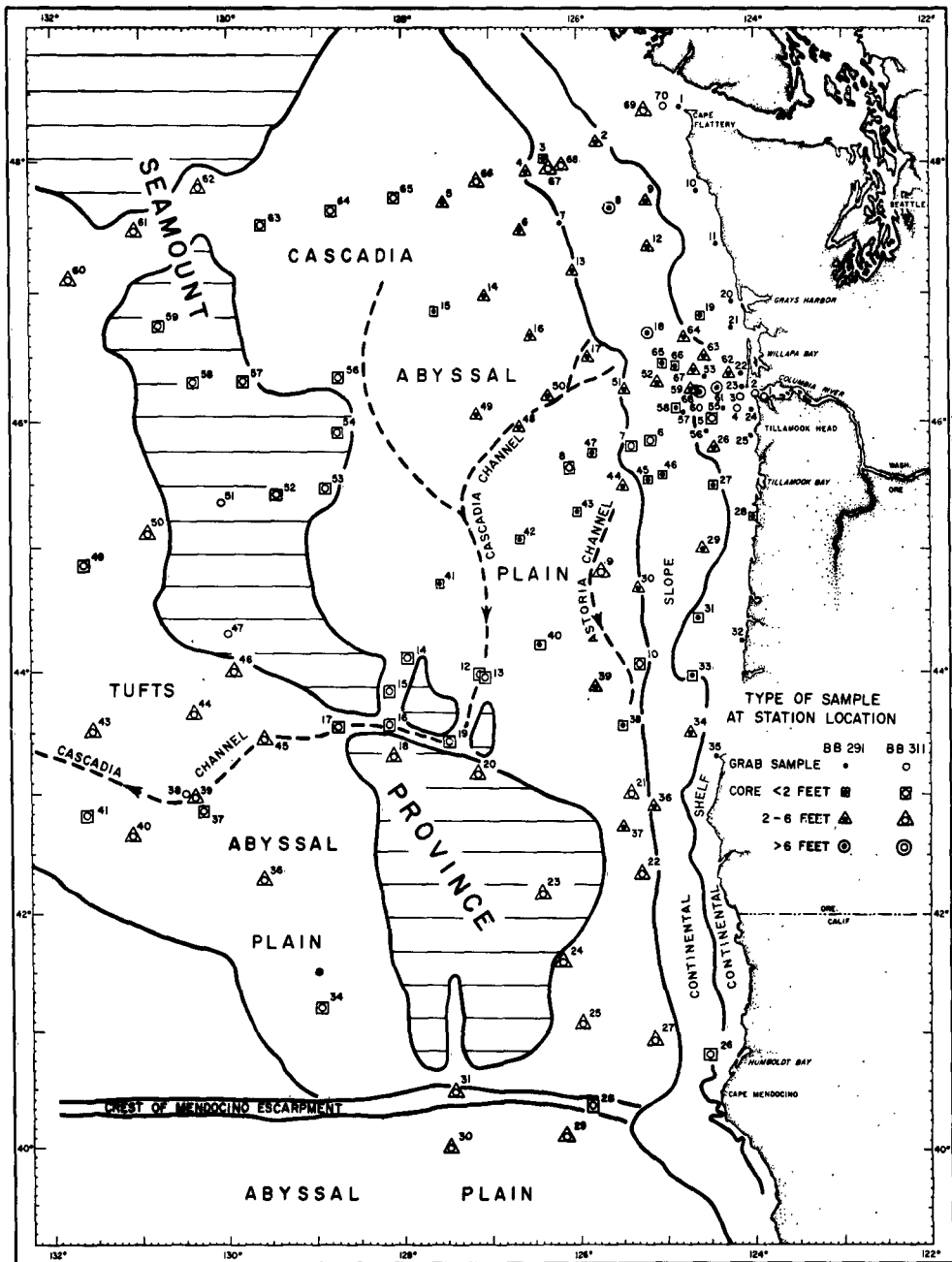


Fig. 1. Index map showing areas sampled during 1961 and 1962 and the physiographic provinces of the Northeast Pacific Ocean along the Oregon-Washington coast.

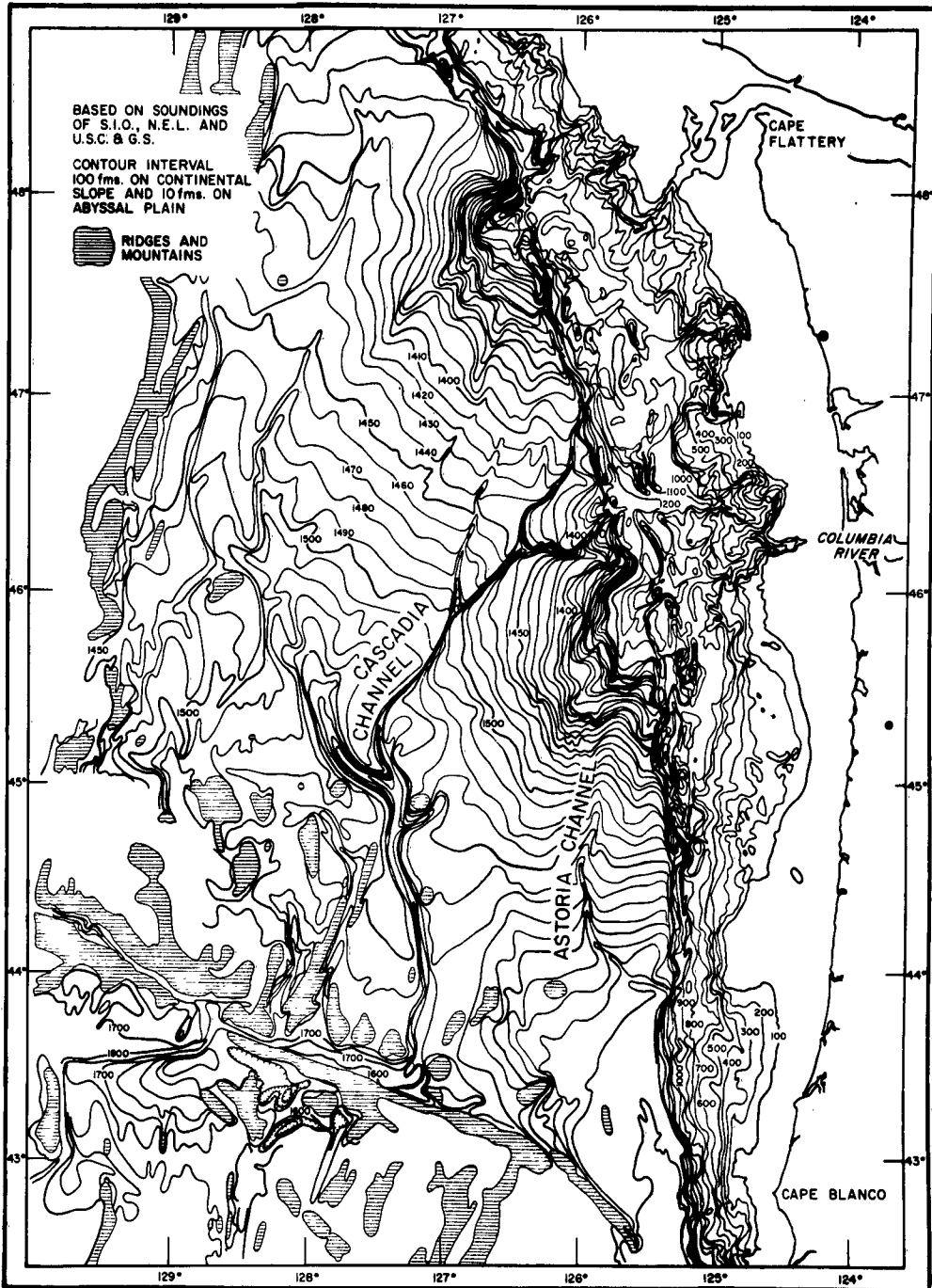


Fig. 2. Bathymetric chart of Cascadia Abyssal Plain and adjacent areas (after Hurley, 1960).

Sediment Variation

In order to recognize sediments derived from the Columbia River, it is necessary to study the shelf, slope and abyssal plain sediments over a wide area (Fig. 3). In general, the sediment in the Columbia River Estuary consists of clean, medium-grained sand¹ with few granules. Sands also occur seaward of the mouth of the river, and comprise most of the sedimentary material bordering Astoria Canyon and covering the inner part of the continental shelf, except locally where the sediment coarsens to gravel e.g. near the Strait of Juan de Fuca; or where rock is exposed, e.g. in the area of irregular topography south of the "mouth" of Astoria Canyon. The inner-shelf sand is, in general, finer grained and darker than the Columbia River estuary sand. The remainder of the shelf is covered with gray to green clayey silt. This gray to green clayey silt also forms a thin deposit overlying hard, gray clay in the inter-ridge areas of the continental slope, such as those off the Oregon coast, and in some of the submarine canyons adjacent to, but excluding Astoria Canyon, which in contrast contains thick silt deposits.

The most common bottom sediment type found on the abyssal plains is silty clay, which covers most of the Cascadia Abyssal Plain, the Tufts Abyssal Plain, and the abyssal plain south of the Mendocino Escarpment. However, there are a few areas occurring between the medial part of Cascadia Plain and the eastern part of Tufts Plain (Fig. 3), where the sediment coarsens to clayey silt. The coarsening of the (bottom) sediment is caused mainly by the abundance of tests of organisms, particularly radiolarian and globigerinid tests. Radiolarian tests account for coarsening of particle size in most of the areas of clayey silt. However, at one location in Cascadia Channel, in the central part of the study-area where a narrow band of clayey silt extends through a passage in the Seamount province (Fig. 3), the coarsening is due to the presence of fine sand-sized quartz grains. Quartz sand grains were not present in quantity in any other surface samples from the channel. In addition to clayey silt, the coarser sediment is also present as silt, but only on Tufts Abyssal Plain at the southwestern end of the central part of the Seamount Province. Here, the globigerinids are more common than the radiolarians, and the silt is part of the Globigerina-rich sediment discussed by Enbysk (1960).

Seamount Province as Sediment Source Area

Although the sediment in the eastern part of Cascadia Abyssal Plain is probably derived mainly from the continent to the east, the sediment in the western part of the plain may contain some material from the adjacent seamounts to the west. The sediment in the Seamount Province and along the western margin of Cascadia Plain consists of Globigerina-rich silts and clays (Nayudu, 1959; Enbysk, 1960). To what extent the silts or clays may represent material derived from the Seamount Province is unknown. An attempt must be made to evaluate the significance of the Seamount Province as a possible source area for sediment on the Cascadia Plain.

¹ The sediment nomenclature proposed by Shepard (1954) will be used in this report.

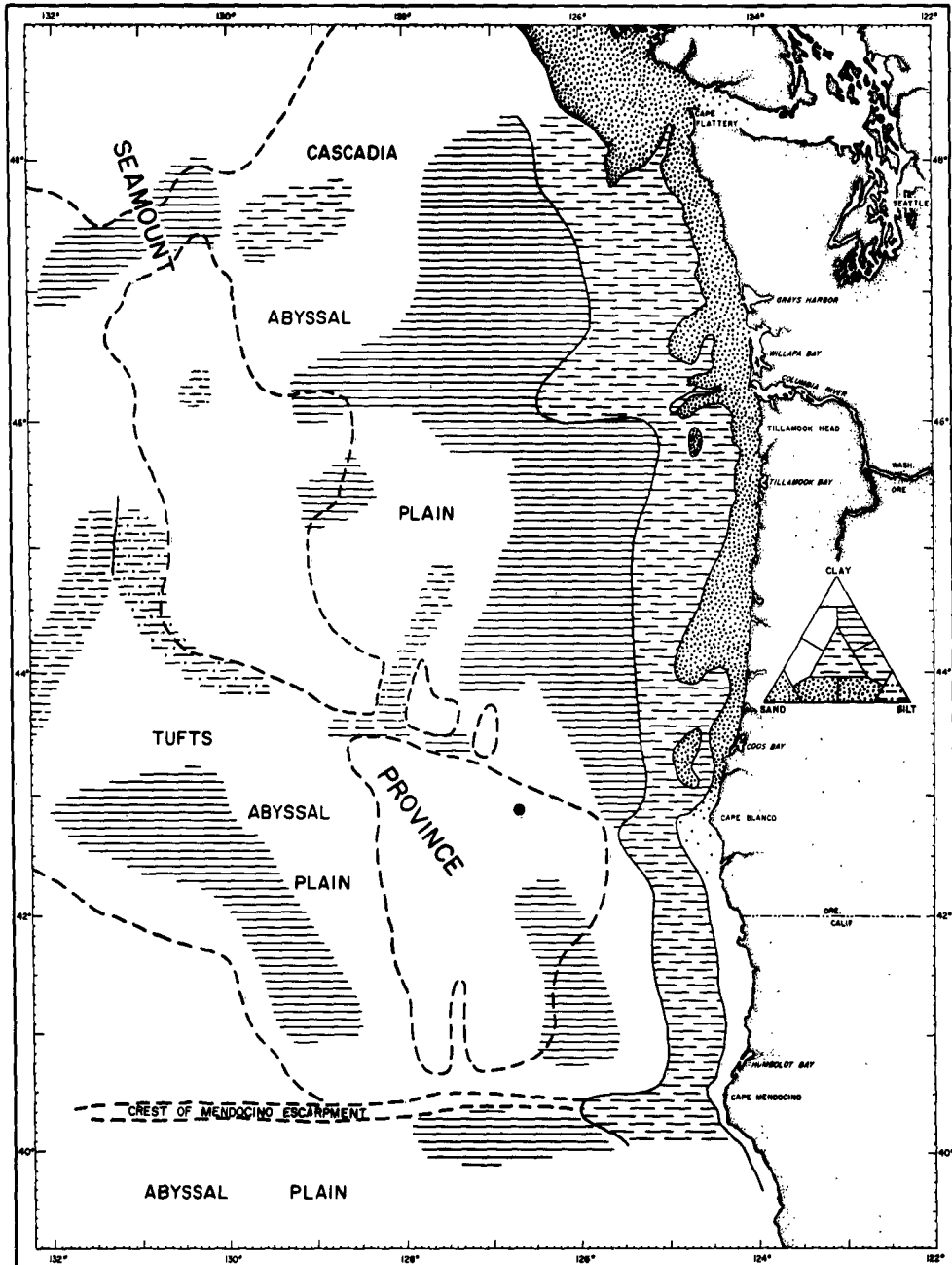


Fig. 3. Distribution of sediments along the Oregon-Washington coast in adjacent offshore areas.

Channels Across Cascadia Abyssal Plain

The most outstanding physiographic feature on the Cascadia Plain is the channels, in particular, Cascadia Channel (Fig. 2), which, along with adjacent channels, was sampled at 6 locations between $43^{\circ} 58.5' N.$, $127^{\circ} 09' W.$ and $43^{\circ} 28.5' N.$, $129^{\circ} 36' W.$ These channels have been proposed as routes for turbidity current movement (Menard, 1955) and estimates of the volume and speed of possible flow have been made (Hurley, 1960). However, sediments sampled from these channels have not yet shown any detectable difference from those of the non-channeled parts of the plains, although a sample from the extension of Cascadia Channel onto the Tufts Abyssal Plain contained sandy silt. This sandy silt was the coarsest sediment sampled in this part of the study area, excluding those samples from the areas of rough topography, such as the Seamount Province, where rock fragments are the most abundant material.

The sandy silt, with sand laminae, is suggestive of turbidity current deposition in this part of the channel. Hurley (1960) states that most of the bottom sediments of Cascadia Plain, including the channel and adjacent areas, are turbidity current deposits. At the present time, analyses merely show little difference in sediment from the two areas, but these analyses are only in the initial stage. However, it is certain, that the radioactive sediment presumably derived from the Columbia River was not restricted to the channels.

Astoria Canyon and the Dispersal Pattern

Samples from the canyon, including 18-foot piston cores, are being analyzed with the object of identifying and separating, if possible, the Columbia River sediment from the shelf sediment. At present, no such successful division has been accomplished, but the presence of abundant clayey silt in the cores, rather than sand, indicates that unusual environmental conditions may exist in and adjacent to Astoria Canyon.

Rate of Deposition

The rate of sedimentation has been estimated at only a few locations in the Northeast Pacific. These locations are very scattered and are for the most part in areas of pelagic sedimentation at some distance from the coast. However, no determinations have been made of the rate of sedimentation in the Cascadia Plain, although a core in the Globigerina-rich silt and clay near Cobb Seamount indicated a rate of two centimeters per one thousand years (Nayudu, 1959, p. 45). At the present time selected cores from each of the major sediment types and sedimentary environments have been selected for dating various layers by means of carbon-14. The cores from which the samples are taken have been chosen to provide the maximum useful information on sedimentation rates in Astoria Canyon, on certain parts of Astoria Fan, and in and near the deep-sea channels.

At the moment the carbon-14 method appears to offer the best possibility for obtaining significant dates for various layers in the sediment. The method is limited to approximately 30,000 years BP so that it can be used for areas of rapid sedimentation or for the upper layers in the cores from areas of slow sedimentation.

With better information about the radionuclides associated with the sediment and their behavior in the marine environment, it may be possible to use the radioactivity derived from the Columbia River or from atmospheric fallout to date the sediments.

SURVEY OF COASTAL SEDIMENTS

In addition to the large area off the Oregon-Washington coast, sampled by the two named Brown Bear cruises, the sedimentary environments immediately adjacent to the Columbia River mouth are being studied. These areas, Grays Harbor, Willapa Bay, Washington, and the ocean beaches between Tillamook Head, Oregon, and Point Brown at Grays Harbor, which are predominantly regions of sand deposition, were studied using small boats from nearby harbors. Separate reports on each area are expected to be completed in the near future. These accounts will deal with the details of the distribution, source areas and transporting processes involving the sediments. Preliminary surveys of the radioactivity of the sediments in each of these environments have been completed and these results are reported in the section on radioactivity.

RADIOACTIVITY OF THE SEDIMENTS

Radioactivity in Columbia River Water

As a result of the operations at the U. S. Atomic Energy Commission's Hanford Plant, approximately 1000 curies per day of radioactive materials (Table 1) were transported into the Columbia River estuary during 1960 and 1961 (Nelson, 1961, 1962).

TABLE 1

Annual average rate of transport of selected major radionuclides past Vancouver, Washington, in 1960 and 1961.

	<u>1960</u>	<u>1961</u>
Chromium-51	850 Curies/day	840 Curies/day
Neptunium-239	72 " "	67 " "
Zinc-65	38 " "	44 " "
Phosphorus-32	17 " "	29 " "

The amount of radioactivity of the river water is dependent upon the level of operation of the reactors and the river flow at Hanford (Clukey, 1957). By taking the average discharge of radionuclides during the past three years, the amount of radioactivity in the Columbia River estuary and adjacent Pacific Ocean in equilibrium with the Columbia River discharge can be estimated as follows (Nelson, 1962):

Chromium-51	33,000 curies
Zinc-65	14,000 "
Phosphorus-32	600 "
Neptunium-239	280 "

Except for phosphorus-32, a beta-emitter, these nuclides emit gamma rays during their decay processes. Appendix II lists the radionuclides, their daughters and modes of decay.

The effect of the radioactivity in the Columbia River on the surrounding environment has been intensively studied (Rostenbach, 1959; Foster and Junkins, 1960; Junkins *et al.*, 1960; Nelson, 1961, 1962). These reports summarize much of the information on the radioactivity of the Columbia River water with particular emphasis on the biological implications in the vicinity of the Hanford plant itself. The radioactivity in the river has been used as a tracer to study mixing processes (Honstead, 1957) and the processes by which radionuclides are removed from the water (Nielsen and Perkins, 1962). Nielsen and Perkins showed that copper-64, zinc-65 and phosphorus-32 are mostly associated with the silt and clay particles in the river. Their data indicate that most of the radioactivity in the sediments at McNary Reservoir is due to the presence of chromium-51 and zinc-65.

Some work has been done on the distribution of radioactivity in marine organisms from the Pacific Ocean near the Columbia River mouth. These studies have shown that chromium-51, zinc-65 and rarely cesium-137 occur in the plankton (Seymour, 1961; Osterberg, 1962a, 1962b) and in various benthic organisms (Watson *et al.*, 1961; Seymour, 1961).

Seymour (1961) also analyzed some sediments from the Columbia River estuary and Willapa Bay. No other analyses of marine sediments near the Columbia River are available.

A study of the distribution of radioactive sediments in the Irish Sea (Dunster, 1958) showed that fission products (predominantly ruthenium-106, strontium-90 and cerium-144) were more abundant in the silt than in the sand fraction of the sediments.

The literature on the sedimentation of radioactive wastes in the sea has been summarized by Waldichuk (1961), and Robinson (1962) has summarized the literature concerning ion-exchange minerals and the disposal of radioactive wastes in the sea. The available information on the problems of radioactive waste disposal in the sea is summarized and specific recommendations for monitoring such disposal has been presented by the International Atomic Energy Agency (1961). The processes affecting the distribution of various radionuclides in the sea have been discussed by Revelle (1957). Certain aspects of the problem of radioactive waste disposal in the sea have been considered by Carritt (1959), Pritchard (1959), and Isaacs (1962).

Surface sediments collected during 1961 from the Oregon-Washington coast and the Pacific Ocean adjacent to the Columbia River (Fig. 1) have been analyzed to determine the amount and identity of any gamma-emitting radionuclides present. The analyses constitute a preliminary survey to determine the distribution of radioactive sediments and the distribution of radionuclides within the sediments. With these data, future sampling operations can be planned to provide a better picture of the distribution of the radioactive sediments and to permit a study of the processes by which radioactive materials are incorporated in the sediments.

Experimental Techniques

The gamma-ray activity of the unwashed total sediment was determined and no attempt was made to separate or concentrate the radioactive phases. The gamma-ray spectrum from 0' to 2 mev of each sediment sample was determined by a multichannel analyzer, employing a 3" x 3" NaI (Tl) crystal as detector (Crouthamel, 1961). The chromium-51 and zinc-65 activities reported have been corrected for radioactive decay between the time the sample was collected and the time it was analyzed.

The 95 percent confidence level has been determined for the net count on all samples. All samples in which the counting rate was less than the uncertainty at the 95 percent confidence level were considered to have no detectable activity. With this criterion the minimum detectable activity for chromium-51 is approximately 20 picocuries² per gram of dry sediment and for zinc-65 approximately 1 picocurie per gram of dry sediment.

Grays Harbor, Washington

Twenty sediment samples collected in July 1961 from Grays Harbor, Washington were analyzed and the results (Table 2) are plotted in Figure 4. The sediments are predominantly sand, mixed with minor amounts of silt and clay. Potassium-40, chromium-51 and zinc-65 were detected in the sediments. The activity due to potassium-40, detected in all the samples, ranged from 11 to 82 picocuries per gram of dry sediment. Zinc-65 was present in 15 of the samples with activities less than 15 picocuries per gram of dry sediment. Chromium-51 was detected in only one of the sediments where its activity was 94 picocuries per gram of dry sediment. Failure to analyze the samples promptly after collection may account in part for the apparent absence of chromium-51 from many of the sediments.

Willapa Bay Sediments

A preliminary survey of the Willapa Bay sediments was conducted May 12, 1962¹, in which seven samples were collected for gamma spectrum analyses (Table 3, Figure 5). These are the only sediments which contain the nuclides most typical of atmospheric fallout: zirconium-95, ruthenium-103, and -106, and cerium-141 and -144. In three of the samples the amount of activity from fallout is greater than the natural radioactivity due to the presence of potassium-40.

Most of the other samples were collected before the resumption of atmospheric testing (September 1, 1961) or before the fallout had been deposited in the sediments. It should be noted that the presence of this fallout greatly complicates the analyses of the sediments and results in values of radioactivity considerably higher than that which can reasonably be ascribed to the Columbia River.

Despite the large amount of sand in these sediments, the potassium-40 content was nearly the same as that of the other sediments of the area, ranging between 16 and 21 picocuries per gram of dry sediment. Zinc-65 was detected in all seven samples with activities from 0.7 picocuries up to 6.3 picocuries per gram of dry sediment. No chromium-51 was detected in these samples.

² A picocurie is 10^{-12} curies or 2.22 disintegrations per minute.

TABLE 2

RADIOACTIVITY OF GRAYS HARBOR SEDIMENTS
 (Activities in picocuries per gram of dry sediment on date of collection
 9-29 July 1961)

Sample No.	Latitude	Longitude	Potassium-40	Zinc-65	Chromium-51	Sediment Type
GH-4	46°58.7' N	124°06.6' W	22 ± 5.3	3.5 ± 1.2	-	Sand
GH-5	46°59.9' N	124°07.5' W	11 ± 1.8	1.3 ± 0.38	-	Sand
GH-7	47°00.8' N	124°08.6' W	25 ± 4.9	-	-	Silty Sand
GH-8	47°01.0' N	124°07.6' W	21 ± 2.8	1.4 ± 0.50	-	Silty Sand
GH-9	47°01.0' N	124°07.6' W	22 ± 3.8	1.5 ± 0.75	-	Sand
GH-10	47°00.4' N	124°06.6' W	21 ± 5.0	2.9 ± 1.1	-	Sand
GH-11	47°00.8' N	124°04.7' W	23 ± 3.5	3.6 ± 0.73	-	Silty Sand
GH-12	47°01.3' N	124°05.6' W	24 ± 6.6	8.6 ± 1.6	-	Sandy Silt
GH-15	47°00.6' N	124°02.6' W	17 ± 4.2	-	-	Sand
GH-16	47°01.1' N	124°03.3' W	31 ± 5.2	-	-	Sand
GH-18	47°00.9' N	124°01.6' W	21 ± 3.6	3.0 ± 0.74	-	Sand
GH-19	47°00.1' N	124°01.0' W	23 ± 3.2	-	-	Silty Sand
GH-22	46°58.2' N	124°03.6' W	34 ± 17	-	-	Sand
GH-29	46°51.3' N	124°04.5' W	44 ± 8.4	12 ± 1.8	-	Sandy Silt
GH-31	46°31.1' N	124°03.4' W	23 ± 4.2	7.3 ± 0.97	-	Sandy Silt
GH-51	46°57.0' N	123°53.3' W	31 ± 6.0	2.3 ± 1.2	94 ± 39	Silt
GH-53	46°57.4' N	123°50.6' W	36 ± 8.5	3.2 ± 1.6	-	Silt
GH-55	46°58.0' N	123°48.9' W	19 ± 3.4	2.1 ± 0.64	-	Silty Sand
GH-56	46°58.2' N	123°55.0' W	26 ± 4.1	4.8 ± 0.85	-	Sandy Silt
GH-57	46°57.6' N	123°56.0' W	82 ± 18.0	5.7 ± 3.4	-	Sandy Silt

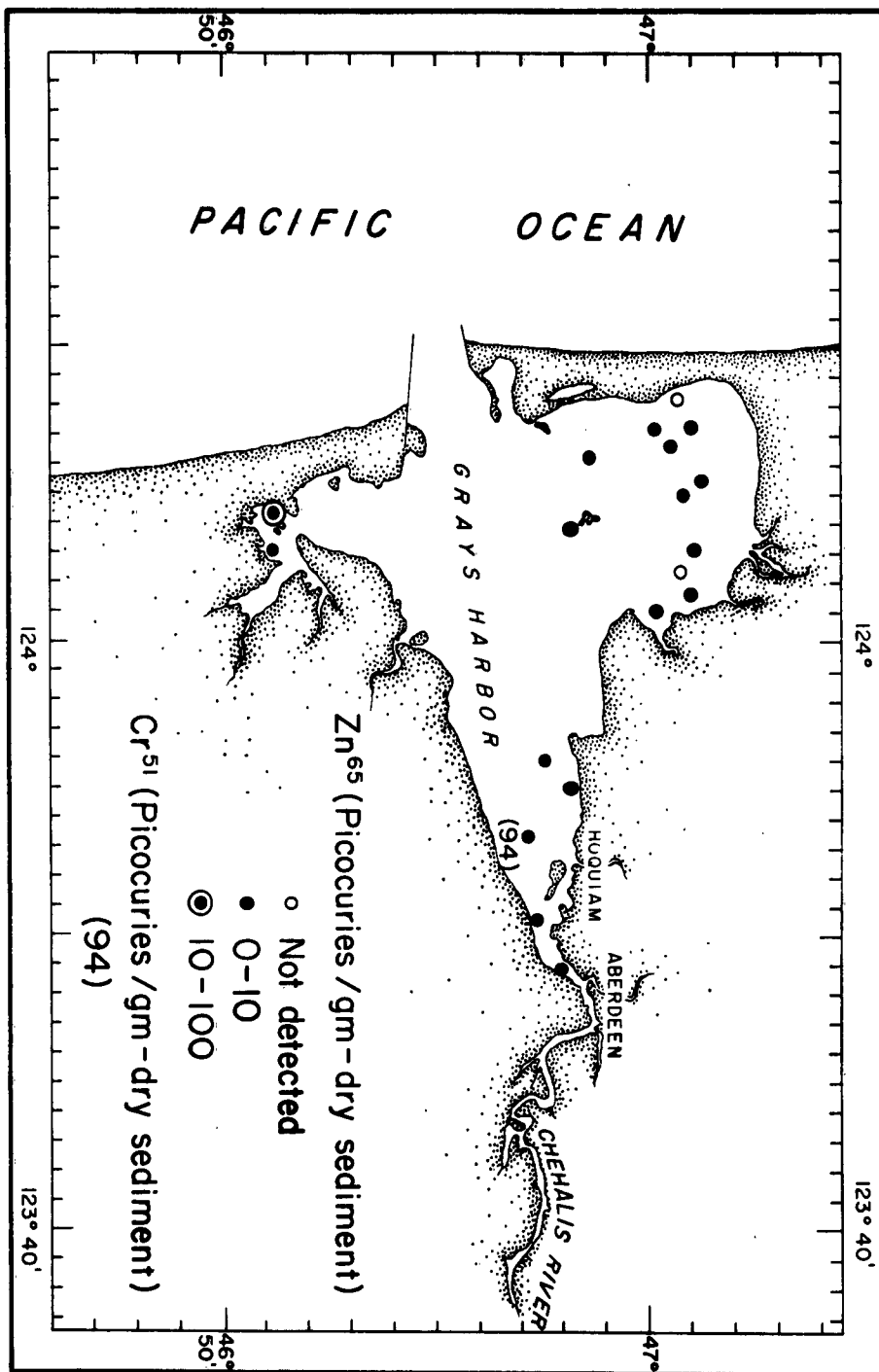


Fig. 4. Radioactivity of sediments in Grays Harbor, Washington, July 1961.

TABLE 3

RADIOACTIVITY OF WILLAPA BAY SEDIMENTS, 12 MAY, 1962
(Activities in picocuries per gram of dry sediment on 12 May, 1962)

Sample No.	Potassium-40	Zinc-65	Zirconium-95 Niobium-95	Ruthenium-103-106*	Cerium-141-144*	Sediment Type
WB-3	21 ± 2.1	6.3 ± .43	13 ± .27	2.1 ± .17	10 ± .41	Silty Sand
WB-4	16 ± 1.3	.74 ± .22	4.4 ± .12	.63 ± .092	4.0 ± .23	Sand
WB-5	19 ± 1.5	1.1 ± .28	2.7 ± .16	.71 ± .12	3.2 ± .32	Silty Sand
WB-7	18 ± 1.4	5.1 ± .32	18 ± .22	2.0 ± .14	1.0 ± .035	Silty Sand
WB-8	21 ± 1.6	2.0 ± .33	3.0 ± .17	.68 ± .14	4.0 ± .36	Clayey Silt
WB-11	16 ± 1.2	.70 ± .20	1.9 ± .097	.33 ± .08	2.4 ± .20	Sand
WB-14	16 ± .98	2.8 ± .21	16 ± .17	2.3 ± .10	11 ± .25	Sand

* Values not corrected for decay between date of collection, 12 May, 1962 and date of analysis, 6 June, 1962.

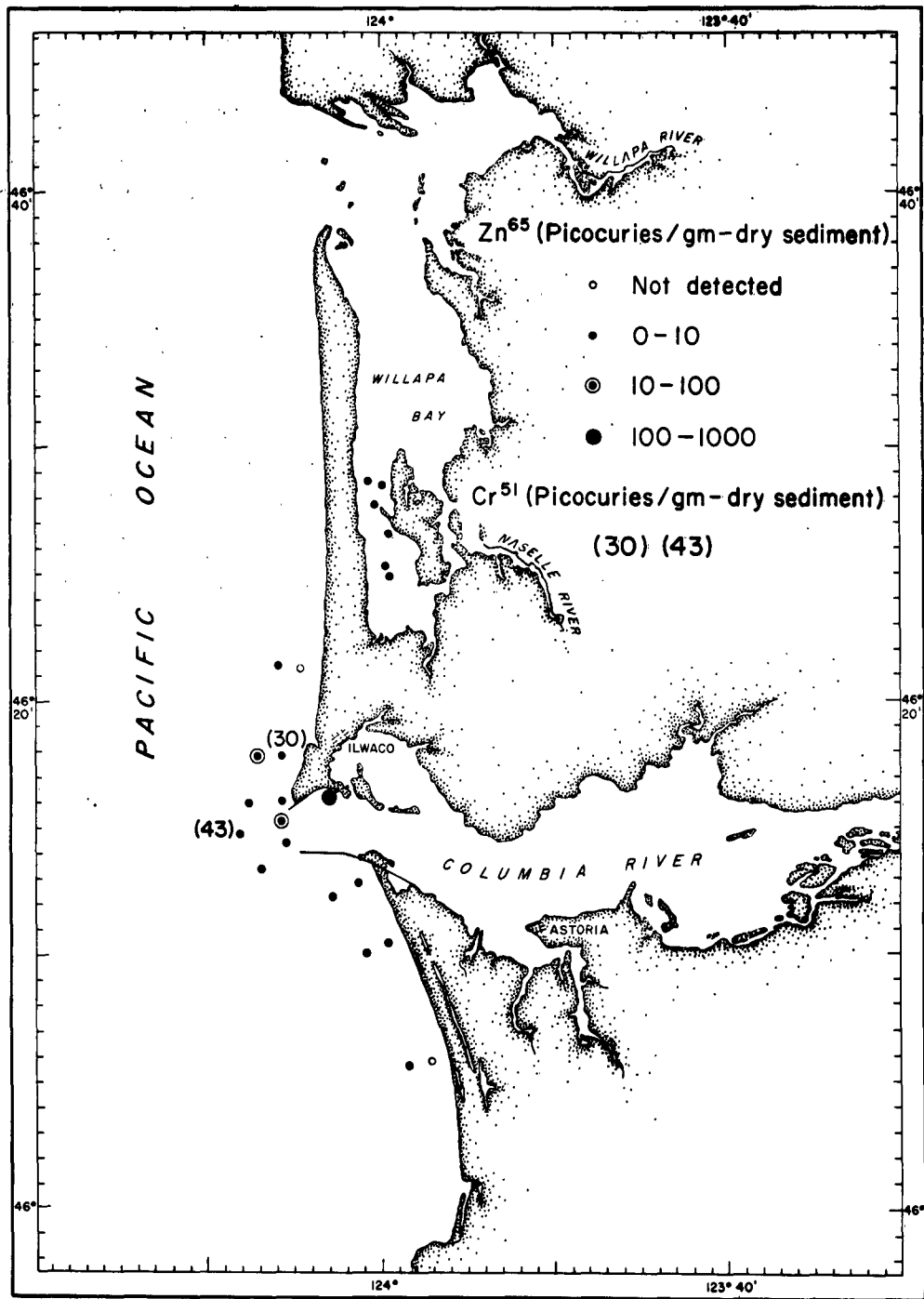


Fig. 5. Radioactivity of sediments near the Columbia River mouth, July-September, 1961.

Coastal and Beach Sediments

Ten sediment samples (Table 4, samples 79, 80, 87 to 89; Fig. 5), collected from the Oregon-Washington coast at depths of 30 fathoms or less were analyzed. As in the case of Grays Harbor, only potassium-40, chromium-51 and zinc-65 were detected in the sandy sediments. Potassium-40 was present in all the samples with activities between 16 and 23 picocuries per gram of dry sediment. Zinc-65 was detected in all but two of the samples with activities less than 15 picocuries per gram of dry sediment. In general the concentration of zinc-65 decreases toward the shore and no zinc-65 has been detected in beach sands taken from nearby stations. Chromium-51 was detected in one sample immediately north of the Columbia River mouth with an activity of 30 picocuries per gram.

In general these sediments were well sorted sands containing from one to two percent silt- and clay-sized material.

Columbia River Mouth and Estuary

The sands recovered from just outside the Columbia River mouth (Table 4, samples 81 to 86, Fig. 5) contained zinc-65 with activities between 2 and 11 picocuries per gram of dry sediment. Chromium-51 was detected in only one sample (No. 83-A) where its activity was 43 picocuries per gram of dry sediment. Potassium-40 occurred in all samples with activities between 17 and 30 picocuries per gram of dry sediment.

Just inside the river mouth, sands and silts (Sample 93) were recovered in which the zinc-65 activity was 450 picocuries per gram and in which both cobalt-57 and cobalt-60 were detected. This is the only sediment in which the cobalt isotopes were detected and it also exhibits the greatest concentration of the zinc-65. A silty sand collected from approximately 4 miles off the mouth of the Columbia River (Table 5, Sample BB291-23) had a total zinc-65 activity estimated at 200 picocuries per gram of dry sediment and in the silt- and clay-size fraction the zinc-65 activity was approximately 450 picocuries per gram.

The only sediments in which the zinc-65 activities were found to be greater than 100 picocuries were within five miles of the Columbia River mouth.

Offshore Sediments

During Brown Bear Cruise 291 (July and August 1961), 68 sediment samples were collected; of these 39 were analyzed (Table 5, Figs. 6 and 7). Radioactivity was detected in 12 samples, of which 10 contained detectable amounts of zinc-65 (2.8 to 460 picocuries per gram) and five contained detectable amounts of chromium-51 (20 to 730 picocuries per gram). Potassium-40 was detected in only two samples where its activity was between 30 and 50 picocuries per gram of dry sediment.

TABLE 4

RADIOACTIVITY OF SEDIMENTS NEAR THE MOUTH OF
THE COLUMBIA RIVER*

(Activities in picocuries per gram of dry sediment, on date of collection
7-15 September 1961)

Sample No.	Latitude	Longitude	Potassium-40	Zinc-65	Chromium-51
OW-79-A	46°21.2' N	124°03.5' W	22 ± 5.5	2.2 ± 1.0	-
OW-79-B	46°21.2' N	124°03.5' W	20 ± 3.6	-	-
OW-80-A	46°17.7' N	124°04.5' W	19 ± 6.2	13 ± 1.5	-
OW-80-B	46°17.7' N	124°04.5' W	23 ± 5.5	2.2 ± 1.0	30 ± 18
OW-81	46°15.9' N	124°07.8' W	18 ± 3.9	5.8 ± 0.82	-
OW-82	46°16.1' N	124°05.9' W	19 ± 6.0	1.8 ± 1.1	-
OW-83	46°14.8' N	124°08.2' W	18 ± 4.1	5.0 ± 0.84	43 ± 13
OW-84	46°15.2' N	124°06.1' W	22 ± 4.5	11.0 ± 1.0	-
OW-85	46°13.5' N	124°07.0' W	17 ± 3.9	6.4 ± 0.88	-
OW-86	46°14.3' N	124°05.7' W	20 ± 5.1	3.0 ± 1.0	-
OW-87-A	46°13.2' N	124°00.5' W	16 ± 1.8	1.5 ± 0.46	-
OW-87-B	46°13.2' N	124°00.5' W	21 ± 5.5	16 ± 0.99	-
OW-88-A	46°10.2' N	123°58.7' W	17 ± 2.6	2.2 ± 0.69	-
OW-88-B	46°10.2' N	123°58.7' W	15 ± 2.5	2.1 ± 0.65	-
OW-89-A	46°05.8' N	123°56.1' W	18 ± 2.2	1.3 ± 0.60	-
OW-89-B	46°05.8' N	123°56.1' W	16 ± 4.5	-	-
OW-92	46°16.3' N	124°03.1' W	22 ± 2.3	14.0 ± 0.81	-
OW-93**	46°16.3' N	124°03.1' W	29 ± 3.6 30 ± 3.7	460.0 ± 3.0 450.0 ± 3.1	- -

* All samples are sand, except OW-93 which is a silty sand.

** This sample also contained cobalt-57 (12 ± 0.35 pc/gram dry sediment) and cobalt-60 (11 ± 0.4 pc/gram dry sediment).

TABLE 5

RADIOACTIVITY OF MARINE SEDIMENTS FROM BROWN BEAR CRUISE 291*
(Activities in picocuries per gram of dry sediment, on 29 July-12 August, 1961)

Sample No.	Latitude (North)	Longitude (West)	Potassium-40	Zinc-65	Chromium-51	Sediment Type
BB 291-19	46°48.2'	124°35.8'	---	7.4 ± 3.3	---	Sandy Silt
BB 291-23	46°14.8'	124°10.0'	45**	195**	270**	Silty Sand
BB 291-23a	46°14.8'	124°10.0'	30 ± 11	80.0 ± 3.4	83 ± 13	Sand Fraction
BB 291-23b	46°14.8'	124°10.0'	54 ± 27	460 ± 12	730 ± 45	Silt Fraction
BB 291-27	45°27.9'	124°28.8'	---	5.5 ± 4.3	---	Clayey Silt
BB 291-28	45°14.4'	124°00.0'	29 ± 12	2.8 ± 1.8	---	Sand
BB 291-35	43°18.4'	124°27.5'	---	---	20 ± 17	Sand
BB 291-38	43°34.6'	125°31.6'	---	5.0 ± 4.1	74 ± 28	Silty Clay
BB 291-40	44°13.0'	126°27.5'	---	9.0 ± 8.5	---	Silty Clay
BB 291-41	44°42.6'	127°36.0'	---	13.0 ± 9.1	160 ± 56	Clayey Silt
BB 291-42	45°00.5'	126°42.5'	---	7.2 ± 5.5	---	Silty Silt
BB 291-44	45°30.2'	125°30.5'	---	---	47 ± 38	Silty Clay
BB 291-49	46°03.0'	127°12.0'	---	8.9 ± 5.4	---	Silty Clay
BB 291-59	46°14.5'	124°41.9'	---	27.0 ± 4.3	---	Clayey Silt

* Neither chromium-51 nor zinc-65 was detected in the following samples from Brown Bear Cruise 291:

1, 2, 3, 5, 6, 7, 9, 12, 13, 14, 15, 17, 18, 30, 31,

34, 36, 37, 39, 43, 45, 46, 47, 52, 54, 59 and 66.

** Estimated values based on the activity of each size fraction and its abundance in the sediment.

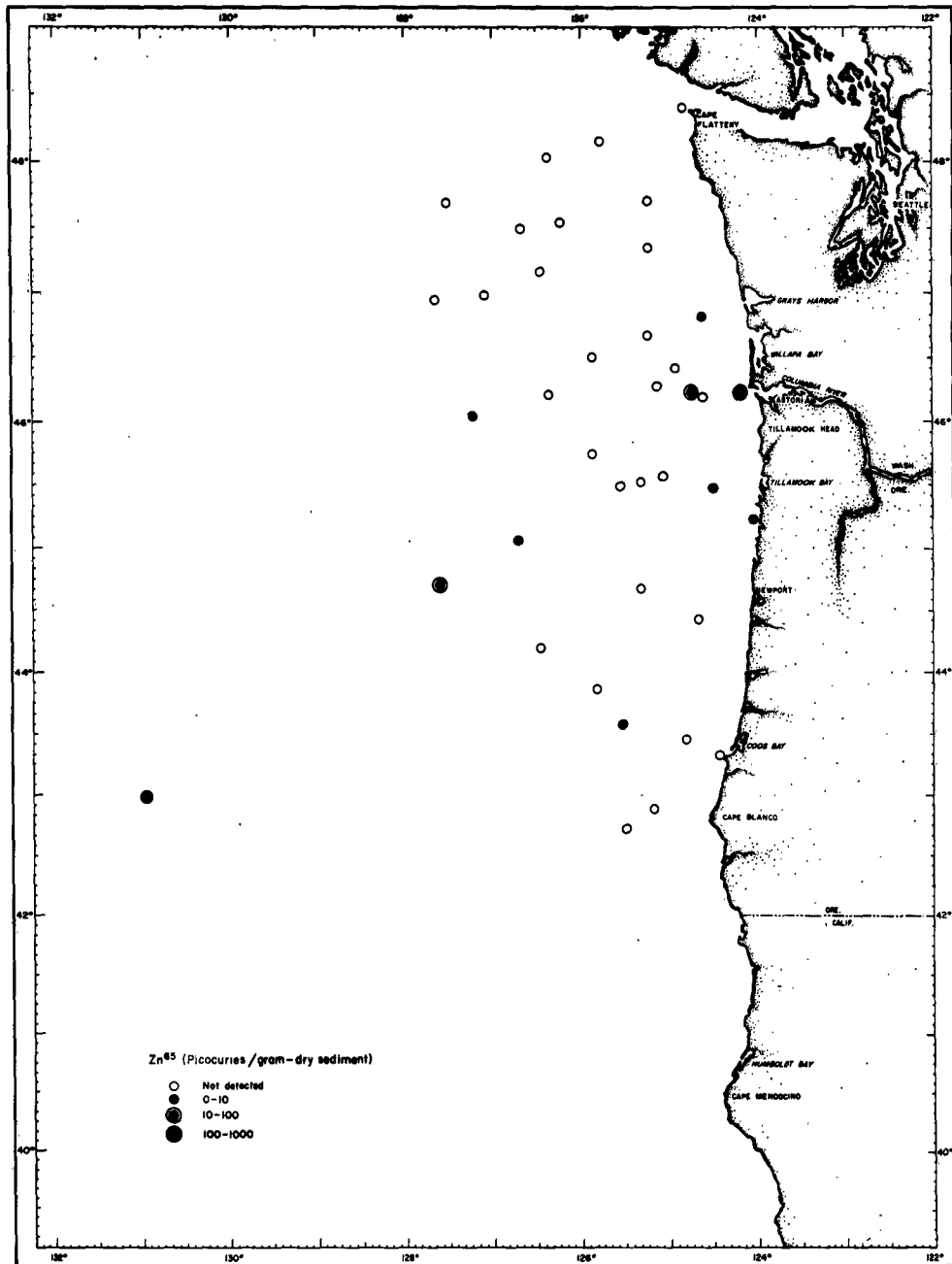


Fig. 6. Distribution of zinc-65 in marine sediments of the Northeast Pacific Ocean, July-August, 1961.

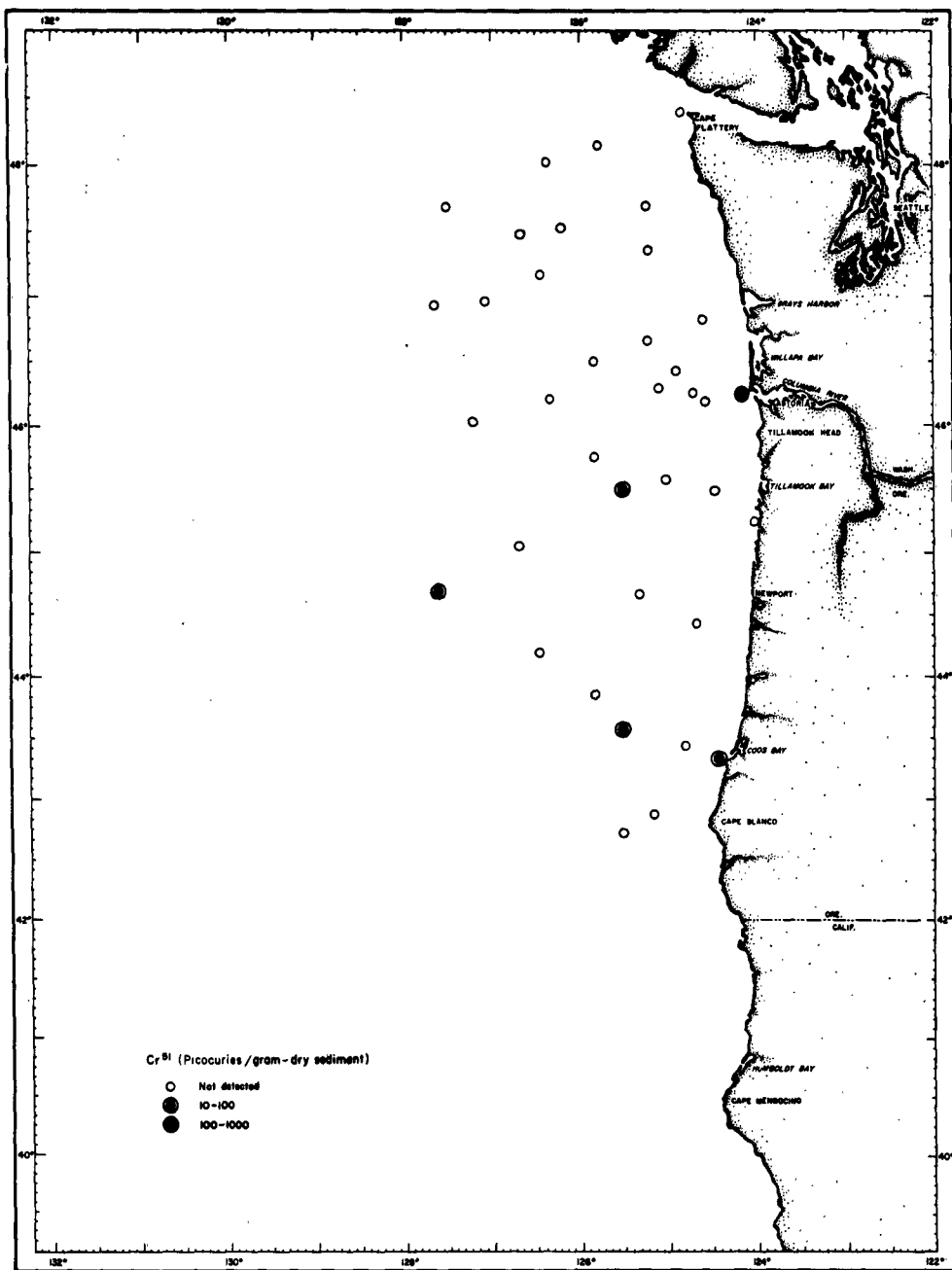


Fig. 7. Distribution of chromium-51 in marine sediments in the Northeast Pacific Ocean, July-August, 1961.

Radioactivity of Different Sized Particles

Experimental work to determine the distribution of the radioactivity within the sediments has been severely limited by the small size of the samples collected during 1961 and the low levels of activity found in them. However, in those sediments collected near the Columbia River mouth, sufficient activity was found in a few, relatively large samples to permit crude experiments involving sieving the sediments and determining the radioactivity of each size fraction. This was done on sample BB291-23 and several samples taken during Brown Bear Cruise 293. The results shown in Table 6 indicate that silt- and clay-size fractions of these sediments contain up to 10 times the amount of zinc-65 present in the sand-size fraction. This is consistent with the results obtained from the other analyses in which the radioactivity of the finer grained samples tended to be higher than the radioactivity of the coarse-grained samples.

Other efforts to separate physically the radioactive phases from the bulk of the sediment have not been successful. It is hoped that with centrifugation it will be possible to separate the various sized fractions with greater facility and to obtain a better picture of the amount of radioactivity in each size fraction.

Other simple experiments have shown that it is possible to separate part of the zinc-65 from the sediments by strong complexing agents (such as EDTA), by strong acids, and by exchanging the zinc with magnesium or aluminum solutions. Oxidation of the sediment in boiling sulfuric acid and potassium dichromate completely removed the zinc-65 from the sediment.

Clay Mineralogy

Because of the possible effect of the clay-mineral composition on the distribution of the radionuclides, a preliminary survey was made of the clay-sized material (finer than four microns) in 30 samples to determine the minerals present in sediments and to obtain some idea of their relative abundance. The fraction of the sediment finer than four microns was sedimented and dried on glass slides which were exposed to ethylene glycol vapor. The slides were then analyzed by standard X-ray diffraction procedures (Brown, 1961). The preliminary data indicate that chlorite, illite, montmorillonite, and quartz are the most abundant clay-sized minerals in the samples analyzed. Kaolinite, if present, is not a major constituent.

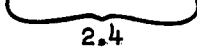
Discussion

Potassium-40, detected in the majority of the sediments analyzed, is the naturally occurring radioisotope of potassium. It occurs in the mineral structure, and as a result of incomplete removal of salt from the samples. No correlation was found between the potassium-40 activity in the samples and the zinc-65 or chromium-51 activity.

These data indicate that during the late summer of 1961 there was relatively little radioactivity in the sandy sediments of Grays Harbor and the Oregon-Washington beaches. The highest activities of chromium-51 and

TABLE 6

DISTRIBUTION OF ZINC-65 IN SEDIMENTS NEAR THE COLUMBIA RIVER
 (Activity expressed in counts per minute per gram of dry sediment)

	<u>Sand</u>	<u>Silt</u>	<u>Clay-size</u>
BB 293-2-1	0.17	2.4	2.4
BB 293-2-2	0.42	1.6	---
BB 293-3	2.30	2.2	5.0
BB 291-23	0.30		

zinc-65 were found in sediments immediately adjacent to or within the Columbia River estuary. From the sediment inside the river mouth, cobalt-57 and cobalt-60 were detected. Both cobalt isotopes occur in the Hanford effluent (Nelson, 1961, 1962).

Figures 3 and 4 show that the sediments containing chromium-51 and zinc-65 are generally restricted to the area south of the Columbia River mouth. Since the samples analyzed were taken largely from the surface layers of cores, it is possible that part of the discontinuous distribution is the result of loss, during sampling and storage, of the upper part of the core which contained the radioactive materials. This possibility is currently being checked by analyzing the grab samples taken from stations in which no radioactivity was detected in the core samples.

It is apparent that the offshore sampling program in 1961 did not cover a large enough area to delineate the distribution of radioactive sediments because the samples from the edges of the area surveyed, 180 nautical miles from the mouth of the Columbia River, contained detectable amounts of chromium-51 and zinc-65.

The data suggest that much of the radioactivity in the sediment along the Oregon-Washington coast comes from the Columbia River. The zinc-65 activity of the sediments is highest near the mouth of the river and chromium-51 tends to be more abundant in the vicinity of the river mouth. The distribution of both chromium-51 and zinc-65 in the sediments in the later part of the summer of 1961 is strikingly similar to the distribution of the plume of Columbia River water during the same period.

It should be pointed out that, except for potassium-40, the activities reported here are minimum values. In every case a large amount of sediment is analyzed, of which the surface layer probably contains the bulk of radioactivity. With better sampling and analytical techniques, it may be possible to analyze only the surface layers of sediment which have been deposited within the last few months or years. This may well show that the most recently deposited sediment surface has much higher activities than are reported here.

The chromium-51 in the sediments must have come from the Columbia River. Chromium-51 is not a fission product and even neutron activation of the chromium in sea water by an underwater detonation would not result in an appreciable amount of chromium-51 in the water (Klement, 1951, p. 92-94). Furthermore, the majority of the sediments were collected before the resumption of atmospheric testing in 1961 (Machta *et al.*, 1962), and the previous series of atmospheric tests were concluded in 1958 (Hearings of the Joint Committee on Atomic Energy, 1959). Any chromium-51 produced in tests before 1958 would have decayed because of its short half life (27.8 days).

Some of the zinc-65 in the sediments might have originated from earlier weapons tests. (Kawabata, 1955; Yamada *et al.*, 1955; Gong *et al.*, 1957; Lowman, 1960). However, the complete absence of zinc-65 in the sediments north of the Columbia River mouth, its abundance near the river mouth, and the similarity between the distribution of zinc-65 in the sediments and the plume of river water strongly suggest that much of the zinc-65 in the sediments analyzed is derived from the Columbia River.

The samples taken during 1962 will reveal the magnitude of the zinc-65 contribution from atmospheric weapons testing as the sediments will have been exposed to the fallout from the 1961-2 series of tests.

Assuming that the Columbia River is its major source, the zinc-65 activity of the offshore sediments is plotted against the distance of the sediments from the mouth of the Columbia River in Figure 8. The observed distribution shows that within 100 miles of the coast the zinc-65 activity is reduced by a factor of 10 or more.

Many investigators (Boroughs et al., 1957; Krumholz et al., 1957; Chipman et al., 1958; Lowman, 1960) have demonstrated the importance of biological processes on the distribution of zinc-65 in the oceans and from analysis of their data it appears likely that this theory will hold true for sedimentation of the zinc-65 in the Columbia River area. Therefore, the amount of organic carbon was determined in several of the offshore sediments in which zinc-65 was detected, but there was no obvious correlation between the amount of organic carbon and the zinc-65 activity of the sediment. Again the data are too few to permit any conclusions and the evaluation of the data is continuing.

Krauskopf (1957) demonstrated that clay minerals can very effectively remove zinc from solution in sea water. Elgablay and Jenny (1943) and Elgablay (1950) showed that ionic, divalent zinc as well as its monovalent complexes may be exchanged by montmorillonites, part of the zinc being fixed in the octahedral layers of the clay mineral structure.

Additional Work Required

1. Future sampling programs should be extended to determine the outer limits of the area receiving chromium-51 and zinc-65 from the Columbia River. This will have profound implications not only for determining the areal limits of dispersal of the Columbia River sediment but will greatly assist in identifying the sedimentary processes involved.
2. More sensitive methods of sample analysis are needed for both chromium-51 and zinc-65. The activities are near the limits of our present methods of detection and more sensitive methods would greatly facilitate analysis and interpretation of the data.
3. The presence of radionuclides derived from atmospheric fallout complicates the problems of gamma-ray spectrum analysis and it may be necessary to investigate a limited number of samples with radiochemical techniques in order to specifically identify certain of the nuclides. Certainly the presence of radionuclides with short half lives requires more rapid sediment preparation and analysis than were available during 1961. Multichannel analyzers aboard ship may be necessary to achieve this end.
4. Much work is required on the problem of sediment sampling. In areas of low sedimentation rates, the major part of the radioactive upper layers of sediment may be lost during coring operations or in subsequent handling and storage of the cores. An in situ detector which could be lowered to the bottom may provide at least a partial solution.

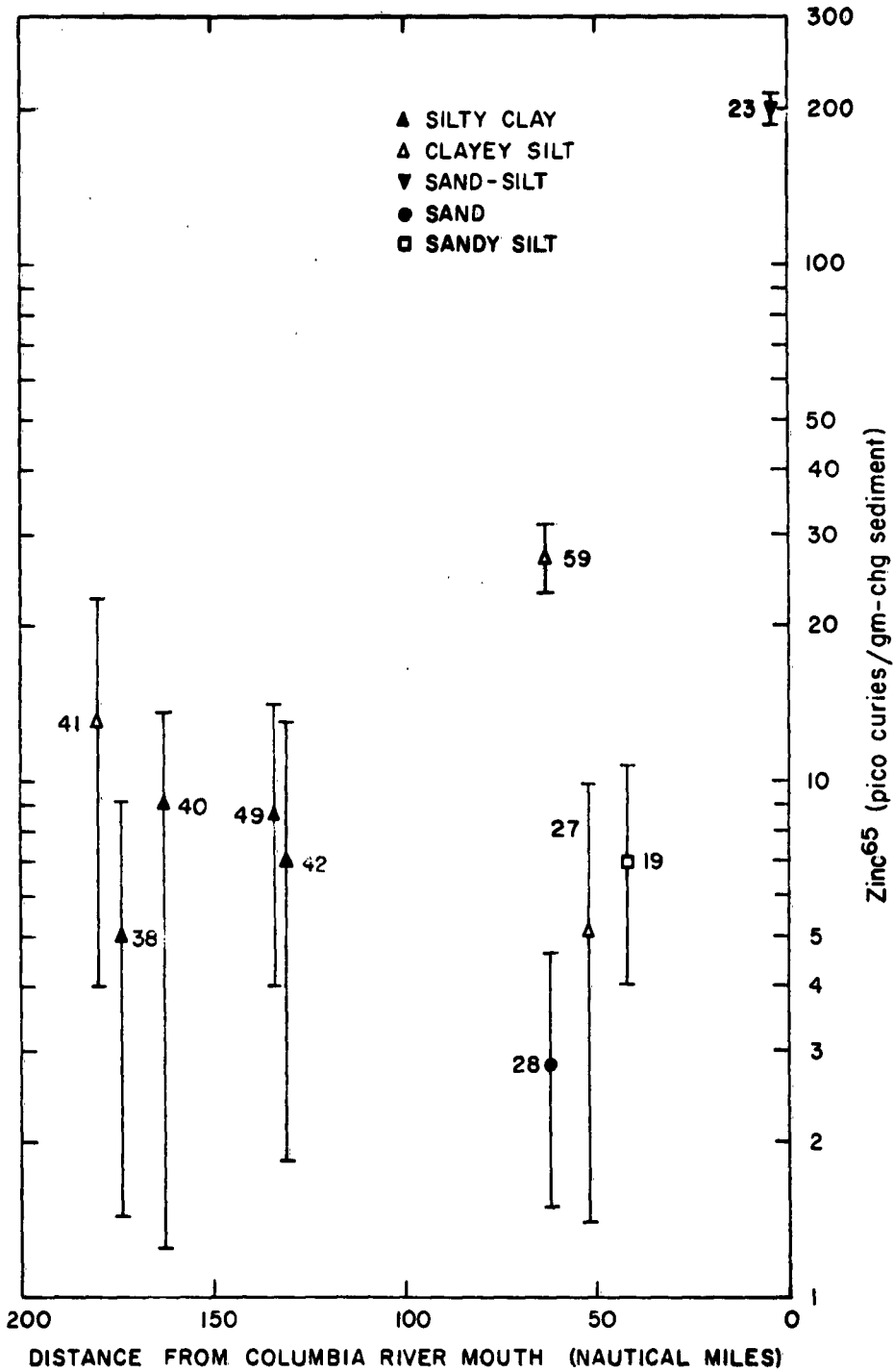


Fig. 8. Zinc-65 content of marine sediments as a function of distance from the Columbia River mouth, July-August, 1961.

5. Additional work is needed to determine whether the activity is associated with the organic or the mineral phases of the sediment and to investigate the processes by which the radionuclides are incorporated in the sediments.

SUMMARY AND CONCLUSIONS

Bottom samples taken off the coast of Washington and Oregon during Brown Bear Cruises 291 and 311 are expected to provide a basis for interpreting the seaward extent of Columbia River sediments and sedimentation. A total of 133 samples have been collected from the continental terrace, the Cascadia Abyssal Plain, and the seamount regions forming the western perimeter of the plain, about 300 miles offshore. In addition, sediments have been collected from three coastal areas: 1) 76 samples from Gray's Harbor, Washington; 2) 55 samples from Willapa Bay, Washington; and 3) 422 samples from 60 miles of beach along the coast of southwestern Washington and northwestern Oregon.

These samples are being analyzed for particle size and heavy mineral content. Where the following analyses are applicable, determinations are being made of the clay mineral content, carbonate content, organic carbon content, and Foraminifera content.

In general, the sediment in the Columbia River Estuary consists of clean, medium-grained sand with a few granules. Sands also occur seaward of the mouth of the river, and comprise most of the sedimentary material bordering Astoria Canyon and covering the inner part of the continental shelf, except locally where the sediment coarsens to gravel or where rock is exposed. The inner-shelf sand is mostly finer grained and darker than the Columbia River sand. The remainder of the shelf is covered with grey to green clayey silt.

The most common type of bottom sediment found on the abyssal plains is silty clay; however, there are a few areas where the sediment coarsens to clayey silt. This coarsening of the sediment is caused mainly by numerous tests of organisms, particularly radiolarian and globigerinid tests. One sample of clayey silt from a deep-sea channel, however, contained fine sand-sized quartz grains.

Detectable amounts of radioactivity are present in the Columbia River at Vancouver, Washington where it is estimated that approximately 1000 curies per day are transported in the river water. The most abundant radionuclides in the river water are chromium-51, neptunium-239, phosphorus-32 and zinc-65; all but phosphorus-32 emit gamma rays.

Analyses of the gamma-ray spectra of sediments collected near the Columbia River mouth in 1961 indicate that radionuclides, other than the naturally occurring potassium-40, are present in the sediments. The sandy sediments from Grays Harbor and the beaches adjacent to the Columbia River had very low activities although chromium-51 (40 pc/gm), zinc-65 (2-15 pc/gm), cobalt-57 (12 pc/gm), and cobalt-60 (11 pc/gm) were detected. The fine-grained sediments in and near the river mouth had the highest concentrations of chromium-51 (270 pc/gm) and zinc-65 (450 pc/gm) which decrease by a factor of 10 within 100 miles of the river mouth. Detectable amounts of chromium-51 (20 pc/gm) and zinc-65 (5 pc/gm) were present in sediments 180 miles south and west from the river mouth. In sediments near the river the activity of the silt and clay fractions was five to ten times greater than the activity of the sand fraction.

Because the highest activities in the sediments due to chromium-51 and zinc-65 occur near the river mouth and because the distribution of the radioactive sediments resembles the distribution of the low-salinity river plume, it is postulated that during the summer of 1961 the Columbia River was the major source of radioactivity in the sediments off the Oregon-Washington coast. All of the radionuclides detected are known to be present in Columbia River water. Some of the zinc-65 may have come from previous series of atmospheric weapons testings.

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APPENDIX II

HALF-LIVES AND TYPES OF RADIATION FROM NUCLIDES PRESENT
IN COLUMBIA RIVER WATER AND FALLOUT
(Sullivan, 1957)

Radio-nuclide	Half-life*	Daughter**	(Energies in mev.)			Electron Capture
			β^+	β^-	γ	
Na ²⁴	15h	Mg ²⁴		1.39	1.37, 2.75	
P ³²	14.3d	S ³²		1.71		
Cr ⁵¹	27.8d	V ⁵¹			0.32	X
Mn ⁵⁶	2.58h	Fe ⁵⁶		2.86, 1.05, 0.75	0.84, 1.81, 2.13	
Fe ⁵⁵	2.94y	Mn ⁵⁵				X
Fe ⁵⁹	45.1d	Co ⁵⁹		0.46, 0.27, 1.56	1.10, 1.29, 0.19	
Co ⁵⁷	270d	Fe ⁵⁷			0.12, 0.13, 0.14	X
Co ⁵⁸	72d	Fe ⁵⁸	0.475		1.62, 0.81	X
Co ⁶⁰	5.27y	Ni ⁶⁰		0.31, 1.48	1.17	
Cu ⁶⁴	12.8h	Ni ⁶⁴ , Zn ⁶⁴	0.657	0.57	1.35	X
Zn ⁶⁵	245d	Cu ⁶⁵	0.325		1.11	X
As ⁷⁶	26.4h	Se ⁷⁶		2.97, 2.41, 1.76	0.56, 0.64, 1.2, 1.4	
Sr ⁹⁰	28y	<u>Y⁹⁰</u>		0.61		
Zr ⁹⁵	63.3d	<u>Nb⁹⁵</u>		0.36, 0.40, 0.88	0.75, 0.72	
Nb ⁹⁵	35d	Mo ⁹⁵		0.162	0.76	
Ru ¹⁰³	41d	Rh ¹⁰³		0.22, 0.14, 0.70	0.49, 0.61	
Ru ¹⁰⁶	1.0y	<u>Rh¹⁰⁶</u>		0.0039		
I ¹³¹	8.05d	Xe ¹³¹		0.61, 0.34, 0.25	0.36, 0.28, 0.64	
Cs ¹³⁷	30y	Ba ¹³⁷		0.52, 1.19		
Ce ¹⁴¹	32.5d	Pr ¹⁴¹		0.43, 0.58		
Ce ¹⁴⁴	290d	<u>Pr¹⁴⁴</u>		0.30, 0.17, 0.22	0.13, 0.08, 0.09	
Np ²³⁹	2.33d	<u>Pu²³⁹</u>		0.32, 0.44	0.10, 0.28	

* h-hours, d-days, y-years

** Radioactive daughters underlined

APPENDIX I

STATUS OF SAMPLING AND ANALYTICAL PROGRAMS
1 December 1962

SAMPLES	GRAYS HARBOR	WILLAPA BAY	OREGON-WASHINGTON BEACHES	OFFSHORE CRUISES
Grab Samples	73	48	422	35
Cores	3	7	-	98
Total	76	55	422	133

Analyses completed:

Size Analyses	76	13	112	178
Heavy Mineral Analyses	17	-	60	37
Clay Mineral Analyses	-	-	-	30
Calcium Carbonate	-	-	-	7
Organic Carbon	15	-	-	13
Gamma Spectrum Analyses	17	41	18	101
Foraminifera	60	-	-	110

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