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Distribution Coefficients for Radionuclides in Aquatic Environments

II. Studies on Marine and Freshwater Sediments Systems Including the Radionuclides 106-Ru, 137-Cs, and 241 Am

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DISTRIBUTION COEFFICIENTS FOR RADIONUCLIDES IN AQUATIC ENVIRONMENTS.

II. Studies on Marine and Freshwater Sediment Systems Including the Radionuclides 106-Ru, 137-Cs and 241-Am.

Second Annual Research Progress Report

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ABSTRACT

The objective of this study was to obtain information that can be used to predict the fate of radionuclides that may enter the aquatic environment from nuclear power plants or fuel reprocessing plants. Constant shaking experiments were used to determine distribution coefficients for pure clay and natural sediments. Dialysis experiments were used to study the distribution of radionuclides among suspended sediments, phytoplankton, organic detritus, and filtered seawater. The value of distribution coefficients varies considerably from one environment to another and for different experimental conditions such as pH. These differences probably result from the presence of different physico-chemical states of the radionuclides. Studies on the uptake of radionuclides and phytoplankton indicated that the time required to reach equilibrium may be longer, greater than 100 hours, than previously assumed. Some radionuclides (e.g., ^{241}Am) have an especially high affinity for organic detritus. Additional experiments to compliment the results of this study were identified.

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DISTRIBUTION COEFFICIENTS FOR TRANSURANIC ELEMENTS IN AQUATIC ENVIRONMENTS.

2. Studies on Marine and Freshwater Sediment Systems Including the Radionuclides 106-Ru, 137-Cs and 241-Am.

1.0 INTRODUCTION

This second annual progress report presents results of our investigations on the distribution coefficients of radionuclides in aquatic environments. It covers the tenth through the twenty-first month of a proposed 3-year program and provides the foundation for the third year's proposal. The principal sections are Introduction, Materials and Methods, Results and Discussion, and summary.

The program has been supported by contract AT(49-1)-0352 in the amount of $101,821 for the period 1 August 1977 to 31 July 1978.

1.1 Background

The distribution, transport, and eventual fate of radionuclides in aquatic environments is affected by numerous physical, chemical, and biological processes including the adsorption of radionuclides to suspended particulate matter in the water column. The relative affinity of radionuclides for sorption onto particulates can be described by use of a distribution coefficient, K_d*, a ratio that compares the quantity of radionuclide bound to particulate matter to the quantity in solution.

In August 1976 we began to investigate adsorption and desorption of ^{237}Pu and ^{241}Am by sediments in freshwater and marine water-sediment systems. During the first year experimental procedures were developed to determine the K_d values using constant shaking and thin layer techniques as described in the 1976-1977 Annual Progress Report (May, 1977). Distribution coefficients of ^{237}Pu and ^{241}Am were determined for one marine, one freshwater and one anoxic marine water-sediment system. We also began dialysis experiments to study the affinity of radionuclides for phytoplankton and suspended sediments. These studies were designed to obtain information on factors that control the distribution of radionuclides between water and particulate matter, since the fate of radionuclides in aquatic environments depends upon partitioning between soluble and particulate phases.

$$*K_d = \frac{\text{concentration of bound radionuclides in dry sediment (dpm/g)}}{\text{concentration of radionuclides in water (dpm/ml)}}$$

The first-year studies showed that the K_d values may vary considerably for different water-sediment systems. Therefore, we cannot extrapolate K_d values from one environment to another, without knowing more about the comparisons between systems. Rather, it is necessary to acquire information from a variety of different environments and determine which parameters are responsible for the variations in K_d values.

1.2 Objectives

The objective of this program is to obtain new and better information for predicting the fate of specific radionuclides in aquatic environments through their interactions in different sediment-water systems. As stated in the 1977-1978 proposal, specific objectives were to: 1) extend the distribution studies to include ^{244}Cm and to include sediments from non-local sources, and 2) extend the dialysis experiments to include organic detritus as well as phytoplankton and suspended sediments. We proposed originally to determine K_d values of ^{244}Cm in addition to ^{241}Am, and to investigate sediment-water systems from the Great Lakes, the Savannah River reservation, and estuaries of the Skagit and Hudson Rivers. However, as we began the contract year, the Nuclear Regulatory Commission requested that we also determine K_d values for ^{90}Sr, ^{106}Ru and ^{137}Cs in these systems and that experiments be conducted with water and sediment from Cattaraugus and Buttermilk Creeks in New York State and from the Clinch River in Tennessee rather than from the Savannah River reservation.

We proposed to expand the dialysis experiments by using two additional dialysis sacs. One sac with filtered seawater served as a blank to determine the effect of dialysis *per se*. The other sac was inoculated with organic detritus which was prepared in our laboratory from *Zostera* sp., a common macrophyte in Puget Sound. A materials balance of the radionuclides in the dialysis experiments was proposed to determine the extent of adsorption of radionuclides to the walls of the container and to the dialysis membrane.

During the first year of the program we evaluated K_d values under equilibrium conditions. In the past year, we have also investigated the rate that radionuclides approach equilibrium in both constant shaking and dialysis experiments. Thus we can evaluate the kinetics of radionuclide transfer between particulate and aqueous phases.

1.3 Personnel

From 1 August 1977 until 31 January 1978, Drs. A. H. Seymour and W. R. Schell served as co-principal investigators. Dr. Schell was named principal investigator as of 1 February 1978. Dr. A. Nevissi, radiochemist, and Mr. A. Sanchez, a Ph.D. candidate, have worked on the project since 1 August 1976.

Michael Brown, Willis McConnaha, and John Clayton have served as research associates at different times during the contract. In February 1978, Dr. T. Sibley was hired as a post-doctoral research associate. The Scientific Programmer, Engineering Technician, and research and laboratory assistants have been utilized as required for preparation and analysis of samples, and data reduction.

2.0 MATERIALS AND METHODS

Sediment and water samples from a variety of natural environments have been used to investigate, in the laboratory, the distribution of radionuclides among water, suspended sediments, phytoplankton, and organic detritus. The experimental methods used for the determination of K_d values and for dialysis experiments were adapted from the works of Duursma and Bosch (1) and Dawson and Duursma (2).

2.1 Sources and Types of Samples

During the past year we have collected and prepared large quantities of "standard" sediment and water samples from three locations: anoxic, marine sediments from Saanich Inlet, Vancouver Island, British Columbia, Canada; marine sediments from the Skagit River estuary in Puget Sound; and samples of organic-rich sediments from Sinclair Inlet in Puget Sound. All sediment samples were obtained with a coring device and the cores were immediately capped for transportation to the laboratory. Deep-water samples were collected with a Battelle Large Volume Water Sampler and filtered through 0.45 μm Millipore filters. Both water and sediment samples were stored at 4°C.

With the cooperation of other laboratories, Oak Ridge National Laboratory (ONL), Argonne National Laboratory (ANL), Battelle Northwest Laboratories (BNL), and Lamont-Doherty Geological Laboratory (LDGL), we have also obtained freshwater samples and sediments from three non-local locations: the Clinch River, Tennessee, approximately 1 1/2 miles downstream from the site of the proposed breeder reactor; one from near the east shore of Lake Michigan from a depth of 67 m; and several samples from Cattaraugus Creek, New York. Samples from the Hudson River Estuary, New York, provided by LDGL, arrived in early June 1978. Information about these samples is given in Tables 1 and 2.

In addition to the natural sediment samples, we have also obtained pure clay minerals from the Georgia Kaolin Company for use in our experiments. Specific clay minerals which have been received are shown in Table 3.

2.2 Determination of Distribution Coefficients in Laboratory Water-Sediment Systems

Duursma and Bosch (1) described three different experimental approaches for the determination of distribution coefficients, namely: thin-layer technique, sedimentation technique, and suspension technique. Each of these techniques and the extent to which we have utilized them is described below.

Thin-layer technique consists of depositing sediment on filter paper, submerging the sediment and filter into a solution of radionuclides

Table 1. Location of seawater and marine sediment samples used in distribution coefficient studies.

SAMPLE TYPE	TEMP °C	SALINITY °/oo	DEPTH m	SEDIMENT DEPTH m	REMARKS
			MARINE SAMPLES		
Saanich Inlet					
Station 48° 35.4' N 123° 30.2' W	8.9	31.243	224	225	Anoxic sediment and water
Skagit River Estuary					
Station 1 48° 18.0' 122° 29.0'	10	29.62	2	9	Sandy sediment
Station 2 48° 17.5' 122° 29.5'	10	30.25		30	Sandy sediment
Station 3 48° 17.0' 122° 30.0'	10	30.25		19	Silt and clay
Station 4 48° 15.5' 122° 32.5'	10	31.05		59	Silt and clay
Sinclair Inlet					
Station 1	9.6	28.91	30	31	Oxic organic rich sediment
Station 2	9.5	28.89	14.5	15.1	Oxic organic rich sediment
Station 3	9.5	28.81	3.5	4.5	Oxic organic rich sediment

Table 2. Location of sediment and freshwater samples used for distribution coefficient studies.

SAMPLE TYPE	LOCATION	REMARKS
Cattaraugus Creek		
BC-4	Thomas	Samples collected by BNL-group
CC-2	Felton Bridge	Samples collected by BNL-group
CC-11	Mouth of Cattaraugus Creek	Samples collected by BNL-group
Lake Michigan		
Station 5	Near the east shore 67 m deep	Sample collected by ANL
Clinch River		
Station 1	1.5 miles downstream from the breeder reactor site. 3 m deep	Sample collected with cooperation of ONL
Hudson River		
Station		Samples collected by LDGL
Sediments SLOSH*III	mp# 43	June 11, 1975
SLOSH II	mp 18	June 2, 1975
SLOSH V	mp 0	September 15, 1975
Water	mp 0	15°/oo salinity
	mp 18	2-3°/oo salinity
	mp 40	Fresh water

* Sediments are internal standards from Lamont-Doherty Geological Laboratory.

mp represents miles upstream from the southern tip of Manhattan.

Table 3. Clay minerals for distribution coefficient studies

Commercial name	Type of clay	Median particle size (Microns)	Remarks
Hydrite UF	pure kaolin	0.20	
Hydrite R	pure kaolin	0.77	
Hydrite Flat D	pure kaolin	4.5	
Gelwhite L	calcium montmorillonite		inorganic colloid
Mineral Colloid BP	sodium montmorillonite		inorganic colloid
Volclay	bentonite		from American Colloid Co.

and determining the accumulation of radionuclides by the sediments. In 1976-1977, we conducted a few experiments to evaluate this technique. The results of these experiments indicated that during sorption, agitation of the water while sampling may cause significant variations in the data due to loss of sediment from the filter. We believe this technique provides less reliable results than other techniques.

Sedimentation technique includes addition of radionuclides to filtered seawater and aging until equilibrium is reached between stable and radioactive isotopes. Then 1-ml samples are removed at specified time intervals (generally 1 day) for radioactive measurements and replaced with 1 ml of a stock sediment suspension. As the sediment settles, radionuclides are sorbed by the particles and removed from solution. The procedure of sampling 1 ml of suspension and replacing with 1 ml of stock sediment solution is continued daily until there is a constant reduction in the percent of radionuclides removed with each addition of sediment.

Suspension (constant shaking) technique has been used for the majority of our experiments. Briefly, the method consists of:

1. adding radionuclides to a filtered water sample, adjusting the pH with a NaOH or HCl solution, and aging to obtain equilibrium among the different physico-chemical species. Aging requires at least one week.

2. adding a known concentration of sediments to a specified volume of the spiked water sample.

3. shaking or stirring the samples at a constant temperature.

4. removing a sample aliquot, filtering, and measuring the concentration of radionuclides retained on the filter paper and in the filtrate.

Controls consisting of spiked water without sediments were processed and analyzed with the samples.

2.3 Dialysis Experiments

Figure 1 is a schematic representation of our dialysis apparatus. The equipment consists of a 1000-ml beaker containing a spiked, filtered water sample. Dialysis sacs, containing 50-70 ml of the same filtered water without the radionuclides, are then submerged in the outside compartment. One dialysis sac contains only filtered water and serves as a control while the other sacs contain known quantities of phytoplankton, sediment, or detritus. The outer compartment is mixed with a magnetic stirrer and the contents of the dialysis sacs are stirred and aerated with a glass stirring rod connected to a small electric motor. Dialysis membranes permeable to molecules smaller than 6000-8000 molecular weight have been used for all the experiments.

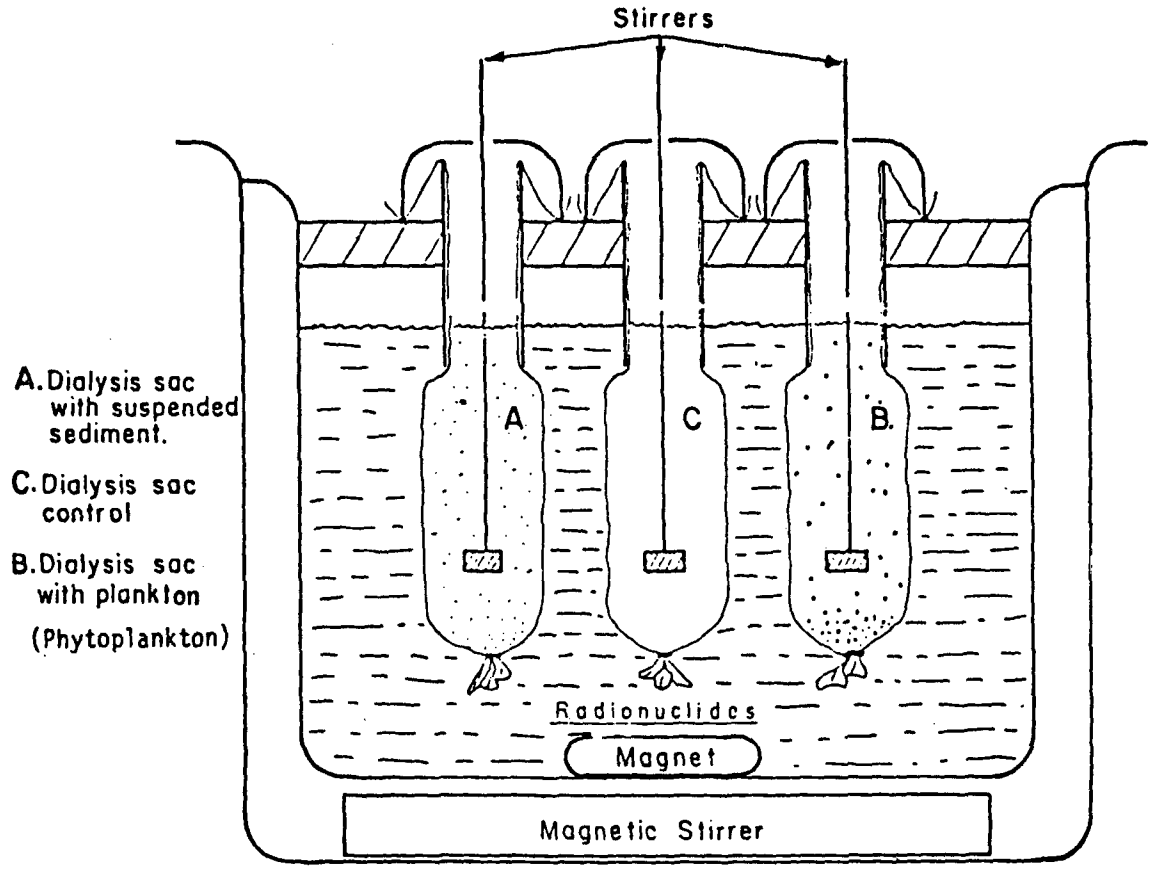

Figure 1. Schematic diagram of apparatus used for dialysis experiments.

Samples are removed from the dialysis sacs and the outside chamber at predetermined times and filtered through Millipore or Nucleopore membrane filters. The filter and a portion of the filtrate are then measured for radionuclides.

When the concentrations of radionuclides in the soluble and particulate phases are calculated, corrections must be made for the radioactivity on the filter that represents soluble radionuclides. We have shown that for ^{137}Cs and ^{60}Co as much as 85-90% of the activity retained on the filter may actually be sorbed from solution rathern than being present in the particulate phase.

2.4 Counting Techniques

The samples from distribution coefficient and dialysis experiments were placed into 1.3 x 5 cm standard sample holders (2 dram polyethylene vials) for gamma counting. The gamma-emitting radionuclides were measured using one of two calibrated Ge(Li) detectors, pulse-height and analyzers and computerized data reduction systems. We also used a new Ge(Li) detection system with a computer-based multi-channel analyzer which integrates the counts for each gamma energy peak, subtracts background and prints out gross and net counts. The resolution (FWHM) of this Ge(Li) detector is 1.88 kev and the efficiency is 14% relative to 3 x 3" NaI(Tl) detector. For the measurement of ^{241}Am and ^{237}Pu in samples, we have used a counting system consisting of a 2" NaI(Tl) well crystal and two single-channel analyzers. The efficiency of this system for these two radionuclides is 65-70%.

In 1976-1977, we used ^{237}Pu which has low energy x- and gamma-ray radiation. During 1977-1978, however, this isotope was not available and we used ^{238}Pu and $^{239,240}Pu$ in our experiments. Measurements of these alpha-emitting radionuclides required extensive radiochemical separation and alpha counting techniques. After gamma-counting, the samples which were designated for plutonium analysis were spiked with ^{242}Pu for chemical yield determination and the chemical separation and purification of plutonium was made. The plutonium was electroplated onto a stainless steel disc and measured with a surface barrier alpha-detector and pulse-height analyzer (3).

3.0 RESULTS AND DISCUSSION

In 1977-1978 distribution coefficients were obtained for sediment-water systems from the Skagit River estuary and Saanich Inlet. Also, we evaluated the sedimentation technique for determining K_d values and the effect of pH on the K_d value of ^{241}Am in a marine sediment-water system. Dialysis experiments were used to investigate the distribution of radionuclides among 1) phytoplankton, sediment, and filtered seawater from the Eastern Pacific, and 2) phytoplankton, sediments, organic detritus, and filtered water from the Skagit River estuary. Results from each of these studies are discussed separately below.

3.1 Evaluation of the Sedimentation Technique

The sedimentation technique was used to determine the K_d value for ^{239}Pu and ^{241}Am in a sterile marine system and the K_d value for ^{241}Am in a non-sterile marine system. The concentration of ^{241}Am in the non-sterile marine system is plotted against time in Fig. 2. Results from the first 7 days (before the addition of sediments) provide an estimate of the loss of radionuclides due to adsorption on the cylinder walls. Approximately 4-6% of ^{241}Am is removed by adsorption. In the sedimentation technique the distribution coefficient is determined by the extent of removal of radionuclides for each sediment addition; that is, from the slope of ^{241}Am concentration versus time for days 8 through 14. Higher distribution coefficients have steeper slopes. Graphs for the activity of ^{241}Am and ^{239}Pu in a sterile marine system are given in Figs. 3 and 4. Since the distribution coefficients are lower and the counting errors are relatively greater for ^{239}Pu the range of K_d values is greater than for ^{241}Am. However, replicate samples produce similar values for the K_d value of ^{239}Pu, $\simeq 3 \times 10^3$. Therefore, it appears that the results for ^{239}Pu are reliable despite the large range of values found for any given experiment.

The K_d value for ^{241}Am in the sterile marine system is approximately seven times higher than the K_d value in the non-sterile system. A similar trend between sterile and non-sterile systems was observed using the constant shaking technique (Annual Report 1976-1977).

Distribution coefficients for ^{241}Am and ^{239}Pu determined by the sedimentation technique are one or two orders of magnitude lower than the distribution coefficients determined by the constant shaking technique. Duursma and Bosch (1) pointed out that the K_d value is dependent upon the analytical technique used to determine the K_d. However, the results for a given technique are generally consistent. One reason for the lower K_d values using the sedimentation technique compared to the constant shaking technique may be the shorter contact time between the sediments and the radionuclides in solution. We would expect that K_d values from the constant shaking and sedimentation techniques would be more similar for radionuclides with rapid sorption rates such as ^{137}Cs.

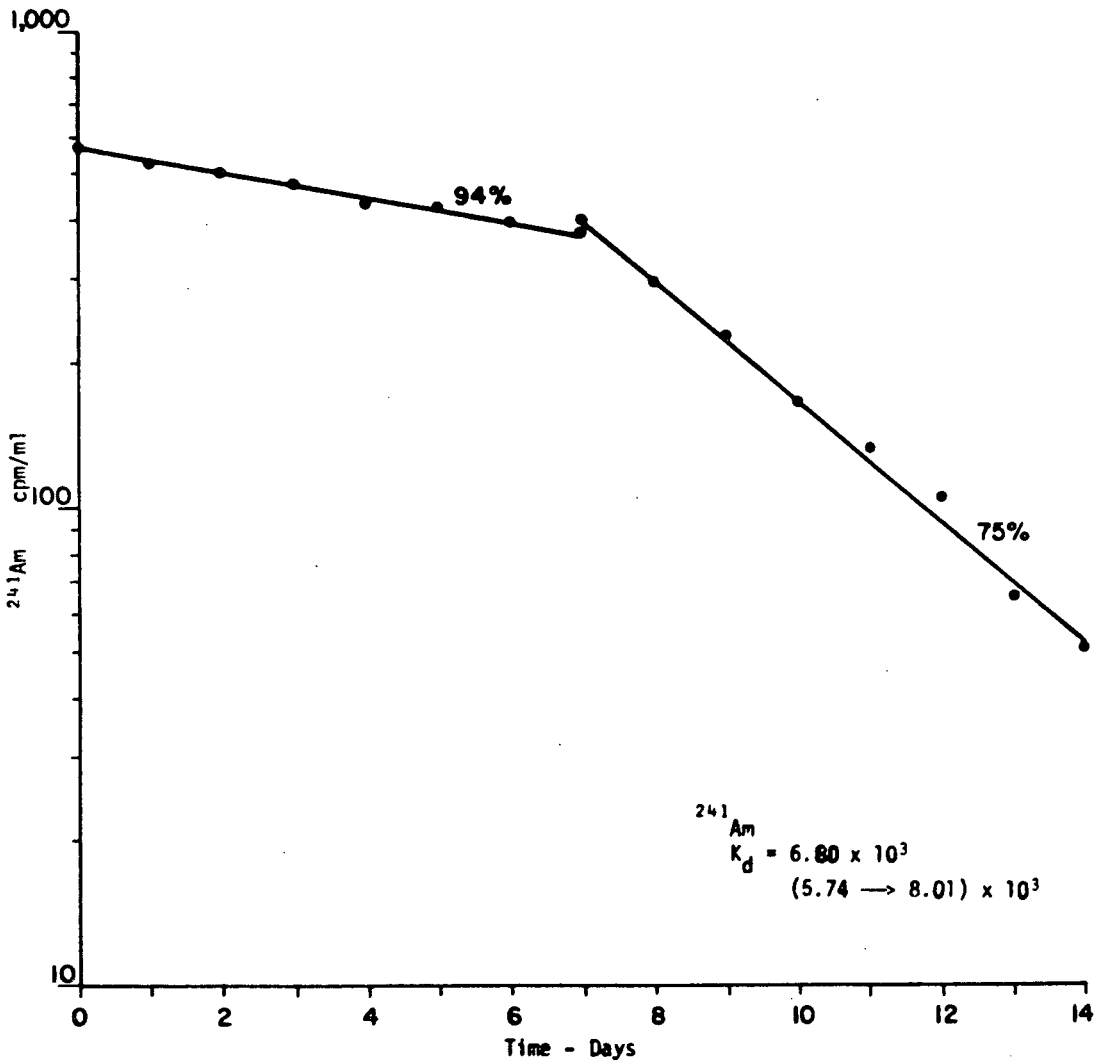

Figure 2: Results of sedimentation technique experiment for determining the distribution coefficient of ^{241}Am in a marine sediment-water system. One ml additions of a stock sediment suspension were added to the experiment each day beginning on day 7. The % values indicate the concentration of ^{241}Am remaining in solution on successive days. The range of K_d values, numbers in parentheses, was obtained by calculating K_d values for each sediment addition.

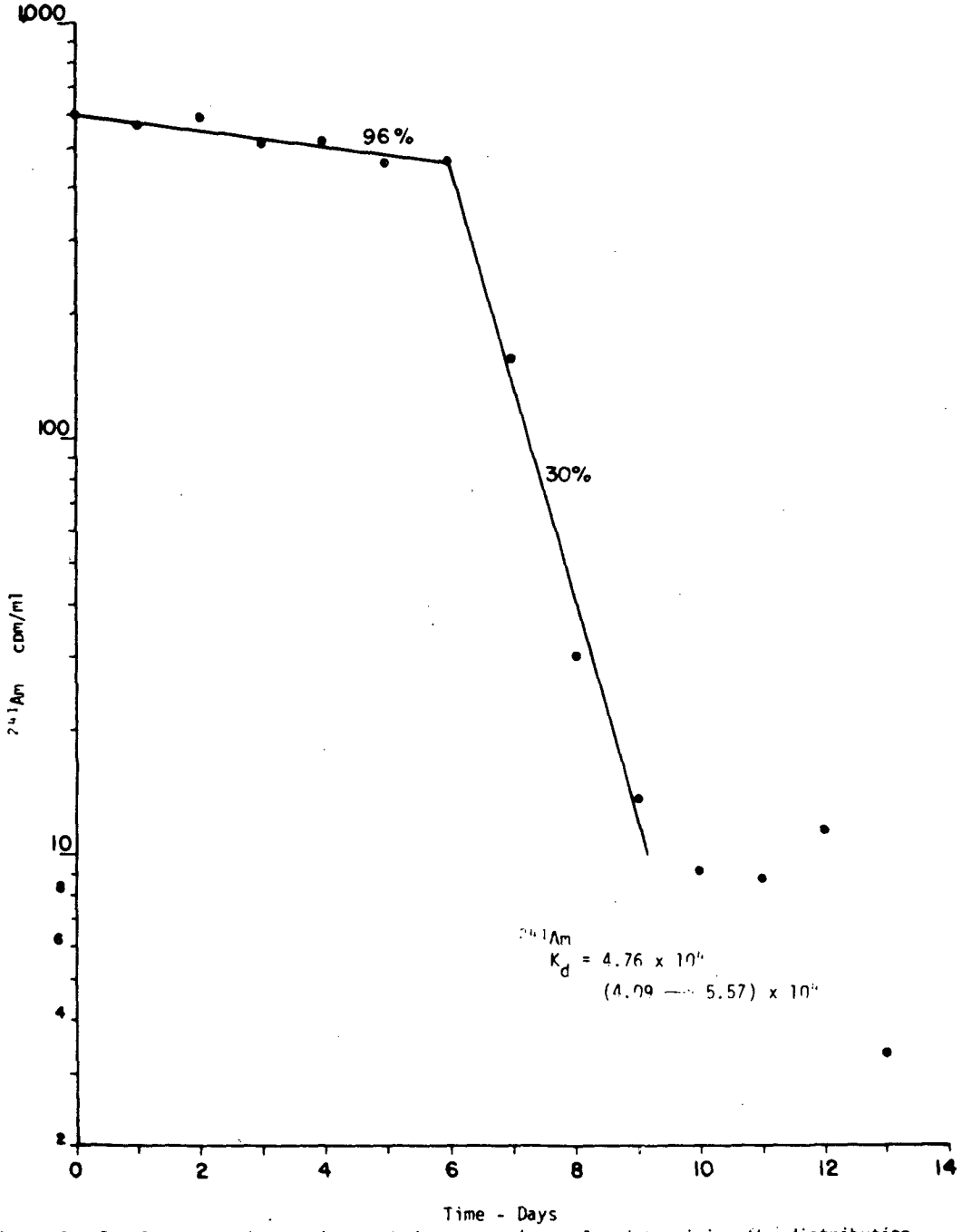

Figure 3: Results of sedimentation technique experiment for determining the distribution coefficient of ^{241}Am in a sterile marine sediment-water system. One ml additions of a stock sediment suspension were added to the experiments each day beginning on day 7. The % values indicate the concentration of ^{241}Am remaining in solution on successive days. The range of K_d values, numbers in parentheses, was obtained by calculating a K_d value for each sediment addition.

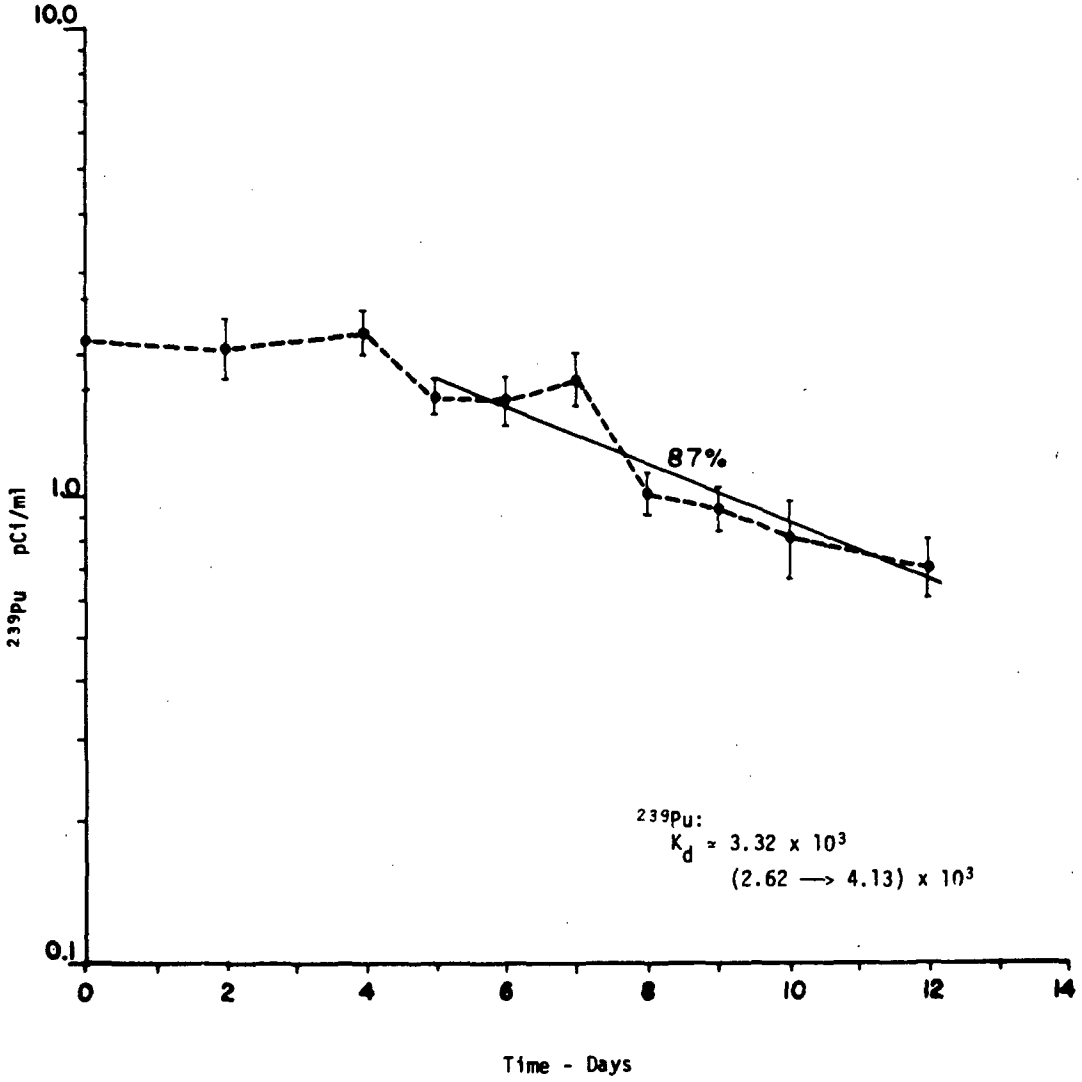

Figure 4: Results of sedimentation technique experiment for determining the distribution coefficient of ^{237}Pu in a sterile marine sediment-water system. One ml additions of a stock sediment suspension were added to the experiments each day beginning on day 7. The % values indicate the concentration of ^{241}Am remaining in solution on successive days. The range of K_d values, numbers in parentheses, was obtained by calculating a K_d value for each sediment addition.

Replicate experiments using the sedimentation technique gave consistent results for K_d values as shown in Table 4. This may be a preferred method of determining distribution coefficients for some sediment types; for example, coarse sediments which in the natural environment are only briefly suspended in the water column.

3.2 Evaluation of the Effects of pH on K_d Values

The effect of pH on the distribution coefficient of ^{241}Am was determined for sterile and non-sterile water-sediment systems for Lake Washington (freshwater) and Lake Nitinat (anoxic-marine). Values of the distribution coefficient were determined by using the constant shaking technique at 4°C for pH values ranging from 4 to 10. The pH was found to vary from 0.2 to 0.4 units during an experiment.

Figure 5 shows the relationship between K_d and pH for both the sterile and non-sterile Lake Washington systems. The values for the sterile system are greater than for the non-sterile system by a factor of at least 3.5. Both systems show a significant increase in K_d values between pH 4 and 6 but remain relatively constant at higher pH values. Starik and Ginzburg (4) reported that ^{241}Am exists in ionic form at low pH, but with an increase in pH, it hydrolyzes and forms a colloid. At pH values of 6.5-7.0, ^{241}Am should occur primarily in the colloidal form. However, colloidal particles are much smaller than the pore size (0.45 μm) of the filters used for filtration and should not be retained. Therefore, it appears that colloidal ^{241}Am is more readily adsorbed to the sediment particles than ionic ^{241}Am. The larger K_d values for the sterile freshwater system compared to the non-sterile system agree with results reported in the 1976-1977 Annual Report. However, when pH is controlled the differences between the sterile and non-sterile systems are much less. It may be that the differences reported earlier, result in part from pH differences between the sterile and non-sterile experiments. Changes in the physico-chemical state of the radionuclides with pH may be a major factor influencing K_d values.

Figure 6 shows the distribution coefficient of ^{241}Am as a function of pH for the anoxic Lake Nitinat sediment-water system. As with the freshwater system, K_d values increase between pH 4 and pH 10. At pH values of 4-7 the K_d values are greater for the sterile system than for the non-sterile system. However, in the pH range of 7-10 the K_d values for the sterile and non-sterile systems overlap.

The effect of pH is much greater in the marine system than in the freshwater system. Between pH 4 and pH 10 there is an increase in K_d values of nearly three orders of magnitude compared to one order of magnitude in the freshwater system. Differences between the freshwater and marine systems may result from the presence of 1) different soluble species of americium, 2) ionic strength effects on sorption, and 3) mineralogical

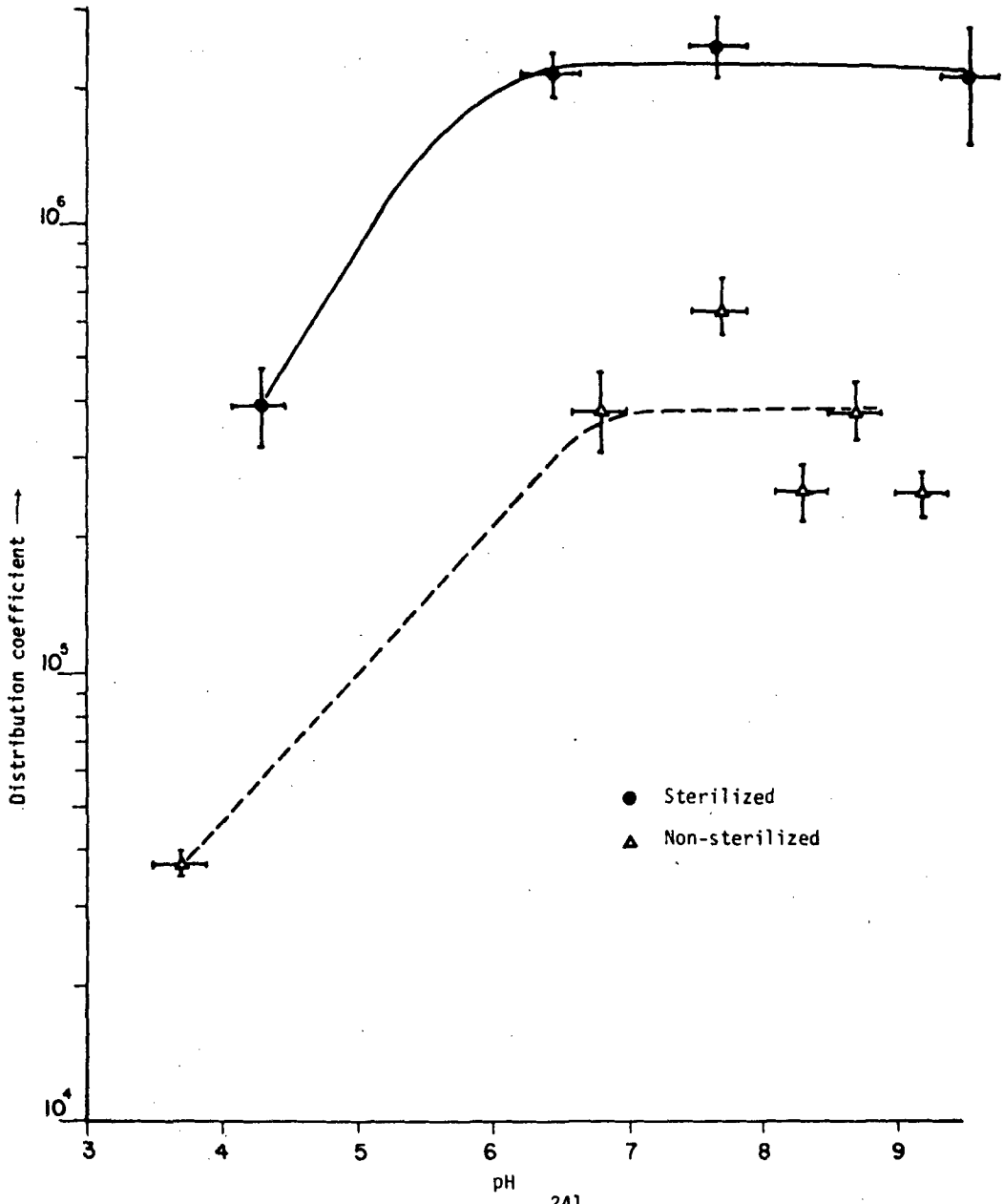

Figure 5. Distribution coefficient for ^{241}Am as a function of pH in sterilized and non-sterilized freshwater-sediment system. Horizontal bars indicate range in pH values and vertical bars indicate the range in K_d.

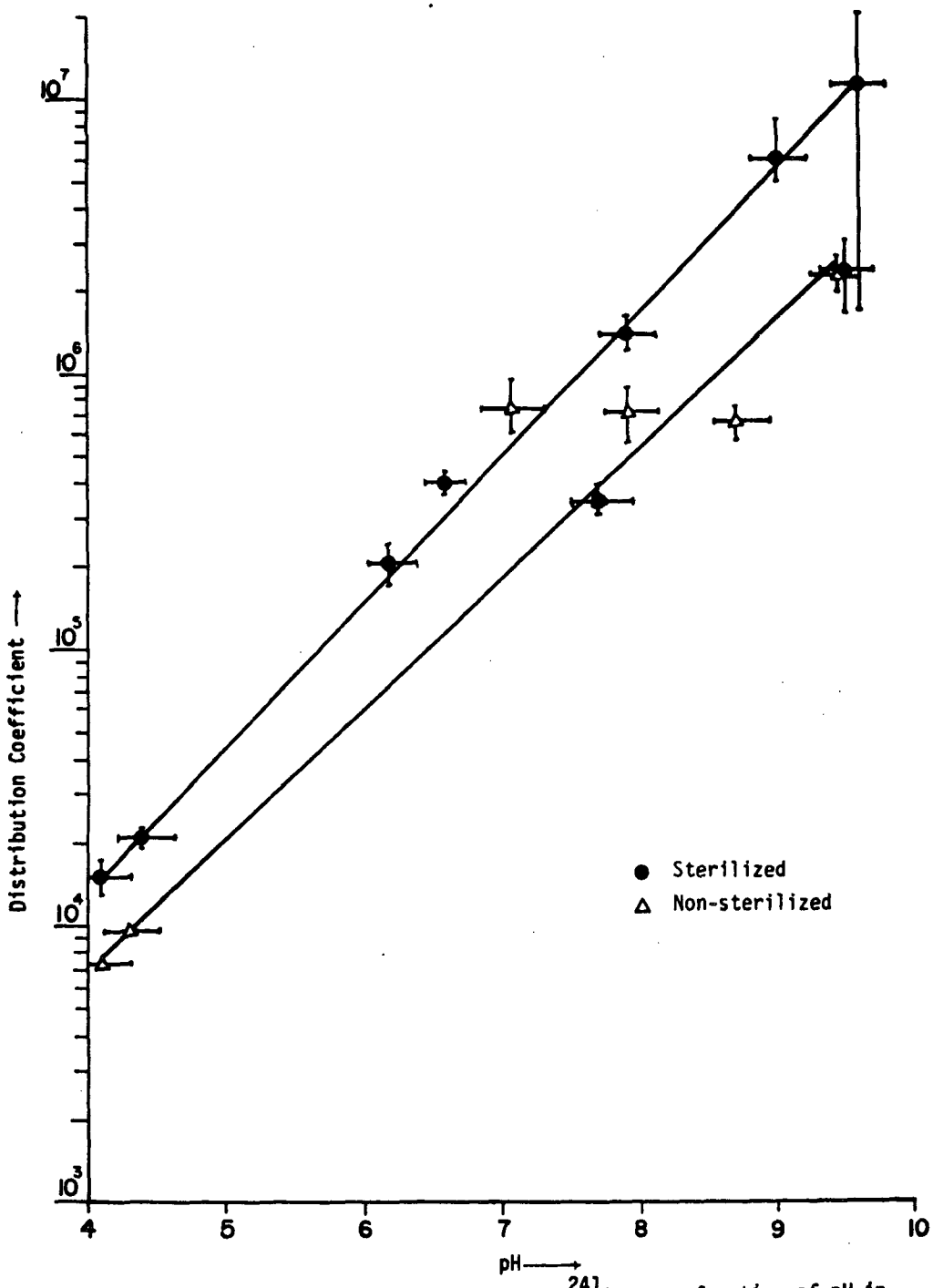

Figure 6. Distribution coefficients for ^{241}Am as a function of pH in sterilized and non-sterilized Lake Nitinat (marine) water-sediment system. Horizontal bars indicate the range in pH values and vertical bars indicate the range in K_d.

Table 4: Distribution coefficients for ^{241}Am and ^{237}Pu using the sedimentation technique for a water-sediment system from the Eastern Pacific Ocean. The range of values for each replicate indicates the results obtained by calculating a K_d value for each sediment addition. The average is the arithmetic average of all sampling times. For the sterile experiments the sediments and water samples were sterilized with a dose of 10 rads of gamma radiation.

		Distribution Coefficients	
Americium-241		Average	Range
Non-sterile	Replicate 1	6.44×10^3	$(5.42----7.61) \times 10^3$
	Replicate 2	6.80×10^3	$(5.74----8.01) \times 10^3$
Sterile	Replicate 1	4.76×10^4	$(4.09----5.57) \times 10^4$
	Replicate 2	4.34×10^4	$(3.73----5.06) \times 10^4$
Plutonium-237			
Sterile	Replicate 1	3.32×10^3	$(2.62----4.13) \times 10^3$
	Replicate 2	3.05×10^3	$(2.38----3.82) \times 10^3$

differences in different sediment types. We have proposed experiments for 1978-1979 to distinguish among these explanations.

3.3 Particle Formation During Constant-Shaking Experiments

Distribution coefficients generally are determined by filtering a solution of radionuclides, measuring the radioactivity on the filter and in the filtrate and calculating the K_d value. The radioactivity measured on the filter is assumed to be from radionuclides adsorbed to the suspended particulate matter. However, even in filtered seawater without sediments, ^{106}Ru and ^{241}Am occur primarily in the particulate phase.

Controls consisting of spiked seawater without sediments have been analyzed with our samples. Figure 7 shows the activity on the filters and in solution for a control used in the Skagit River estuary sediment-water experiment. There was a continuous increase in ^{106}Ru on the filter for the first 70-80 hours with a corresponding decrease of ^{106}Ru in solution. After about 80 hours there was a small simultaneous increase in ^{106}Ru for both the soluble and the particulate phases. The increase in activity after 80 hours probably results from desorption of ^{106}Ru from the walls of the experiment vessel. At the beginning of an experiment when the concentration of soluble ^{106}Ru is high, the radioisotope could adsorb to the container walls. The formation of particulate ^{106}Ru, however, will significantly reduce the concentration of dissolved species. Therefore, ^{106}Ru which had adsorbed to the container walls may desorb and distribute into both the soluble and particulate phases. In a second control from the Skagit River estuary ^{106}Ru in the particulate phase increased and the soluble phase decreased after 320 hours (15 days). This indicates that for some radionuclides the time necessary to reach steady state conditions may be significantly greater than the 24-40 hours that was assumed previously for ^{241}Am and ^{237}Pu. After the first 80 hours, the small changes in the concentration of soluble and particulate ^{106}Ru probably do not alter the K_d values significantly.

For ^{241}Am in salt water approximately 90% of the total radioactivity in the controls is retained on the filter. Furthermore, the conversion of soluble ^{241}Am in the spike to particulate ^{241}Am occurs quite rapidly. After only 1 hour 75-80% of the total ^{241}Am activity is present in the particulate phase. There is, however, a slight increase in the concentration of particulate ^{241}Am throughout the experiment. Figure 8 shows the percent of the ^{241}Am in the particulate phase as a function of time for two controls from the Skagit River estuary.

The exact physico-chemical state of particulate ^{106}Ru and ^{241}Am is uncertain at this time. Since the spiked seawater had been filtered, the particulates that are present must be forming during the experiment either by precipitation from solution or by aggregation of colloidal sized particles.

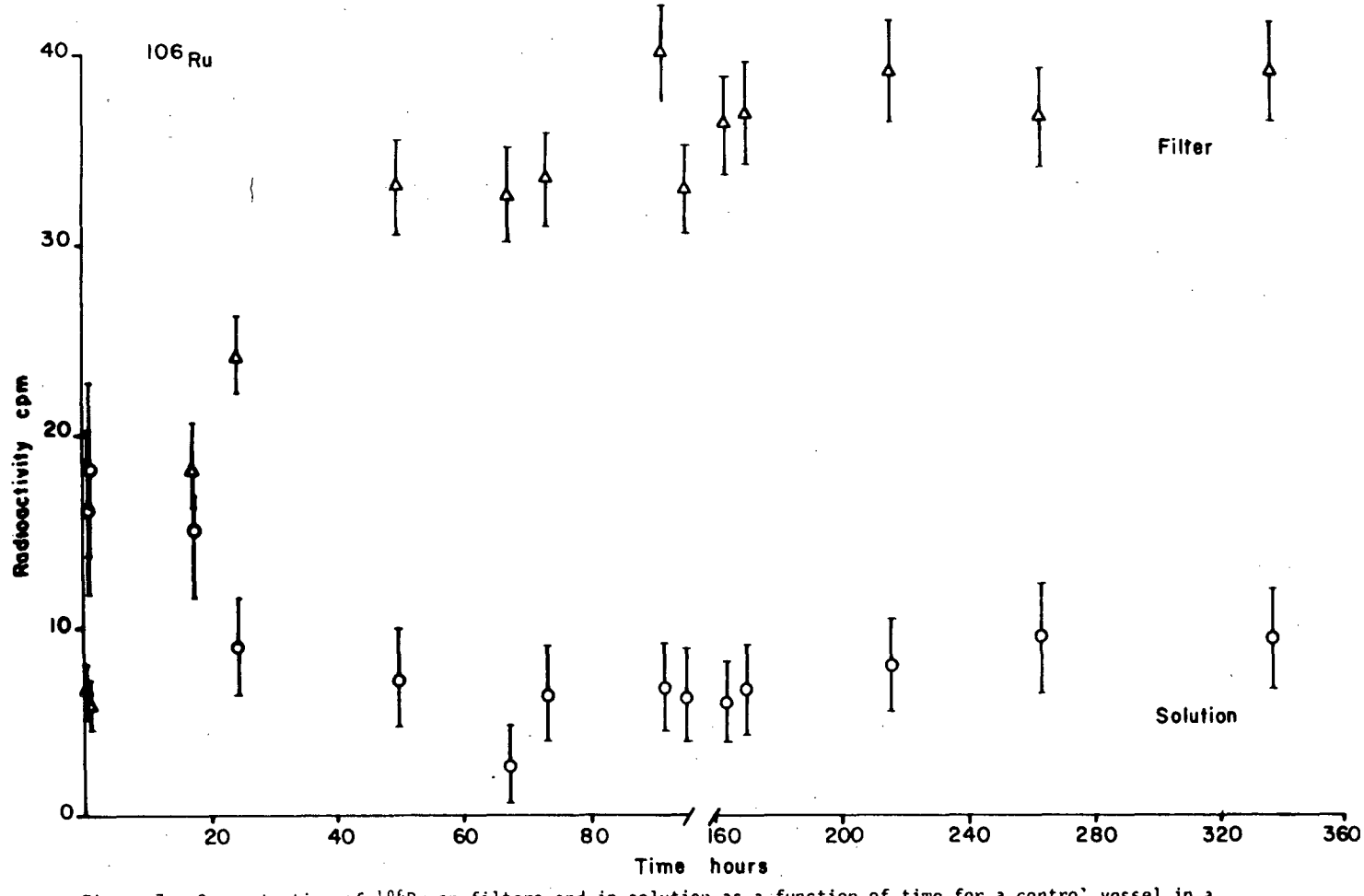

Figure 7. Concentration of ^{106}Ru on filters and in solution as a function of time for a control vessel in a constant shaking experiment from the Skagit River estuary.

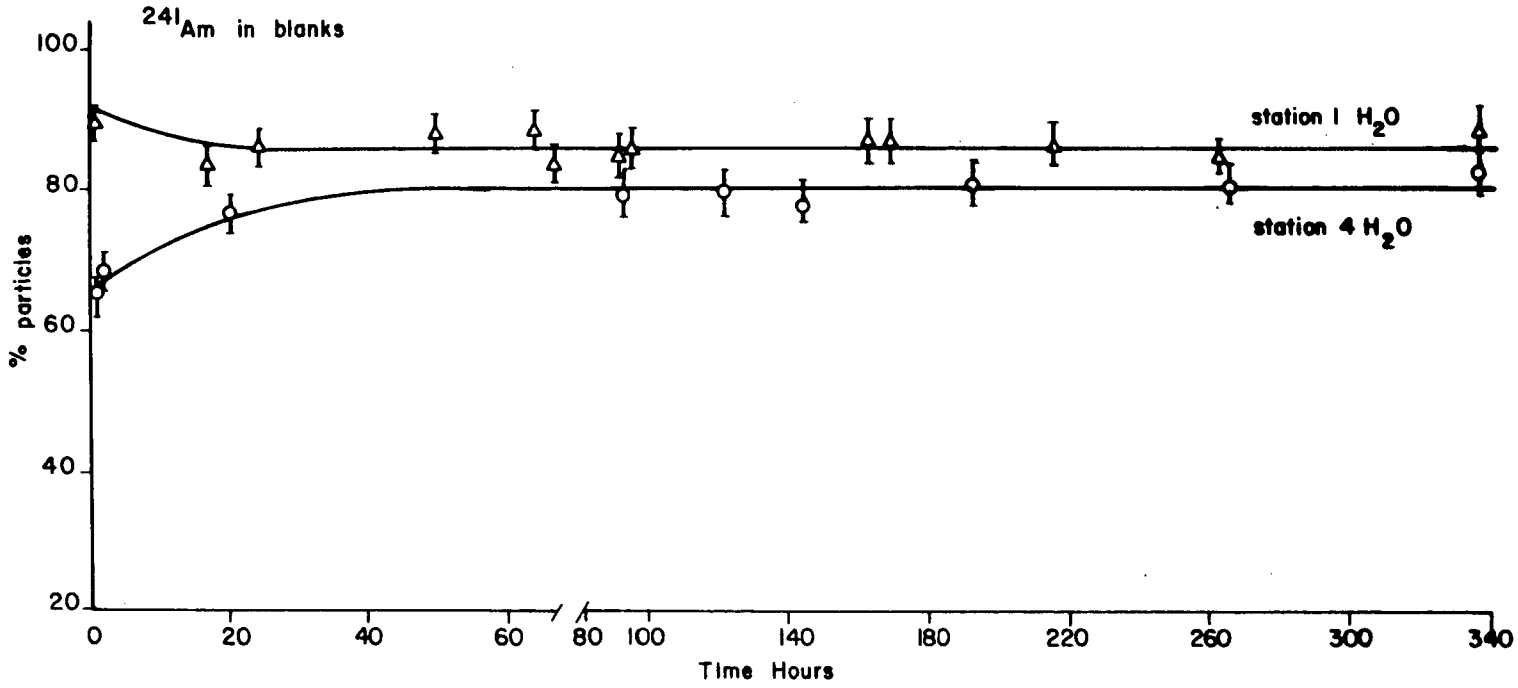

Figure 8. Percent of ^{241}Am in the particulate phase as a function of time for two control vessels from the Skagit River estuary.

Starik and Ginsburg (1961) showed that americium hydolyzes and forms colloids at the pH of this experiment: such colloids may then combine to form filterable particulates. It seems more likely that the particulate phases of ^{241}Am and ^{106}Ru present in these experiments are adsorbed to other precipitates such as manganese and ferric hydroxides. Further experiments are required to explain the formation of particulate ^{106}Ru and ^{241}Am in the filtered samples since these processes strongly influence the K_d values measured in different sediment-water systems.

3.4 Distribution Coefficients for Skagit River Estuary

Distribution coefficients of ^{106}Ru, ^{137}Cs, and ^{241}Am were determined for water-sediment systems from three different locations in the Skagit River Estuary. In each case, the constant-shaking technique was used to determine K_d values for filtered (<0.45μm) deep-water samples and sediments from the top 5 cm of a sediment core.

Distribution coefficients for these experiments are given in Table 5. The K_d value for ^{241}Am is approximately 1-2 x 10^5: this value is higher than the K_d values for other radionuclides in this experiment but it is lower than the K_d values we have reported previously for sediment-water systems from Lake Washington, Lake Nitinat, and the Washington continental shelf. The difference in K_d values between these experiments may result from differences in particle sizes of the sediments. We have not yet performed a particle size analysis on the sediments from the Skagit River Estuary. However, it is clear from visual observations that a high percentage of the sediment consists of sand-sized particles. On a weight basis these particles adsorb less radionuclides than smaller-grained sediments so the K_d value will be lower. There may also be mineralogical differences that account for the lower K_d values for sediments from the Skagit River Estuary. Experiments are currently underway to determine K_d values for the clay fraction (<63 μm) from the Skagit River Estuary cores. When these experiments are analyzed they may help to differentiate between the effects of particle size and mineral composition.

Distribution coefficients for ^{106}Ru in sediment-water systems from the Skagit River Estuary is approximately 2 x 10^4, nearly an order of magnitude less than the K_d value for ^{241}Am. If we do not consider the particle formation which was found in the controls, there are no significant differences between the K_d values measured for ^{106}Ru in any of these experiments. As was discussed above, however, 75-85% of the total activity of ^{106}Ru in the controls is found in the particulate phase. It may be important, therefore, to discriminate between the radionuclides which are adsorbed to the sediments and those that appear in the particulate phase due to precipitation from solution. If we subtract the fraction of the particulate ^{106}Ru that may be due to particle formation from the total in the particulate phase and then recalculate K_d values, we obtain the values indicated in column 4 or Table 5. For core #1, all of the particulate

Table 5: Summary of distribution coefficient experiments for sediment-water systems from the Skagit River Estuary.

Water-Sediment System	Radionuclide	Distribution Coefficient[c]		Approximate Percentage Distribution of Isotopes		Slope for Regression line[e]	
		Total	Less Particle Formation	Soluble	"Sorbed"[d]	Water	Sediment
Skagit Core #1	^{241}Am	$(1.67 \pm 0.58)\ 10^5$	$(2.67 \pm 0.94)\ 10^4$	3	97 (84% particles)	<0	0
	^{106}Ru	$(2.9 \pm 1.2)\ 10^4$	- - -	16	84 (100% particles)	0	0
	^{137}Cs	90 ± 27		93	7 (78% on filters)	0	0
Skagit Core #3	^{241}Am	$(2.09 \pm 0.64)\ 10^5$	$(7.6 \pm 2.4)\ 10^4$	4	96 (64% particles)	<0	<0
	^{106}Ru	$(1.97 \pm 0.57)\ 10^4$	$(8.5 \pm 2.9)\ 10^3$	29	71 (57% particles)	<0	0
	^{137}Cs	145 ± 38		93	7 (78% on filters)	0	0
Skagit Core #4	^{241}Am	$(1.07 \pm 0.26)\ 10^5$	$(4.4 \pm 1.2)\ 10^4$	2	98 (59% particles)	<0	0
	^{106}Ru	$(1.72 \pm 0.38)\ 10^4$	$(1.08 \pm 0.31)\ 10^4$	13	86 (39% particles)	0	0
	^{137}Cs	134 ± 24		90	10 (53% on filters)	0	0
Skagit Blank #1[a]	^{241}Am	- - -	- - -	8	92 (100% particles)	0	>0
	^{106}Ru	- - -	- - -	15	85 (100% particles)	>0	>0
	^{137}Cs	- - -	- - -	94	6 (100% on filters)	>0	0
Skagit Blank #2[b]	^{241}Am	- - -	- - -	13	87 (100% particles)	<0	>0
	^{106}Ru	- - -	- - -	22	78 (100% particles)	<0	>0
	^{137}Cs	- - -	- - -	95	5 (100% on filters)	<0	0

a,b Skagit Blank #1 = water from Sta. #1, S = 30.184 ‰; Skagit Blank #2 = water from Sta. #4, S = 30.224 ‰.

c K_d's in ml/g, differentiated into total K_d which includes particles formed for both ^{241}Am and ^{106}Ru, and K_d values which do not include particle formation. Cesium-137 does not form particles.

d "Sorbed" isotopes is the percent of the total activity measured on the filters. This could be further divided into the amount formed as particles/colloids for ^{241}Am and ^{106}Ru, or the amount sorbed on the filter in the case of ^{137}Cs, both given in parenthesis. The remaining activity is the amount removed by the sediments.

e t tests were performed to determine 95% confidence intervals on the slopes for each regression line. If slope = 0 is within the confidence band, equilibrium distribution was assumed to be obtained for the radionuclide (see text for discussion).

^{106}Ru could be accounted for by particle formation; however, particle formation cannot account for all of the particulate ^{106}Ru in cores #3 and #4.

Assigning geochemical significance to the "corrected" distribution coefficient is difficult. For core #1 we could assign all the activity in the particulate phase to particle formation. However, some ^{106}Ru probably is adsorbed to the sediments. The particulate ^{106}Ru could also be precipitated with iron and manganese hydroxides. Therefore, in subsequent experiments water samples will be filtered consecutively through filters with different nominal pore sizes. The radioactivity on the filters will be measured to discriminate better between the uptake of radionuclides by precipitation and adsorption to the suspended sediments.

The distribution coefficient for ^{137}Cs in the sediment-water systems from the Skagit River Estuary is approximately 100 and there are no significant differences among the different cores. The K_d values of ^{137}Cs are the lowest we have determined for any constant-shaking experiment. We did, however, find lower K_d values for ^{137}Cs using the thin layer method.

Equilibration time: Figure 9 shows the concentration of ^{137}Cs accumulated by sediments as a function of time for the Skagit River Estuary core #2. Equilibrium appears to be reached after approximately 40 hours. The concentration of ^{106}Ru on sediments and in solution is plotted as a function of time in Fig. 10. For ^{106}Ru, equilibrium between the soluble and particulate phases was not reached for approximately 80 hours. Subsequent experiments with Skagit estuary cores #3 and #4 were sampled "at equilibrium," i.e., after 90 hours.

"Equilibrium" conditions were analyzed statistically by obtaining a linear regression of radionuclide count rate vs. time and evaluating the slope together with its error. Equilibrium conditions were assumed to exist if the slope was not significantly different from zero. Slopes of the regression line for experimental vessels and controls are given on Table 5. The controls do not appear to be in equilibrium for ^{106}Ru, ^{137}Cs, or ^{241}Am. For the experimental vessels, ^{106}Ru and ^{137}Cs generally are in equilibrium; however, soluble ^{241}Am decreases at a slow rate throughout the sampling period.

3.5 Distribution Coefficients for the Saanich Inlet Sediment-water System

The distribution coefficients were measured for ^{85}Sr, ^{106}Ru, ^{137}Cs, and ^{241}Am using filtered (<0.45μm) deep water and the top 5 cm of sediments from Saanich Inlet, an anoxic marine basin. A control consisting of spiked, filtered seawater from the sampling site was measured with the sample.

In the initial experiment with a Saanich Inlet sediment-water system, we attempted to measure the K_d values of both ^{85}Sr and ^{106}Ru. However, there was counting interference between the main gamma ray energies of

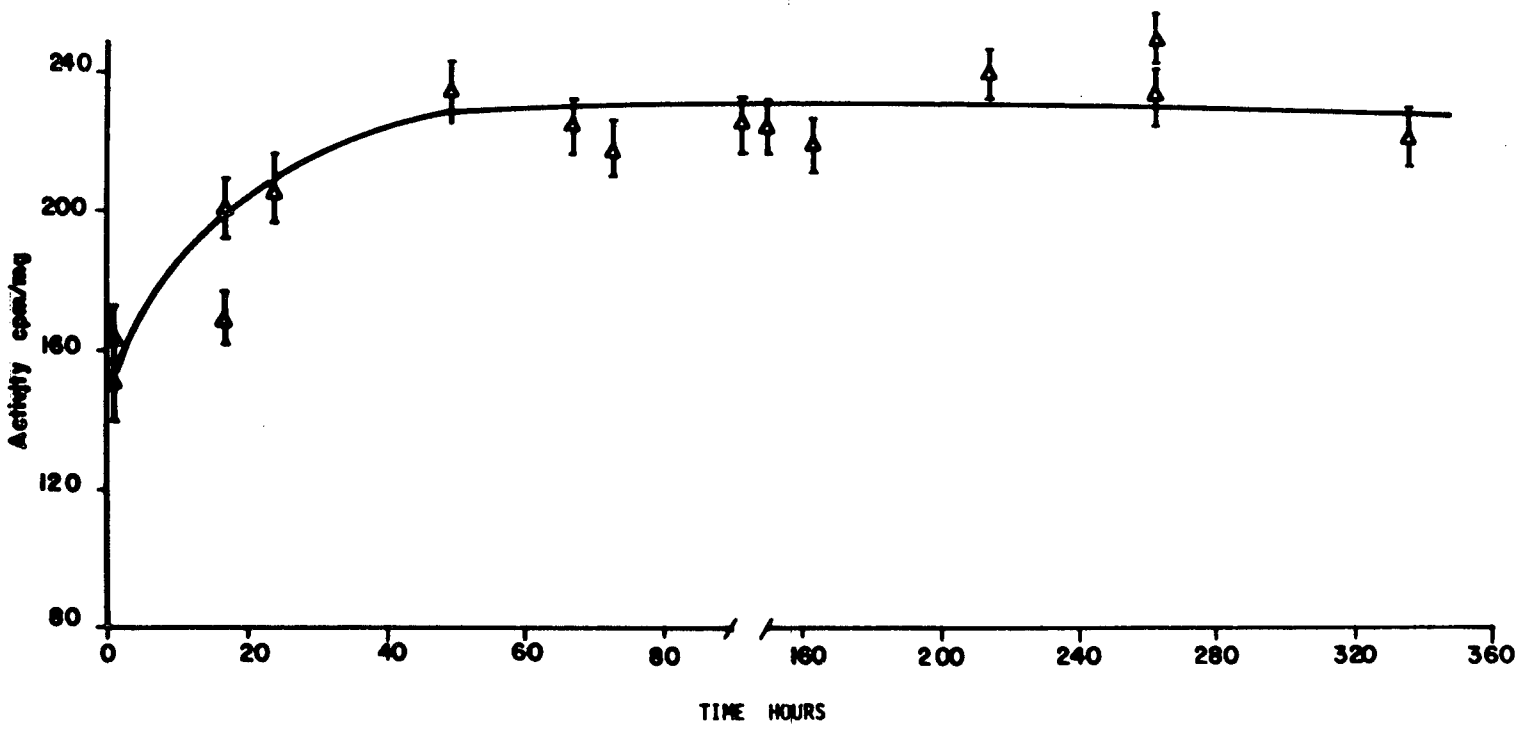

Figure 9. Concentration of ^{137}Cs in suspended sediments as a function of time for the Skagit River estuary core #1. Error bars indicate two standard deviations of propagated counting errors.

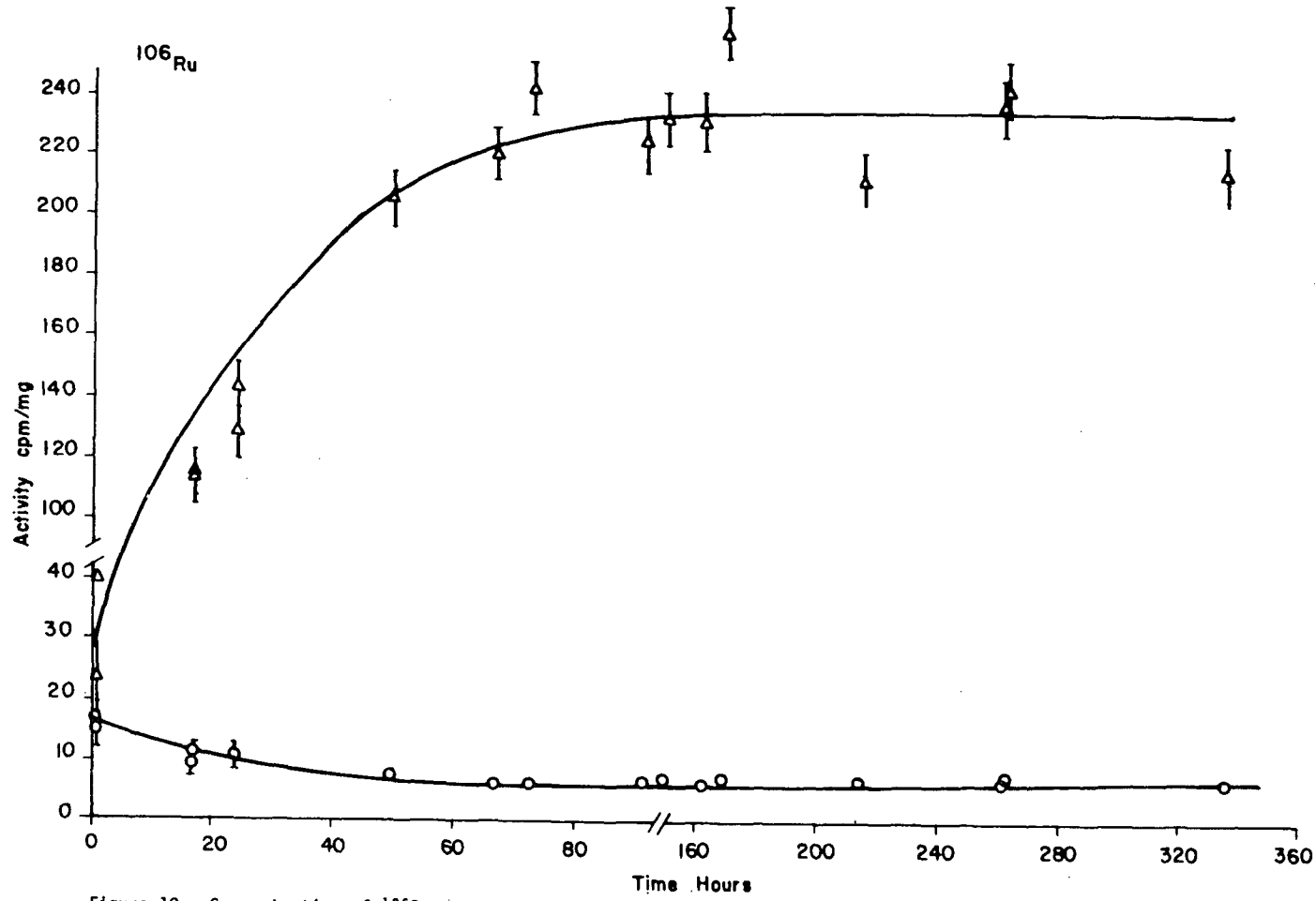

Figure 10. Concentration of ^{106}Ru on the filter and in solution as a function of time for a sediment-water system from the Skagit River estuary. Error bars represent two standard deviations of propagated counting errors.

^{106}Ru and ^{85}Sr. By using a lower abundance gamma ray energy for ^{106}Ru, the two radionuclies were analyzed but the counting error was large. A second experiment without ^{106}Ru, showed that ^{85}Sr generally remained in the soluble phase. There was no significant difference in radioactivity between the experiments and the controls for either the particulate or the soluble phase. Thus, we could not calculate a distribution coefficient value for ^{85}Sr but it must be very small for this water-sediment system.

The distribution coefficient for ^{137}Cs is 61 ± 24; this is the lowest K_d value we have been able to calculate for any radionuclide in any sediment-water system. Only 6% of the total activity of ^{137}Cs was retained by the filter and approximately 85% of this activity was sorbed to the filter rather than being taken up by the sediments.

In both the control and experimental vessels, 100% of ^{241}Am activity was retained on the filters. In fact, measurements of ^{241}Am radioactivity in the soluble phase approached the minimum detection limits of our instruments. Therefore, we can only estimate the K_d value to be between 1×10^5 and 3×10^6. Additional experiments are currently being conducted to obtain more reliable K_d values for ^{241}Am as well as for ^{90}Sr, ^{106}Ru, ^{244}Cm, ^{137}Cs, and ^{239}Pu.

3.6 Dialysis Experiments

During the past year we have conducted dialysis experiments using filtered water from the Eastern Pacific Ocean and the Skagit River Estuary. These experiments provide information on the relative affinity of radionuclides for phytoplankton, suspended sediments, and organic detritus and they complement the constant shaking experiments for the determination of K_d values. Furthermore, they indicate which biogeochemical pathways are most important for determining the transport and eventual disposition of specific radionuclides. The presence of a dialysis membrane prohibits transfer of physico-chemical species with a nominal molecular weight of larger than 6,000-8,000. Therefore, the data from these experiments may indicate which physico-chemical species predominate in a particular environment.

For one experiment, filtered Eastern Pacific seawater (32°/oo, pH = 8.0) collected at 150 m depth off Port Angeles, Washington was spiked with radionuclides and aged. Approximately 3,000 ml of spiked sea-water was placed in the outer compartment and three dialysis sacs with 100 ml of distilled water were submerged in the seawater. Five ml of a *Phaeodactylum tricornutum* culture (2×10^5 cells/ml) were added to the phytoplankton sac and 5 ml of filtrate from the algal culture were added to the control and sediment sacs; the sediment sac contained 765 mg of sediment from Lake Nitinat. The experiment was conducted at pH = 7.5 and the equilibrium salinity was 29.1 °/oo.

Samples were collected from all four compartments at the beginning of the experiment and after 1, 2, 4, and 11 days and analyzed for radioactivity in the soluble (<0.22μm) and particulate phase. The radioactivity of ^{60}Co, ^{65}Zn, ^{137}Cs, ^{207}Bi, and ^{241}Am was determined by counting using the Ge(Li) gamma ray spectroscopy system, and ^{238}Pu was measured by radiochemical separation and counting using the alpha spectroscopy system.

Data are presented in terms of radionuclide concentration in the soluble and particulate phases of each compartment as a function of time. Figure 11 shows the changes in concentration with time measured for ^{65}Zn, ^{137}Cs, ^{60}Co, ^{207}Bi, and ^{241}Am in the outer, spiked seawater compartment. During the first 2 days there is rapid decline in the concentration of all radionuclides. Except for ^{207}Bi, the decrease in concentration results primarily from losses in the outer compartment as the soluble species diffuse across the dialysis membranes and into the dialysis sacs. After the fourth day there are no significant changes, i.e., within counting errors, in the concentration of any of the radionuclides in either the soluble or the particulate phase.

It is important to note the scale used for the particulate and soluble data for ^{65}Zn, ^{137}Cs, and ^{60}Co. For these three radionuclides the radionuclide concentration in the particulate phase is generally less than 1% of that in the soluble phase. Therefore, the observed increase in concentration of particulate ^{60}Co between day 2 and 4 (Fig. 11) is only a negligible fraction of the total ^{60}Co concentration present.

Inside the control sac there was a significant increase in the soluble concentration of ^{65}Zn, ^{137}Cs, ^{60}Co, and ^{241}Am during the first day (Fig. 12) due to the soluble radionuclides diffusing across the membrane. The apparent concentration of ^{241}Am continued to increase slightly until day 4. However, the differences between days 2 and 11 did not exceed the propagated counting errors. For ^{65}Zn, ^{60}Co, and ^{137}Cs the soluble concentrations were unchanged or declined slightly from day 1 until the end of the experiment.

There was no detectable concentration of soluble ^{207}Bi in the control sac until the fourth day. However, the appearance of particulate ^{271}Bi inside the control sac implies that soluble ^{207}Bi from the outer chamber diffused across the dialysis membrane and then precipitated. The decrease in the concentration of particulate ^{207}Bi in the outer chamber also suggests this mechanism. It appears that the diffusion of soluble ^{207}Bi from the outer chamber produces a dissolution of ^{207}Bi from the particulate phase.

Both ^{241}Am and ^{207}Bi inside the control sac are found in the particulate phase after 1 day and reach an apparent equilibrium between the particulate and soluble species by day 4; however, in the same sac the concentrations of particulate ^{65}Zn, ^{137}Cs, and ^{60}Co increased significantly between days 4 and 11, indicating that for these radionuclides equilibrium may not have been attained. As with the outside compartment, the

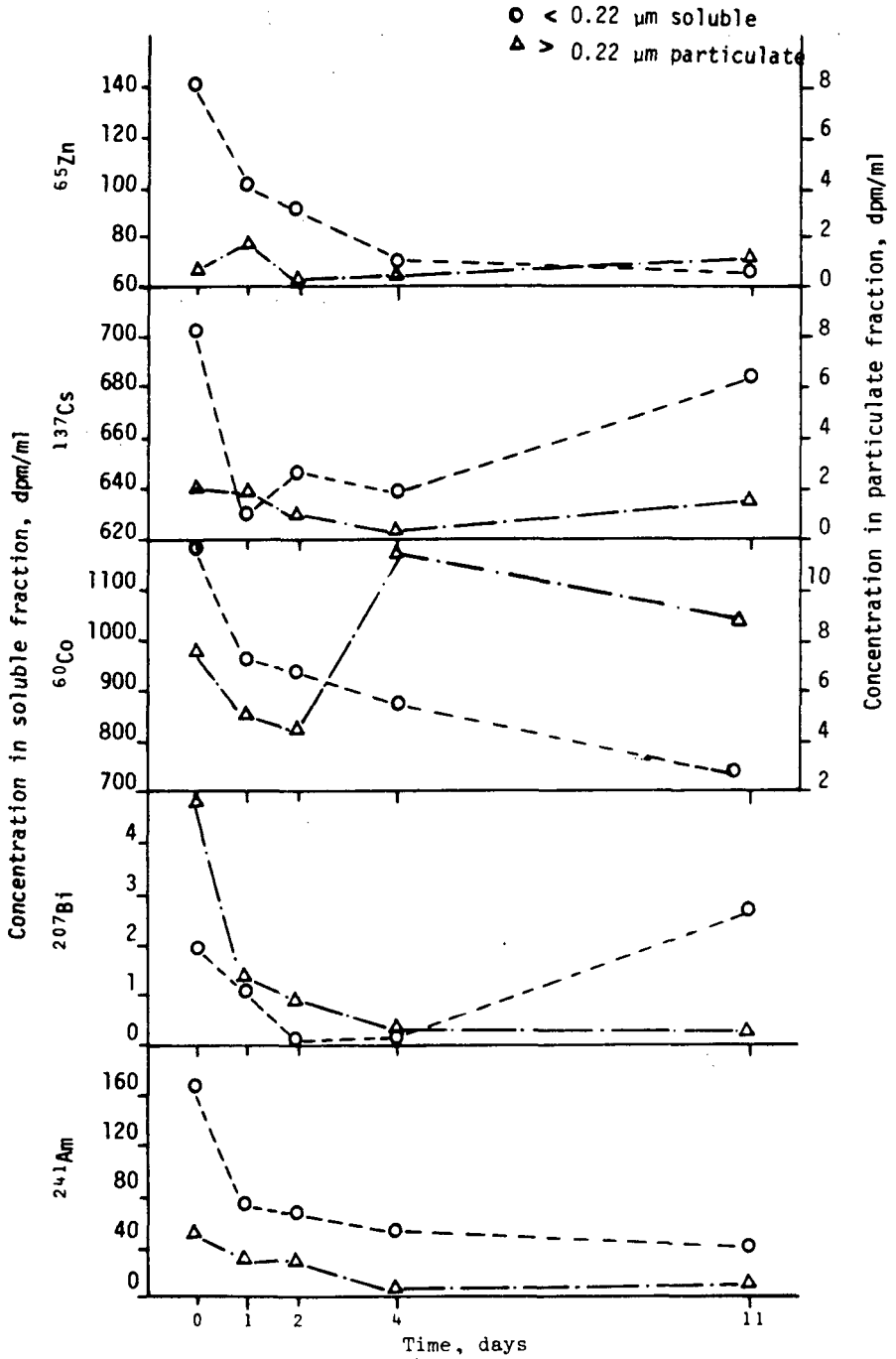

Figure 11. Concentration of radionuclides in soluble and particulate phases outside the dialysis sacs on days 0, 1, 2, 4, and 11 of a dialysis experiment.

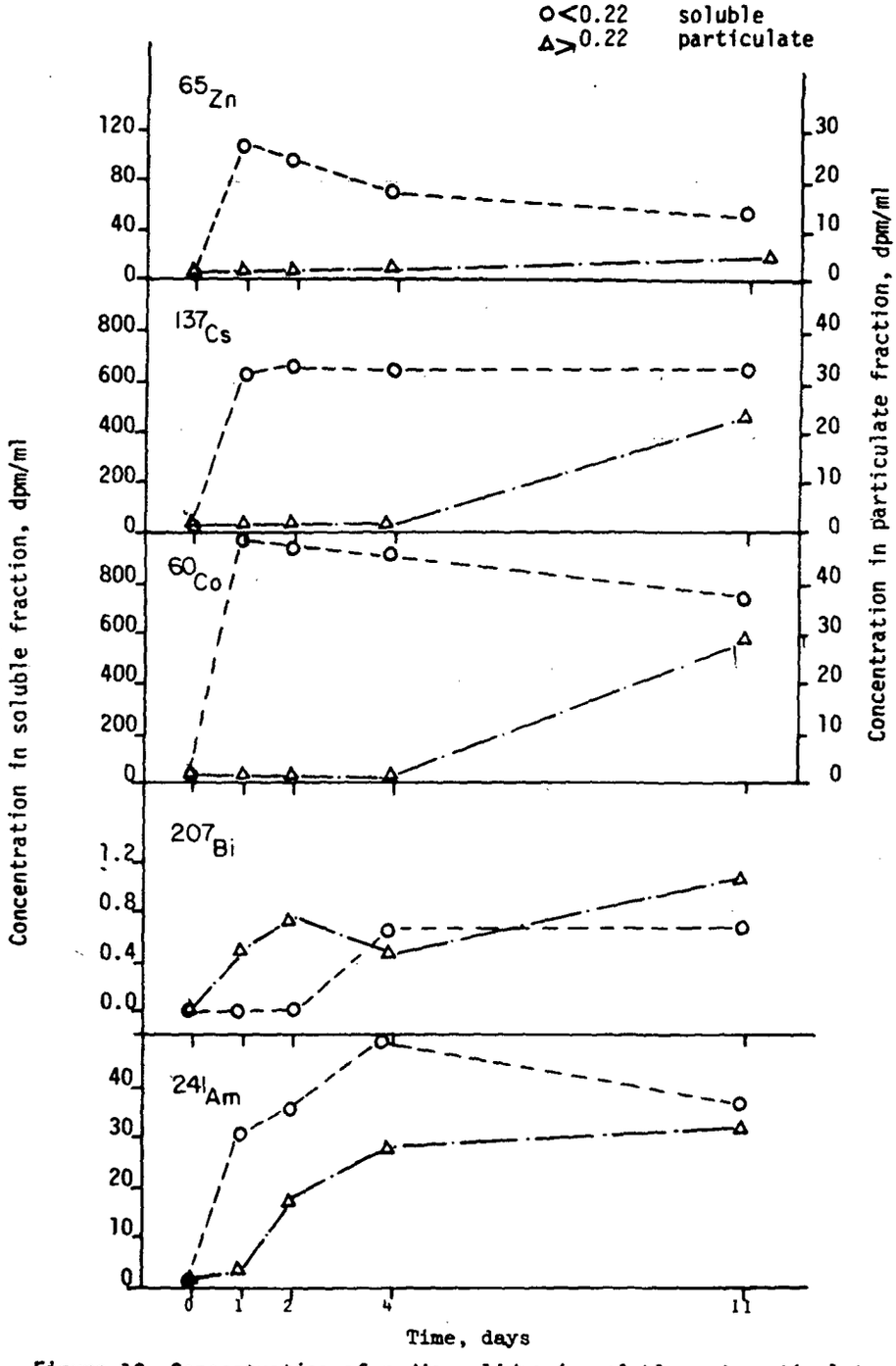

Figure 12. Concentration of radionuclides in soluble and particulate phases within the control dialysis sac on day 0, 1, 2, and 11 of a dialysis experiment.

controlsac contained only a small fraction (<5%) of the total concentration of ^{65}Zn, ^{137}Cs, and ^{60}Co in the particulate phase.

The distribution of ^{65}Zn, ^{137}Cs, ^{60}Co, ^{207}Bi, and ^{241}Am in the plankton and sediment chambers is shown in Figs. 13 and 14. Except for ^{137}Cs, most (>80%) of the activity for each of these radionuclides is found in the particulate phase; in the sediment sac approximately 40% of ^{137}Cs is associated with the particulates. Except for ^{65}Zn which shows similar behavior in both the sediment and plankton chambers, the sediment accumulated significantly higher concentrations of all radionuclides than the phytoplankton did.

The concentration of ^{238}Pu in the three sacs as a function of time is shown in Fig. 15. In the outer compartment and inside the control sac low concentrations of ^{238}Pu are found in the particulate phase. However, in both the suspended sediment and the phytoplankton sacs considerable concentrations of ^{238}Pu are found in the particulate phase as shown in Fig. 16. During the first two days ^{238}Pu accumulates in both plankton and sediments but after that time the concentration (dpm/g) appears relatively constant. Figure 15, however, indicates that ^{238}Pu may not have reached equilibrium in the sediment sac within 11 days.

The concentrations of ^{65}Zn, ^{137}Cs, ^{60}Co, ^{207}Bi, and ^{241}Am per unit weight of sediment and phytoplankton are shown in Fig. 17. After 2 days the radionuclides in solution and on the sediments and phytoplankton have reached equilibrium, except for ^{137}Cs on phytoplankton. The concentrations of ^{241}Am, ^{207}Bi, and ^{60}Co are approximately equivalent for both sediments and phytoplankton. However, significantly greater quantities of ^{137}Cs are accumulated by sediments, while phytoplankton have a slightly higher concentration of ^{65}Zn.

Mass balance calculations were made for the radionuclides in this experiment at day 11. These calculations include the activity in the particulate and soluble phase for each of the compartments at day 11, the activity removed by sampling during the experiment and the activity adsorbed to miscellaneous apparatus. The latter includes activity adsorbed to the dialysis membranes and that removed from the glassware by three washings with hot HNO_3. A complete listing of these calculations is provided in the Appendix[Tables A1-A6] for individual radionuclides. A summary of the results is given in Table 6 which presents the percent of the total recovered activity found in each compartment for each radionuclide. Approximately 100% of ^{60}Co, ^{65}Zn, ^{137}Cs, and ^{207}Bi was recovered. However only 68% of ^{241}Am and 21% of ^{238}Pu was recovered; it seems likely that these radionuclides were strongly adsorbed to the glassware and were not removed by the HNO_3 washes. In terms of the percent recovered, the major portion of ^{207}Bi, ^{60}Co, ^{137}Cs, ^{238}Pu, and ^{65}Zn occurs in the soluble fraction of the outer compartment. However, ^{241}Am has ≃ 47% in the particulate phase of the sediment sac and another 12% in the particulate phase of the plankton sac.

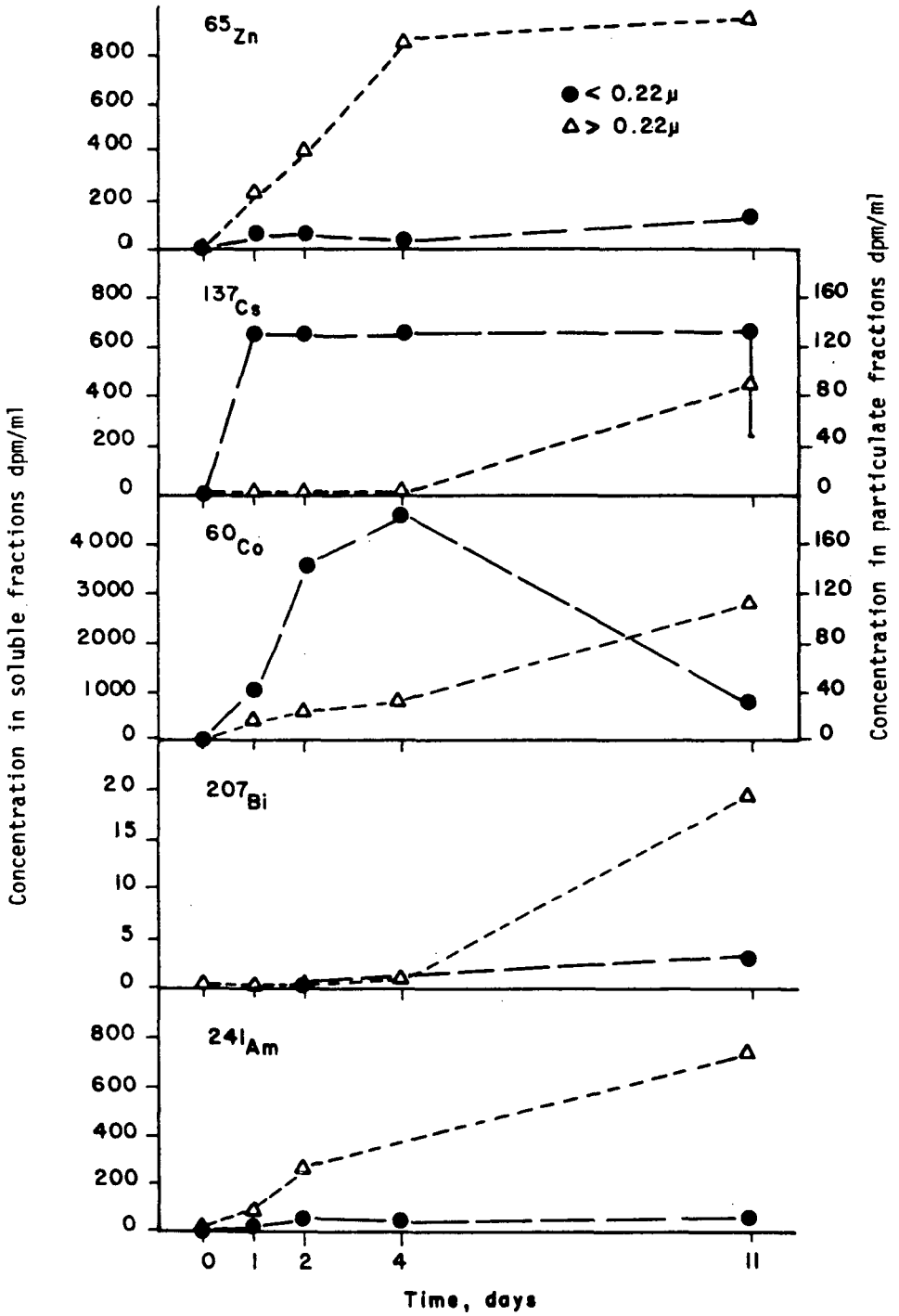

Figure 13. Concentration of radionuclides in the soluble and particulate phases within the plankton dialysis sac on days 0, 1, 2, 4 and 11 of a dialysis experiment.

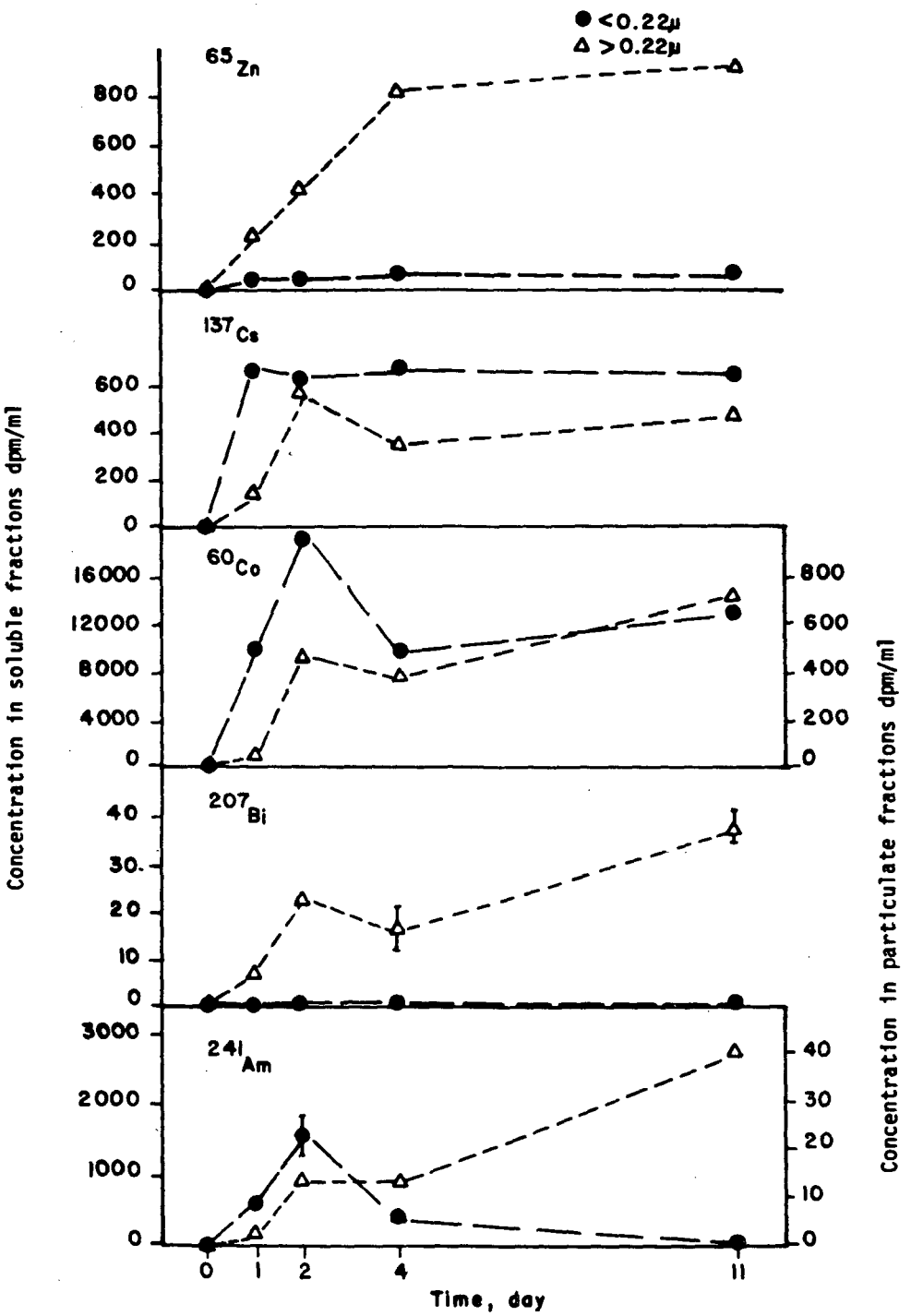

Figure 14. Concentration of radionuclides in the soluble and particulate phases within the sediment sac on days 0, 1, 2, 4 and 11 of a dialysis experiment.

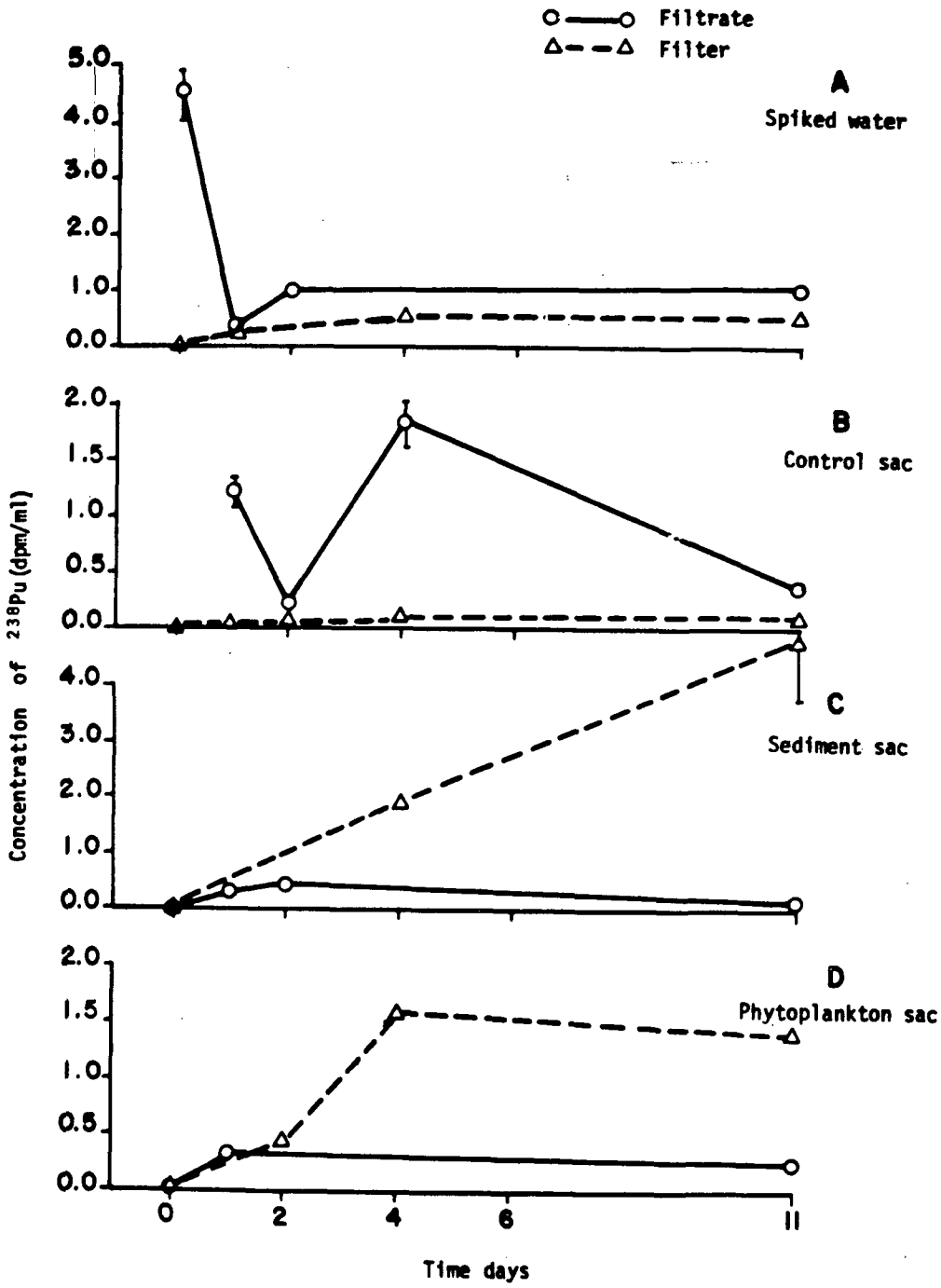

Figure 15. Concentration of ^{238}Pu in the soluble and particulate phases on days 0,1,2,4, and 11 of a dialysis experiment.

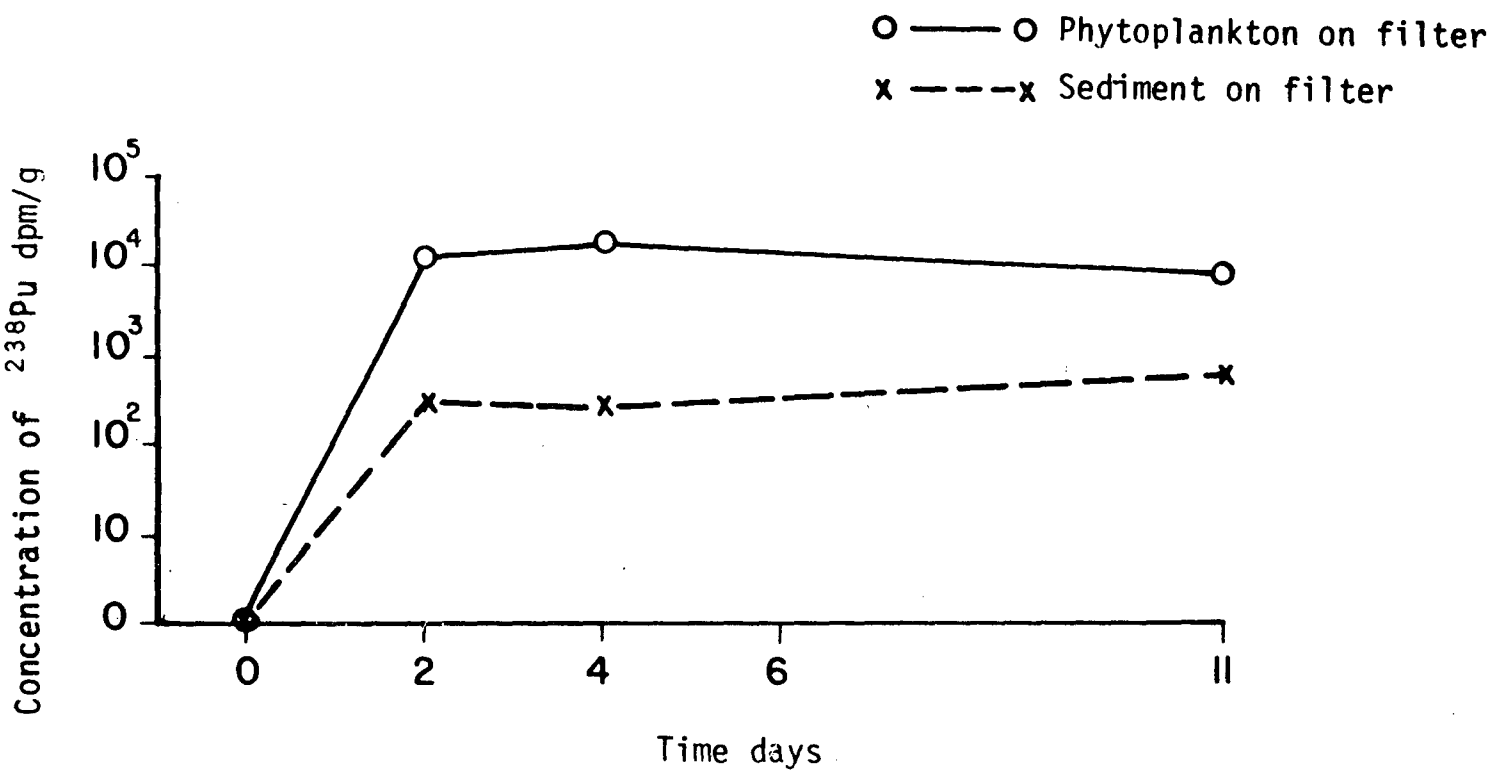

Figure 16. Concentration of ^{238}Pu per unit weight of sediment and plankton on days 0, 1, 2, 4 and 11 of a dialysis experiment.

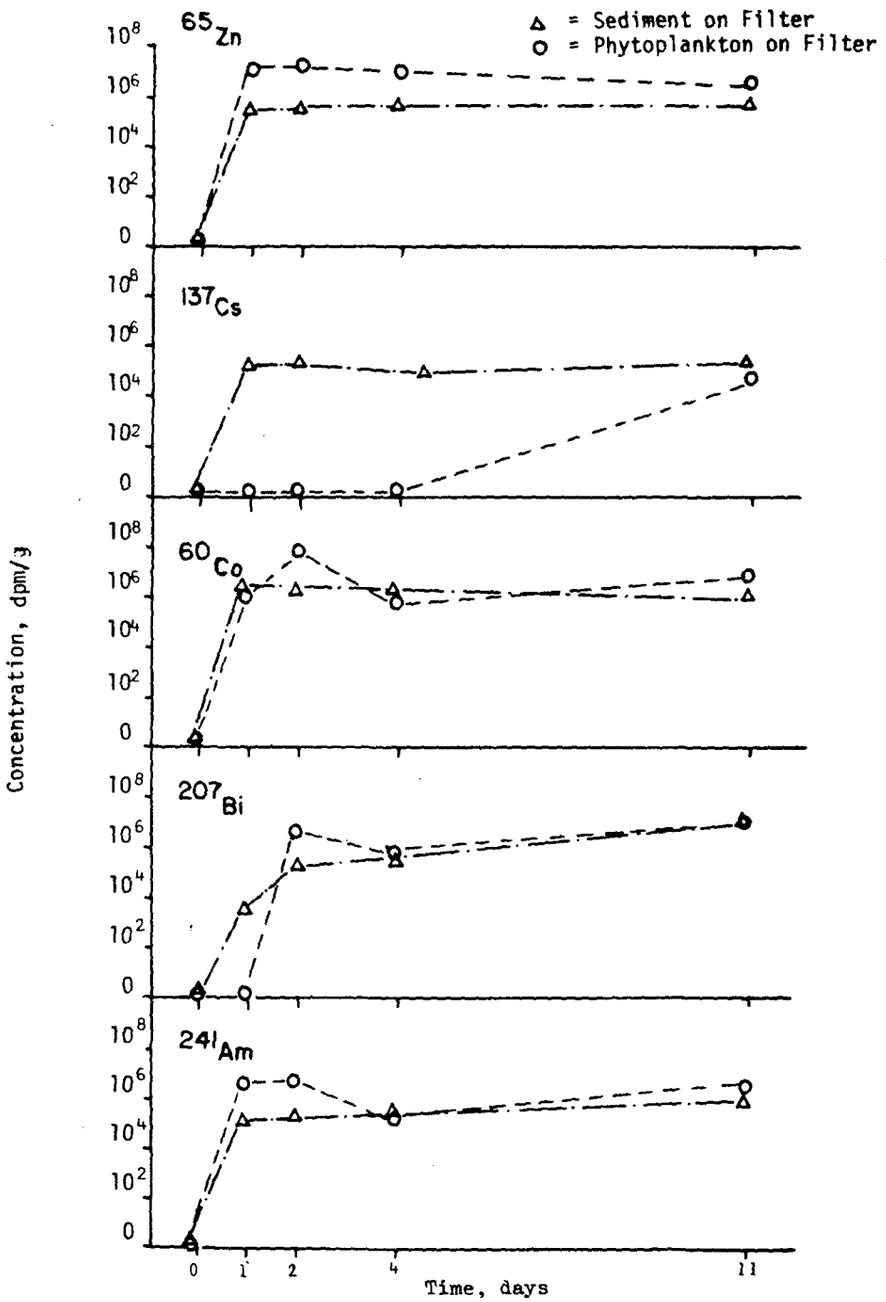

Figure 17. Concentration of ^{65}Zn, ^{137}Cs, ^{60}Co, ^{207}Bi and ^{241}Am per unit weight of sediment and plankton on days 0, 1, 2, 4 and 11 of a dialysis experiment.

Table 6: Percent distribution of radionuclides after 11 days of dialysis experiment.

Compartments		^{241}Am	^{137}Cs	^{238}Pu	^{204}Bi	^{65}Zn	^{60}Co
Spiked water	<0.22m	26.1	86.3	77.7	64.4	44.7	54.2
	>0.22m	8.2	0.0	3.0	0.0	0.8	0.8
Control Sac	<0.22m	0.7	2.2	0.5	0.0	1.0	8.4
	>0.22m	0.5	0.1	0.1	0.6	0.1	0.1
Sediment Sac	<0.22m	0.0	2.1	0.2	0.0	1.1	1.4
	>0.22m	47.1	1.6	9.2	22.3	25.8	30.5
Plankton Sac	<0.22m	0.8	2.2	0.4	1.9	2.4	1.7
	>0.22m	12.4	0.3	2.7	11.5	16.9	0.3
Samples during experiment		4.5	0.7	2.5	2.8	7.3	4.5
Miscellaneous apparatus		0.5	0.0	3.8	0.6	0.0	0.0

Another dialysis experiment was conducted to investigate the effect of organic detritus on the distribution of radionuclides. Filtered (0.45μm) water from the Skagit River Estuary was spiked with ^{59}Fe, ^{60}Co, ^{106}Ru, ^{137}Cs, and ^{241}Am; approximately 625 ml of this solution was placed in the outer compartment. Four dialysis sacs containing 60 ml of filtered, but unspiked, water from the Skagit River Estuary were then submerged in the outer solution. The phytoplankton sac contained 1.2 x 10^7 cells of Phaeodactylum tricornutum, the sediment sac contained 12.8 mg of the pure clay montmorillinite, and the detritus sac contained 6.0 mg of dried and finely ground Zostera sp.; no additions were made to the control dialysis sac. The experiment was conducted at pH = 7.5, salinity = 32 °/oo and temperature = 20°C.

Samples were collected from each chamber at 2, 10, 24, 48, and 96 hours and the activity (cpm/ml) was determined for the soluble (<0.45μm) and particulate (>0.45μm) fractions. The distribution of radionuclides for each of the sampling times is given in Appendix B (Tables B1-B5). By comparing the concentration of soluble radionuclides in each compartment we can estimate the time required to reach equilibrium between the outside chamber and the inside of the dialysis sacs for each radionuclide. Cesium-137 reaches equilibrium after 10 hours and ^{106}Ru reaches equilibrium after 24 hours. However, for the radionuclides, ^{241}Am, ^{60}Co, ^{65}Zn, and ^{59}Fe, at least one dialysis sac does not come to equilibrium during 96 hours of the experiment.

Figures C-1 - C-11 show the concentration of different radionuclides in the soluble or particulate phase for each compartment throughout the experiment. The distribution of ^{137}Cs in the soluble phase is plotted in Fig. C-1. All the dialysis sacs appear to be in equilibrium with the outer compartment after 10 hours and there are only small changes in concentration after that time. Table 7 shows that the concentration of particulate ^{137}Cs never exceeded detection limits and less than 1% of the total ^{137}Cs is in the particulate phase.

There is a rapid decrease in the concentration of ^{241}Am in the outer compartment (Fig. C-2 and C-3) and an increase in both the soluble and particulate phases in all the dialysis sacs. However, the concentration within the dialysis sacs continues to increase throughout the experiment and the soluble species do not come to equilibrium among the different compartments within the 96 hours. The highest concentrations of both particulate and soluble ^{241}Am are found in the detritus chamber followed by the phytoplankton chamber. Therefore, suspended organic matter, especially detritus, may be very important for the transport of ^{241}Am. The affinity of ^{241}Am for organic surfaces indicates that chemical complexation as well as adsorption may contribute to the uptake of ^{241}Am. This is an important observation since there is very little information on the complexation of ^{241}Am by organic ligands. Table 8 indicates that most ^{241}Am occurs in the particulate phase throughout the experiment.

Table 7. Distribution of ^{137}Cs during a 96-hour dialysis experiment

Time (hrs.)		Spiked Seawater cpm/ml±2 S D	Dialysis Chambers: Blank cpm/ml±2 S D	Plankton cpm/ml±2 S D	Clay cpm/ml±2 S D	Detritus cpm/m.±2 S D
0		24.4				
2	Soluble	18.2 (0.92)	14.7 (0.40)	16.2 (0.76)	16.5 (0.76)	16.5 (0.72)
	Particulate	< 0.37	< 0.16	< 0.16	< 0.06	< 0.18
	% Particulate	< 2.0	< 1.1	< 1.0	< 0.4	< 1.1
10	Soluble	21.5 (0.95)	20.3 (0.89)	21.6 (0.91)	20.9 (0.87)	19.2 (0.84)
	Particulate	< 0.30	< 0.19	< 0.20	< 0.19	< 0.19
	% Particulate	< 1.4	< 0.9	< 0.9	< 0.9	< 1.0
24	Soluble	19.9 (0.93)	20.5 (0.90)	22.2 (0.88)	21.1 (0.93)	19.8 (0.93)
	Particulate	< 0.24	< 0.13	< 0.20	< 0.22	< 0.22
	% Particulate	< 1.4	< 0.6	< 0.9	< 1.1	< 1.1
48	Soluble	20.2 (0.92)	18.9 (0.90)	20.9 (0.50)	21.8 (0.93)	21.9 (0.94)
	Particulate	< 0.23	< 0.19	< 0.21	< 0.20	< 0.22
	% Particulate	< 1.1	< 1.0	< 1.0	< 0.9	< 1.0
96	Soluble	21.2 (0.92)	20.3 (0.51)	20.4 (0.95)	23.2 (0.93)	23.0 (0.93)
	Particulate	< 0.19	< 0.20	< 0.23	< 0.15	< 0.24
	% Particulate	< 0.9	< 1.0	< 1.1	< 0.6	< 1.1

Table 8. Distribution of ^{241}Am during a 96-hour dialysis experiment.

Time (hrs.)		Spiked Seawater cpm/ml±2 S D	Dialysis Chambers: Blank cpm/ml±2 S D	Plankton cpm/ml±2 S D	Clay cpm/ml±2 S D	Detritus cpm/ml±2 S D
0		1703				
2	Soluble	152.5 (4.2)	5.56 (0.46)	1.36 (0.84)	2.02 (0.89)	1.08 (0.79)
	Particulate	705.3 (2.5)	1.91 (0.47)	3.90 (0.53)	1.55 (0.23)	0.59 (0.45)
	% Particulate	82.2	25.6	74.1	43.4	35.3
10	Soluble	116.5 (2.2)	15.79 (1.11)	18.19 (1.17)	15.55 (1.08)	18.10 (1.18)
	Particulate	451.5 (3.4)	11.47 (0.84)	49.20 (1.27)	21.81 (0.95)	43.94 (1.20)
	% Particulate	79.5	42.1	73.0	58.4	70.8
24	Soluble	100.5 (2.1)	23.97 (1.26)	25.45 (1.31)	22.40 (1.24)	47.71 (1.55)
	Particulate	195.7 (2.3)	13.16 (0.68)	100.6 (1.7)	74.50 (1.50)	138.3 (1.9)
	% Particulate	66.1	35.4	79.8	76.9	74.4
48	Soluble	101.7 (2.1)	27.73 (1.38)	41.60 (0.88)	26.20 (1.34)	110.5 (2.2)
	Particulate	108.2 (2.4)	39.24 (1.18)	219.1 (2.4)	98.80 (1.73)	272.5 (2.6)
	% Particulate	51.4	58.6	84.0	79.0	71.1
96	Soluble	134.5 (2.3)	33.99 (0.85)	54.75 (1.71)	26.84 (1.40)	134.1 (2.4)
	Particulate	62.7 (1.4)	68.95 (1.40)	357.4 (3.0)	149.4 (1.1)	555.6 (3.7)
	% Particulate	31.8	66.3	86.7	84.8	80.6

The data for ^{106}Ru are plotted in Figs. C-4 and C-5. Throughout the experiment there are higher concentrations of dissolved species than particulate species in all chambers, and the concentration of ^{106}Ru in the particulate phase is generally less than 20% of the total concentration except for the dialysis sac containing plankton.

The concentration of soluble ^{106}Ru species is comparable for each of the dialysis sacs after the first 10 hours, but equilibrium with the outside chamber was not attained during that period. After 4 hours detritus and plankton sacs appear to be in equilibrium with the outer compartment; the control and clay sacs reach equilibrium after 48 hours. Between 24 hours and 96 hours the dialysis sac with detritus and the outer spiked seawater compartment behave similarly, but the concentration of soluble species in the plankton, clay, and blank dialysis chambers change independently of the outer compartment.

The concentration of particulate ^{106}Ru in the outer compartment decreases throughout the experiment. In the blank dialysis chamber the concentration of particulate ^{106}Ru is constant from 10 hours through 96 hours and appears to be in equilibrium with the outside chamber after 96 hours. There was no significant concentration of particulate ^{106}Ru in any of the dialysis sacs after 2 hours and detectable concentrations in the clay chamber were not apparent until 24 hours. The clay, detritus, and plankton chambers all showed significant increases in the concentration of ^{106}Ru in particulate phases during the experiment and had significantly higher concentrations of particulate ^{106}Ru than either the blank dialysis sac or the outer compartment. Thus, each of the suspended solids take up ^{106}Ru. Phytoplankton showed the greatest affinity for ^{106}Ru, followed by clay and detritus.

Of the radionuclides included in this experiment the data for ^{65}Zn are most variable. Figure C-6 indicates that only the clay chamber reaches equilibrium with the outer chamber during the course of the experiment although the blank dialysis sac is approaching equilibrium at 96 hours. Throughout the experiment there are significant differences in the concentration of soluble species among the different dialysis sacs. In the compartment and each of the dialysis sacs. Therefore, it is unlikely that differential diffusion from the outer compartment into the dialysis sacs contributes significantly to the observed differences in concentration. Rather, the speciation within each dialysis sac must be independent of the other chambers. The similarity between the plankton and the detritus chambers suggests that organic complexes are being formed in those chambers which then diffuse out; this process would explain the lower concentration of soluble species found within these sacs.

The particulate species of ^{65}Zn (Figure C-7) behave more predictably than the soluble species. All of the dialysis chambers have detectable concentrations of particulate species after 10 hours, and, except for the blank chamber, the concentration of particulate species increases throughout the experiment. Initially, the concentration of particulates in the

detritus chamber is significantly higher than for the clay or plankton chamber, although they all have comparable concentrations after 96 hours. Throughout the experiment the concentration of particulate species is significantly less than the concentration of soluble species. Generally, less than 10% of the total concentration is in the particulate phase.

Concentrations of soluble ^{60}Co are shown in Fig. C-8. The blank and detritus chambers reach equilibrium with the outer compartment within 24 hours but the clay and plankton chambers do not reach equilibrium within 96 hours. The concentration of soluble ^{60}Co in the clay chamber is slightly less than in the blank and detritus chambers, but the concentration of soluble ^{60}Co in the plankton dialysis sac suggests the formation of an organic complex which diffuses back out of the plankton dialysis sac.

Clay, plankton, and detritus all take up ^{60}Co into the particulate phase (Fig. C-8) but except for plankton, the concentration of particulate ^{60}Co is generally less than 1% of the total concentration. Since the percent particulate is significantly higher in the plankton chamber than in any other compartment (Table B-5), plankton may represent the principal component of particulate ^{60}Co in marine systems.

4.0 SUMMARY

1. The sedimentation technique for determining distribution coefficients of ^{106}Ru and ^{241}Am produced K_d values approximately two orders of magnitude lower than the values determined by the constant shaking technique. Therefore, to evaluate distribution coefficients for different radionuclides or different water-sediment systems it is necessary to compare values that were obtained using the same experimental techniques.

2. Distribution coefficients were determined for ^{241}Am as a function of pH for sterile and non-sterile water-sediment systems from Lake Washington and Lake Nitinat. The K_d values are higher at higher pH values for all systems, increasing by three orders of magnitude between pH 4 and 10 for the sterile Lake Nitinat systems. Similar experiments should be conducted to determine the effect of pH on the K_d values of other radionuclides in both freshwater and marine systems.

3. In filtered seawater significant amounts of ^{106}Ru and ^{241}Am are found in the particulate phase. The exact nature of this phenomenon has not yet been determined although the radionuclides may be adsorbed on iron or manganese solids which precipitate from solution. Additional experiments are recommended to determine if other radionuclides are present in the particulate phase under similar conditions.

   Also, experiments should be conducted to determine if these radionuclides can occur as particulates in filtered freshwater, distilled water, and 0.7 M NaCl solution.

4. Distribution coefficients for sediment-water systems from the Skagit River Estuary were determined for ^{241}Am ($1\text{-}2 \times 10^5$), ^{106}Ru ($\simeq 2 \times 10^4$), and ^{137}Cs ($\simeq 1 \times 10^2$). Distribution coefficients were found to vary significantly at stations that were expected to have comparable K_d values. Additional experiments are required to determine if particle size differences among the different stations are contributing to the observed differences in K_d values.

5. Experiments were conducted to determine distribution coefficients for an anoxic sediment-water system from Saanich Inlet. The K_d value for ^{137}Cs was 61 ± 24. Americium-241 occurred almost entirely in the particulate phase while nearly all of the ^{85}Sr was soluble.

6. Dialysis experiments with 6000-8000 molecular weight exclusion size dialysis membranes indicated that most radionuclides are transported across the dialysis membrane quite rapidly. Cesium-137 comes to equilibrium after 10 hours and ^{106}Ru after 24 hours but ^{241}Am, ^{60}Co, ^{65}Zn, ^{59}Fe, and ^{207}Bi do not reach equilibrium within 4 days. It

appears that interactions between the different chambers, especially the formation of organic complexes with the radionuclides, may account for the slow approach to equilibrium. Additional experiments using different pore size dialysis membranes should provide valuable information on the different physico-chemical species in these experiments.

7. Organic detritus was shown to be an important factor in the distribution of radionuclides. Compared to suspended sediments and phytoplankton, detritus has a very high affinity for ^{241}Am. Other radionuclides, ^{59}Fe, ^{65}Zn, and ^{106}Ru are also accumulated by detritus but the uptake corresponds to the uptake by sediments and phytoplankton. Additional experiments with pure clays and different types of detrital particles will provide more information on the relative affinity of these surfaces for different radionuclides.

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APPENDIX A

Mass balance calculations for radionuclides after 11 days of dialysis experiment.

6 Tables

Table A-1. Mass balance calculations for ^{241}Am after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside	filtrate	1.14×10^5	1.10×10^4	26.1	2.6
dialysis sacs	filter	3.60×10^4	2.16×10^3	8.2	0.5
b, inside	filtrate	2.84×10^3	2.88×10^2	0.6	0.1
control sac	filter	2.31×10^3	1.28×10^2	0.5	0.0
(2) Dialysis sac contents					
a, sediment	filtrate	0.00	0.00	0.0	0.0
sac	filter	2.06×10^5	1.14×10^3	47.1	1.3
b, plankton	filtrate	3.49×10^3	3.70×10^2	0.8	0.1
sac	filter	5.4×10^4	6.52×10^2	12.3	0.4
(3) Subsamples		1.95×10^4	1.47×10^2	4.4	0.1
(4) Flask washdown and dialysis sacs		2.18×10^3	4.71	0.5	0.0
Total of (1), (2), (3), and (4)		4.37×10^5	1.13×10^4	100.7	3.0
Amount added†		6.45×10^5	2.13×10^4		
% Accounted for				67.8	2.8
% Unaccounted for				-32.2	2.8

*Single sample error values are one-sigma, propagated, counting error.

†Amount added = spiked water at day 0.

Table A-2. Mass balance calculations for ^{137}Cs after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside dialysis sacs	filtrate	2.00×10^6	5.21×10^4	89.3	3.1
	filter	0.00	0.00	0.0	0.0
b, inside control sac	filtrate	4.97×10^4	1.32×10^3	2.2	0.1
	filter	1.71×10^3	1.85×10^2	0.1	0.0
(2) Dialysis sac contents					
a, sediment sac	filtrate	4.73×10^4	1.11×10^3	2.1	0.1
	filter	3.48×10^4	1.30×10^3	1.6	0.1
b, plankton sac	filtrate	4.95×10^4	1.16×10^3	2.2	0.1
	filter	7.17×10^3	3.03×10^3	0.3	0.1
(3) Subsamples		4.64×10^4	3.12×10^2	0.7	0.0
(4) Flask washdown and dialysis sacs		1.02×10^1	1.40	0.0	0.0
Total of (1), (2), (3), and (4)		2.24×10^6	5.23×10^4	98.5	3.1
Amount added†		2.12×10^6	4.77×10^4		
% Accounted for				105.7	3.4
% Unaccounted for				+ 5.7	3.4

*Single sample error values are one-sigma, propagated, counting error.

†Amount added = spiked water at day 0.

Table A-3. Mass balance calculations for ^{238}Pu after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside dialysis sacs	filtrate	3.17×10^3	2.85×10^2	77.7	9.2
	filter	1.22×10^2	3.27×10	3.0	0.8
b, inside control sac	filtrate	2.09×10	3.08×10	0.5	0.1
	filter	3.31	3.10×10^{-1}	0.1	0.4
(2) Dialysis sac contents					
a, sediment sac	filtrate	7.01	1.93	0.2	0.0
	filter	3.76×10^2	9.63×10	9.2	2.5
b, plankton sac	filtrate	1.59×10	3.08	0.4	0.1
	filter	1.08×10^2	7.37×10	2.6	1.8
(3) Subsamples		1.00×10^2	2.73	2.5	0.2
(4) Flask washdown and dialysis sacs		1.57×10^2	3.25×10	3.9	0.3
Total of (1), (2), (3), and (4)		4.08×10^3	3.13×10^2	100.0	9.7
Amount added†		1.93×10^4	2.46×10^3		
% Accounted for				21.1	3.1
% Unaccounted for				-78.9	3.1

*Single sample error values are one-sigma, propagated, counting error.

†Amount added = spiked water at day 0.

Table A-4. Mass balance calculations for ^{207}Bi after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside dialysis sacs	filtrate	7.73×10^3	3.26×10^3	64.4	15.7
	filter	0.00	0.00	0.0	0.0
b, inside control sac	filtrate	0.00	0.00	0.0	0.0
	filter	7.70×10^1	2.28×10^1	0.6	0.2
(2) Dialysis sac contents					
a, sediment sac	filtrate	0.00	0.00	0.0	0.0
	filter	2.85×10^3	2.75×10^2	22.3	6.1
b, plankton sac	filtrate	2.46×10^2	8.55×10^1	1.9	0.8
	filter	1.47×10^3	1.21×10^2	11.5	3.1
(3) Subsamples		3.62×10^2	2.81×10^1	2.8	0.3
(4) Flask washdown and dialysis sacs		7.40×10^1	9.7×10^{-1}	0.6	0.2
Total of (1), (2), (3), and (4)		1.28×10^4	3.27×10^3	104.1	17.1
Amount added†		1.47×10^4	6.24×10^2		
% Accounted for				87.1	22.6
% Unaccounted for				-12.9	22.6

*Single sample error values are one-sigma, propagated, counting error.

†Amount added = spiked water at day 0.

Table A-5. Mass balance calculations for ^{65}Zn after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside	filtrate	1.92×10^5	7.78×10^3	44.7	2.0
dialysis sacs	filter	3.27×10^3	6.80×10^2	0.8	0.2
b, inside	filtrate	4.10×10^3	1.90×10^2	0.9	0.1
control sac	filter	3.62×10^2	4.11×10^1	0.1	0.0
(2) Dialysis sac contents					
a, sediment	filtrate	4.84×10^3	1.67×10^2	1.1	0.0
sac	filter	1.11×10^5	6.69×10^2	25.8	0.5
b, plankton	filtrate	1.04×10^4	2.44×10^2	2.4	0.1
sac	filter	7.28×10^4	4.37×10^2	16.9	0.3
(3) Subsamples		3.15×10^4	1.01×10^2	7.3	0.1
(4) Flask washdown and dialysis sacs		1.05×10^2	1.27	0.0	0.0
Total of (1), (2), (3), and (4)		4.30×10^5	7.85×10^3	100.0	2.1
Amount added†		4.27×10^5	9.39×10^3		
% Accounted for				100.7	2.9
% Unaccounted for				+0.7	2.9

*Single sample error values are one-sigma, propagated, counting error.

†Amount added = spiked water at day 0.

Table A-6. Mass balance calculations for ^{60}Co after 11 days of dialysis experiment.

		Activity (dpm)	± SD*	% Total accounted for	± SD*
(1) Water					
a, outside dialysis sacs	filtrate	2.13×10^6	7.45×10^4	54.2	2.5
	filter	2.59×10^4	5.57×10^3	0.8	0.2
b, inside control sac	filtrate	2.84×10^5	1.01×10^4	8.4	0.3
	filter	1.87×10^3	2.78×10^2	0.0	0.0
(2) Dialysis sac contents					
a, sediment sac	filtrate	4.71×10^4	1.56×10^3	1.4	0.1
	filter	1.20×10^6	6.59×10^3	30.5	0.7
b, plankton sac	filtrate	5.80×10^4	1.71×10^3	1.7	0.1
	filter	8.69×10^3	4.82×10^2	0.3	0.0
(3) Subsamples		1.76×10^5	7.18×10^2	4.5	0.1
(4) Flask washdown and dialysis sacs		5.45×10^1	8.2×10^{-1}	0.0	0.0
Total of (1), (2), (3), and (4)		3.93×10^6	7.57×10^4	101.8	2.7
Amount added†		3.62×10^6	9.13×10^4		
% Accounted for				108.6	3.5
% Unaccounted for				+ 8.6	3.5

*Single sample error values are one-sigma, propagated, counting error.

†Amount added = spiked water at day 0.

APPENDIX B

Distribution of radionuclides at selected times during a 96-hour dialysis experiment.

5 Tables

Table B-1. Distribution of ^{241}Am, ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{60}Co, and ^{59}Fe after two hours in a dialysis experiment

		Spiked Seawater	Dialysis Chambers			
			Blank	Plankton	Clay	Detritus
		cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D
^{241}Am	Soluble	152.5 (4.2)	5.56 (0.46)	1.36 (0.84)	2.02 (0.89)	1.08 (0.79)
	Particulate	705.3 (2.5	1.91 (0.47)	3.90 (0.53)	1.55 (0.23)	0.59 (0.45)
	% Particulate	82.2	25.6	74.1	43.4	35.3
^{137}Cs	Soluble	18.2 (0.92)	14.7 (0.40)	16.2 (0.76)	16.5 (0.76)	16.5 (0.72)
	Particulate	< 0.37	< 0.16	< 0.16	< 0.06	< 0.18
	% Particulate	< 2.0	< 1.1	< 1.0	< 0.4	< 1.1
^{106}Ru	Soluble	9.26 (0.76)	2.02 (0.21)	1.70 (0.41)	2.41 (0.45)	1.55 (0.43)
	Particulate	2.70 (0.37)	< 0.14	< 0.14	< 0.06	< 0.14
	% Particulate	22.6	< 6.5	< 7.6	< 2.3	< 8.3
^{65}Zn	Soluble	25.51 (0.97)	9.41 (0.32)	7.69 (0.55)	10.37 (0.62)	6.91 (0.52)
	Particulate	1.02 (0.30)	< 0.14	< 0.13	< 0.8	< 0.19
	% Particulate	3.8	< 1.15	< 1.7	< 7.2	< 2.7
^{60}Co	Soluble	45.98 (1.19)	16.32 (0.39)	8.79 (0.73)	17.09 (0.72)	17.11 (0.71)
	Particulate	2.61 (0.33)	< 0.12	< 0.13	< 0.06	< 0.14
	% Particulate	5.4	< 0.8	< 0.2	< 0.4	< 0.8
^{59}Fe	Soluble	11.01 (0.80)	0.31 (0.20)	0.81 (0.35)	0.44 (0.38)	0.45 (0.35)
	Particulate	30.31 (0.82)	< 0.10	< 0.11	< 0.05	0.15 (0.11)
	% Particulate	73.4	<24.4	<12.0	<10.2	25.0

Table B-2. Distribution of ^{241}Am, ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{60}Co and ^{59}Fe after ten hours in a dialysis experiment.

			Dialysis Chambers			
		Spiked Seawater cpm/ml±2 S D	Blank cpm/ml±2 S D	Plankton cpm/ml±2 S D	Clay cpm/ml±2 S D	Detritus cpm/ml±2 S D
^{241}Am	Soluble	116.5 (2.2)	15.79 (1.11)	18.19 (1.17)	15.55 (1.08)	18.10 (1.18)
	Particulate	451.5 (3.4)	11.47 (0.84)	49.20 (1.27)	21.81 (0.95)	43.94 (1.20)
	% Particulate	79.5	42.1	73.0	58.4	70.8
^{137}Cs	Soluble	21.5 (0.95)	20.3 (0.89)	21.6 (0.91)	20.9 (0.87)	19.2 (0.84)
	Particulate	< 0.30	< 0.19	< 0.20	< 0.19	< 0.19
	% Particulate	< 1.4	< 0.9	< 0.9	< 0.9	< 1.0
^{106}Ru	Soluble	5.45 (0.73)	4.65 (0.56)	4.31 (0.60)	4.24 (0.55)	4.01 (0.58)
	Particulate	1.86 (0.33)	0.17 (0.16)	1.46 (0.22)	< 0.18	0.35 (0.20)
	% Particulate	25.4	3.5	25.3	< 4.1	8.0
^{65}Zn	Soluble	23.65 (0.97)	17.67 (0.80)	5.63 (0.59)	13.66 (0.70)	7.28 (0.62)
	Particulate	0.33 (0.25)	0.21 (0.17)	0.41 (0.19)	0.51 (0.18)	1.45 (0.25)
	% Particulate	1.4	1.2	6.8	3.6	16.6
^{60}Co	Soluble	46.10 (1.20)	36.75 (1.07)	5.68 (1.10)	30.52 (0.97)	36.99 (0.97)
	Particulate	1.00 (0.28)	< 0.19	0.36 (0.21)	< 0.19	0.37 (0.21)
	% Particulate	2.1	< 0.5	6.0	< 0.6	1.0
^{65}Fe	Soluble	8.01 (0.74)	0.24 (0.13)	0.53 (0.25)	0.63 (0.47)	< 0.53
	Particulate	19.50 (0.66)	< 0.13	0.41 (0.14)	< 0.15	0.22 (0.16)
	% Particulate	70.9	<35.1	43.6	<19.2	> 29.3

Table B-3. Distribution of ^{241}Am, ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{60}Co and ^{59}Fe after 24 hours in a dialysis experiment.

		Spiked Seawater	Dialysis Chambers			
			Blank	Plankton	Clay	Detritus
		cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D
^{241}Am	Soluble	100.5 (2.0)	23.97 (1.26)	25.45 (1.31)	22.40 (1.24)	47.71 (1.55)
	Particulate	195.7 (2.3)	13.16 (0.68)	100.6 (1.7)	74.50 (1.50)	138.3 (1.9)
	% Particulate	66.1	35.4	79.8	76.9	74.4
^{137}Cs	Soluble	19.9 (0.93)	20.5 (0.90)	22.2 (0.88)	21.1 (0.93)	19.8 (0.93)
	Particulate	< 0.24	< 0.13	< 0.20	< 0.22	< 0.22
	% Particulate	< 1.4	< 0.6	< 0.9	< 1.1	< 1.1
^{106}Ru	Soluble	5.99 (0.66)	4.51 (0.63)	5.29 (0.60)	4.73 (0.62)	6.18 (0.62)
	Particulate	1.14 (0.25)	0.24 (0.12)	1.81 (0.25)	1.36 (0.21)	0.61 (0.28)
	% Particulate	16.0	5.0	25.5	22.3	9.0
^{65}Zn	Soluble	22.18 (0.92)	11.26 (0.71)	4.60 (0.59)	18.06 (0.85)	2.45 (0.57)
	Particulate	1.19 (0.23)	0.13 (0.12)	0.64 (0.19)	0.82 (0.23)	1.70 (0.24)
	% Particulate	5.1	1.1	12.2	4.3	41.0
^{60}Co	Soluble	41.89 (1.04)	43.63 (0.82)	4.76 (1.13)	36.68 (1.07)	44.28 (1.15)
	Particulate	0.46 (0.22)	< 0.13	0.48 (0.21)	< 0.22	0.37 (0.22)
	% Particulate	1.1	< 0.3	9.2	< 0.6	0.8
^{59}Fe	Soluble	6.76 (0.68)	1.22 (0.55)	1.06 (0.52)	2.20 (0.51)	1.55 (0.59)
	Particulate	8.26 (0.43)	0.44 (0.11)	1.13 (0.20)	1.20 (0.23)	0.86 (0.32)
	% Particulate	54.9	26.5	51.6	31.7	35.7

Table B-4. Distribution of ^{241}Am, ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{60}Co and ^{59}Fe after 48 hours in a dialysis experiment

		Spiked Seawater cpm/ml±2 S D	Dialysis Chambers: Blank cpm/ml±2 S D	Plankton cpm/ml±2 S D	Clay cpm/ml±2 S D	Detritus cpm/ml±2 S D
^{241}Am	Soluble	101.7 (2.1)	27.73 (1.38)	41.60 (0.88)	26.20 (1.34)	110.5 (2.2)
	Particulate	108.2 (2.4)	39.24 (1.18)	219.1 (2.4)	98.80 (1.73)	272.5 (2.6)
	% Particulate	51.4	58.6	84.0	79.0	71.1
^{137}Cs	Soluble	20.2 (0.92)	18.9 (0.90)	20.9 (0.50)	21.8 (0.93)	21.9 (0.94)
	Particulate	<0.23	<0.19	<0.21	<0.20	<0.22
	% Particulate	<1.1	<1.0	<1.0	<0.9	<1.0
^{106}Ru	Soluble	6.15 (0.68)	5.55 (0.62)	5.10 (0.36)	6.29 (0.63)	7.07 (0.67)
	Particulate	0.48 (0.21)	0.22 (0.17)	3.40 (0.31)	1.11 (0.23)	0.82 (0.23)
	% Particulate	7.2	3.8	40.0	15.0	10.4
^{65}Zn	Soluble	23.54 (0.93)	16.48 (0.85)	14.84 (0.44)	22.98 (0.90)	11.13 (0.72)
	Particulate	0.58 (0.19)	0.29 (0.17)	0.87 (0.24)	1.17 (0.24)	1.76 (0.25)
	% Particulate	2.4	1.7	5.6	4.8	13.7
^{60}Co	Soluble	44.28 (1.18)	43.26 (1.14)	15.01 (0.65)	39.40 (1.10)	45.88 (1.19)
	Particulate	<0.23	<0.20	0.70 (0.20)	0.27 (0.22)	0.53 (0.24)
	% Particulate	<0.4	<0.5	4.5	0.7	1.14
^{54}Fe	Soluble	6.40 (0.67)	1.57 (0.59)	1.71 (0.31)	1.81 (0.56)	3.86 (0.66)
	Particulate	4.56 (0.34)	0.93 (0.18)	2.30 (0.28)	2.63 (0.28)	1.90 (0.26)
	% Particulate	41.6	37.2	57.4	59.2	33.0

Table B-5. Distribution of ^{241}Am, ^{137}Cs, ^{106}Ru, ^{65}Zn, ^{60}Co and ^{59}Fe after 96 hours in a dialysis experiment.

		Spiked Seawater	Dialysis Chambers			
			Blank	Plankton	Clay	Detritus
		cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D	cpm/ml±2 S D
^{241}Am	Soluble	134.5 (2.3)	33.99 (0.85)	54.75 (1.71)	26.84 (1.40)	134.1 (2.4)
	Particulate	62.7 (1.4)	68.95 (1.40)	357.4 (3.0)	149.4 (1.1)	555.6 (3.7)
	% Particulate	31.8	66.3	86.7	84.8	80.6
^{137}Cs	Soluble	21.2 (0.92)	20.3 (0.51)	20.4 (0.95)	23.2 (0.93)	23.0 (0.93)
	Particulate	<0.19	<0.20	<0.23	<0.15	<0.24
	% Particulate	<0.9	<1.0	<1.1	<0.6	<1.1
^{106}Ru	Soluble	4.73 (0.68)	5.55 (0.35)	6.05 (0.65)	6.41 (0.62)	5.39 (0.68)
	Particulate	0.30 (0.18)	0.33 (0.20)	3.87 (0.34)	1.58 (0.16)	1.05 (0.26)
	% Particulate	6.0	5.6	39.0	19.8	16.3
^{65}Zn	Soluble	23.11 (0.93)	20.25 (0.48)	16.03 (0.82)	19.48 (0.85)	8.40 (0.69)
	Particulate	<0.19	0.47 (0.47)	1.50 (0.26)	1.87 (0.16)	1.94 (0.27)
	% Particulate	<0.8	2.3	8.6	8.8	18.8
^{60}Co	Soluble	42.04 (1.16)	47.13 (0.69)	16.21 (1.19)	37.76 (1.07)	47.40 (1.12)
	Particulate	<0.21	0.40 (0.22)	1.32 (0.20)	1.03 (0.16)	0.40 (0.23)
	% Particulate	<0.3	0.8	7.53	2.7	0.8
^{59}Fe	Soluble	5.44 (0.66)	1.73 (0.34)	2.49 (0.58)	1.93 (0.58)	3.11 (0.66)
	Particulate	2.96 (0.28)	1.95 (0.23)	5.43 (0.38)	6.53 (0.22)	5.38 (0.35)
	% Particulate	35.2	53.0	68.6	77.2	63.4

APPENDIX C

Distribution of radionuclides in soluble or particulate phase during 96-hour dialysis experiment.

11 Figures

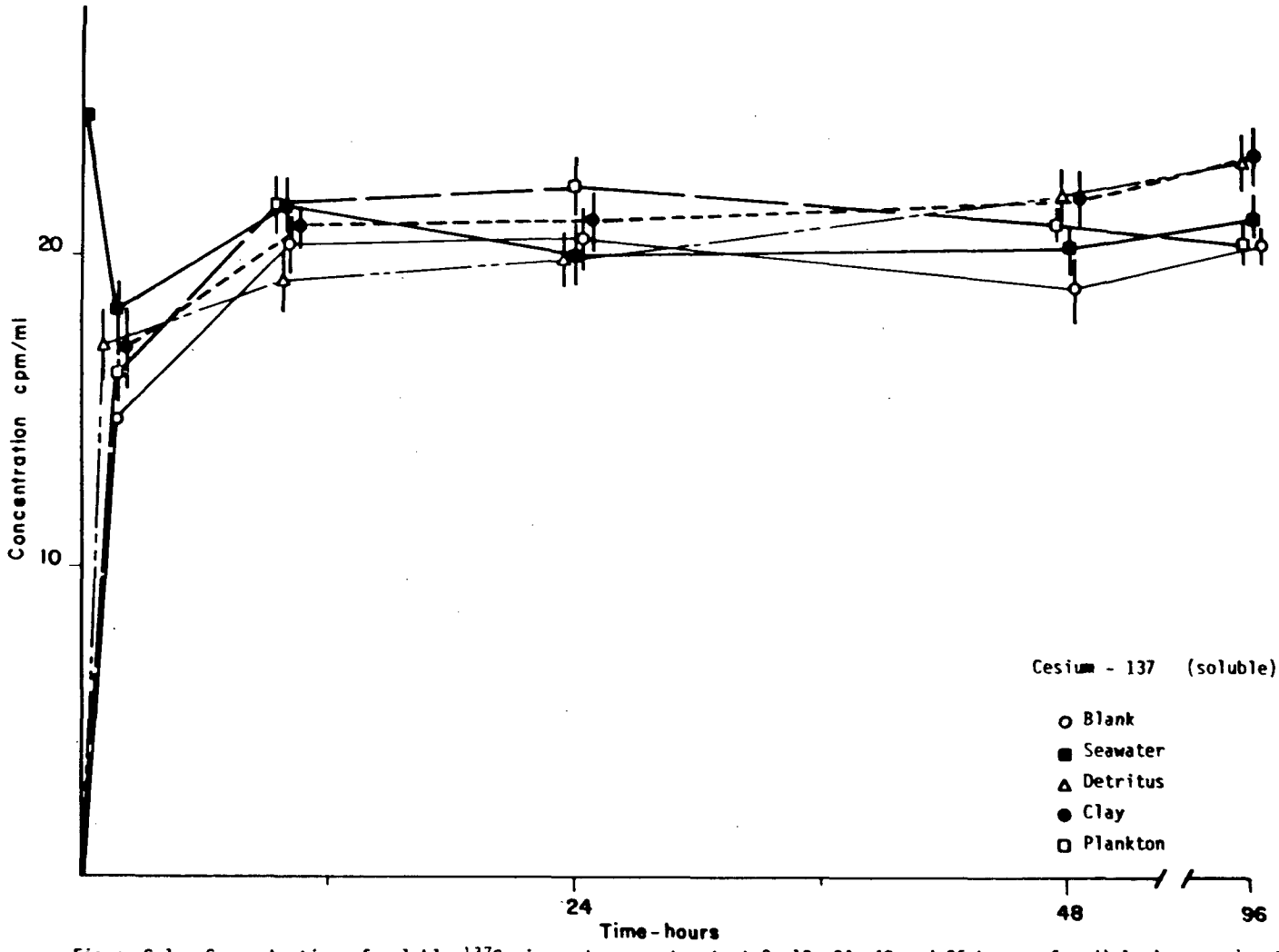

Figure C-1. Concentration of soluble ^{137}Cs in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

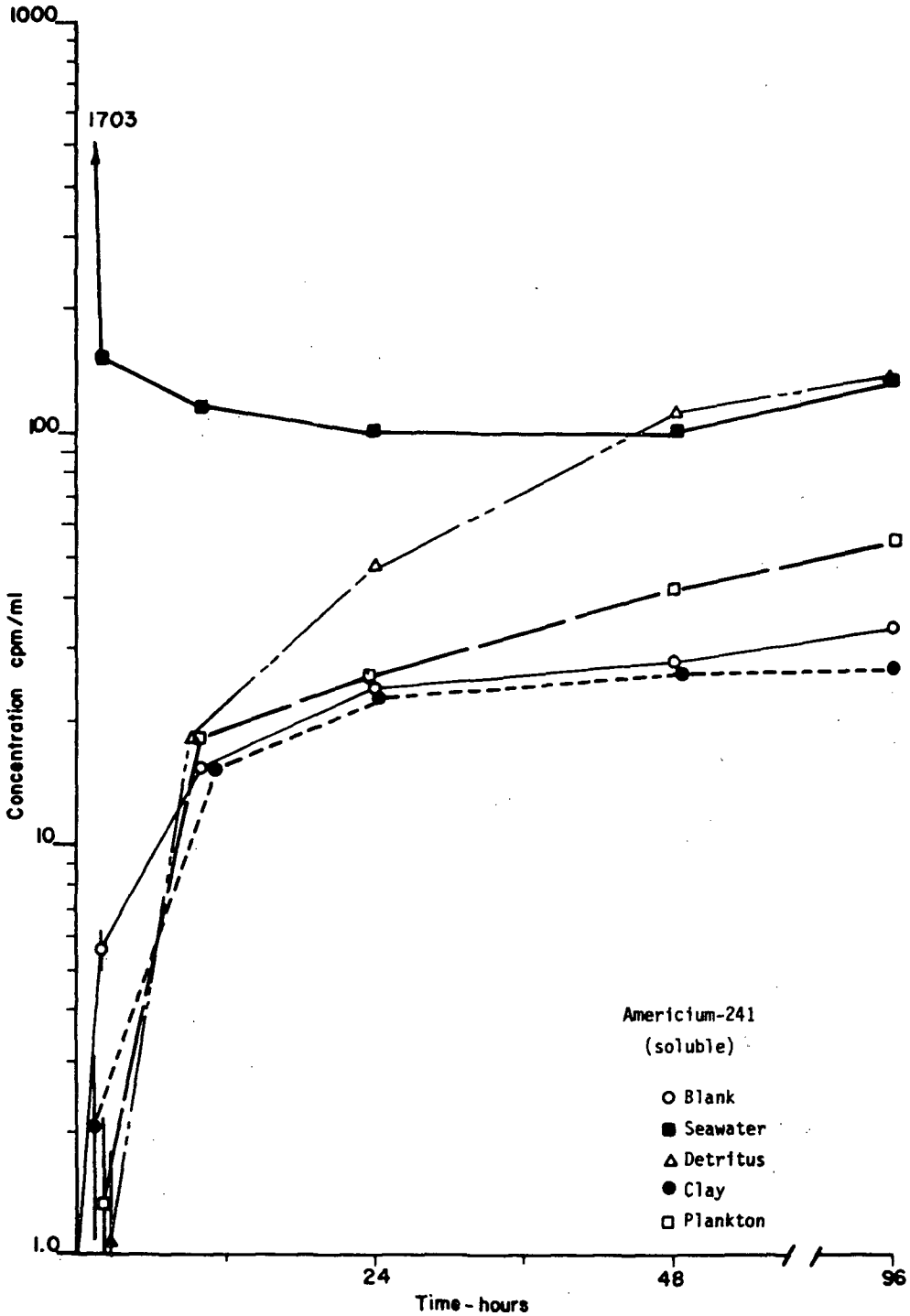

Figure C-2. Concentration of soluble ^{241}Am in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

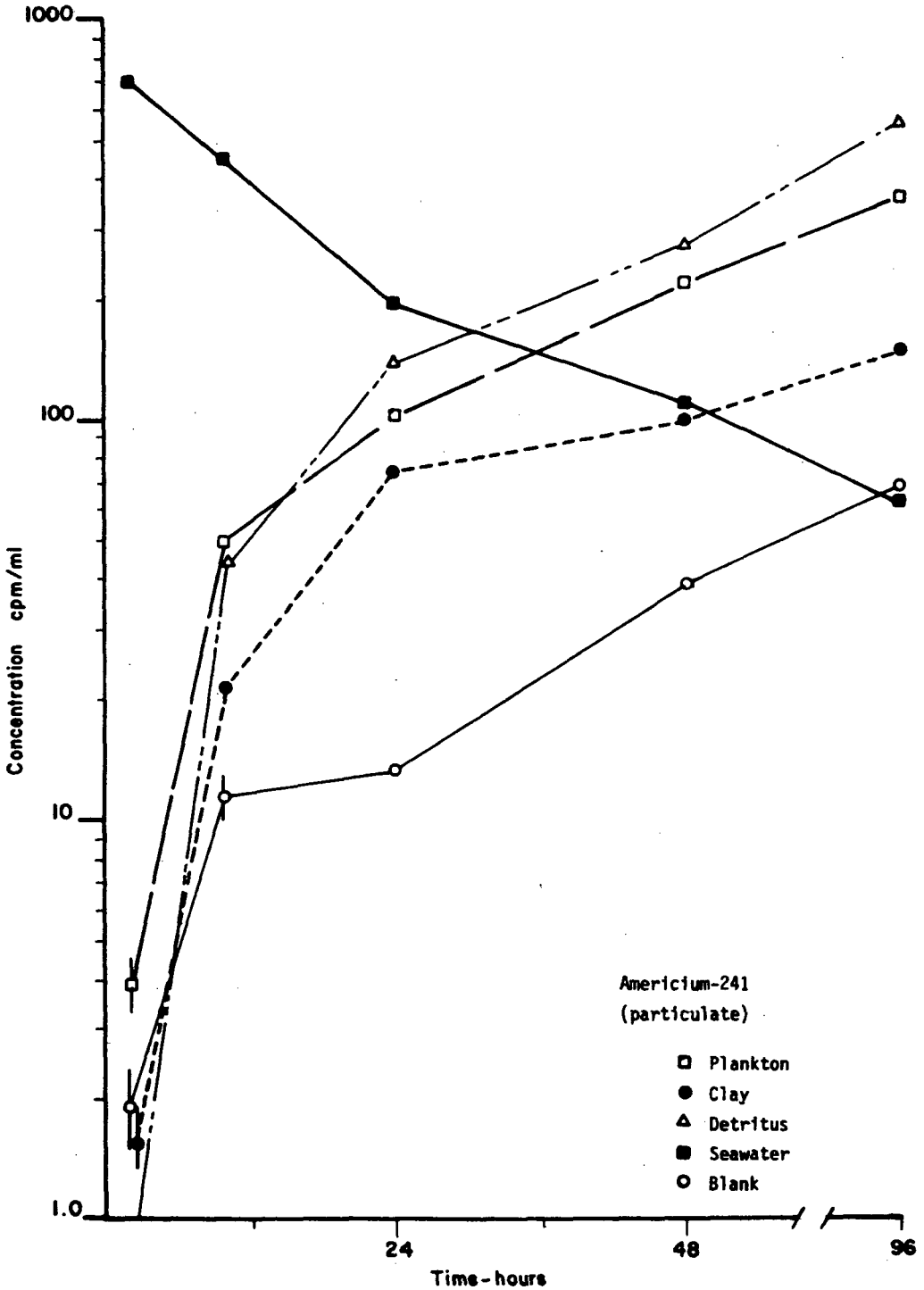

Figure C-3. Concentration of particulate ^{241}Am in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

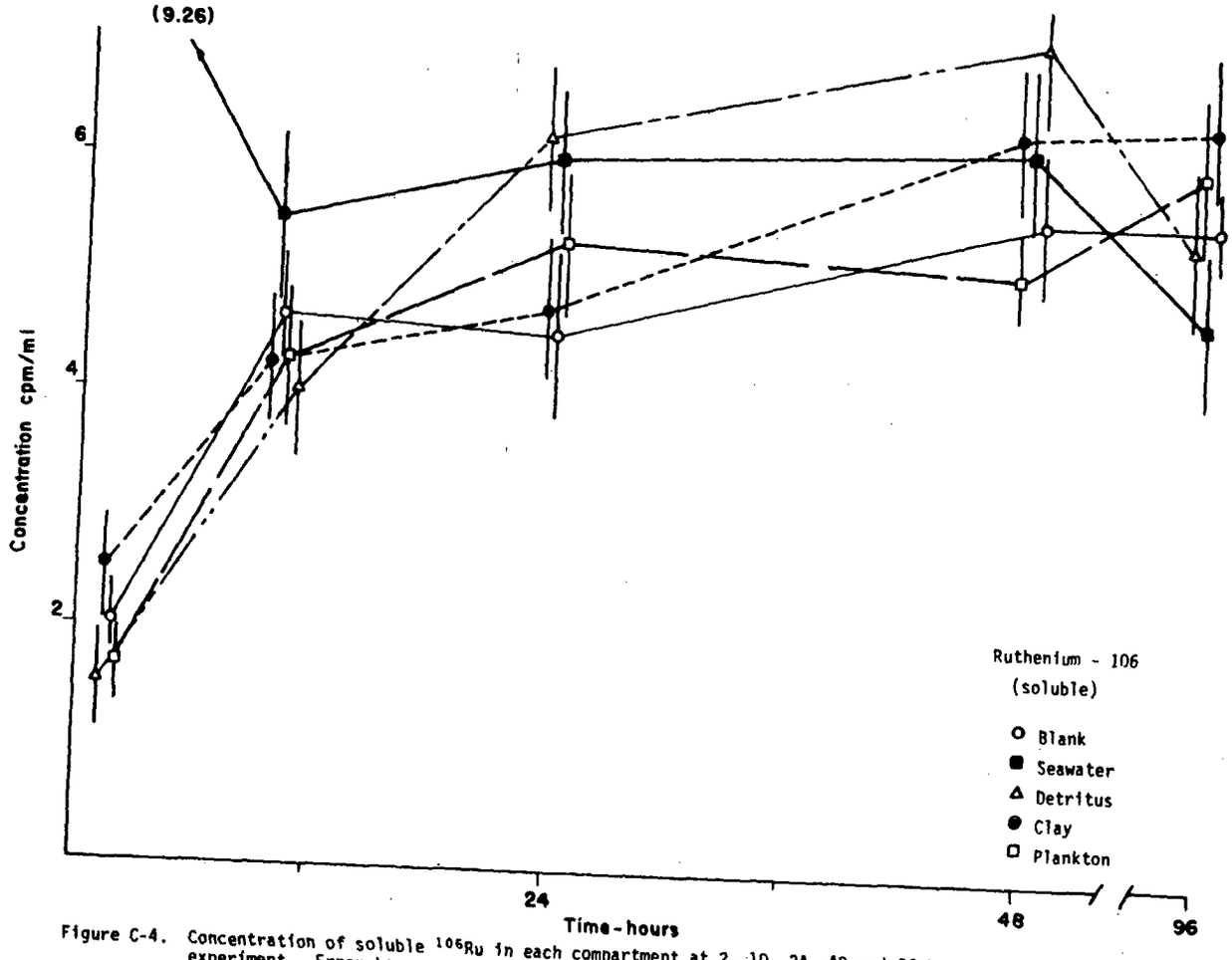

Figure C-4. Concentration of soluble ^{106}Ru in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

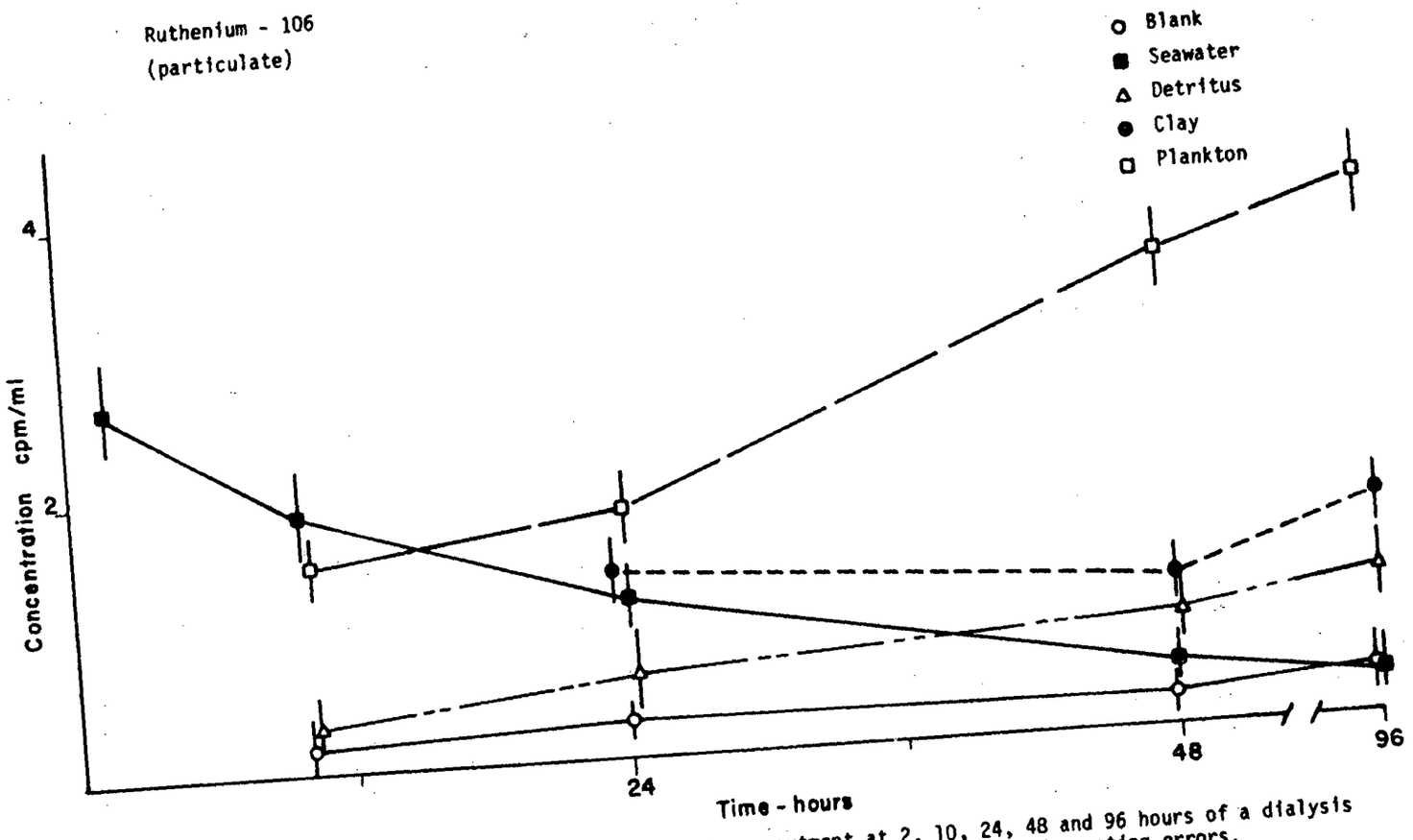

Figure C-5. Concentration of particulate ^{106}Ru in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

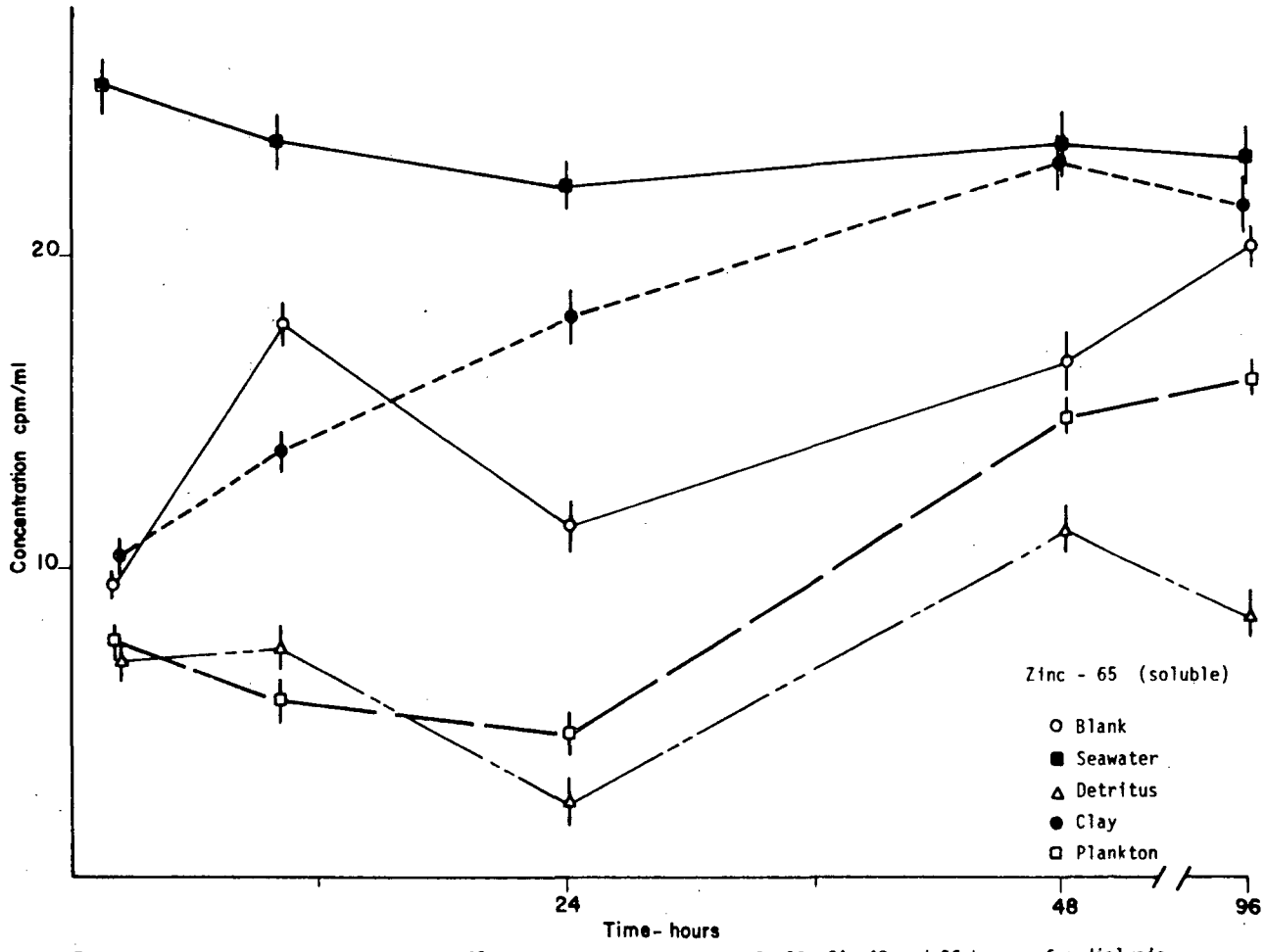

Figure C-6. Concentration of soluble ^{65}Zn in each compartment at 2, 10, 24, 43 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

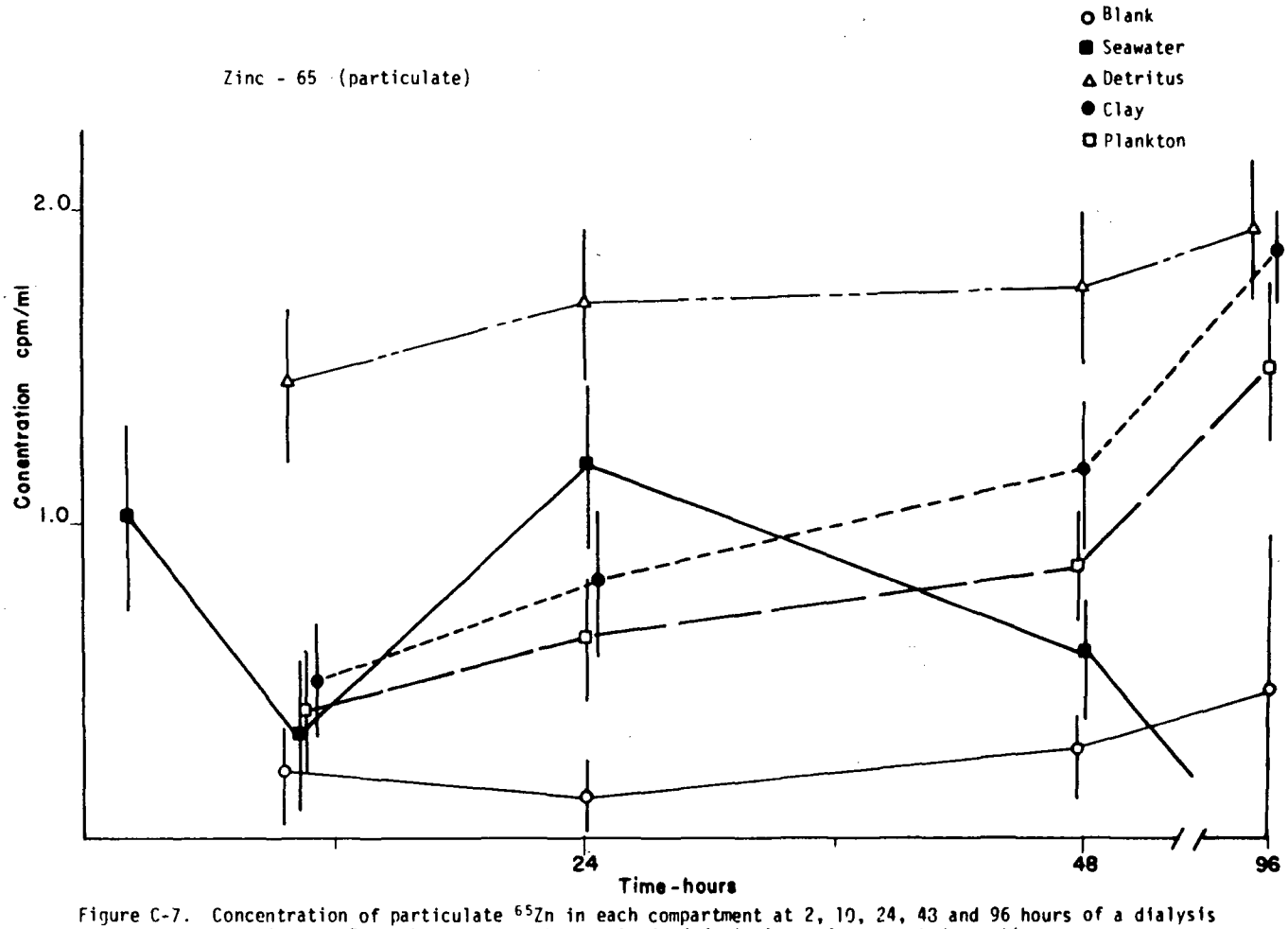

Figure C-7. Concentration of particulate ^{65}Zn in each compartment at 2, 10, 24, 43 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

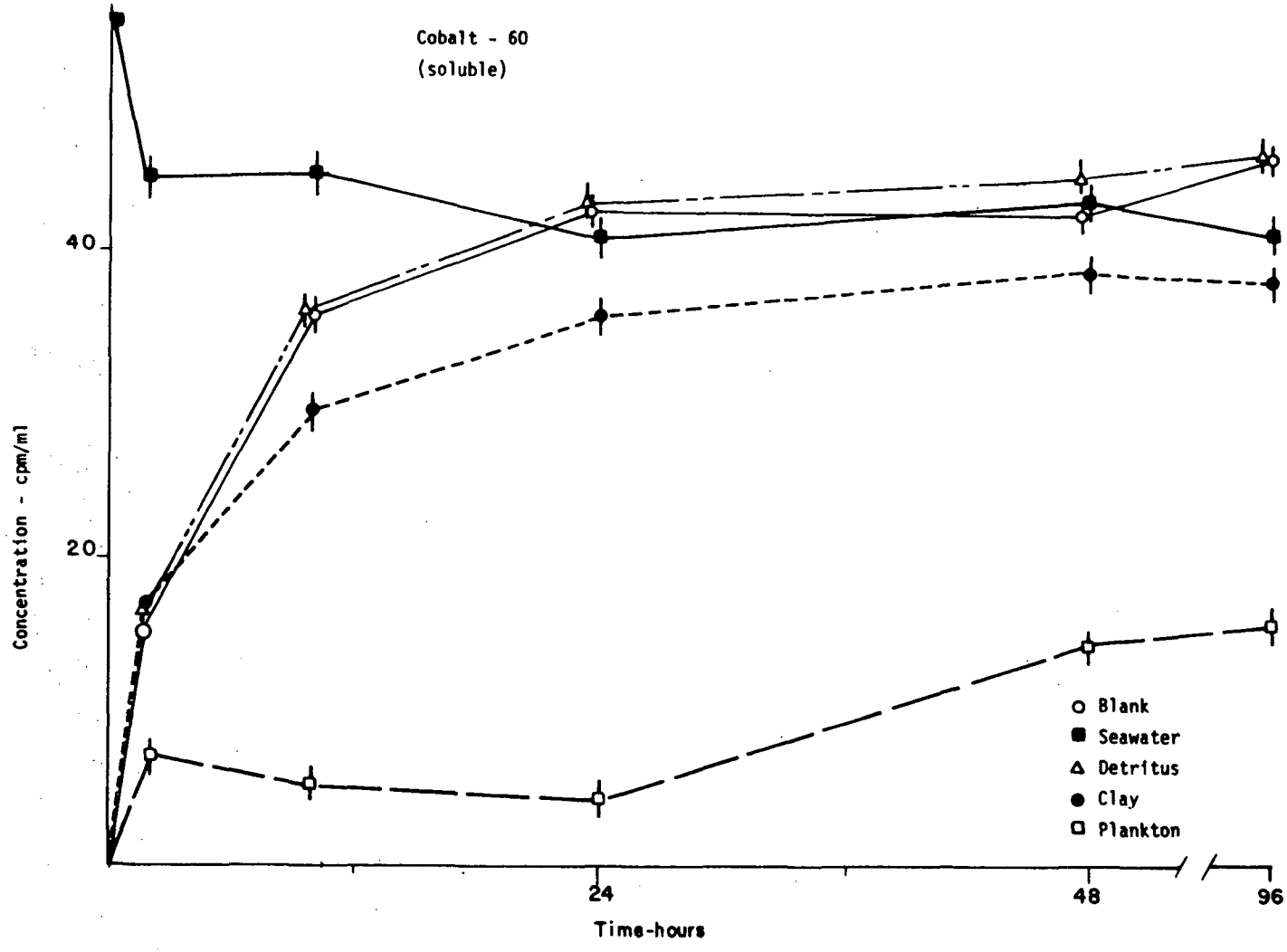

Figure C-8. Concentration of soluble ^{60}Co in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

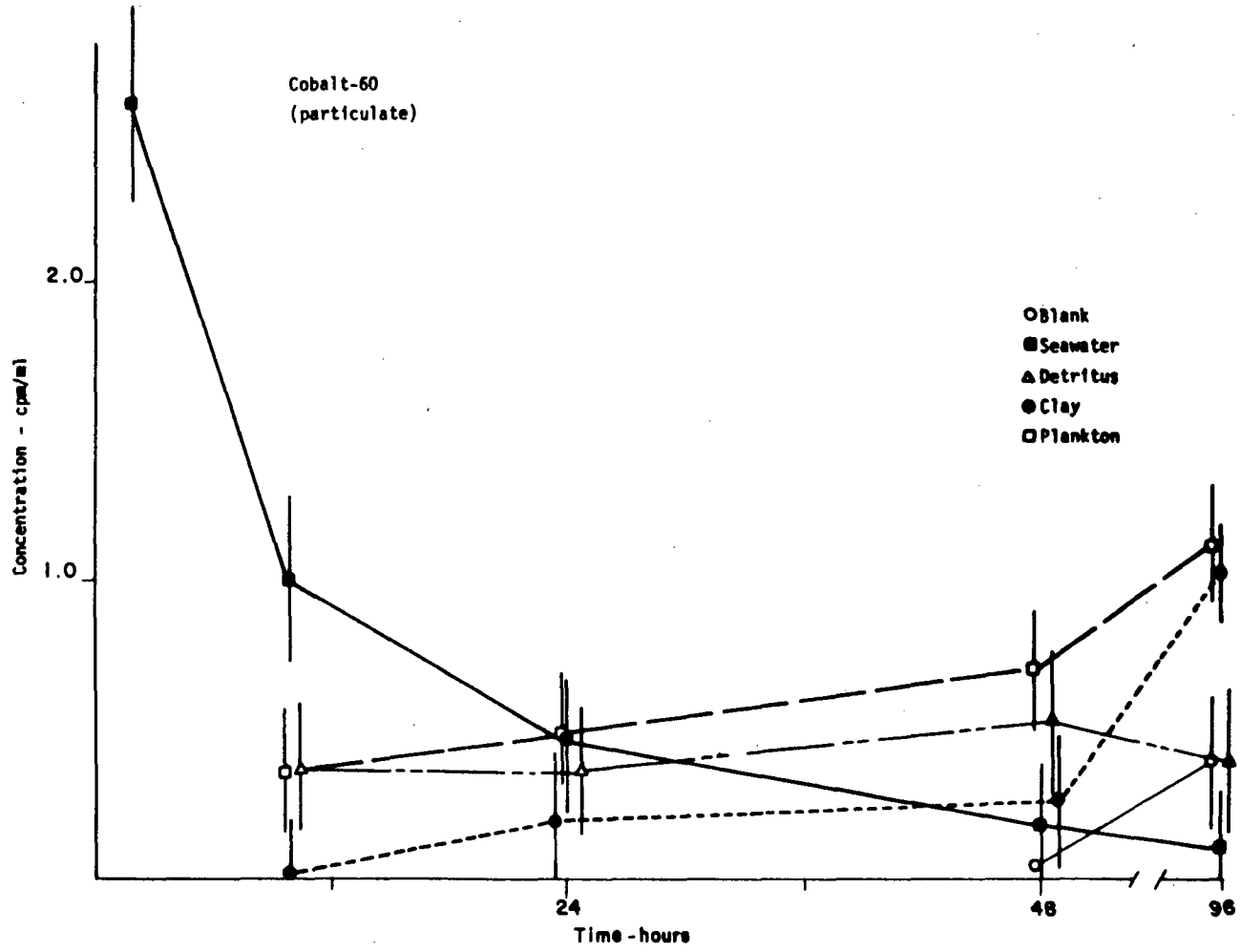

Figure C-9. Concentrations of particulate ^{60}Co in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

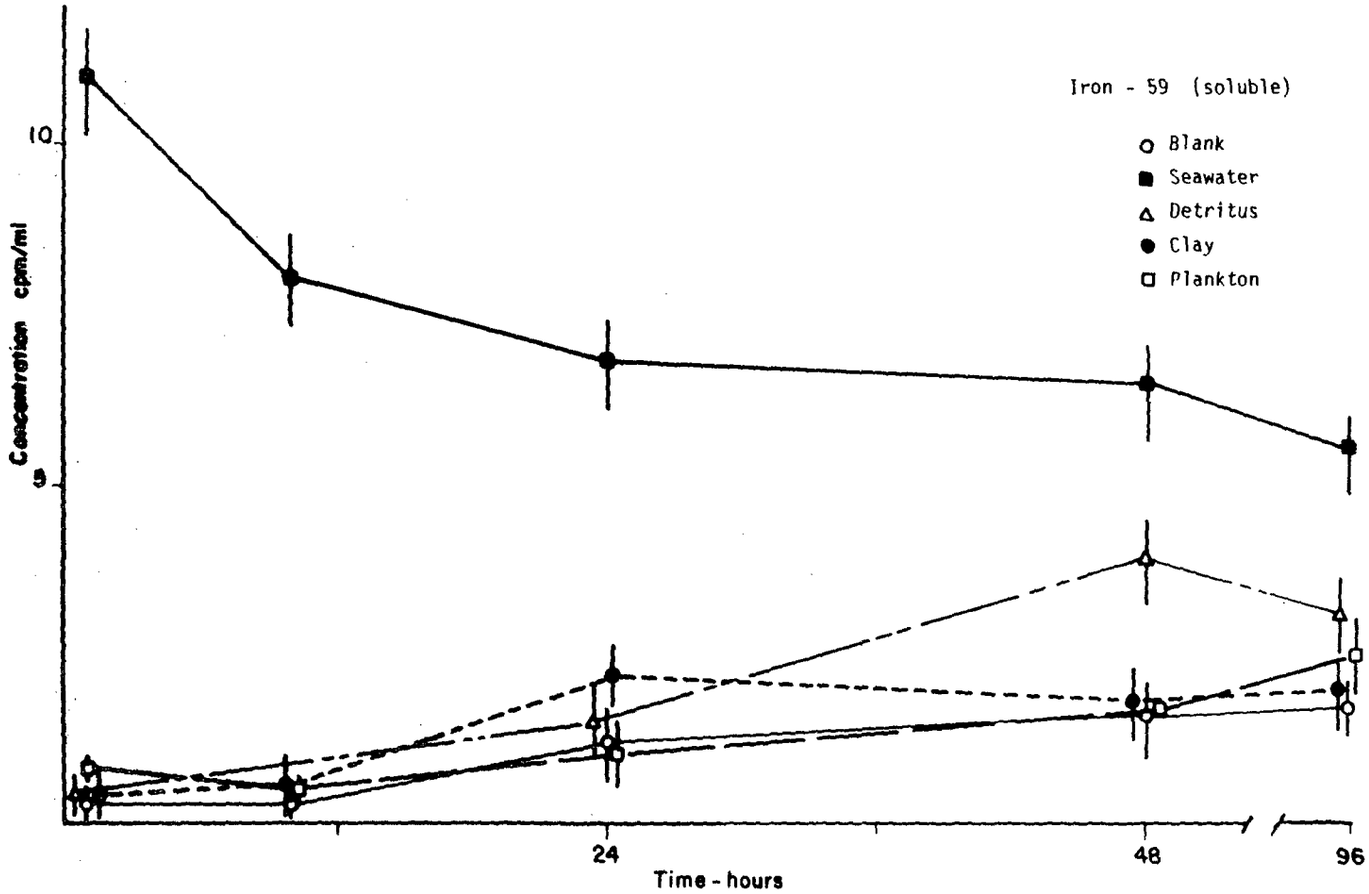

Figure C-10. Concentrations of soluble ^{59}Fe in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

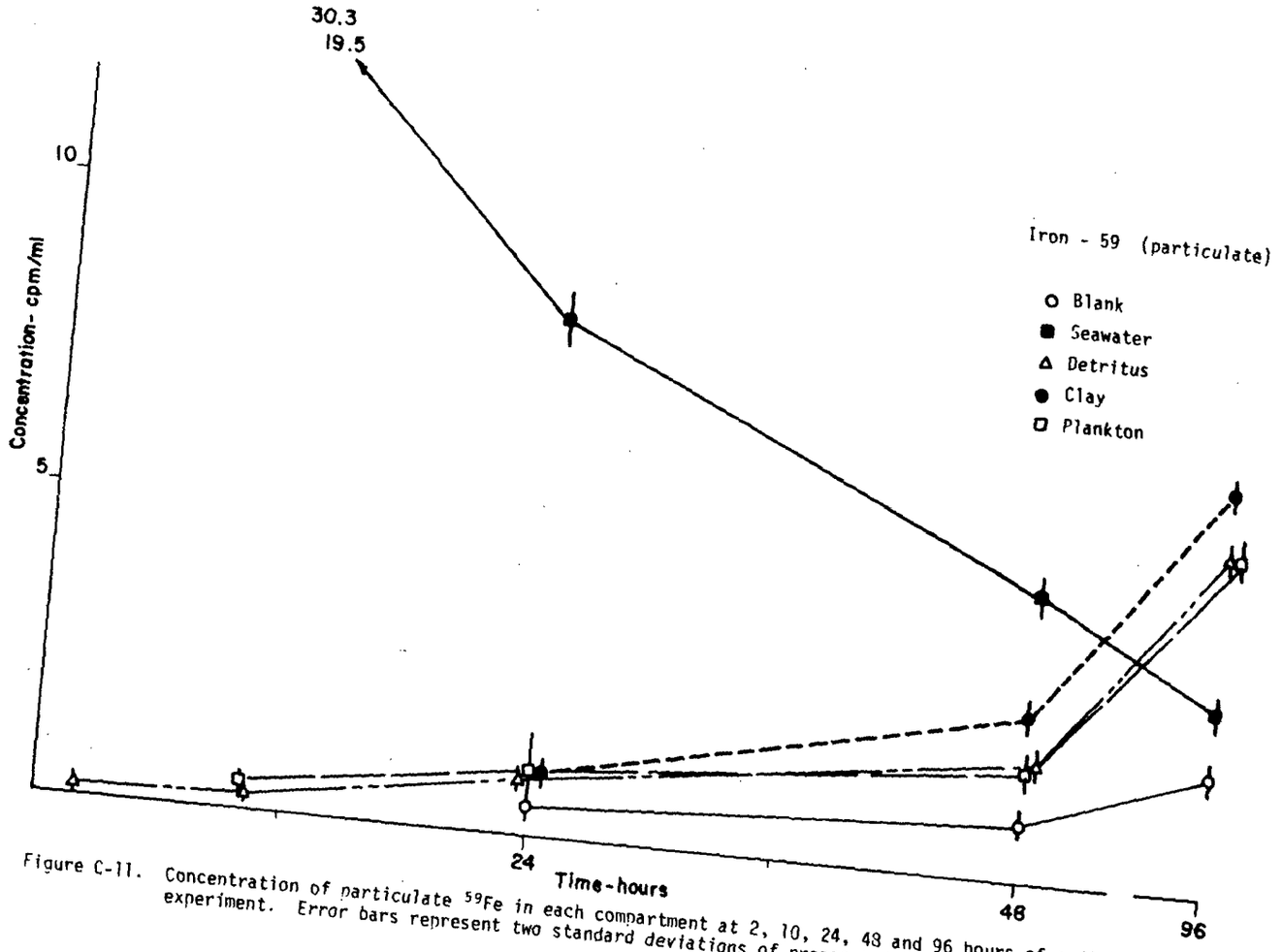

Figure C-11. Concentration of particulate ^{59}Fe in each compartment at 2, 10, 24, 48 and 96 hours of a dialysis experiment. Error bars represent two standard deviations of propagated counting errors.

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16. ABSTRACT *(200 words or less)*

The objective of this study was to obtain information that can be used to predict the fate of radionuclides that may enter the aquatic environment from nuclear power plants or fuel reprocessing plants. Constant shaking experiments were used to determine distribution coefficients for pure clay and natural sediments. Dialysis experiments were used to study the distribution of radionuclides among suspended sediments, phytoplankton, organic detritus, and filtered seawater. The value of distribution coefficients varies considerably from one environment to another and for different experimental conditions such as pH. These differences probably result from the presence of different physico-chemical states of the radionuclides. Studies on the uptake of radionuclides and phytoplankton indicated that the time required to reach equilibrium may be longer, greater than 100 hours, than previously assumed. Some radionuclides (e.g., ^{241}Am) have an especially high affinity for organic detritus Additional experiments to compliment the results of this study were identified.

17. KEY WORDS AND DOCUMENT ANALYSIS

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Marine-Water Sediment
Ru-106

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Am-241
Physico-Chemical states
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