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RW Perkins, JL Nelson, WL Haushild.

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BEHAVIOR AND TRANSPORT OF RADIONUCLIDES IN THE COLUMBIA RIVER BETWEEN HANFORD AND THE OCEAN (a)

R. W. Perkins(b), Jack L. Nelson(b), and W.L. Haushild(c)

ABSTRACT

A study of the behavior during transport by the Columbia River of the eleven radionuclides Sc^{46}, Cr^{51}, Mn^{54}, Co^{58}, Fe^{59}, Co^{60}, Zn^{65}, Zr^{95}-Nb^{95}, Ru^{106}, Sb^{124}, and Ba^{140} was completed in the river reach between Pasco, Washington and Vancouver, Washington during the period January 1964 through January 1965. This extensive study of radionuclide behavior was performed by direct counting of water salts and filter samples on a multidimensional gamma-ray spectrometer and represents the first measurements of the interactions of most of these radionuclides in the river. Through this multiple tracer technique the specific and relative behaviors of the eleven radionuclides in regard to their sorption by and movement with suspended particulates in the Columbia River were determined. Also, depletion of these radionuclides from the river during transport, and the river bottom's radionuclide inventory in the river reach between Pasco, Washington and Vancouver, Washington were determined.

INTRODUCTION

Cooling of the Hanford Reactors with Columbia River water and the subsequent disposal of this water to the river provides a wide spectrum of radioactive tracers to the river. These radionuclides are produced from the river water salts on exposure of the water to the reactor's neutron flux. In addition, significant quantities of some radionuclides enter the river as fallout from the large areas drained by the Columbia River. For example, it was observed during 1963 that the fission products Zr^{95}-Nb^{95} were present in the Snake River and in the Columbia River upstream from the Hanford Works at concentrations comparable to that found downstream from Hanford[1].

Although the concentrations of all radionuclides in the Columbia River are low, and at points of domestic usage fall well within accepted radiological standards

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for drinking water[2], a study of their behavior provides useful information in understanding the processes that take place in rivers as well as information on routes by which Columbia River radionuclides may become available for uptake by man.

As a part of our overall investigation of the behavior of the Columbia River radionuclides a study has been conducted to determine their physical behavior and transport by the River.

Several of the short-lived radionuclides and the long-lived radionuclides Zn^{65} and Cr^{51} which are present in the Columbia River are monitored routinely at Pasco, Washington (see Figure 1) by the Radiological Protection Operation at Hanford[3,4,5,6,7]. Crude measurements, involving only total beta counts are also made at this point by the Public Health Service on a routine basis[8].

Our past studies with a few radionuclides have shown that changes in physical form and in concentration occur during transport by the river[1,9,10,11]. Core sampling of sediments in the McNary Dam reservoir and slack water regions of the river have shown a buildup of sediments containing these radionuclides[9].

As a preliminary part of this work the use of multidimensional gamma-ray spectrometry was explored in our laboratory[12]. This technique proved to be extremely useful and allowed a much more thorough investigation of the problems than had previously been considered possible. By use of this method it was possible to make direct measurement of eleven of the long-lived radionuclides at locations between the Hanford nuclear reactors and the ocean. These radionuclides include Sc^{46}, Cr^{51}, Mn^{54}, Co^{58}, Fe^{59}, Co^{60}, Zn^{65}, Zr^{95}-Nb^{95}, Ru^{106}, Sb^{124}, and Ba^{140}. This multiple tracer technique allowed the rapid accumulation of essential information on the specific and relative behavior of these radionculides in regard to their transport, sorption, and depletion in the river environment. This study is based on radionuclide measurements of river water samples which were collected on a weekly basis at Pasco and Vancouver Washington and at less frequent intervals at Hood River, Oregon. This extensive study has supplied the answers to several basic questions on the behavior of radionuclides in the Columbia River.

EXPERIMENTAL METHOD

The study was designed to include a sufficient number of measurements so that the transport and general behavior of most of the detectable radionuclides in the Columbia River could be determined. This was accomplished by the collection and

analysis of representative samples on a periodic basis at the Columbia River bridges at Pasco, Washington, Hood River, Oregon and Vancouver, Washington.

Sampling

During preliminary studies in 1962-63 sampling from each bridge was performed at 4 or 5 centroids of flow of the river and these samples were analyzed separately to determine the existing variations of the river cross section. Also, samples collected near the surface and near the bottom were analyzed to demonstrate any vertical variations in radionuclide concentrations. From these observations it was concluded that such variations were relatively minor and that a single sample composited from 4 or 5 flow centroids would provide a reasonably representative aliquot of the river for analysis.

Sample Preparation

Six gallon aliquots of river water are collected from the sampling sites and immediately filtered through 0.3 μ membrane filters (AM-7, Gelman Instrument Co., Chelsea, Michigan). In the Laboratory the water is acidified with HNO_3 and evaporated to near dryness. It is then transferred to a small (250 ml) beaker and evaporated to dryness under a heat lamp. The salts are transferred to a 30 ml sample holder for counting which consists of a 0.500 inch long section of a 2 inch diameter pipe with waterproof adhesive tape across the bottom. The salts transfer and dissolution is accomplished by 4 washes of the beaker with 5 ml portions of distilled water during which the inside bottom and sides of the beaker are scrubbed with a rubber policeman. To the sample is added 10 ml of a hot 10% agar agar solution which is uniformly stirred into the sample and which solidifies fairly rapidly to provide a standard geometry for counting. The top of the ring is covered with waterproof adhesive tape for counting. These samples can be stored for long periods in a refrigerator without degradation or can be stored indefinitely in frozen state. The membrane filters, which contain the particulate material removed from the river water samples, are finely divided then mounted in agar agar in a manner similar to that of the river salts for counting.

Instrumental Analysis

Multidimensional gamma-ray spectrometry[12] provides the most sensitive known method for the direct measurement of many trace radionuclides in a complex radionuclide mixture and has proven extremely useful in this study.

The detector system[12] for the multidimensional analyzer consists of two 6-inch diameter by 4-inch thick NaI(Tl) crystals which are enclosed in a 11.5 inch diameter by 12-inch long annular NaI(Tl) crystal which serves as an anticoincidence

shield. The sample (see sample preparation) is located between the two 6-inch diameter detectors and the signals from these detectors are fed to the two respective ADC's of a 4096 channel multiparameter analyzer. When a single photon interacts with one of the detectors, the event is stored on the corresponding X or Y axis of the analyzer memory. When two photons are emitted simultaneously from a radionuclide, and each interacts with a separate detector, the event is stored in an energy-energy plane at a point uniquely characteristic of the two energies. This instrument provides orders of magnitude less interference both from other radionuclides and from the natural background for the measurement of many of the radionuclides.

The gamma-rays which are used in the measurement of the eleven radionuclides in river water are summarized in Table I. In addition, the actual energy areas which are used for the measurements in the multidimensional gamma-ray spectra are summarized in Figure 2. As in normal gamma-ray spectrometry some of the higher energy coincidence gamma-rays are incompletely absorbed and contribute to the counting rate where lower energy coincidence events occur. These corrections are much lower than those required for normal gamma-ray spectrometry but are made in the same manner. The unique energy-energy definition of several of the radionuclides plus the extremely low background at their positions of measurement permits the direct measurement of this large group of radionuclides with reasonably good precision.

TABLE I

SINGLE AND COINCIDENCE GAMMA RAYS USED IN THE MEASUREMENT OF COLUMBIA RIVER RADIONUCLIDES

Radionuclide	Gamma Ray Energies
Sc^{46}	0.802 and 1.118
Cr^{51}	0.322
Mn^{54}	0.835
Co^{58}	0.51 and 0.814
Fe^{59}	1.289
Co^{60}	1.173 and 1.332
Zn^{65}	1.114
Zr^{95}-Nb^{95}	0.762 + 0.760 + 0.768
Ru^{106}	0.513 and 0.624
Sb^{124}	0.603 and 1.69
Ba^{140}	0.487 or 0.815 and 1.597

RESULTS

Radionuclide Concentrations and Physical Form

The observed concentrations of the eleven radionuclides in the Columbia River for the period January 1964 through January 1965 at Pasco, Washington, Hood River, Oregon and Vancouver, Washington are summarized in Figure 3. These values are in

terms of specific activity and the total transport of these radionuclides is of course a function of the river discharge. A curve showing the Columbia River discharge for a one year period at a gauging station (at Priest Rapids Dam) upstream from Pasco,Washington is included for comparison at the bottom of Figure 3. It is immediately apparent from Figure 3 that while the concentrations of most of the radionuclides decrease considerably between Pasco and Vancouver during most of the year, that during the spring and early summer that they are about constant or they actually increase. This is due to scouring action of sediment from the river bed and is considered in detail later. Some of the values for Ru^{106} and Co^{58} are reported as "less than" numbers. This was necessary because sufficient counting time was not available to allow their precise measurement in all samples; however, sufficient values are available to give a reasonably good picture of each radionuclide's behavior. It is interesting that the Ru^{106} concentration did not change much during the spring and summer high water. This radionuclide, as well as other radionuclides in "fallout" is present in the river above the Hanford project and the concentration of such radionuclides would be affected by factors other than river flow rate.

As previously indicated all of the water samples were filtered immediately after collection to permit a measurement of the fractions of each radionuclide associated with particulate material at each sampling point. Figure 4 presents graphs of the percent of each radionuclide which was attached to suspended particulate material in the river at Pasco, Hood River and Vancouver and it allows a direct comparison of the physical changes which occur during transport of these radionuclides. These curves demonstrate the vast differences which exist in the sorption behavior of these elements. The differences range from Sb^{124} which is essentially all in solution at all of the sampling points to that of Fe^{59} which is practically all associated with particulate material. In Table II the physical forms of these eleven radionuclides as they enter the river, (in reactor effluent water) and at the various downstream locations are compared. Major changes do occur both during the original mixing and transport to Pasco, Washington, and during subsequent transport by the river.

This "solution behavior" is of course helpful in understanding the natural depletion of radionuclides during transport by the river. Those radionuclides which are mainly in the particulate form in reactor effluent water or which are readily adsorbed by particulate material in the river include Sc^{46}, Mn^{54}, Co^{58}, Fe^{59}, Co^{60}, Zn^{65}, Zr^{95}-Nb^{95} and they would be expected to undergo natural depletion during their transport by the river. The radionuclides Cr^{51}, Ru^{106}, Sb^{124} and Ba^{140} which enters the river in solution and are not associated to a large extent with the suspended particulate material would be expected to be transported with a minimum of d pletion.

TABLE II

PHYSICAL FORM OF THE HANFORD REACTOR EFFLUENT WATER RADIONUCLIDES ON ENTRY INTO THE RIVER AND AT DOWNSTREAM LOCATIONS

(% particulate)

Radionuclide	Effluent Water*	Pasco **	Hood River***	Vancouver**
Sc^{46}	36	74	85	89
Cr^{51}	2.4	6.4	4.0	7.6
Mn^{54}	2.6	20	88	83
Co^{58}	4.2	27	~83	~88
Fe^{59}	64	85	80	80
Co^{60}	1.8	26	80	91
Zn^{65}	1.8	14	64	76
Zr^{95}-Nb^{95}	~69	69	68	85
Ru^{106}	32	24	15	17
Sb^{124}	1.1	3.4	--	~5.9
Ba^{140}	2.3	9.0	--	37

* February 25, 1965

** Average from last half of January 1965

*** Average of January through March 1965

The change in physical form during transport is perhaps best illustrated by comparing the concentrations of soluble radionuclides to that of soluble Cr^{51} at Pasco, Hood River and Portland. For this comparison the radionuclide concentrations at Hood River and Vancouver have been corrected for radioactive decay[11] for their river travel time from Pasco. The comparison is presented in Figure 5 and shows the large drop in the ratios of many of the radionuclides to that of Cr^{51} which occurred during their transport in the river. In addition to showing annual changes, these curves show rather clearly that the large increase in total activity of the river, which results from the resuspension of sediments by the spring freshet, (see Figure 7) is not accompanied by a similar large increase in the soluble radionuclide concentrations. This would indicate that these resuspended sediments are not significantly redissolved during transport.

The relative degree of depletion, or scouring can be obtained by a similar comparison in which the total (solution plus particulate) concentration of each radionuclide is compared to that of Cr^{51}. This comparison is made in Figure 6 and again uses values for the radionuclide concentrations at Hood River and Vancouver which are decay corrected for the river's travel time from Pasco. The relative magnitudes of depletion or resuspension of radionuclides are indicated respectively by their

lower or higher ratios relative to Cr^{51} at the downstream locations.

Radionuclide Transport

Figure 7 presents the transport rates of the eleven radionuclides at Pasco, Hood River and Vancouver during the period January 1964 through January 1965. The transport rates were calculated directly from the published river discharge[13], and the radionuclide concentrations at the sampling locations (see Figure 3) and are not corrected for the radioactive decay which occurred during the river flow time between Pasco and Hood River or Vancouver. This radioactive decay reduces the Ba^{140} and Cr^{51} concentrations between Pasco and Vancouver by as much as 30% and 65% respectively and is discussed in a later section of this paper. Radioactive decay of the other 9 radionuclides amounts to less than 10%.

The average daily discharge at Pasco can be determined from the Priest Rapids Dam gauging station which is located about 50 miles upstream by adding the contribution from the Yakima River. However, during lower flow periods (about 7 months out of the year) the flow rate past the Hanford project and Pasco, Washington undergoes a large diurnal variation. The Priest Rapids Dam is a "power peaking dam" and in meeting power requirements during low flow, when the entire river flow is channeled to the power plant turbines, the Columbia River discharge is varied by a factor of 2 to 4. With the Hanford reactors adding radioactivity to the river at essentially a constant rate, the effect of this river flow variation is to produce peaks and valleys of activity in the river water flowing past our first sampling point at Pasco, Washington. Although the river flow variation at the Priest Rapids Dam is about a factor of 3 the flow variation at Pasco is damped considerably and normally amounts to only about $\pm$ 20% variation from the average. Also the variation in the radioactivity of the river is relatively small. This is partially due to dispersion phenomena in the river itself but is also due to the fact that the reactor effluent water was added to the river from 8 reactors which are distributed along a 15 mile stretch of the river. In order to assess the degree of diurnal variation of radionuclide concentrations in the river at Pasco, Washington the concentrations of the two radionuclides Na^{24} and Cr^{51} were measured from samples collected at two hour intervals. These measurements were made during a period of large diurnal variation in river input but of fairly constant input of the radionuclides into the river. The variations in concentrations at Pasco are compared with the flow rates at Priest Rapids Dam and at Pasco in Figure 8. While the discharge at Priest Rapids Dam change by four-fold, the variation in both the Na^{24} and Cr^{51} concentrations from their average was about $\pm$ 15%. It is evident that the Na^{24} (T 1/2-15 hr) concentration change is less than those of Cr^{51}. This is due to the fact that the flow

time to Pasco is longer for radionuclides which are introduced into the river during low river discharge at the reactors and although their initial concentrations are high there is a longer period for decay before reaching Pasco. The Na^{24} decay may amount to 75% between the reactors and Pasco while that of Cr^{51} is insignificant (2-3%) as is the case for the other radionuclides which are included in this study.

The diurnal variation in concentrations of the radionuclides complicates the precise measurement of the radionuclide transport at Pasco; however, it is apparent from Figure 8 that the sampling which was performed for this study during the s between 0900 and 1500 daily would provide values which were reasonably close to the average value of the river water's radioactivity levels. Therefore in view of uncertainties involved in trying to estimate better values for the average daily river concentrations, the measured values were assumed to be the average concentration and were used in calculating the total radionuclide transport. It was realized in so doing that any given values may be in error by as much as ± 15%.

Radionuclide Depletion and Inventory

On comparison of the transport of radionuclides at Pasco, Hood River, and Vancouver (see Figure 7) it is evident that those radionuclides which associated themselves with particulate material (see Figure 4) are depleted from the river water by a factor of 5 to 10 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 of course is due to scouring action which resuspends deposited sediments which contain large amounts of radionuclides. During river transport, the radionuclides Cr^{51}, Ru^{106}, Sb^{124}, and Ba^{140} which are not associated to a large extent with suspended particulate material (see Figure 4) are transported with a relatively small depletion (see Figure 7).

The total amount of radionuclides which are deposited in the Columbia River sediments, or are removed by stationary river biota, etc., can be determined for the stretch of river between Pasco and Vancouver from the difference in radioactivity passing these two locations. The percent depletion of each radionuclide through this stretch of river was calculated for the period January 1964 through January 1965 by the equation:

$$\text{Depletion} = \frac{\text{Pasco transport} - \text{Vancouver transport}^{*}}{\text{Pasco transport}} \times 100$$

* Vancouver value corrected for decay for river travel time between Pasco and Vancouver[11].

Depletion values for the various radionuclides are presented in Figure 9. The negative depletion values indicate that the river scouring action has exceeded the

deposition rate.

If it is assumed that the depletion and the input rates of radionuclides to the river have been constant for the past several years their total inventory in this stretch of the river can easily be obtained. Such an inventory would be changing continuously and would be a function of both the depletion rate and the decay rate of the radionuclides. Figure 10 presents the estimated radionuclide inventory in the river bottom materials for each month of the year, as calculated from the preceding measurements and assumptions. For convenience, the January 1, 1965 inventories of these radionuclides are summarized in Table III. The exact location of the large radionuclide inventory cannot be determined from the data presented here; however, from examination of transport data at Pasco, Hood River, and Vancouver it appears that more than 75 percent of the inventory is upstream from Hood River. Previous studies have shown that a large deposit of sediments containing these radionuclides is located in the McNary Dam reservoir[9] and this appears to be a major deposition area of the Columbia River.

TABLE III

INVENTORY OF DEPOSITED RADIONUCLIDES IN THE COLUMBIA RIVER BETWEEN PASCO AND VANCOUVER, WASHINGTON

Radionuclide	Inventory	
	dpm x 10^{-14}	Curies
Sc^{46}	11.5	518
Cr^{51}	261	11,800
Mn^{54}	7.2	320
Co^{58}	3.1	140
Fe^{59}	3.1	140
Co^{60}	10.0	450
Zn^{65}	179	8060
Zr^{95}-Nb^{95}	2.4	110
Sb^{124}	0.11	5.0
Ba^{140}	0.67	30

DISCUSSION

The transport and behavior pattern of the Columbia River radionuclides observed in this study provide a substantial background of basic knowledge and a basis for further study and interpretation of river phenomena. The behavior in the Columbia River of this large group of elements can serve as a basis in predicting the interaction of other elements in this and other river systems. The magnitude of the inventories of the various radionuclides and their seasonal variation has been establish

for the major stretch of the Columbia River. The observed uptake of the radionuclides by suspended particulate material and their depletion by deposition provide basic data which will require, for its detailed explanation, studies of the rates and mechanisms of sorption, desorption and biological assimilation of the radionuclides.

Since the basic behavior of this large group of radionuclides is well known, general behavior at other river locations or in other rivers could be inferred from studies of just a few of these radionuclides.

General questions which have not been answered by this study include the precise locations of radionuclide deposition on the river bottom, the position of redeposition of sediments which are resuspended during the spring freshet, and the behavior pattern in the river of the biologically important radionuclides such as P^{32} and Sr^{90}. Also, the mechanisms by which the radionuclide become associated with suspended particulate material, inorganic and organic, require detailed study.

R. W. Perkins
Jack L. Nelson
W. L. Haushild
May 17, 1965

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FIGURE 1

ITE MAP OF COLUMBIA RIVER FROM PRIEST RAPIDS TO MOUT

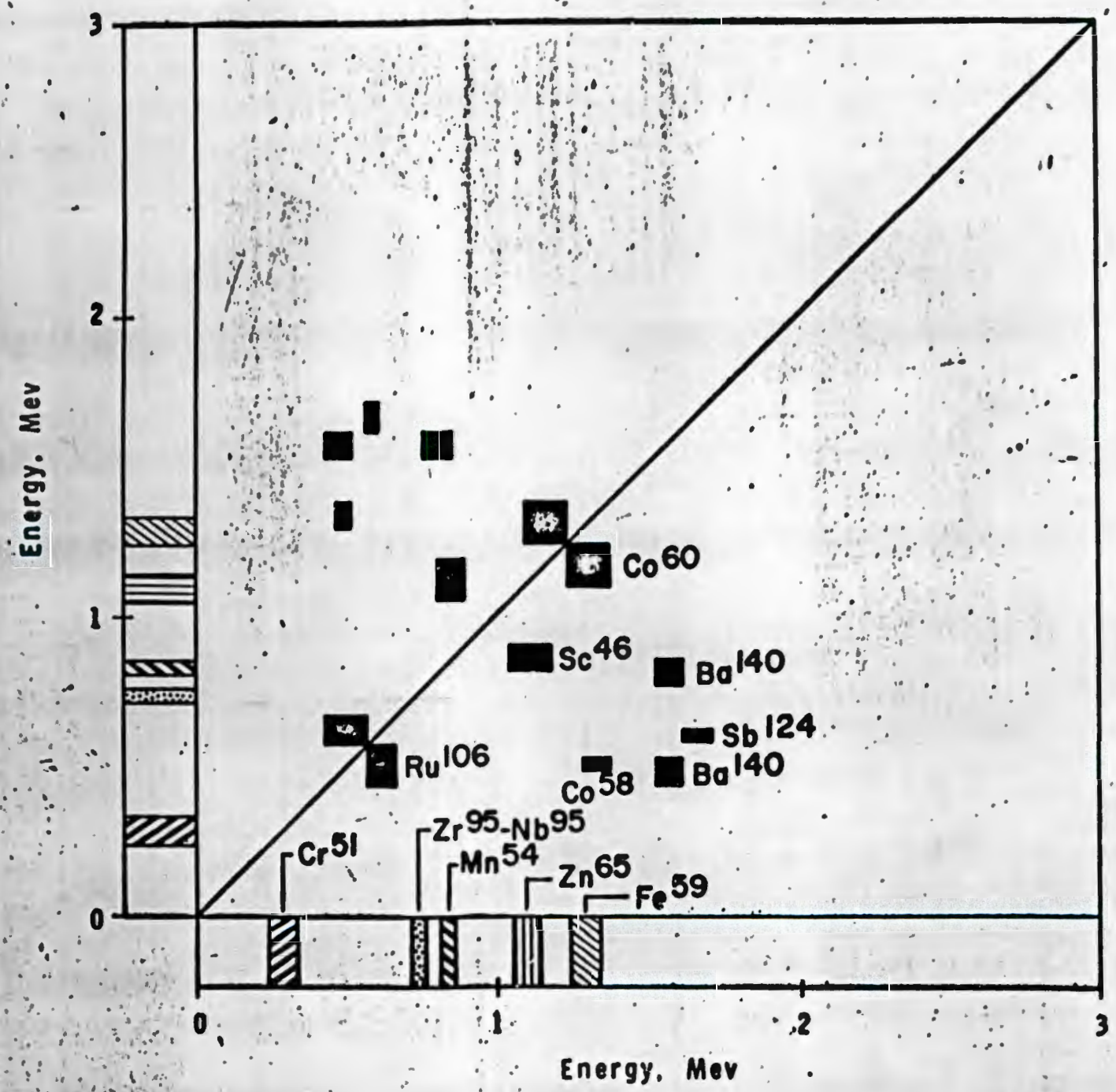


FIGURE 2

ENERGY AREAS USED IN THE DIRECT MEASUREMENT OF 11 RADIONUCLIDES IN COLUMBIA RIVER WATER

FIGURE 3
RADIONUCLIDE CONCENTRATION IN THE COLUMBIA RIVER AT PASCO, HOOD RIVER, AND VANCOUVER FOR THE PERIOD JANUARY 1964 THROUGH JANUARY 1965

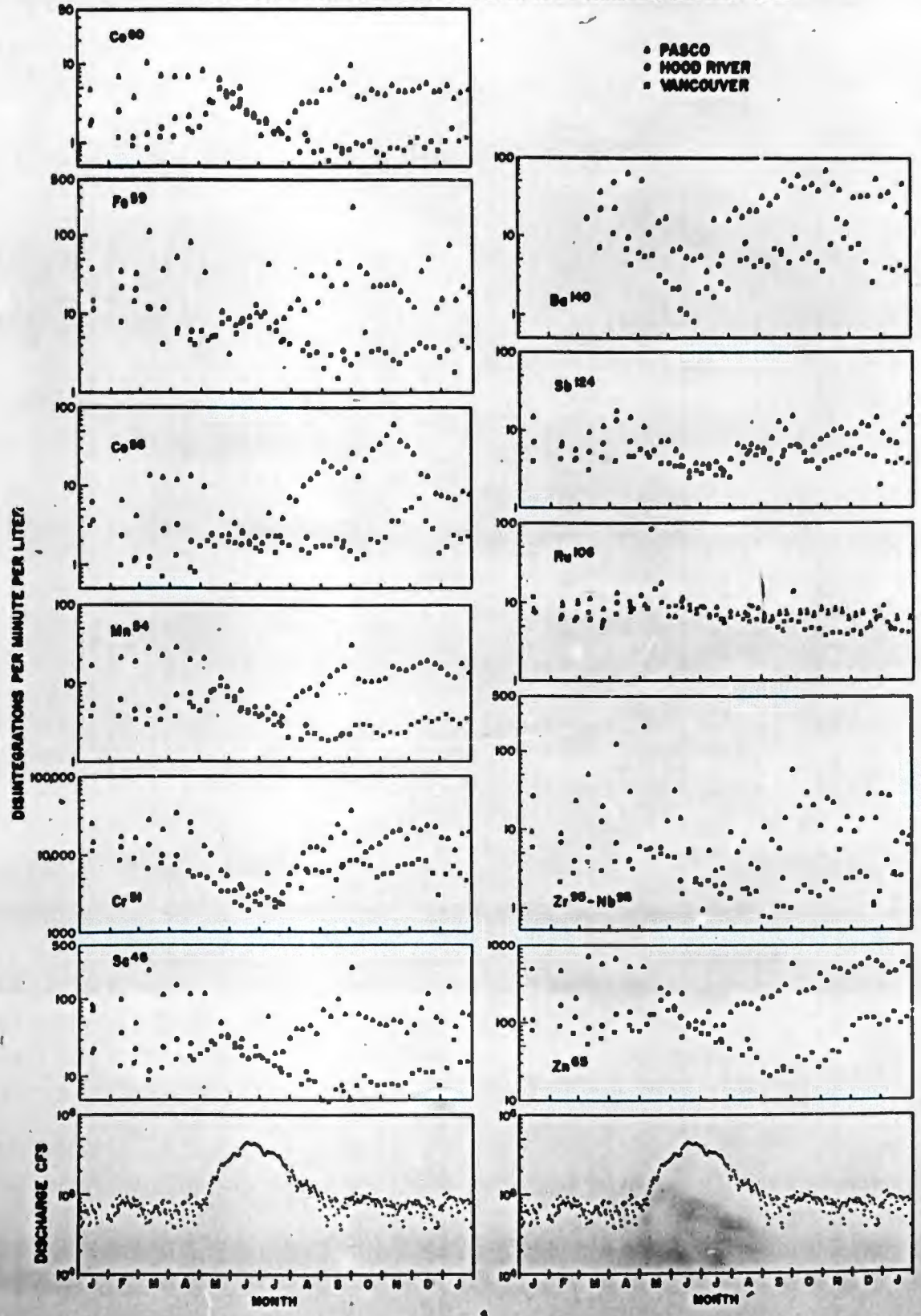


FIGURE 4

PHYSICAL FORM OF THE COLUMBIA RIVER RADIONUCLIDES AT PASCO, HOOD RIVER AND VANCOUVER DURING THE PERIOD JANUARY 1964 THROUGH JANUARY 1965

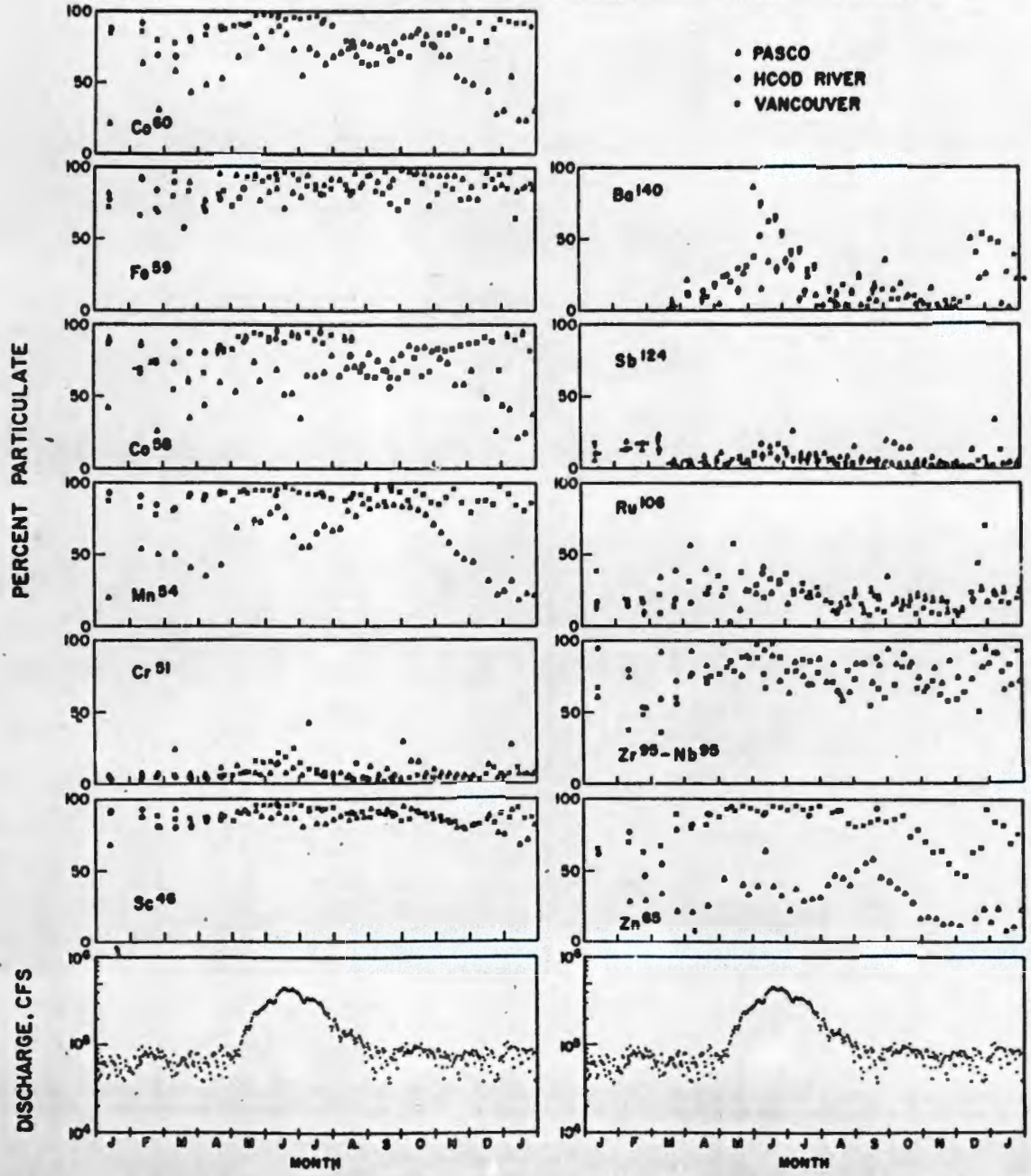


FIGURE 5

RATIOS OF THE CONCENTRATIONS OF COLUMBIA RIVER RADIONUCLIDES TO Cr^{51}, IN SOLUTION, AT PASCO, HOOD RIVER, AND VANCOUVER FOR THE PERIOD JANUARY 1964 THROUGH JANUARY 1965

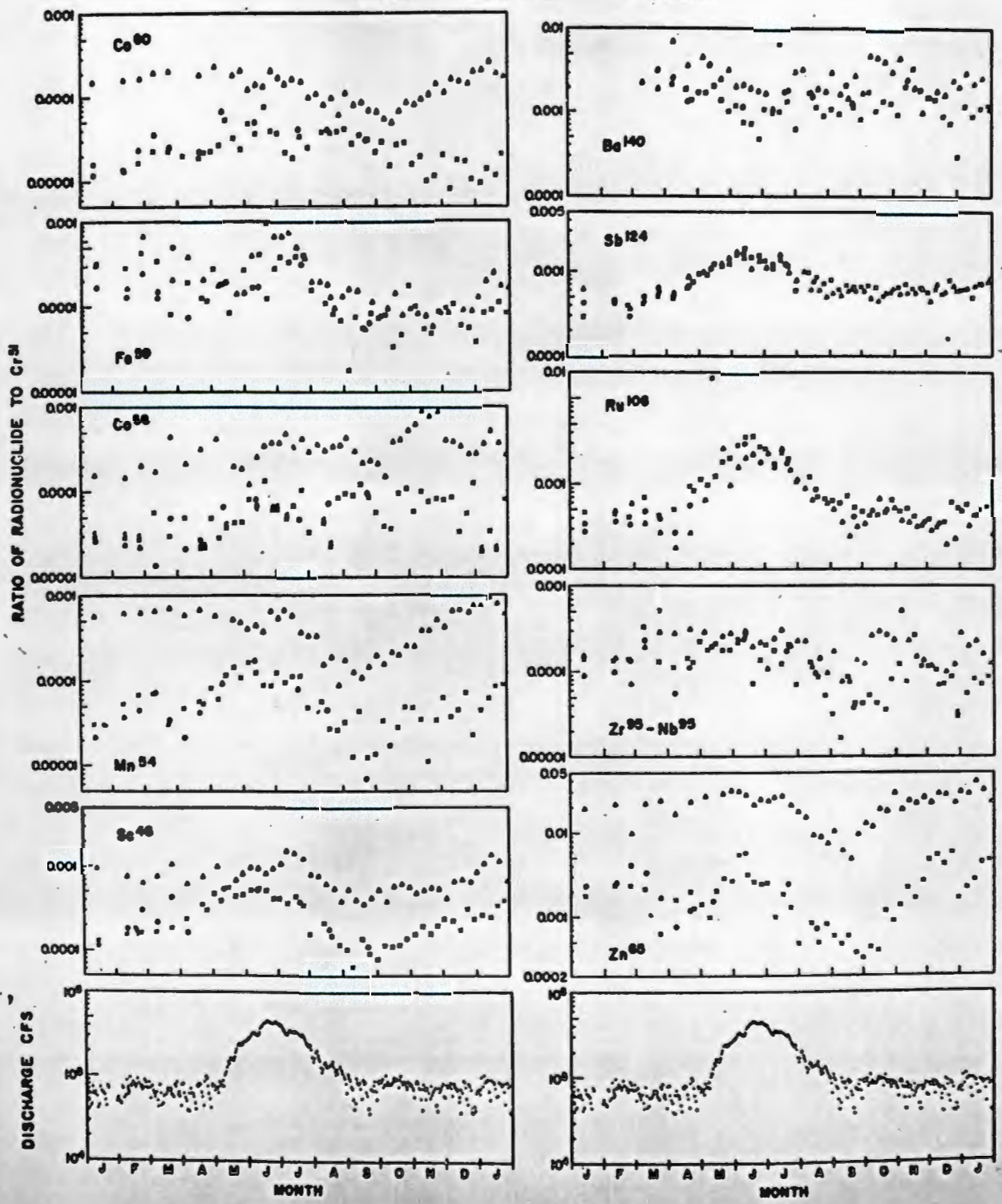


FIGURE 6

RATIO OF THE TOTAL (SOLUTION PLUS PARTICULATE) COLUMBIA RIVER RADIONUCLIDE CONCENTRATION TO THAT OF Cr^{51} AT PASCO, HOOD RIVER AND VANCOUVER FOR THE PERIOD JANUARY 1964 THROUGH JANUARY 1965

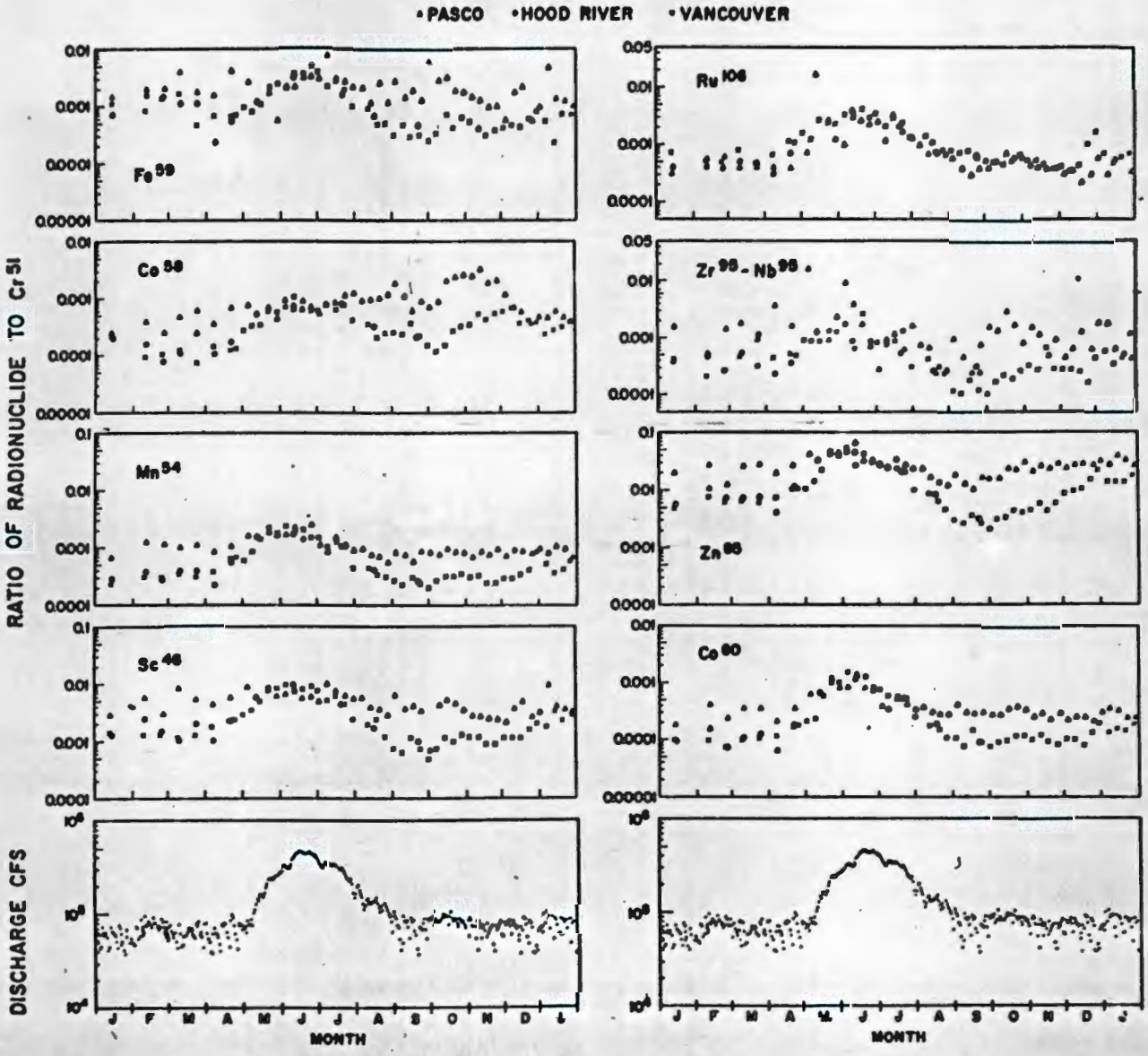


FIGURE 7

TRANSPORT OF RADIONUCLIDES BY THE COLUMBIA RIVER AT PASCO, WASHINGTON, HOOD RIVER, OREGON AND VANCOUVER, WASHINGTON FOR THE PERIOD JANUARY 1964 THROUGH JANUARY 1965

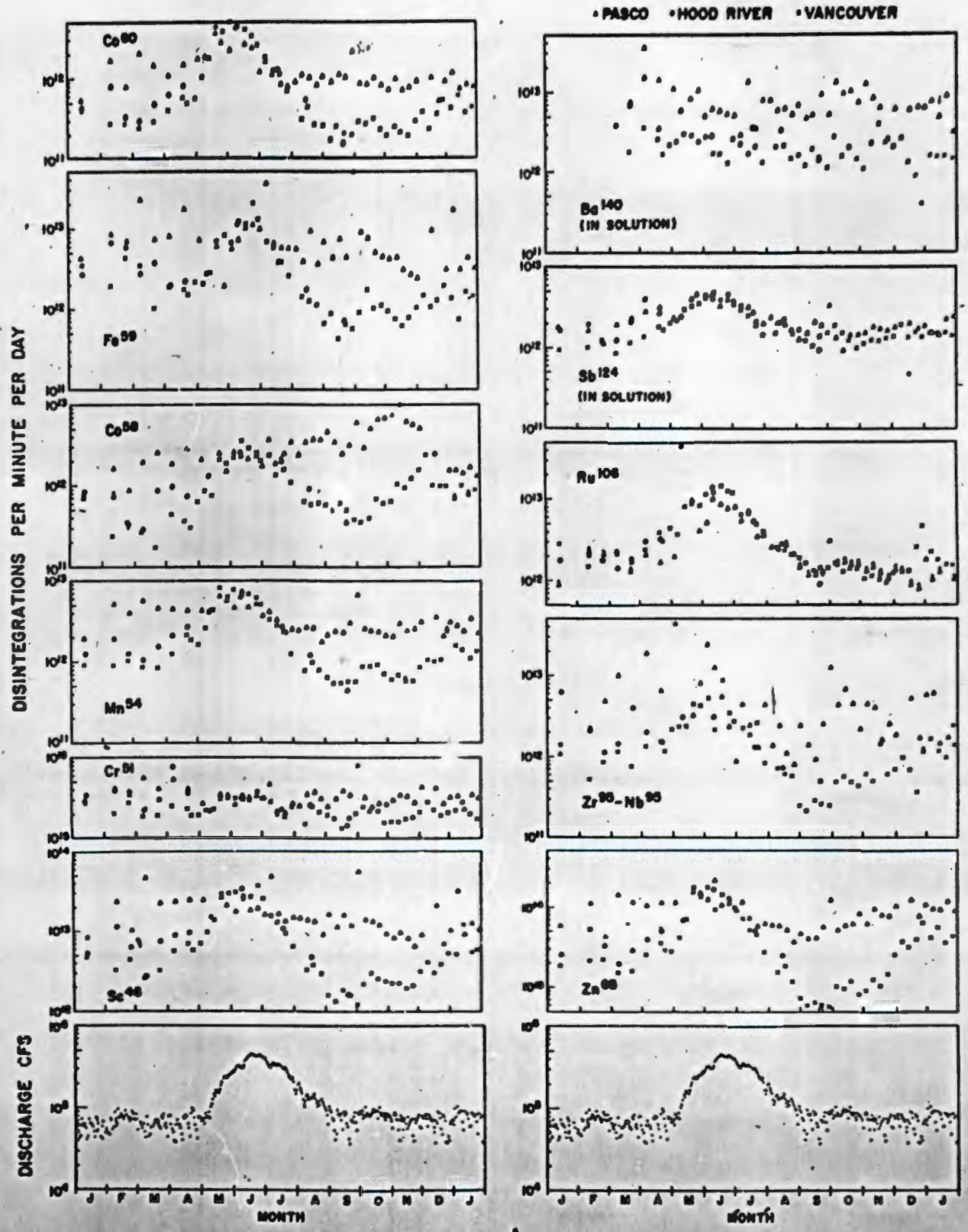
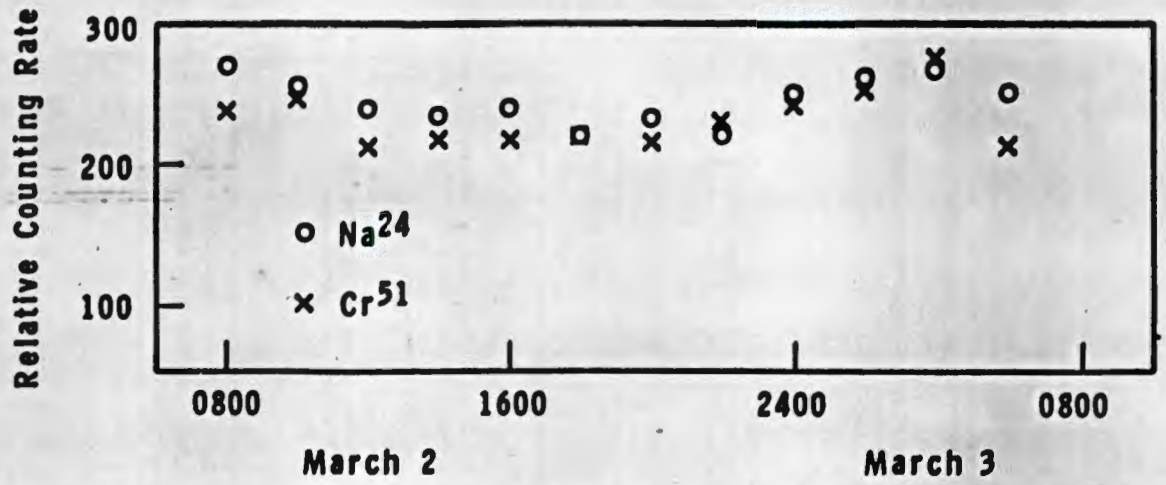


FIGURE 8

DIURNAL VARIATION IN COLUMBIA RIVER DISCHARGE AND RADIONUCLIDE CONCENTRATIONS



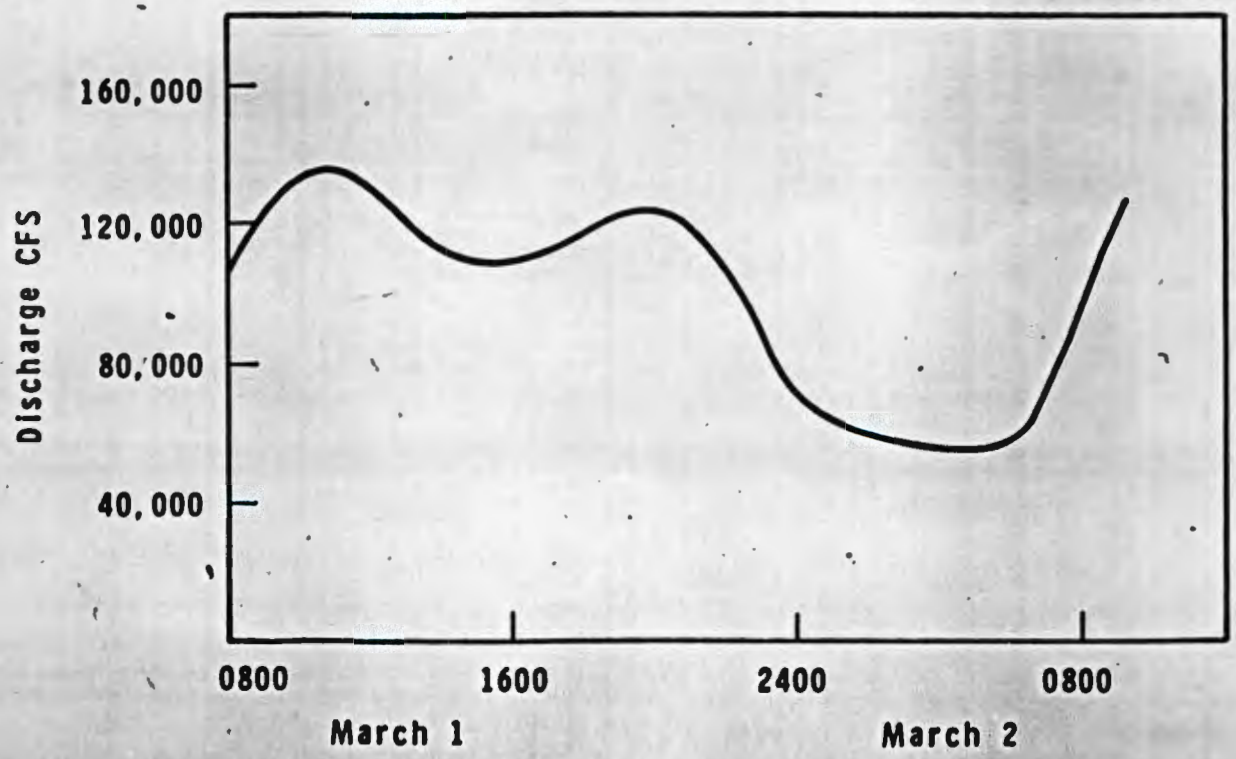


FIGURE 9

DEPLETION OF COLUMBIA RIVER RADIONUCLIDES BETWEEN PASCO AND VANCOUVER DURING THE PERIOD JANUARY 1964 THROUGH JANUARY 1965

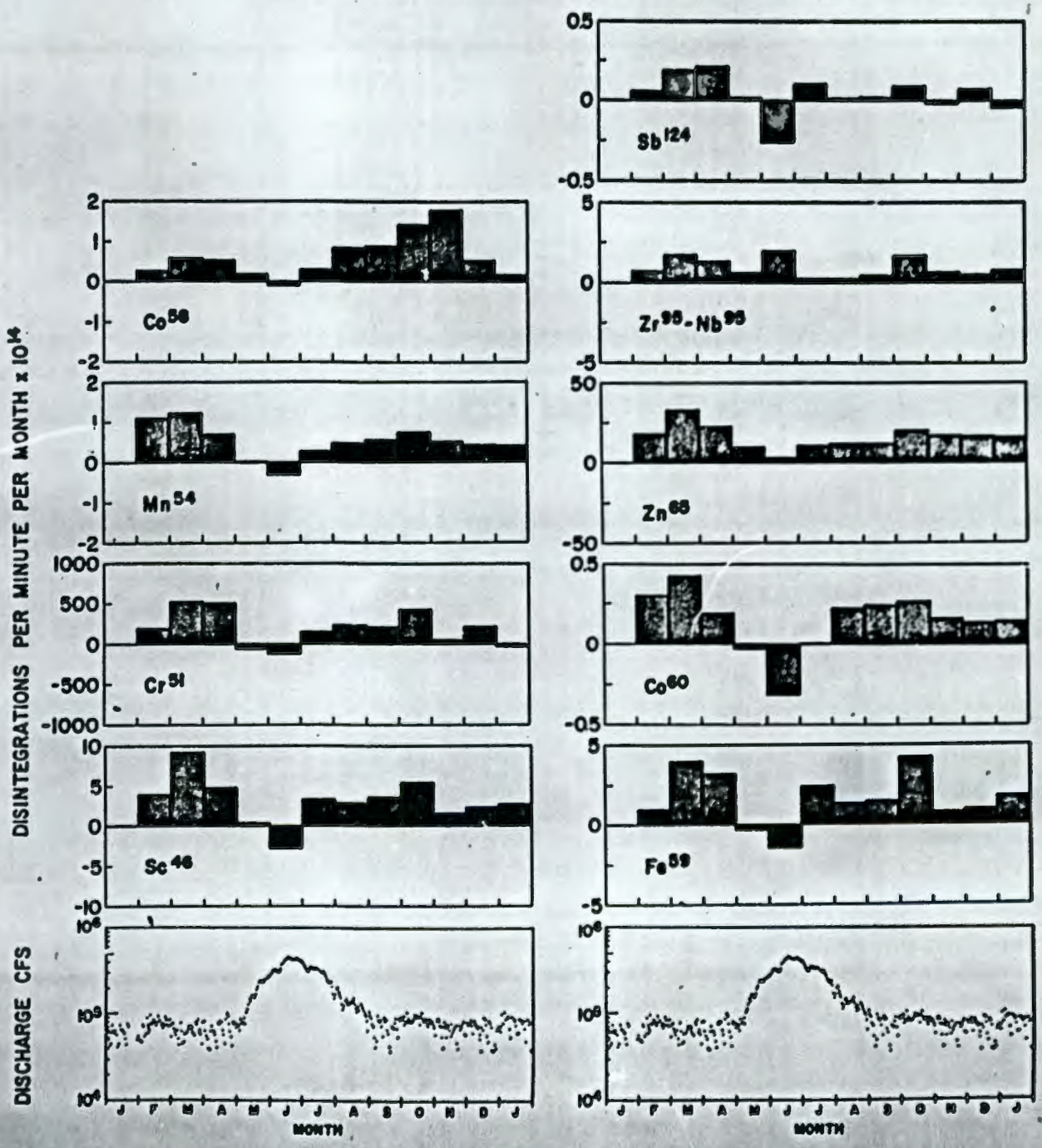
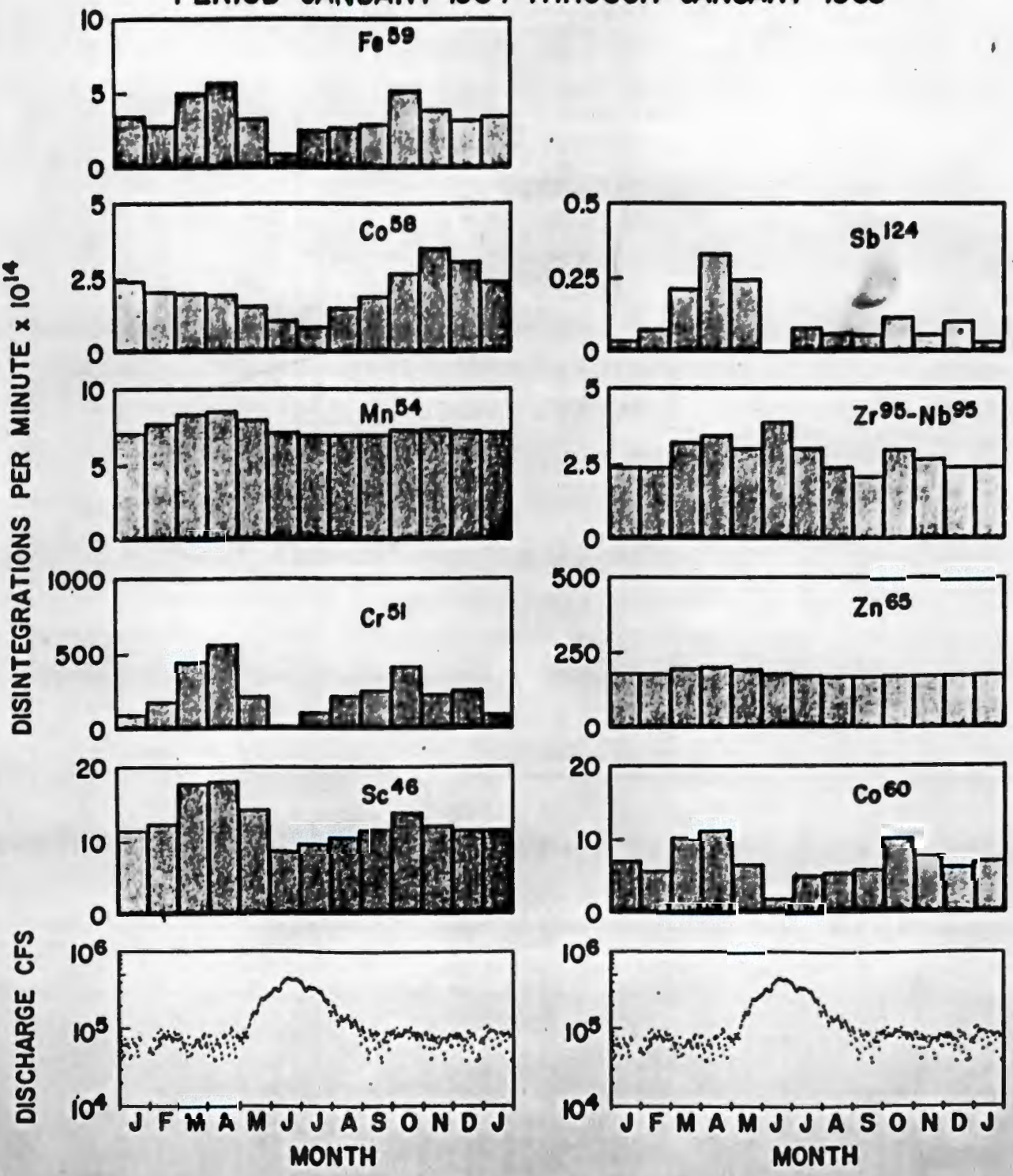


FIGURE 10

RADIONUCLIDE INVENTORY IN COLUMBIA RIVER SEDIMENTS BETWEEN PASCO AND VANCOUVER FOR THE PERIOD JANUARY 1964 THROUGH JANUARY 1965