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ASSOCIATION OF LONG-LIVED RADIOACTIVITY WITH SEDIMENT ALONG THE COLUMBIA RIVER SHORELINE, ISLANDS, BOTTOM AND SLOUGH AREAS

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DISCLAIMER

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ABSTRACT

Since deactivation of the last once-through cooling production reactor in January 1971, radioactivity associated with sediments or biota in the Columbia River attributable to past Hanford operations decreased to very low levels. Currently only a few long-lived radionuclides are measurable and generally due to the association of these radionuclides with sediments in the river. The data available from several different monitoring programs provide a preliminary description of the extent, concentration, and physical form of these radionuclides:

- A detailed aerial survey of the Columbia River shoreline, islands, and slough areas by EG&G, Inc. of Las Vegas between March 26 and April 28, 1974, which showed radiation, at several locations, from ^{60}Co (maximum, 0.022 mR/hr) and, at a few locations, from ^{137}Cs (maximum, 0.C03 mR/hr) in addition to natural background radiation (about 0.010) mR/hr).
- Analyses of sediment core samples collected from the bottom of the Columbia River behind Priest Rapids Dam (upstream of the Hanford Reservation) and McNary Dam (downstream of the Hanford Reservation) allow a comparison of the accumulation of long-lived radionuclides (^{65}Zn, ^{60}Co, ^{152}Eu) from Hanford operations behind McNary Dam with the accumulation of fallout radionuclides behind Priest Rapids Dam.
- Analyses of sediment samples collected from the shoreline, island, and slough areas, observed to have the highest levels of radioactivity during the aerial survey, provide measurement of the remaining concentration of long-lived radionuclides due to past Hanford operations.

- Biweekly data obtained with a "filter-resin" method of sampling Columbia River water during most of 1973, 1974, and 1975 allow the separation of observed concentrations of radionuclides into a particulate (collected on filters) or a soluble (adsorbed on resin) fraction.

INTRODUCTION

The Hanford Plant was originally designed, built, and operated to produce plutonium for nuclear weapons. At one time, nine production reactors, eight with once-through cooling, were in operation along the Columbia River. Between December 1964 and January 1971, all eight reactors with once-through cooling were deactivated. N Reactor, the remaining production reactor in operation, has a closed primary cooling loop. During operation of the production reactors with once-through cooling, substantial quantities of activation products were released to the river.

Once released to the river, the radionuclides became a part of the ecology of the river being measurable in water, algae, fish, ducks, sediment, etc. Since January 1971, when the last of these reactors was deactivated, the levels of short-lived radionuclides have decreased to either extremely low levels or the point of nonexistence. Figure 1 shows the sharp drop in gross beta activity measurable in river water to essentially background levels after January 1971. Figure 2 shows the gradual decline, approximately as the radioactive decay rate, in ^{65}Zn activity in muscle tissue of Willipa Bay oysters. Long-lived radionuclides remaining from past Hanford operations are currently measurable along the Columbia River shoreline, islands, bottom, and slough areas. These radionuclides are associated with river sediment and, as a result, the major reservoir of radioactivity is expected to be the sediment accumulated behind McNary Dam, the first dam downstream of the Hanford Reservation. The data available from three separate monitoring programs

- Aerial survey of Columbia River,
- Sediment sampling from Columbia River shoreline, islands, bottom and slough areas,

and

- "Filter-Resin" continuous sampling of river water

provide a preliminary description of the magnitude, distribution, and physical form of long-lived radioactivity along the Columbia River.

AERIAL SURVEY

Between March 26 and April 28, 1974, a detailed aerial survey of Columbia River shoreline, islands and slough areas was conducted by EG&G, Inc. of Las Vegas.(1) The survey covered an area from approximately four kilometers above Vernita Bridge to approximately 10 kilometers below the intersection of the Snake River with the Columbia River. An additional 20 kilometers downstream from McNary Dam was also surveyed. The survey was conducted at an altitude of 45 meters using a Navy helicopter. A minimum of three lines, spaced 60 meters apart, were flown along each bank, starting at the shoreline and moving inland. Similar patterns were flown over the islands. The counting system employed consisted of two pods, each containing twenty 12.5 cm diameter by 5 cm thick NaI (th) detectors, mounted externally on the helicopter. The signals from the detectors were fed into three units: a gross count scaler, a set of five adjustable window single channel analyzers, and a 300-channel multichannel analyzer.

Photographs 1 through 12 show the observed levels of ^{60}Co recorded during the aerial survey. Because of the presence of water which caused large fluctuations in the natural terrestrial background count rate, the gross count information was not very useful. For this reason, only the spectroscopic data were used.

The highest activity observed at areas within the site boundary which are restricted from public use occurred along the slough area between the 100-H and 100-D Areas and along the slough area just north of the old Hanford townsite. Both ^{60}Co and ^{137}Cs were observed in these areas. Radiation levels up to 0.014 mR/hr from ^{60}Co and 0.003 mR/hr from ^{137}Cs were seen in the slough area between 100-H and 100-D. Levels up to 0.022 mR/hr from ^{60}Co and 0.001 mR/hr from ^{137}Cs were seen in the other slough area.

The highest radiation levels observed offsite during the aerial survey occurred on the islands between the old Hanford townsite and the 300 Area.

A maximum reading of 0.014 mR/hr of ^{60}Co was obtained. No ^{137}Cs activity, above background levels, was observed. The average external exposure rate from natural sources in the Hanford Environs is approximately 0.010 mR/hr.

SEDIMENT SAMPLING

Sediment samples collected from the Columbia River are of two types:

1) Core samples collected from the Columbia River bottom.
2) Sediment samples collected from Columbia River shoreline, island, and slough areas observed to have the highest levels of radioactivity during the aerial survey.

Core Samples

Samples of sediment on the Columbia River bottom behind McNary Dam, the first dam downstream of the Hanford Reservation, have been collected during past years to determine the accumulation and variation with depth of radionuclides released from Hanford facilities. Additional samples were collected behind Priest Rapids Dam, the first dam upstream of the Hanford Reservation, during 1973 to allow a determination of background concentrations due to fallout. Table 1 presents a summary of the radionuclides and the range of concentrations observed at the two dams. The

TABLE 1. Range of Radionuclide Concentrations Observed at Priest Rapids and McNary Dams

Radionuclide	Concentration (pCi/g - dry weight)			
	Priest Rapids Dam Range		McNary Dam Range	
	Minimum	Maximum	Minimum	Maximum
Natural				
^{40}K	23	30	9	15
^{226}Ra	2	4	0.2	2.
^{228}Th	0.7	0.9	0.3	0.6
Artificial				
^{46}Sc		<0.005	<0.5	0.8
^{54}Mn		<0.2	<0.5	2.7
^{60}Co	<0.001	0.007	1.8	26.7
^{65}Zn		<0.2	<0.5	16.3
^{137}Cs	<1	7	1.0	7.5
^{152}Eu		<0.05	0.9	28.6

radionuclides present in the greatest amounts relative to background concentrations are ^{60}Co and ^{152}Eu. Figures 3 and 4 show the average variation with depth of these two radionuclides behind McNary Dam from measurements taken during 1973. The figures indicate that radioactivity from Hanford operations is at least 22 inches in depth. The average concentrations near the surface are decreasing due to dilution from additional sediment being transported downstream.

Sediment Samples

Subsequent to the Aerial survey, sediment samples were collected from the river locations which showed the highest levels of radioactivity. These locations are shown in photographs 1 through 12. Generally samples were collected from the top one inch of sediment although in a few locations profile samples to 8 inches in depth were collected. Table 2 presents a summary of the range of values and the average concentration for each radionuclide observed. Except for naturally occurring radionuclides, the radionuclides observed most frequently and in the highest concentrations were ^{60}Co, ^{137}Cs, and ^{152}Eu. Figure 5 and 6 show the average depth profile for ^{60}Co and ^{152}Eu to 8 inches. For both ^{60}Co and ^{152}Eu, a maximum average concentration at a depth of about 2 inches was observed.

TABLE 2. Radionuclides Observed on Columbia River Sediment Samples Collected During 1974.

Radionuclide	Concentration (pCi/g)		
	Minimum	Maximum	Average[a]
Natural			
^{40}K	9	16	12.6±1.2 (41)
^{224}Ra	1	10	2.1±1.2 (41)
^{226}Ra	0.4	1.1	.696±.20 (41)
Artificial			
^{54}Mn	*	0.2	<2.2 E-2 (15)
^{58}Co	*	0.1	<2.9 E-3 (13)
^{60}Co	*	9.6	<1.54 (39)
^{65}Zn	*	0.4	<3.7 E-2 (20)
^{106}Ru	*	0.7	<1.4 E-2 (9)
^{134}Cs	*	0.1	<1.9 E-2 (22)
^{137}Cs	0.1	3.3	1.0±.67 (41)
^{144}Ce	*	1.0	<1.7 E-1 (30)
^{152}Eu	*	3.1	<9.5 E-1 (37)
^{154}Eu	*	6.0	<4.3 E-1 (29)

* Less than detectable.

(a) Average plus or minus one standard deviation calculated if all analyses were positive. Otherwise a less-than number was calculated from all results including less than detectable values. The numbers in parenthesis indicate the number of positive results out of a total of 41 analyses.

Table 3 compares the aerial survey results for ^{60}Co in μR/hr with the concentration of ^{60}Co measured in sediment samples for each sampling location. As expected, the sediment sample results are quite variable and too few in number to allow a quantitative description of the radionuclide concentrations. However, the results do verify the presence of elevated levels of ^{60}Co, consistent with the aerial survey data, when compared to an expected background concentration of approximately 0.01 pCi/g.

TABLE 3. Comparison of Aerial Survey and Sediment Sampling Results

Sample No.	Description	Aerial Survey ^{60}Co μR/hr	Sediment Sample ^{60}Co, pCi/g-dry wt.
1	B Reactor Shoreline	3.5-8.8	0.2
2	N Reactor Shoreline	> 22.4(a)	0.6
3	D Reactor Shoreline	5.6-8.8	2.4
4(b)	High Water Channel	3.5-5.6	9.6
5(b)	White Bluffs Slough	5.6-8.8	1.0
6	White Bluffs Slough	2.2-5.6	0.06
7	100 F Slough	2.2-8.8	1.1
8(b)	100 F Slough	2.2-8.8	7.3
9	Hanford Slough	2.2-8.8	1.0
10	Hanford Slough	2.2-8.8	0.01
11	Savage Island-high water channel	3.5-8.8	0.7
12(b)	Wooded Island	5.6-8.8	1.7
13	Island #345 Above 300 Area	3.5-8.8	6.2
14	Island #344, 300 Area	1-2	2.4
15	Island #342, North Richland	< 1-2	2.1
16	Island #341, Richland Pumphouse	< 1-3	0.6
17(b)	Island #340, Below Richland Pumphouse	3.5-8.8	1.4
18	Island #333, Below Confluence of Yakima River	< 1-5.6	2.5
19	Island #332, Columbia Park	< 1-3.5	2.1
20	Island #333, Columbia Park	< 1-3.5	3.2
21	Sacajawea Park Shoreline	< 1	0.6

(a) Radiation from 1301-N leach trench instead of shoreline.

(b) Location of profile samples to 8-inches.

"FILTER-RESIN" SAMPLING

Beginning in 1973, a new method of sampling river water for radioactivity, refined by BNW's Radiological Chemistry staff,[2] was used. The method results in a much lower detection level and involves running a known quantity of river water through a nylon filter, a series of fiberglass filters, and a mixed bed ion exchange column. The sampler operates continuously during

which the river water flows (~3 liters/hour) through the nylon filter which removes all macro (>5 microns) particles, through the series of fiberglass filters which remove all particles greater than 0.3 microns, and then the filtered water flows through the resin to remove all soluble radionuclides with the exception of tritium. The filters and resin are changed biweekly and directly counted with a high sensitivity multi-dimensional (coincidence) gamma ray spectrometer to measure the radionuclides present.

Table 4 presents a summary of the radionuclides observed, the range of results, and average concentration observed during 1973, 1974 and the first few months of 1975. With the exception of naturally occurring radionuclides, ^{60}Co and ^{106}Ru were observed with the most consistency.

TABLE 4. Radionuclides Observed in Columbia River Water

		Concentration (pCi/l)								
		1973			1974			1975(a)		
Radionuclide	Detection limit	Maximum Observed	Minimum Observed	Average	Maximum Observed	Minimum Observed	Average	Maximum Observed	Minimum Observed	Average
^{40}K	0.009	0.8	0.5	0.7	2.3	*	<1.0	0.8	0.4	0.5
^{54}Mn	0.014	1.0	*	<0.2	<0.03	*	<0.01	0.6	*	<0.1
^{60}Co	0.0005	0.5	0.03	0.09	0.04	*	<0.02	0.1	0.006	0.02
^{65}Zn	0.005	0.2	0.02	0.06	<0.04	*	<0.01	0.1	*	<0.02
$^{95}ZrNb$	0.005	*	*	*	0.6	*	<0.1	0.5	0.05	0.1
^{106}Ru	0.005	1.0	0.05	0.09	0.2	0.02	0.12	0.2	0.09	0.1
^{137}Cs	0.005	*	*	*	<0.07	*	<0.03	0.4	*	<0.1
^{152}Eu	0.02	*	*	*	<0.03	*	<0.02	<0.05	*	<0.02
^{226}Ra	0.002	0.07	*	<0.03	0.14	<0.01	<0.05	0.8	0.003	0.04
^{228}Th	0.0005	0.012	*	0.004	0.04	*	<0.01	0.05	0.004	0.01

* Less than detectable

(a) Data collected from February 26 to July 14, 1975, only.

Figures 7 and 8 show the variability of ^{60}Co and ^{106}Ru results for both soluble and particulate (insoluble) fractions for each beweekly sampling period. The elevated concentrations of radioactivity associated with particulates each spring and early summer are assumed due to higher water levels in the Columbia River and the resulting resuspension of greater quantities of sediment in the water. The sharp peak observed during July 1973 for soluble ^{106}Ru was due to fallout from a June 26, 1973 nuclear

test in the atmosphere by the Peoples Republic of China.

Table 5 is a summary of the average concentrations of each radionuclide observed for both the soluble and particulate fractions. ^{40}K, ^{226}Ra, and ^{106}Ru are primarily observed in a soluble form whereas ^{54}Mn, ^{60}Co, and ^{65}Zn are primarily observed in a particulate form. These latter radionuclides were released in large quantities from past operation of once-through production reactors and the majority of the observed concentrations are attributed to this source. The contribution of fallout to the observed levels is not known but expected to be quite low from analysis of sediment core samples collected from Priest Rapids Dam (Table 1). A similar "filter-resin" sampling system has been installed at Priest Rapids Dam during the first week of September 1975. This unit will measure and identify radionuclides present in the river from background and fallout sources. In any event, all of the observed concentrations are less than 0.01% of the most restrictive ERDAM 0524 Guideline[3] for drinking water as shown in Table 5.

TABLE 5. Soluble and Particulate Fraction Concentrations Observed During 1973, 1974 and 1975 (pCi/ℓ)

Radionuclide	0524[a]	1973		1974		1975[b]	
		Soluble	Particulate	Soluble	Particulate	Soluble	Particulate
Natural							
^{40}K	--	0.7	0.05	0.9	<0.1	0.5	<0.2
^{226}Ra	30	0.02	<0.006	<0.05	<0.01	<0.04	<0.008
^{228}Th	7,000	<0.002	<0.002	<0.01	<0.005	<0.006	0.002
Artificial							
^{54}Mn	100,000	<0.02	0.13	*	*	<0.02	<0.07
^{60}Co	30,000	0.01	0.08	0.007	0.02	0.008	0.02
^{65}Zn	100,000	0.02	0.04	<0.005	<0.007	<0.002	<0.01
$^{95}ZrNb$	60,000	*	*	<0.05	<0.07	<0.04	<0.05
^{106}Ru	10,000	0.07	<0.02	0.11	0.02	0.11	0.05
^{137}Cs	20,000	*	*	*	*	<0.06	<0.02
^{152}Eu	60,000	*	*	*	*	<0.01	<0.006

(a) ERDA Manual Chapter 0524 standards only apply to concentrations due to ERDA facilities in excess of naturally occurring or fallout radioactivity. ERDAM 0524 does not list a concentration standard for ^{40}K, a naturally occurring radionuclide.

(b) Data collected from February 26 to July 14, 1975 only.

CONCLUSION

Long-lived radioactivity from past operation of once-through cooling production reactor operation is currently measurable at several places along the Columbia River shoreline, islands, bottom and slough areas.

A detailed aerial survey of the Columbia River by EG&G, Inc. of Las Vegas showed radiation, at several locations, from ^{60}Co (maximum 0.022 mR/hr) and, at a few locations, from ^{137}Cs (maximum, 0.003 mR/hr) in addition to natural background radiation (about 0.010 mR/hr). Sediment core samples collected from the bottom of the Columbia River behind Priest Rapids Dam (upstream of the Hanford Reservation) and McNary Dam (downstream of the Hanford Reservation) indicate that radioactivity from past Hanford operations is at least 22 inches in depth. Radionuclides present in the greatest amounts relative to background concentrations are ^{60}Co and ^{152}Eu. Analysis of sediment samples collected from the river shoreline, islands, and slough areas showed elevated levels of long-lived radionuclides, primarily ^{60}Co, ^{137}Cs, and ^{152}Eu. The concentration of radioactivity due to past once-through cooling production reactor operation near the surface is decreasing due to dilution.

Continuous river water sampling with a "filter-resin" sampler showed very low concentrations of long-lived radionuclides (^{60}Co, ^{65}Zn, ^{152}Eu) presumably due to past once-through cooling reactor operation. The contribution of fallout to the observed concentrations is not known. An additional "filter-resin" sampler installed at Priest Rapids Dam will allow measurement of the concentration of radionuclides in the river due to fallout. All observed concentrations were less than 0.01% of the most restrictive ERDA Manual Chapter 0524 standard for drinking water.

REFERENCES

1. W. J. Tipton, An Aerial Radiological Survey of the U. S. Atomic Energy Commission's Hanford Reservation, EG&G, Las Vegas, NV, in press.

2. D. E. Robertson, et al, "Transport and Depletion of Radionuclides in the Columbia River," IAEA, Vienna, 1973.

3. "Standards for Radiation Protection," ERDA Manual, Chapter 0524, with Appendix. U. S. Energy Research and Development Administration, Washington, DC, 1963. Revised October 1973.

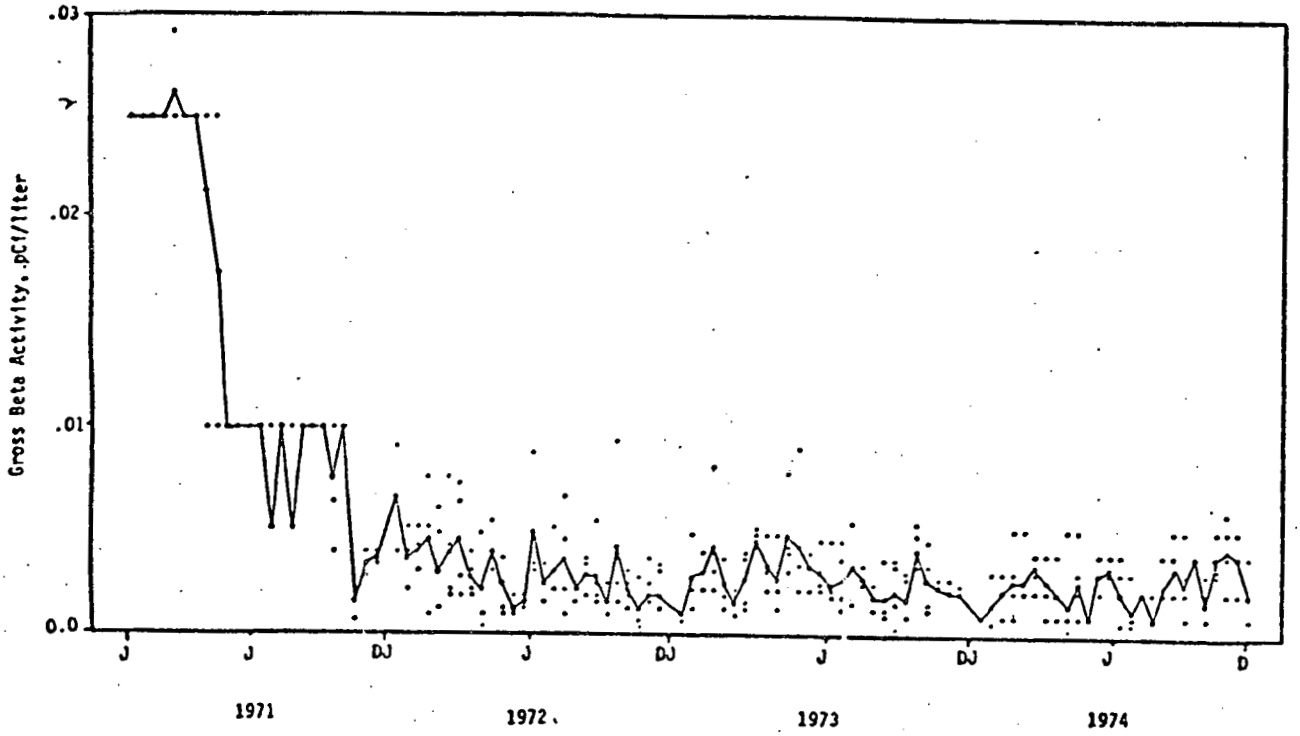


FIGURE 1. Variation of gross beta activity in the Columbia River.

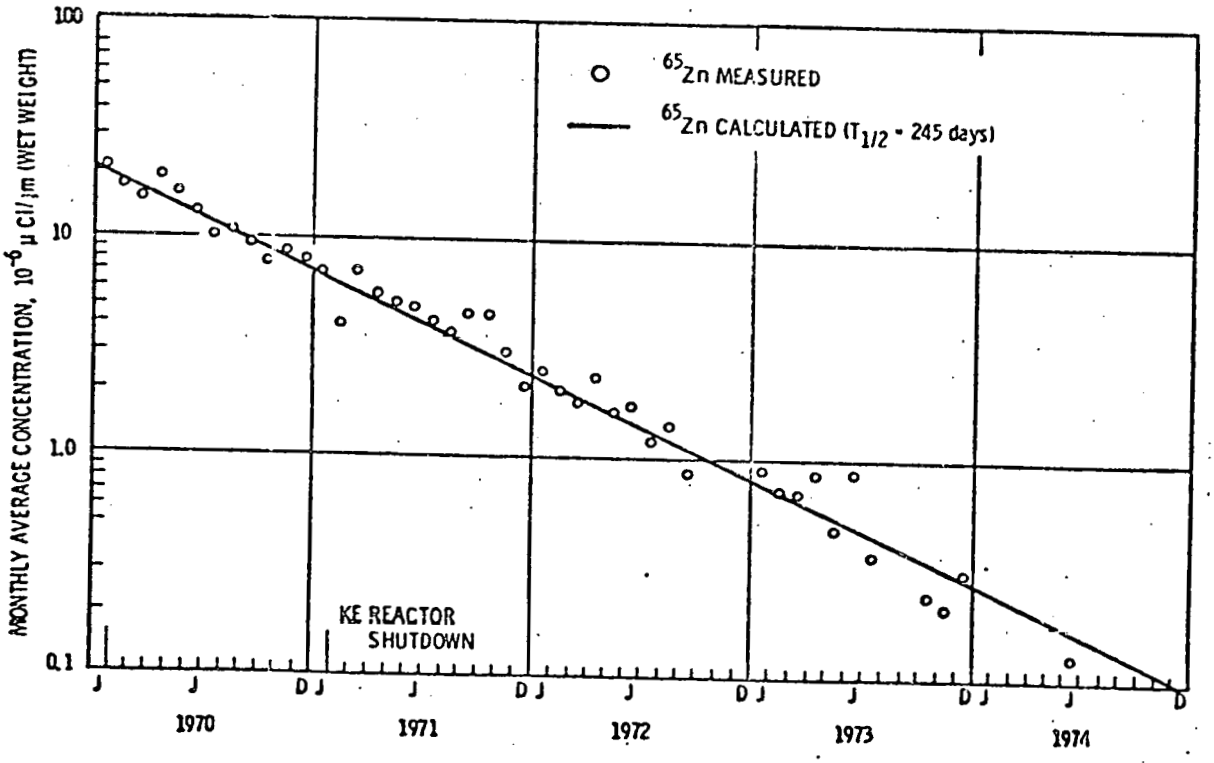


FIGURE 2. Decline of ^{65}Zn activity in Willapa Bay oysters.

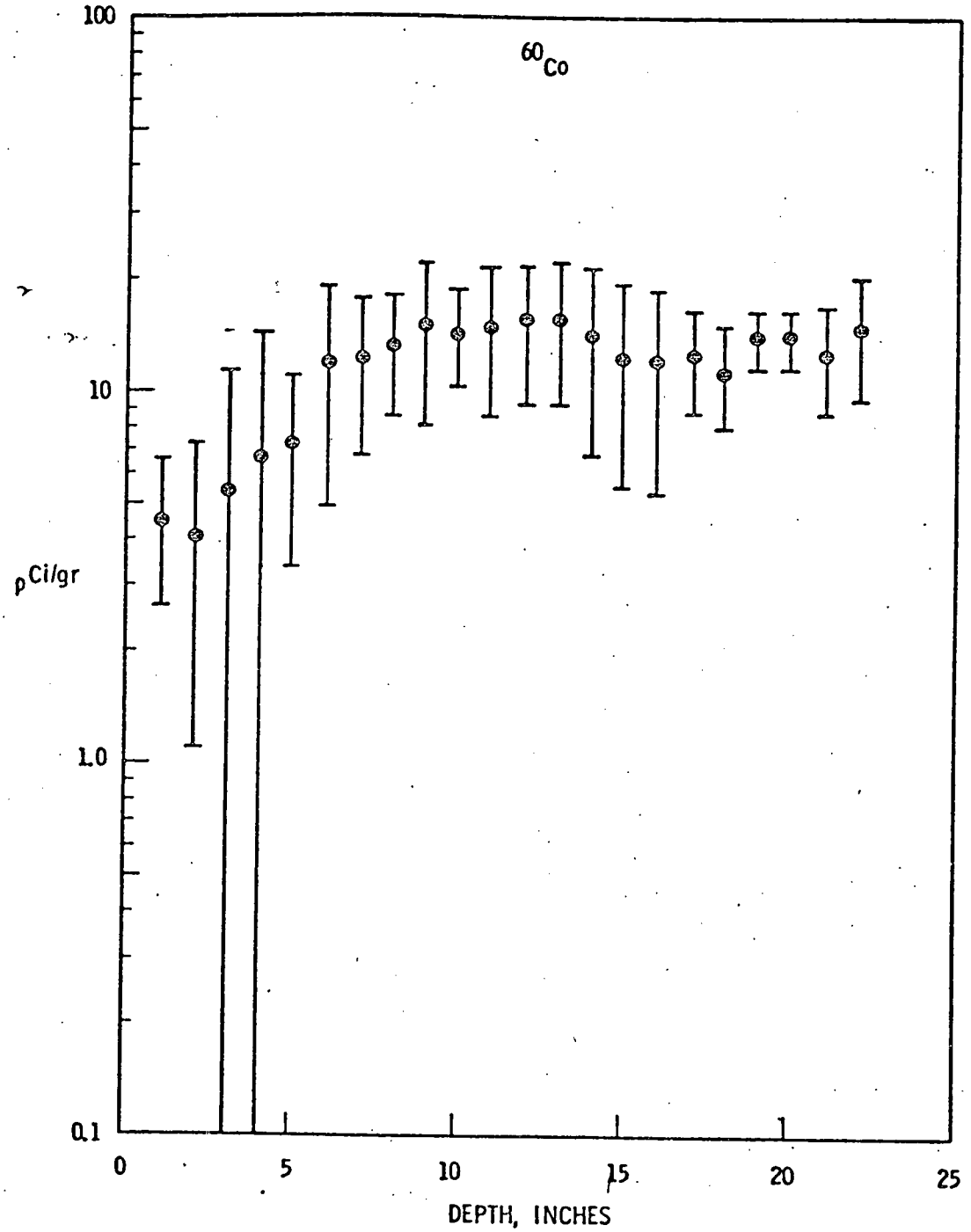


FIGURE 3. Depth profile of ^{60}Co in sediment behind McNary Dam from measurements during 1973.

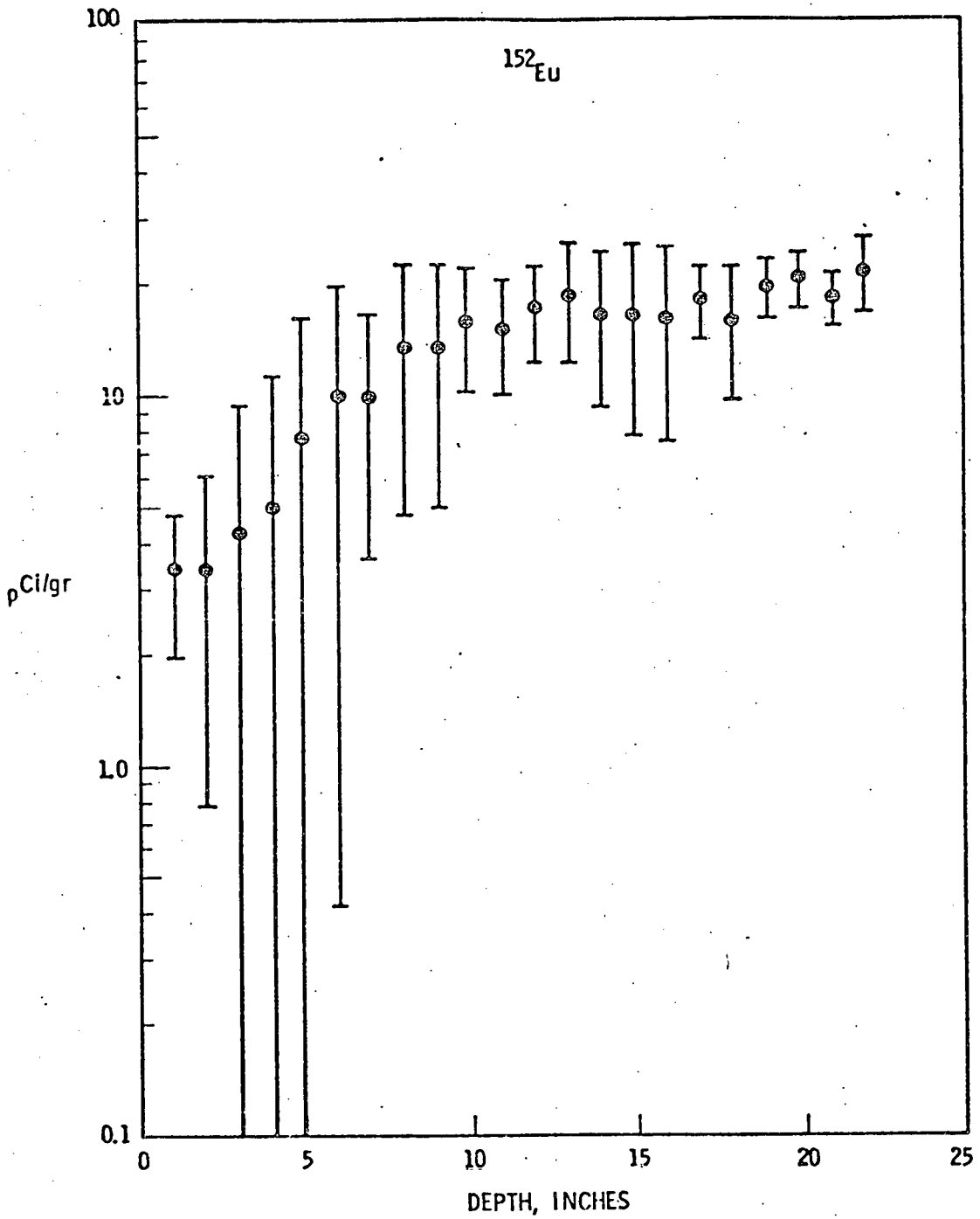


FIGURE 4. Depth profile of ^{152}Eu in sediment behind McNary Dam from measurements during 1973.

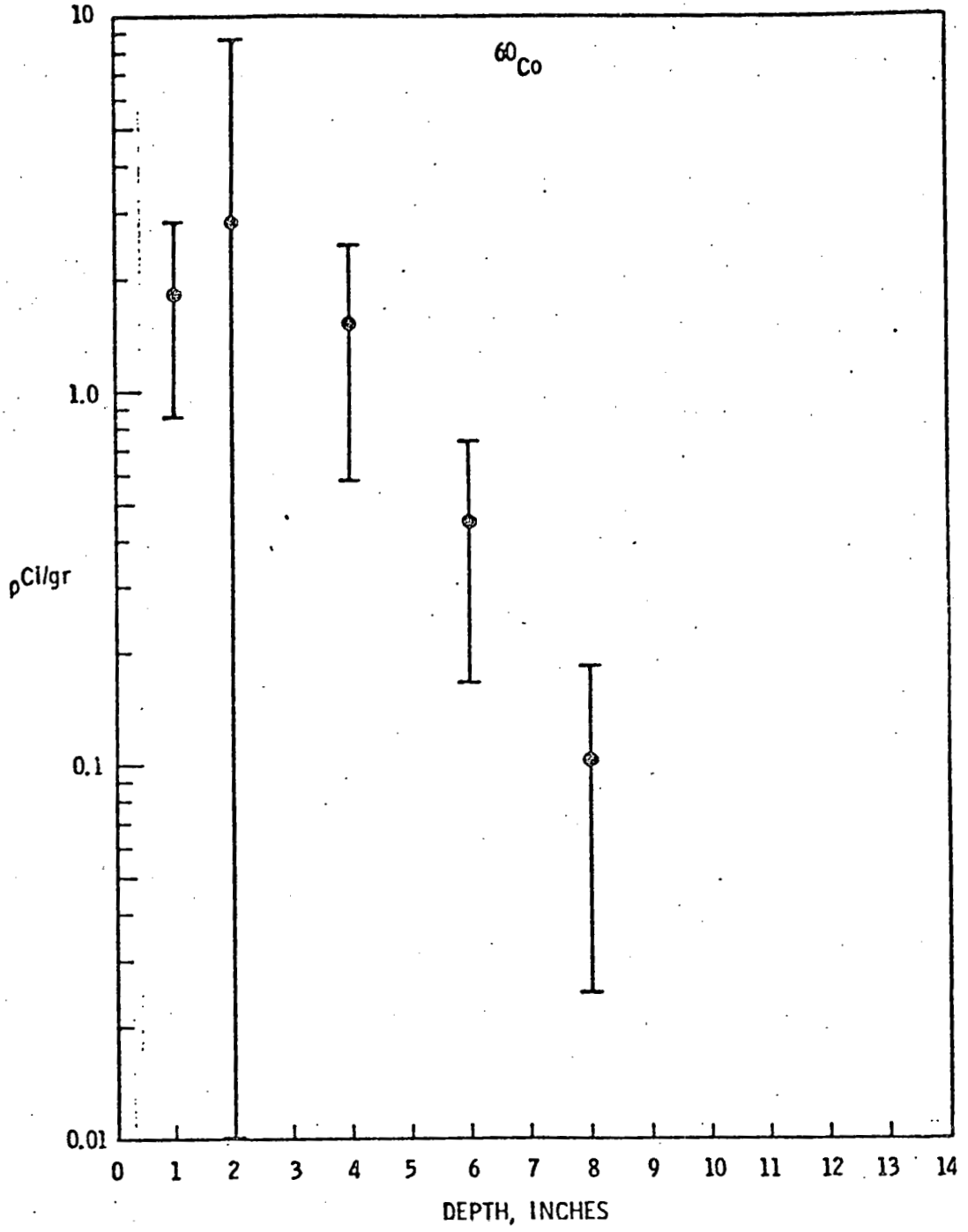


FIGURE 5. Depth profile of ^{60}Co in sediment from Columbia River shoreline, islands and slough areas.

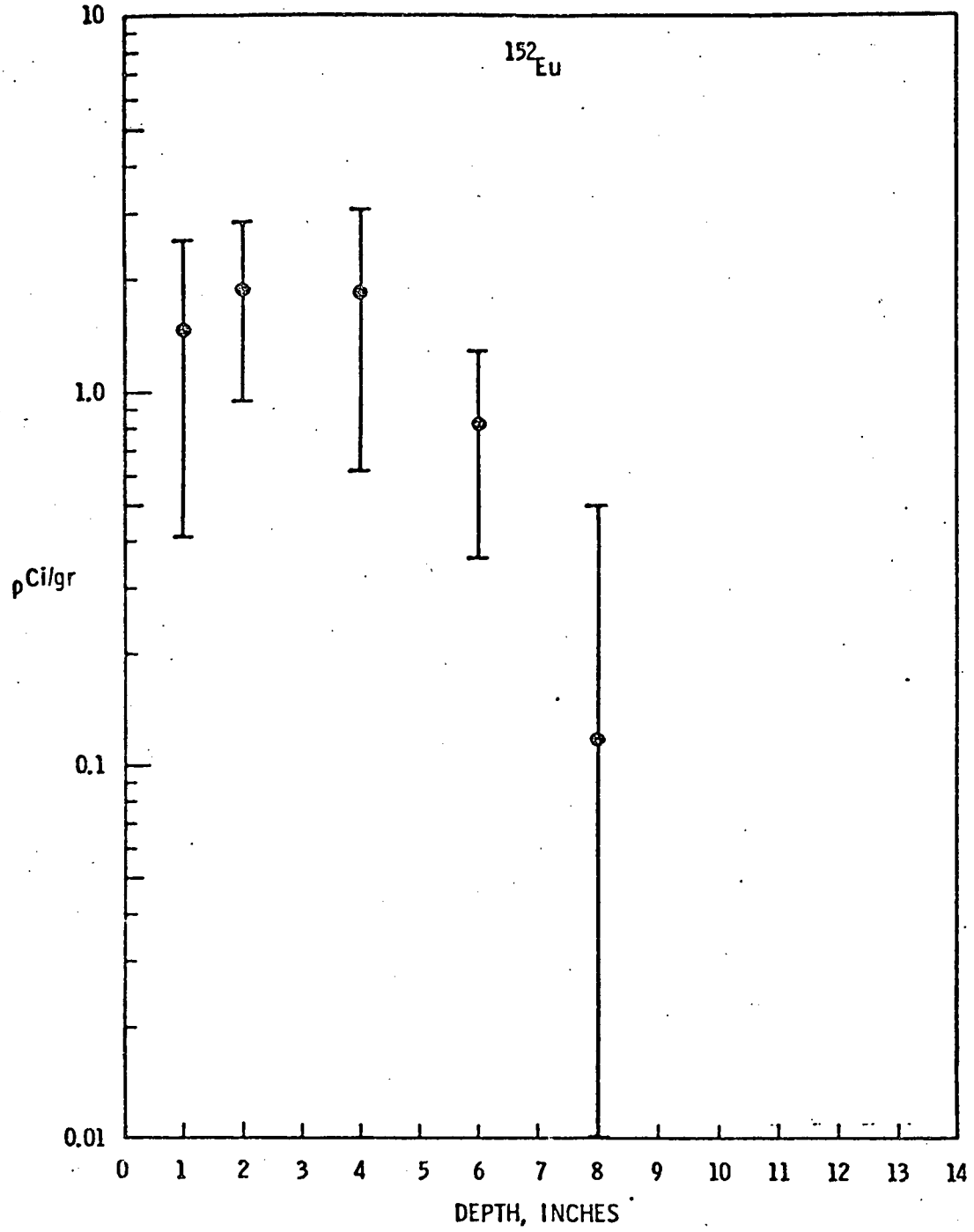


FIGURE 6. Depth profile of ^{152}Eu in sediment ~~samples collected~~ from Columbia River shoreline, islands, and slough areas.

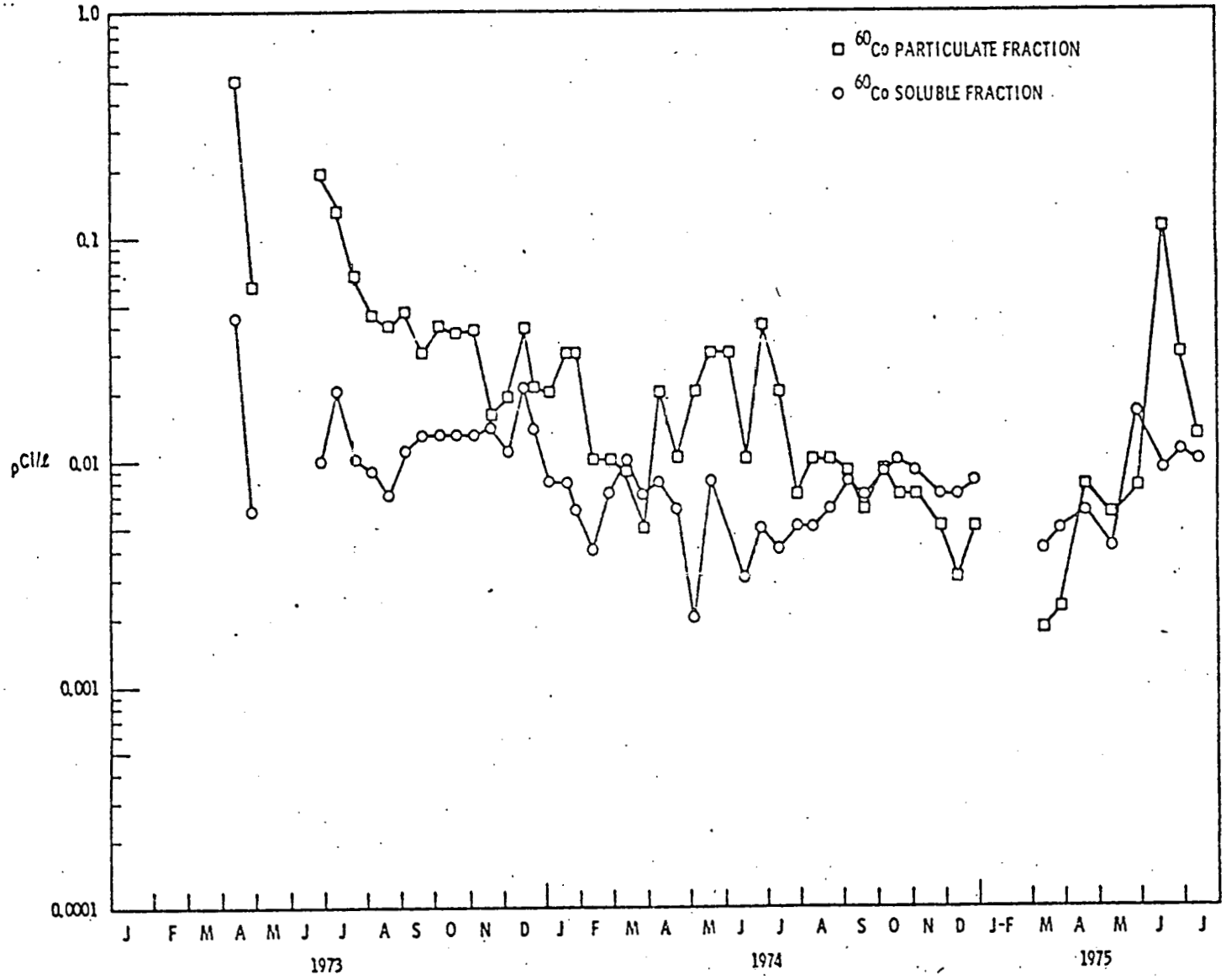


FIGURE 7. ^{60}Co activity in Columbia River, soluble versus insoluble fractions.

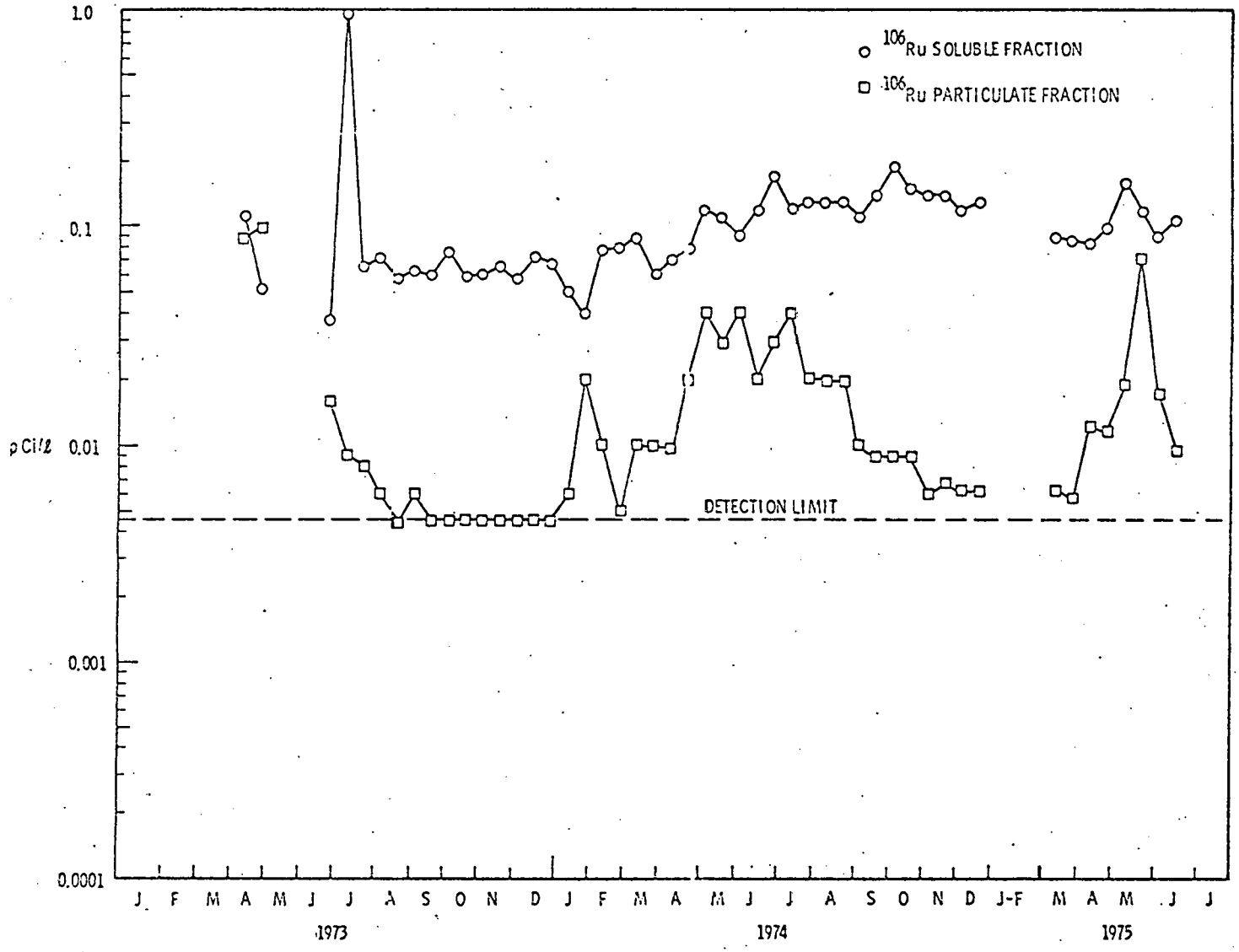
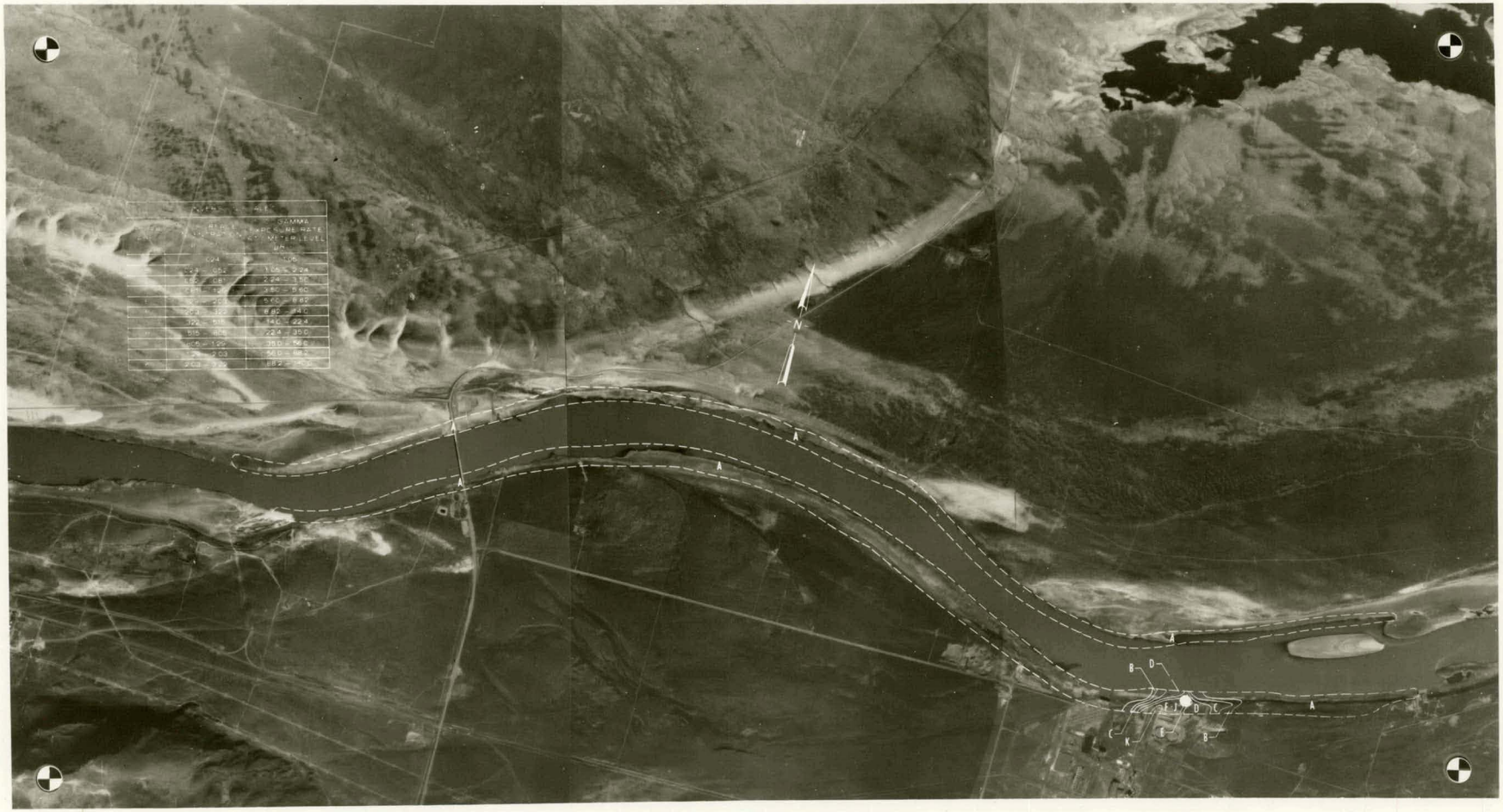


FIGURE 8. ^{106}Ru activity in Columbia River, soluble versus insoluble fractions.



1 MILE
2 KILOMETERS
SCALE

PHOTO TAKEN 21, MARCH 1974

● SEDIMENT SAMPLING LOCATION

COLUMBIA RIVER PHOTO I
VERNITA BRIDGE TO 100B AREA

PHOTO NO. 75590-12

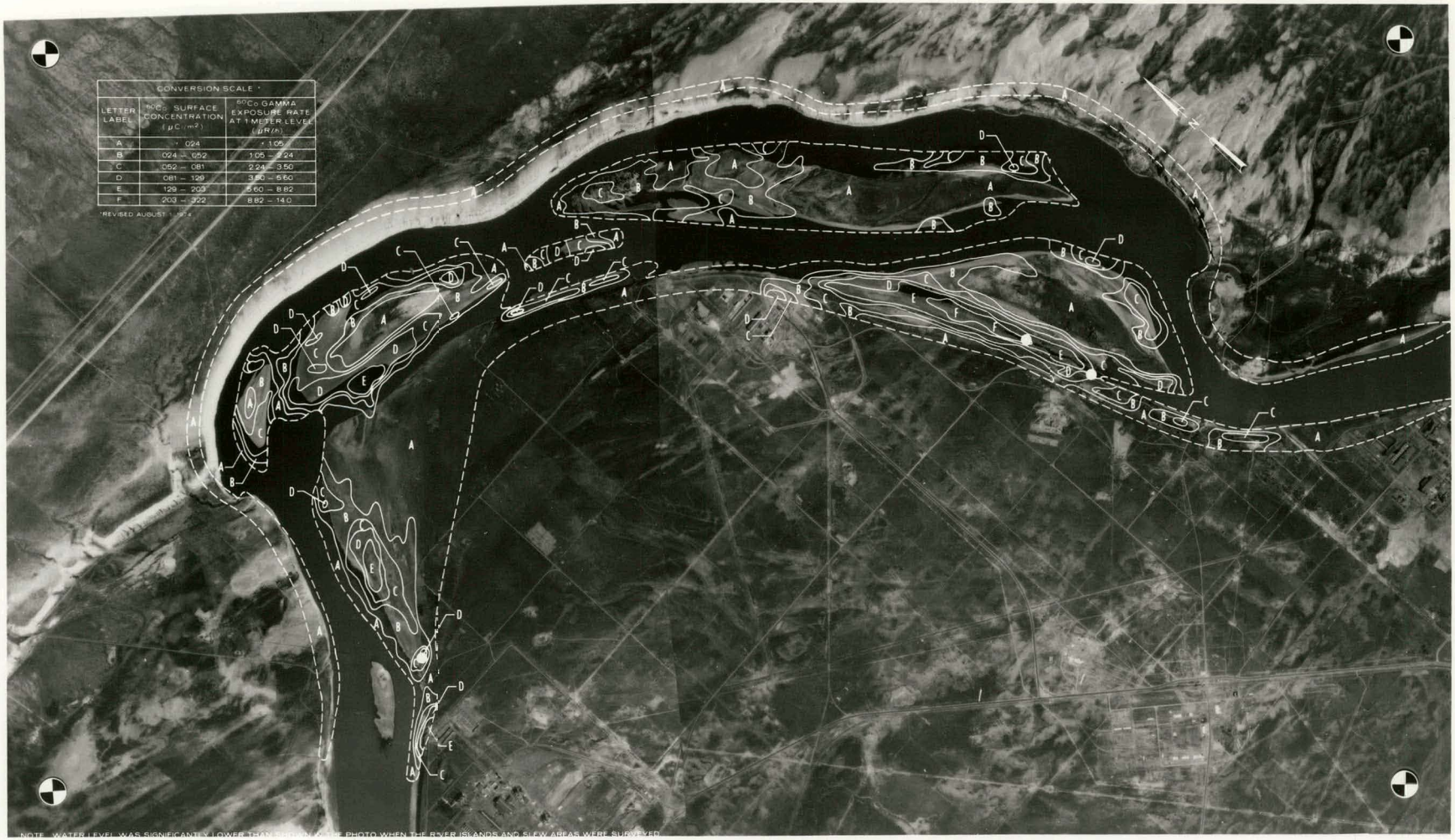
SEATTLE ARTS-WEST PHOTOGRAPHY
SEATTLE WASHINGTON

1 MILE
2 KILOMETERS
SCALE

PHOTO TAKEN 21, MARCH 1974

● SEDIMENT SAMPLING LOCATION

COLUMBIA RIVER - PHOTO II
100B TO 100D AREAS

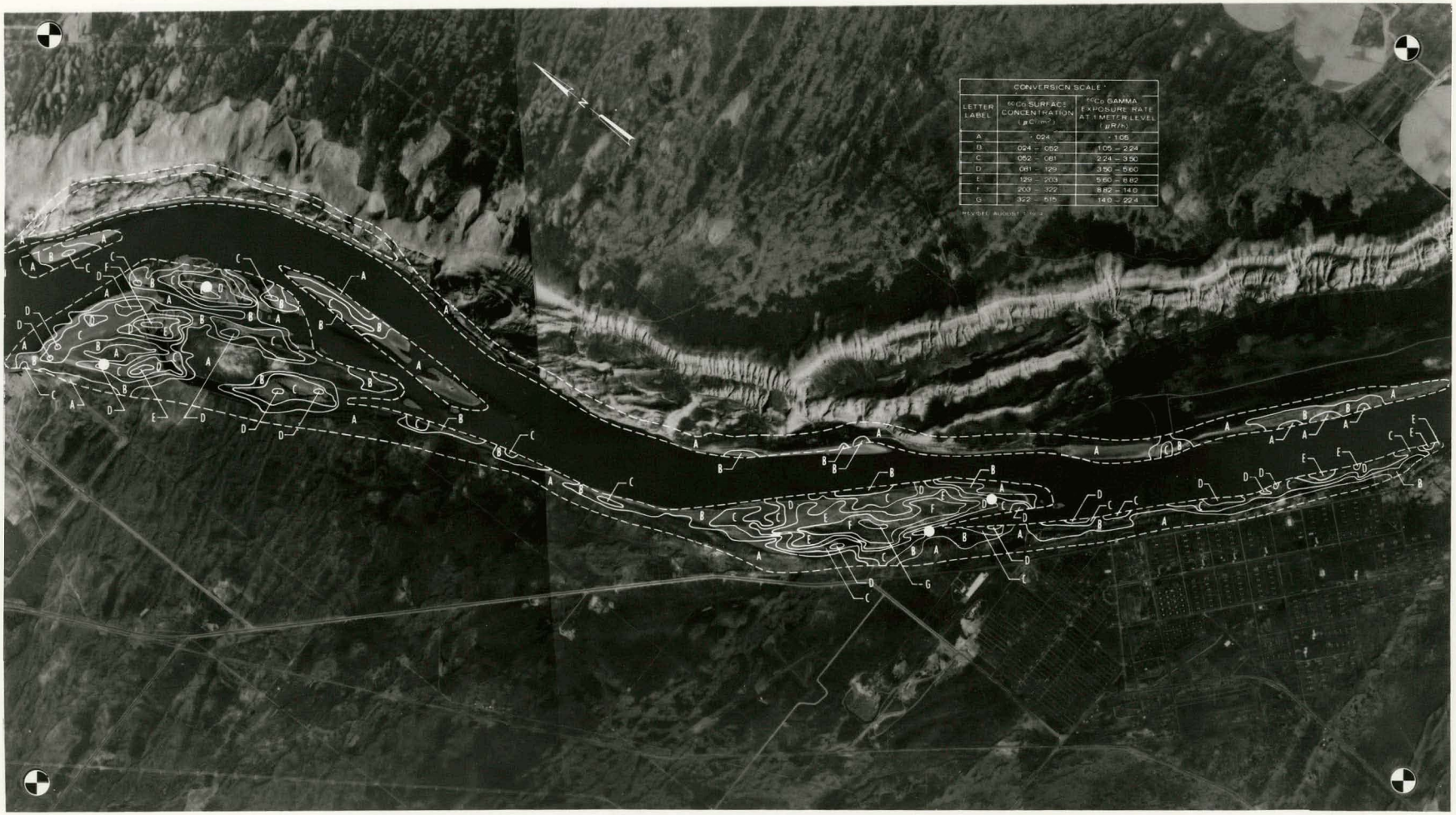


1 MILE
2 KILOMETERS
SCALE

PHOTO TAKEN 21, MARCH 1974

● SEDIMENT SAMPLING LOCATION

COLUMBIA RIVER - PHOTO III
100D TO 100F AREAS



1 MILE
2 KILOMETERS
SCALE

PHOTO TAKEN 21, MARCH 1974

● SEDIMENT SAMPLING LOCATION

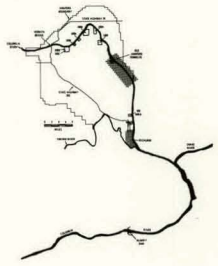
COLUMBIA RIVER - PHOTO IV
100F AREA TO HANFORD

1 MILE
2 KILOMETERS
SCALE

PHOTO TAKEN 21. MARCH 1974

● SEDIMENT SAMPLING LOCATION

COLUMBIA RIVER - PHOTO V
SAVAGE ISLAND AREA

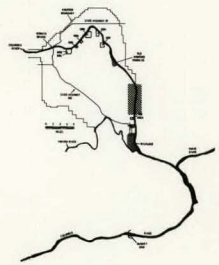


1 MILE
2 KILOMETERS
SCALE

PHOTO TAKEN 21, MARCH 1974

● SEDIMENT SAMPLING LOCATION

COLUMBIA RIVER - PHOTO VI
WOODED ISLAND AREA



1 MILE
2 KILOMETERS
SCALE

PHOTO TAKEN 21, MARCH 1974

● SEDIMENT SAMPLING LOCATION

COLUMBIA RIVER - PHOTO VII
300 AREA TO NORTH RICHLAND

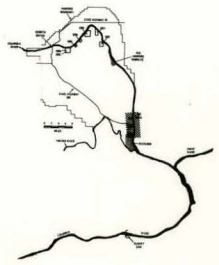


PHOTO NO. 755990-2

PHOTO TAKEN 21, MARCH 1974

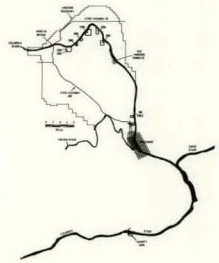
1 MILE

2 KILOMETERS

SCALE

● SEDIMENT SAMPLING LOCATION

COLUMBIA RIVER - PHOTO VIII
INTERSECTION WITH YAKIMA RIVER



1 MILE
2 KILOMETERS
SCALE

PHOTO TAKEN 21. MARCH 1974

● SEDIMENT SAMPLING LOCATION

COLUMBIA RIVER - PHOTO IX
PASCO - KENNEWICK AREA

755990-7

1 MILE
2 KILOMETERS
SCALE

PHOTO TAKEN 21, MARCH 1974

COLUMBIA RIVER - PHOTO X
INTERSECTION WITH SNAKE RIVER

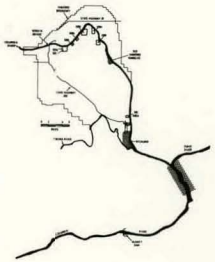


PHOTO NO. 755990-4

1 MILE
2 KILOMETERS
SCALE
PHOTO TAKEN 21, MARCH 1974

COLUMBIA RIVER - PHOTO XI
McNARY DAM - UMATILLA AREA

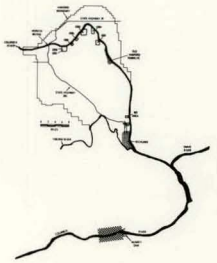
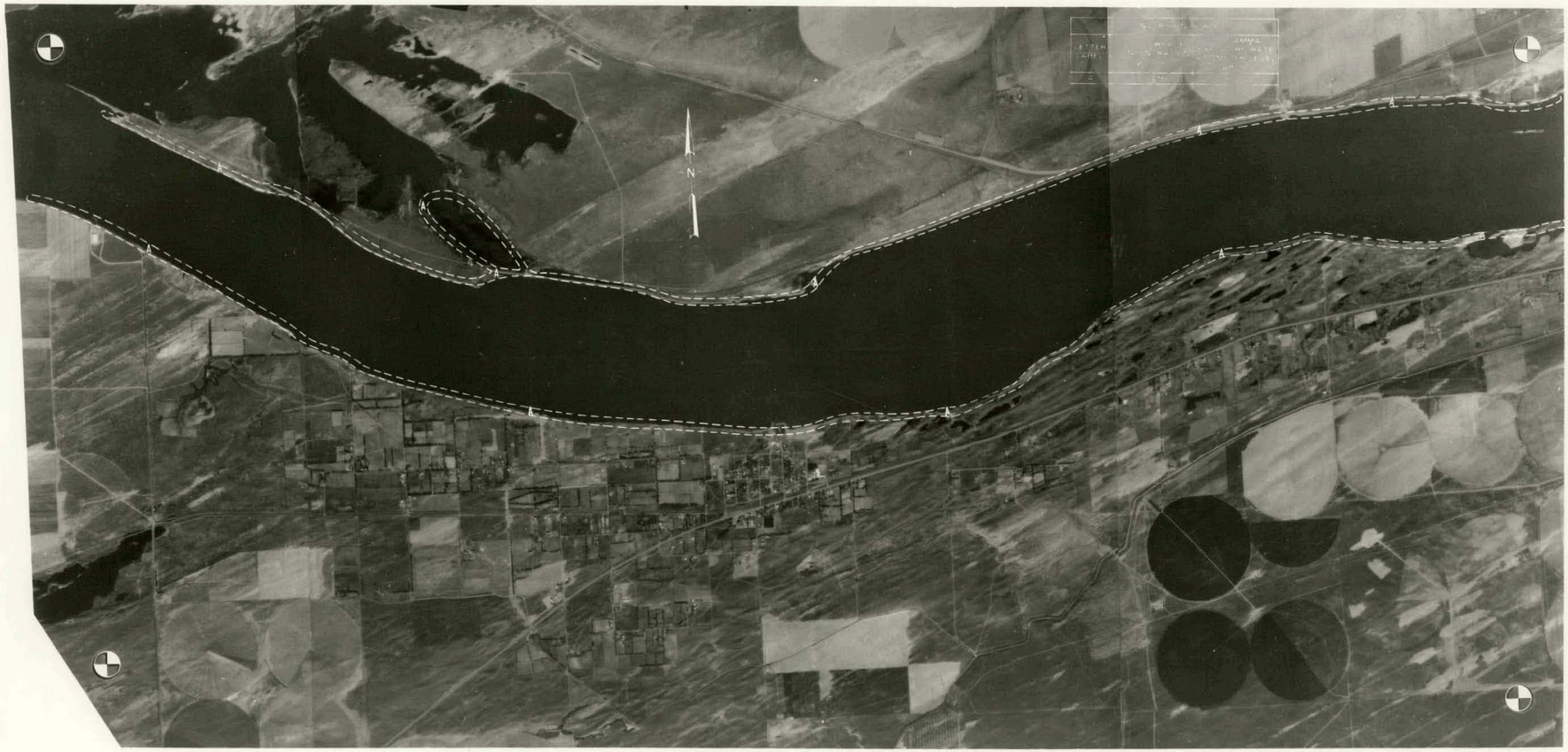


PHOTO NO. 755990-5



1 MILE

2 KILOMETERS

SCALE

PHOTO TAKEN 21. MARCH 1974

COLUMBIA RIVER - PHOTO XII
IRRIGON AREA

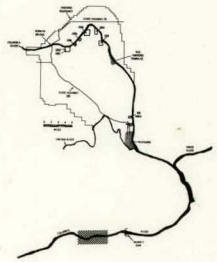


PHOTO NO. 75 5990-6