
Distribution Coefficients for Radionuclides in Aquatic Environments

III. Adsorption and Desorption Studies of ^{106}Ru , ^{137}Cs , ^{241}Am , ^{85}Sr and ^{237}Pu in Marine and Freshwater Systems

Annual Report: August 1978 - July 1979

Prepared by W. R. Schell, A. L. Sanchez, T. H. Sibley, J. R. Clayton, Jr.

University of Washington

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Commission

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ABSTRACT

Sediments potentially serve as important sinks and sources for radionuclides released into aquatic systems. In this regard, we have collected data over the past year to evaluate mechanisms affecting particle-water distributions of selected radionuclides (^{57}Co , ^{60}Co , ^{85}Sr , ^{106}Ru , ^{137}Cs , ^{237}Pu , ^{241}Am , and ^{244}Cm). Samples used in these studies were obtained from several natural freshwater, estuarine, and marine locations around the continental United States. Particle-water distribution coefficients (K_d) were found to vary from 10^2 to 10^6 depending on the specific radionuclide, the type of particle(s), and the overall physical and chemical properties of a given test system. In general, K_d values increased as follows: $^{85}\text{Sr} < ^{137}\text{Cs} < ^{106}\text{Ru}$ or $^{237}\text{Pu} < ^{241}\text{Am}$. However, both the absolute values of these K_d 's and their magnitudes relative to other radionuclides were found to be dramatically and nonuniformly affected by certain variables. The variables tested and discussed in this report include: the quantity and chemical composition of the particles, the degree of reversibility of radionuclide-sediment interactions, the pH of the water, the abundance and composition of organic ligands and the chemical speciation (i.e., molecular weight size associations) of the radionuclides. This data will contribute to the formulation of predictive models to describe distributions and eventual fates of radionuclides in aquatic environments.

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LIST OF PREVIOUS REPORTS

Distribution Coefficients for Radionuclides in Aquatic Environments.

I. Development of Methods and Results for Plutonium and Americium in Fresh and Marine Water-Sediment Systems. Annual Report: August 1976 - July 1977. A.H. Seymour, A. Nevissi, W.R. Schell and A. Sanchez. NUREG/CR-0801.

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 . Annual Report: August 1977 - July 1978. W.R. Schell, T.H. Sibley, A. Nevissi and A. Sanchez. NUREG/CR-0802.

DISTRIBUTION COEFFICIENTS FOR
RADIONUCLIDES IN AQUATIC ENVIRONMENTS

3. Adsorption and Desorption Studies of ^{106}Ru , ^{137}Cs ,
 ^{241}Am , ^{85}Sr , and ^{237}Pu in Marine and Freshwater Systems

1.0 INTRODUCTION

This report presents experimental results obtained between August 1, 1978 and July 31, 1979 to determine distribution coefficients of selected radionuclides (^{57}Co , ^{60}Co , ^{85}Sr , ^{106}Ru , ^{137}Cs , ^{237}Pu , ^{241}Am , and ^{244}Cm) in laboratory sediment-water systems. The report covers the third year of a proposed 4-year research program, started in August 1976, to obtain new and better information for predicting the fate of radionuclides in aquatic ecosystems. Between August 1978 and 31 July 1979 this program has been supported by contract No. NRC-04-76-352 for \$140,000.

There are four principal sections in the report: Introduction, Materials and Methods, Results and Discussion, and Conclusions and Recommendations.

1.1 Background

The transport and eventual fate of radionuclides in aquatic ecosystems is affected by numerous physical, chemical, and biological processes including adsorption to inorganic and organic particulate matter. The relative affinity of radionuclides for various types of suspended particulates can be described by the use of distribution coefficients, K_d 's,* a ratio that compares the quantity of radionuclide in the particulate phase to the quantity in solution. Since K_d values provide a measure of the uptake and accumulation of radionuclides by particulates, they are an important parameter for predicting both the hydrological transport and the eventual fate of radionuclides released into aquatic environments.

Although a great deal of research has been conducted to determine K_d values of some radionuclides, there is remarkably little information for the transuranic elements. Therefore, this program has started to obtain K_d values for plutonium and americium in selected marine and freshwater environments. It has since been expanded to include the radionuclides ^{85}Sr , ^{106}Ru , ^{137}Cs and ^{244}Cm , in sediment-water systems that are of particular concern to the Nuclear Regulatory Commission (NRC).

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

In August 1976 we began to investigate adsorption and desorption phenomena of ^{237}Pu and ^{241}Am in freshwater and marine sediment-water systems. During the first year experimental methods and analytical techniques were developed to determine K_d values using the constant shaking and thin-layer techniques (1). Results for preliminary experiments using ^{65}Zn and ^{137}Cs were compared to published results (2) in order to evaluate our experimental techniques. Sediments and water collected from Lake Washington (freshwater), Lake Nitinat (anoxic marine) and the mouth of the Columbia River (oxic marine) were used to determine distribution coefficients for ^{237}Pu and ^{241}Am in laboratory sediment-water systems. Dialysis experiments were used to study the distribution of ^{60}Co , ^{65}Zn , ^{137}Cs , ^{239}Pu and ^{241}Am among suspended sediments, phytoplankton and filtered seawater. Since diffusion across the dialysis membranes is limited to chemical species of sizes less than 6000-8000 nominal molecular weight, these experiments provide information on the effect of different chemical states on the uptake of radionuclides by sediments and phytoplankton. Results from the first year of research are included in NUREG/CR0801 (3).

During FY 77-78 the project was extended to include new sediment types and additional radionuclides. We also evaluated the sedimentation technique (1) for determining K_d 's of ^{237}Pu and ^{241}Am , the effect of pH on the distribution coefficient of ^{241}Am in sediment-water systems from Lake Nitinat and Lake Washington and particle formation by ^{106}Ru and ^{241}Am in the absence of suspended sediments. These results are available in NUREG/CR0802 (4).

Constant shaking experiments utilized ^{85}Sr , ^{106}Ru , ^{137}Cs and ^{241}Am in sediment-water systems from three locations in Skagit Bay, an estuary in Western Washington, and a single sample from Saanich Inlet, an anoxic fjord in British Columbia. In addition, sediments and water were obtained from several nonlocal sources as requested by NRC. These included Clinch River in Tennessee, Lake Michigan, three locations in the Hudson River Estuary, and Buttermilk and Cattaraugus Creeks downstream from the Nuclear Fuel Services Waste Storage Facility in West Valley, New York. Distribution coefficients for radionuclides in sediment-water systems from these locations have been determined during the past year and are included in this report.

Dialysis experiments were expanded in FY 77-78 to include the radionuclides ^{59}Fe , ^{60}Co , ^{65}Zn , ^{137}Cs , ^{207}Bi , ^{238}Pu and ^{241}Am . Additional sacs were added containing a blank of filtered seawater and organic detritus as well as the sediment and plankton. During the past year, analysis experiments have utilized sediment and water from Lake Michigan and the radionuclides ^{85}Sr , ^{60}Co , ^{106}Ru , ^{137}Cs , ^{239}Pu , and ^{241}Am .

This project was originally proposed as a 3-year program with the third year used for reducing and evaluating data and preparing a final report. However, with the addition of new radionuclides and an

increase in the number of sediment-water systems, an additional year was added in order to complete the experimental program.

1.2 Objectives

The general objective for this program is to obtain new and better information for predicting the fate of radionuclides that may be released from nuclear power plants or waste storage facilities into aquatic environments.

In particular, these studies are intended to obtain information on the accumulation of radionuclides by suspended particulate matter. These data will contribute important parameters for modeling the hydrological transport of radionuclides in marine, estuarine and freshwater environments. Therefore, it will be easier to predict the eventual environmental distribution.

The principal objectives for the past year have been:

1. To obtain distribution coefficients for all the sediment-water systems that we have available.
2. To begin investigating the effects of various physico-chemical parameters including pH and organic ligands on distribution coefficients.

1.3 Personnel

The senior persons for this project were Drs. W. R. Schell, principal investigator, and T. H. Sibley, research associate. Arthur Sanchez and John Clayton have served as pre-doctoral research associates and other staff personnel have been utilized as required for preparation and analysis of samples and data reduction.

2.0 MATERIALS AND METHODS

The experimental procedures used in this research were adapted from published reports of other workers and modified as needed for our experiments. In particular, the methods developed by E.K. Duursma and his colleagues at International Atomic Energy Agency (IAEA) Marine Laboratory, Monaco, have been used extensively. Our application of these techniques has been described previously (3, 4) and will be discussed briefly below.

2.1 Sources and Types of Samples

For the first 2 years of this program, experiments were conducted with sediments and water from freshwater and marine environments in Western Washington. A few of these samples were also used for experiments during the past year. Information for these samples is provided in Table 1.

The majority of our experiments during the past year have utilized sediments and water from non-local environments. Many of these samples were collected in collaboration with other laboratories. In particular, Argonne National Laboratories (ANL) provided sediments and water from Lake Michigan and Lamont-Doherty Geological Laboratories (LDGL) provided samples from three different locations in the Hudson River estuary. Several samples from Cattaraugus and Buttermilk Creeks were provided by Battelle Pacific Northwest Laboratories (PNL) or obtained during joint sampling trips between PNL and University of Washington, Laboratory of Radiation Ecology (LRE). Finally, samples from the Clinch River, Tennessee, were collected by Dr. Ahmad Nevissi of our laboratory with the cooperation of Oak Ridge National Laboratories (ONL). Specific information for nonlocal samples is provided in Table 2. Although many of these samples were collected in FY 77-78, they were not utilized for experiments until the past year.

A question has arisen regarding the representative nature of K_d values obtained from samples which were frozen or freeze-dried before use. These values also must be compared to what is occurring naturally in the field. Ideally, measurements should be made under natural conditions but this is often difficult because of the low radionuclide concentrations or the unavailability of suitable field equipment. Thus, any measurement made in the laboratory is a compromise, often with several major uncertainties. The purpose of these studies is to use representative natural sediment and water to measure the K_d values. In most cases the samples have been used fresh or used after storage at -40°C to minimize alterations by chemical or microbial activity. Several samples were frozen to limit changes in sediment or water properties and to decrease microbial activity until used. The Hudson River sediment samples were freeze-dried and then rehydrated with water collected above the sediments. This would maintain the

Table 1. Sampling stations in local environments for sediment and water samples used in distribution coefficient studies.

Sample station	Temperature °C	Salinity 0/00	Depth m	Sediment depth m	Sediment treatment	Remarks
<u>MARINE SAMPLES</u>						
Saanich Inlet						
<u>Station</u> 48° 35' 4" N 123° 30' 2" W	8.9	31.24	224	225	Fresh	Anoxic sediment and water
Skagit River Estuary						
<u>Station 1</u> 48° 18' 0" N 122° 29' 0"	10	29.62	2	9	Fresh	Sandy sediment
<u>Station 4</u> 48° 15' 5" 122° 32' 5"	10	31.05		59	Fresh	Silt and clay
Sinclair Inlet						
<u>Station 1</u>	9.6	23.91	30	31	Fresh	Oxic organic rich sediment
<u>Station 2</u>	9.5	28.89	14.5	15.1	Fresh	Oxic organic rich sediment
<u>Station 3</u>	9.5	28.81	3.5	4.5	Fresh	Oxic organic rich sediment

Table 2. Sampling stations in non-local environments for sediment and water samples used in distribution coefficient studies.

Sample station	Location	Remarks	Sediment treatment
Cattaraugus Creek BC-4 CC-11	Mouth of Cattaraugus Creek	Samples collected by PNL Samples collected by PNL	Fresh
Lake Michigan <u>Station 5</u>	Near the east shore 67 m deep	Sample collected by ANL	Frozen
Clinch River <u>Station 1</u>	1.5 miles downstream from the breeder reactor site 3 m deep	Sample collected with cooperation of ONL	Fresh
Hudson River <u>Station</u>		Samples collected by LDGL	
Sediments	SLOSH* III mp# 43	June 11, 1975	Freeze-dried
	SLOSH II mp# 18	June 2, 1975	"
	SLOSH V mp# 0	Sept. 15, 1975	"
Water	mp# 0	15 o/oo salinity	
	mp# 18	2-3 o/oo salinity	
	mp# 40	Fresh water	

* SLOSH = Standard Lamont Observatory Sediments from the Hudson.

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

#mp represents miles upstream from the southern tip of Manhattan.

elements present in pore water with the sediment and would dehydrate and maintain the clay lattice in an expanded form. It is expected that freeze drying would maintain the sediment structure better than the 80°C heat evaporation-dehydration.

In addition to natural sediments we have also used Gelwhite L, a reference calcium montmorillonite, supplied by the Georgia Kaolin Company.

2.2 Constant Shaking Experiments

Adsorption K_d Values. Experiments were conducted to determine distribution coefficients in laboratory sediment-water systems using the constant shaking technique (1, 5). Briefly, the method consists of:

1. Adding the radionuclides to a known volume of filtered (0.45 μm or 0.2 μm) water sample and adjusting the pH to the initial pH of the water sample with 0.2 M NaOH.
2. Adding sediments from a stock sediment suspension to make a final predetermined sediment concentration, generally 200 mg/l.
3. Shaking the sediment-water mixture at 200 rpm in a constant temperature (5-8°C) cold room.
4. Collecting samples at designated time intervals during the experiment, filtering to separate particulate and dissolved radionuclides and measuring the concentration of radionuclides in the dissolved and particulate phases.

The effects of pH were studied by altering the pH with NaOH or HCl and determining K_d at different pH values. Similarly, the effect of sediment concentration was studied by adding different amounts of suspended sediments. Preliminary experiments with organic ligands were conducted by adding known concentrations of selected ligands to radionuclide spiked sediment-water systems.

Desorption K_d Values: Following adsorption of radionuclides on to sediments the spiked suspensions were centrifuged to separate the sediments. The centrifuged sediments were then resuspended in a non-radioactive water sample to make a sediment concentration of 200 mg/l. The suspensions were then shaken in the cold room and sampled at designated intervals as for the adsorption experiments.

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 with vanes connected to a small electric motor. Dialysis membranes permeable to molecules smaller than 6000-8000 molecular weight have been used for all the experiments.

Samples are removed from the dialysis sacs and the outside chamber at predetermined times and filtered through Millipore or Nuclepore membrane filters in order to separate the particulate and soluble phases. Both the filtrate and filtrate are then measured to determine the concentration of particulate and dissolved radionuclides in each chamber of the experiment.

2.4 Measurement of Radioactivity

The samples from constant shaking and dialysis experiments were placed in standard sample holders, 1.3 cm x 5 cm (2 dram) polyethylene vials, and sealed for gamma counting. The gamma emitting radionuclides, ^{57}Co , ^{60}Co , ^{106}Ru , ^{137}Cs , and ^{241}Am , were measured on a Ge(Li) detector connected to a computer based multi-channel analyzer which integrates the counts for each peak, performs a linear background subtraction and prints out the 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 ^{85}Sr and ^{237}Pu , we used a 2" NaI(Tl) well crystal and two single-channel analyzers. Appropriate corrections were made to account for the contribution of each radionuclide to the other's analyzer.

Initial attempts to determine ^{244}Cm concentrations by alpha spectroscopy using the method of Bojanowski et al. (6) proved unsatisfactory because of low yields of the ^{243}Am tracer and the considerable amount of time required for the radiochemical separations. Therefore, an ongoing effort is being made to perfect an analytical procedure for the quantitative determination of curium. Because of the large number of samples to be analyzed and the small sizes of individual samples, it is desirable to be able to analyze both curium and plutonium from a single sample in a minimum of investigator's time.

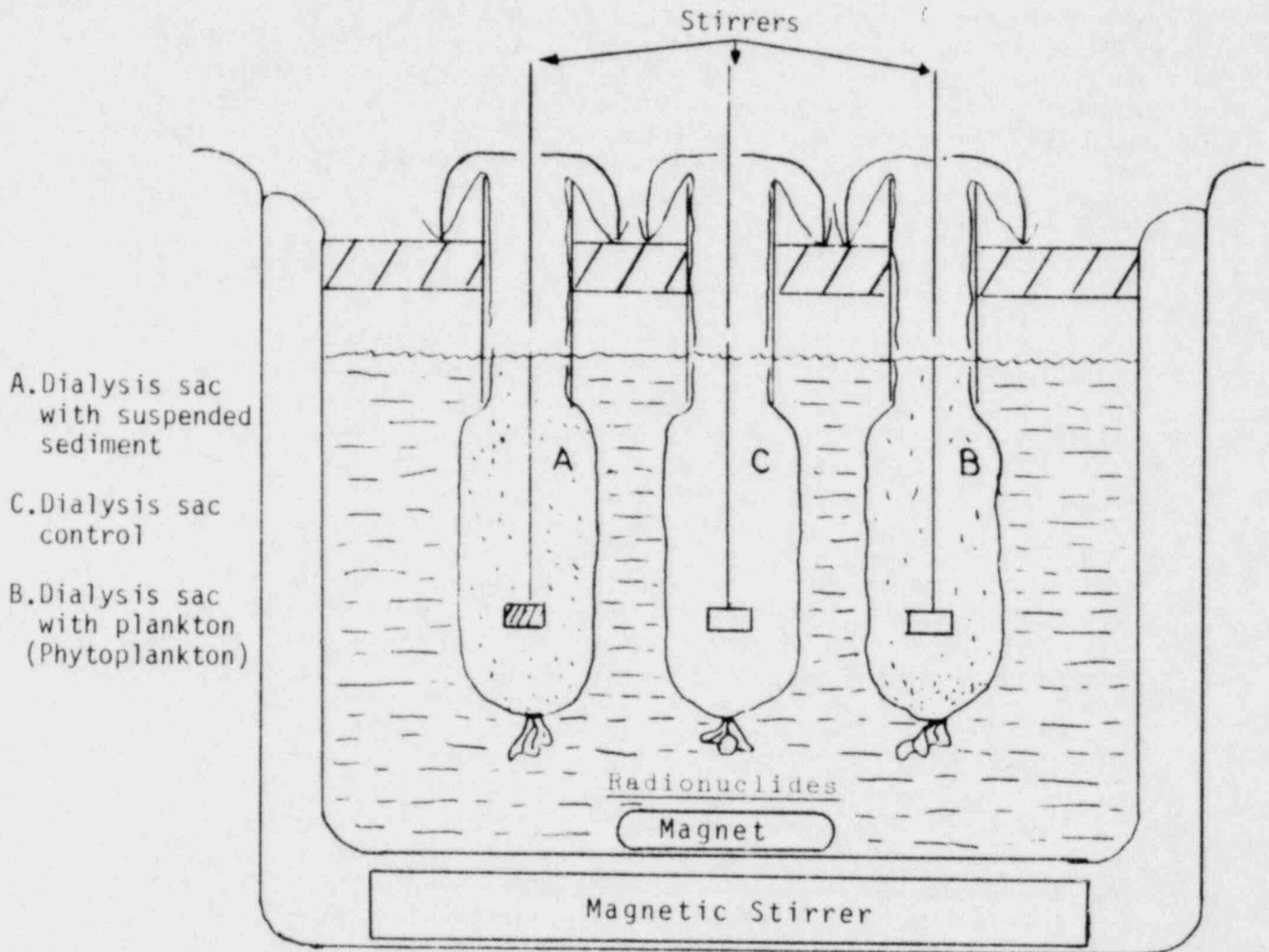


Figure 1. Schematic diagram of apparatus used for dialysis experiments.

Because the chemical properties of curium and americium are generally considered to be similar, the procedure of Bojanowski et al. (6), for americium was initially used for our curium studies. In this procedure, the transuranics (i.e., curium, plutonium, and americium) are initially leached from particulate samples with hot concentrated nitric acid and concentrated hydrochloric acid. The leachate is then evaporated to dryness and appropriate amounts of radionuclide tracers (^{243}Am , ^{236}Pu , or ^{242}Pu) are added to the sample beakers with 200 ml of 8M HNO_3 and approximately 0.1 g NaNO_2 . The NaNO_2 is added to convert all plutonium to the +4 valence state for subsequent ion exchange chromatography. Americium - 243 is being used at the present time as the analytical tracer for curium since we have been unable to obtain an appropriate curium isotope tracer. Previous studies have indicated that americium is an acceptable yield tracer for curium. For future work, however, we shall attempt to obtain ^{248}Cm from the Oak Ridge National Laboratories to be used as the curium tracer.

Results of tests have shown that the procedure of Bojanowski et al. (6) is satisfactory for measuring Am and Cm but the chemical yields were not as high as we would like. To minimize these analytical difficulties, modifications of Holm and Fukai (7) have been incorporated in our analytical procedure. Results of recovery experiments with the modified procedure (Table 3) show that the recoveries of both ^{243}Am and ^{244}Cm are relatively high (i.e., between 54 and 75% in most cases). The exception is the 12-22% recovery of ^{244}Cm in the Cm-Th series of samples. More measurements are needed to evaluate the reasons for low recovery in that series. Both ^{243}Am and ^{244}Cm are recovered exclusively in their anticipated fraction with no detectable contamination of the plutonium fraction. However, substantial amounts of ^{236}Pu were observed in the americium-curium fraction. This undesired contamination was presumably due to the elution of ^{236}Pu through the Dowex 1-X8 column. To convert all plutonium to the desired +4 oxidation state for sorption onto the Dowex 1-X8 column, 0.1 g of NaNO_2 is now added to the sample solution (NaNO_2 is not used in the initial Holm and Fukai procedure). It should be noted that the apparent incomplete separation of ^{228}Th from americium and curium may not be acceptable for the analysis of environmental samples. Large amounts of thorium are present in soil and the alpha decay energies of ^{228}Th are almost identical to ^{241}Am (5.43, 5.34 MeV for ^{228}Th vs. 5.49, 5.44 MeV for ^{241}Am). These differences cannot be resolved by the alpha spectrometer system.

In tracer experiments using only ^{244}Cm , the simplest analytical method is to measure the gamma or X-ray radiation emitted in the decay. This method does not require the complex and lengthy chemical separation procedures discussed previously. However, gamma or X-ray emitted radiation is of low abundance in the decay of curium. This fact, coupled with the low detection efficiency with Ge(Li) or NaI(Tl) detectors, limits the utility of the gamma and X-rays for analysis. We have attempted to develop a procedure for measuring the 18.3 KeV L_β

Table 3. Results of Am, Cm, & Pu recoveries with Holm-Fukai procedure.

Sample No.	Spike				Am-Cm Fraction ^a Recovery							Pu Fraction ^a Recovery						Comments		
	²⁴³ Am (dpm)	²⁴⁴ Cm (dpm)	²³⁶ Pu (dpm)	Th(NO ₃) ₄ 4H ₂ O (g)	²⁴³ Am		²⁴⁴ Cm		²³⁶ Pu		²⁴¹ Am ^b (dpm)	²⁴³ Am		²⁴⁴ Cm		²³⁶ Pu			²⁴¹ Am ^b (dpm)	
					dpm	%	dpm	%	dpm	%		dpm	%	dpm	%	dpm	%			
Am-Cm-16	115	93	0	0	77.6	67.5	64.8	69.7			48.6	0.0	0.0	0.0	0.0			0.0		
Am-Cm-17	115	93	0	0	74.2	64.5	57.9	62.3			353.3	0.0	0.0	0.0	0.0			0.1		
Am-Cm-18	115	93	0	0	68.8	59.8	68.9	74.1			71.5	0.0	0.0	0.0	0.0			0.0		
\bar{x}^c						63.9		68.7					0.0		0.0					
s^c						3.9		6.0					0.0		0.0					
Pu-Am-3	58	0	46	0	36.6	63.1					11.1	24.1	8.5	0.0	0.0			1.5	3.3	0.0
Pu-Am-4	58	0	46	0	33.2	57.2					7.5	16.3	4.5	0.0	0.0			1.9	4.1	0.2
Pu-Am-5	58	0	46	0	31.4	54.1					10.9	23.7	4.5	0.0	0.0			1.5	3.3	0.1
\bar{x}^c						58.1													3.6	
s^c						4.6													0.5	
Cm-Th-1	0	93	0	0.51	0.0		12.0	12.9			10.8	0.0		0.0	0.0					0.1
Cm-Th-2	0	93	0	0.65	0.0		20.0	21.5			49.7	0.0		0.0	0.0					0.2
Cm-Th-3	0	93	0	0.49	0.0		22.2	23.9			6.2	0.0		0.0	0.0					0.0
\bar{x}^c								19.4												
s^c								5.8												
Am-Cm-19	0	0	0	0	0.0		0.0				2.1	0.0						0.0		0.2
Am-Cm-20	0	0	0	0	0.1		0.0				3.6	0.1						0.5		0.2
\bar{x}^c					0.0		0.0				2.8	0.0						0.2		0.2

- a) Appropriate values from the analytical blanks (i.e., Am-Cm-19 and 20) have been subtracted from absolute activity values of all spiked samples in this table.
- b) "²⁴¹Am" - apparent ²⁴¹Am impurity obtained in samples.
- c) \bar{x} and s are the mean and one standard deviation unit, respectively.

decay X-rays with a germanium intrinsic detector and pulse height analyzer. We have found that it is necessary to use ^{244}Cm alone rather than with several other radionuclides in these experiments. In this procedure, particulate ^{244}Cm is counted directly while soluble ^{244}Cm is coprecipitated with $\text{Fe}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$ and ^{244}Cm in the precipitate is measured. The preliminary data that have been obtained to date indicate that the K_d values for ^{244}Cm in the freshwater-sediment systems are approximately 10^5 .

3.0 RESULTS AND DISCUSSION

3.1 K_d Values by Constant Shaking Experiments

Constant shaking experiments were conducted for several water-sediment systems, including Clinch River, Cattaraugus Creek, Lake Michigan, and Sinclair Inlet (an organic rich area in Puget Sound) and for the pure clay calcium montmorillonite in water from the Skagit River Estuary. Experiments were also completed for three different water-sediment systems representing different salinities from the Hudson River Estuary and for the $<63 \mu\text{m}$ sediment fraction for cores from the Skagit River Estuary and Saanich Inlet. The radioisotopes included in these experiments are shown in Table 4.

Adsorption of ^{106}Ru , ^{137}Cs , ^{241}Am . Distribution coefficients have been calculated for ^{106}Ru , ^{137}Cs , and ^{241}Am for sediment-water systems from the Skagit River Estuary, Saanich Inlet, Lake Michigan, and for calcium montmorillonite. These results are given in Table 5. The distribution coefficients for all three radionuclides are highest for the Lake Michigan sediment-water system and lowest for calcium montmorillonite-Skagit Estuary water system. The differences between systems are within an order of magnitude for all of the radionuclides and correspond to the values we have reported earlier for other sediment-water systems. The high K_d values for ^{106}Ru , ^{137}Cs , and ^{241}Am in the Lake Michigan system are expected since in freshwater, there are fewer ligands available for complexation than in marine systems so that adsorption of radionuclides by suspended sediments is relatively more important in freshwater than in marine water systems. Also, adsorption by sediments is less at the higher ionic strength of the marine systems because of saturation of the sorption sites by sodium and other ions. This may contribute to the low values of distribution coefficients with calcium montmorillonite, a clay with a large base exchange capacity ($\sim \frac{100 \text{ meq}}{100 \text{ g}}$). However, adsorption to

natural sediments in estuaries may actually be due to the metal oxides or organic coatings that cover the surfaces of clay minerals in natural environments. The absence of such coatings on pure calcium montmorillonite in this test system may contribute to the lower values found for the distribution coefficients of ^{106}Ru , ^{137}Cs , and ^{241}Am .

The data for ^{137}Cs from Cattaraugus Creek (Fig. 2) indicate that the K_d values increase as a function of time for all sediment concentrations. This suggests that at least two processes may be contributing to the observed uptake by sediments, namely, a rapid initial adsorption on the sediment surface followed by an ion-exchange reaction in the mineral lattice. Distribution coefficients of ^{137}Cs are also found to increase with time for the sediment-water system

Table 4. Radioisotopes used in constant shaking experiments.

	^{85}Sr	^{106}Ru	^{137}Cs	^{237}Pu	^{241}Am
Skagit Core #1	x	x	x	x	x
Core #2	x	x	x	x	x
Saanich Inlet	x	x	x	x	x
Lake Michigan	x	x	x	x	x
Clinch River		x	x	x	x
Cattaraugus Creek*	x		x	x	x
Sinclair Inlet		x	x		x
Hudson River Estuary					
15% salinity mp 0	x	x	x	x	x
2-3% salinity mp 18	x	x	x	x	x
freshwater mp 43	x	x	x	x	x
Montmorillonite	x	x	x	x	x

*Cattaraugus Creek experiments did not have ^{90}Sr or ^{239}Pu added to the constant shaking experiments but those radionuclides were included in dialysis experiments.

Table 5. Adsorption distribution coefficients (ml/g) for ^{241}Am , ^{106}Ru and ^{137}Cs .

AMERICIUM - 241			
	n*	Average K_d **	Range***
Skagit Estuary			
Core #1	15	$(2.68 \pm 0.74) \times 10^5$	$(1.50 \pm 0.79) - (4.08 \pm 1.69) \times 10^5$
Core #2	10	$(3.80 \pm 1.06) \times 10^5$	$(2.87 \pm 1.24) - (5.84 \pm 3.57) \times 10^5$
Gelwhite	6	$(1.91 \pm 0.41) \times 10^5$	$(1.29 \pm 0.26) - (2.39 \pm 0.45) \times 10^5$
Saanich Inlet	14	$(3.71 \pm 0.84) \times 10^5$	$(2.69 \pm 0.86) - (5.24 \pm 3.28) \times 10^5$
Lake Michigan	12	$(5.48 \pm 3.73) \times 10^5$	$(2.43 \pm 1.05) - (13.0 \pm 9.8) \times 10^5$
RUTHENIUM - 106			
Skagit Estuary			
Core #1	21	$(3.61 \pm 0.63) \times 10^4$	$(2.56 \pm 0.64) - (4.88 \pm 0.79) \times 10^4$
Core #2	12	$(4.51 \pm 0.76) \times 10^4$	$(3.48 \pm 0.86) - (5.65 \pm 2.22) \times 10^4$
Gelwhite	12	$(1.93 \pm 0.38) \times 10^4$	$(1.29 \pm 0.21) - (27.3 \pm 11.1) \times 10^4$
Saanich Inlet	17	$(4.42 \pm 0.52) \times 10^4$	$(3.56 \pm 0.25) - (5.24 \pm 0.89) \times 10^4$
Lake Michigan	12	$(5.23 \pm 1.68) \times 10^4$	$(2.98 \pm 0.54) - (7.95 \pm 1.10) \times 10^4$
CESIUM - 137			
Skagit Estuary			
Core #1	9	$(1.87 \pm 0.15) \times 10^2$	$(1.66 \pm 0.17) - (2.09 \pm 0.12) \times 10^2$
Core #2	5	$(2.43 \pm 0.07) \times 10^2$	$(2.34 \pm 0.18) - (2.50 \pm 0.18) \times 10^2$
Gelwhite	6	65.1 ± 7.22	$(56.3 \pm 12.0) - (75.6 \pm 13.8)$
Saanich Inlet		Did not reach equilibrium	$(1.89 \pm 0.17) - (3.15 \pm 0.15) \times 10^2$
Lake Michigan	17	$(5.09 \pm 0.31) \times 10^2$	$(4.44 \pm 0.24) - (5.54 \pm 0.31) \times 10^2$

- * n = the total number of K_d values determined for a radionuclide in a given sediment-water system.
- ** Average K_d is the mean for all K_d determinations. Error terms are one standard deviation from the mean of the replicate determinations.
- *** Range in K_d is the lowest and highest K_d values obtained in each sediment-water system. Error terms are 2 σ propagated counting errors.

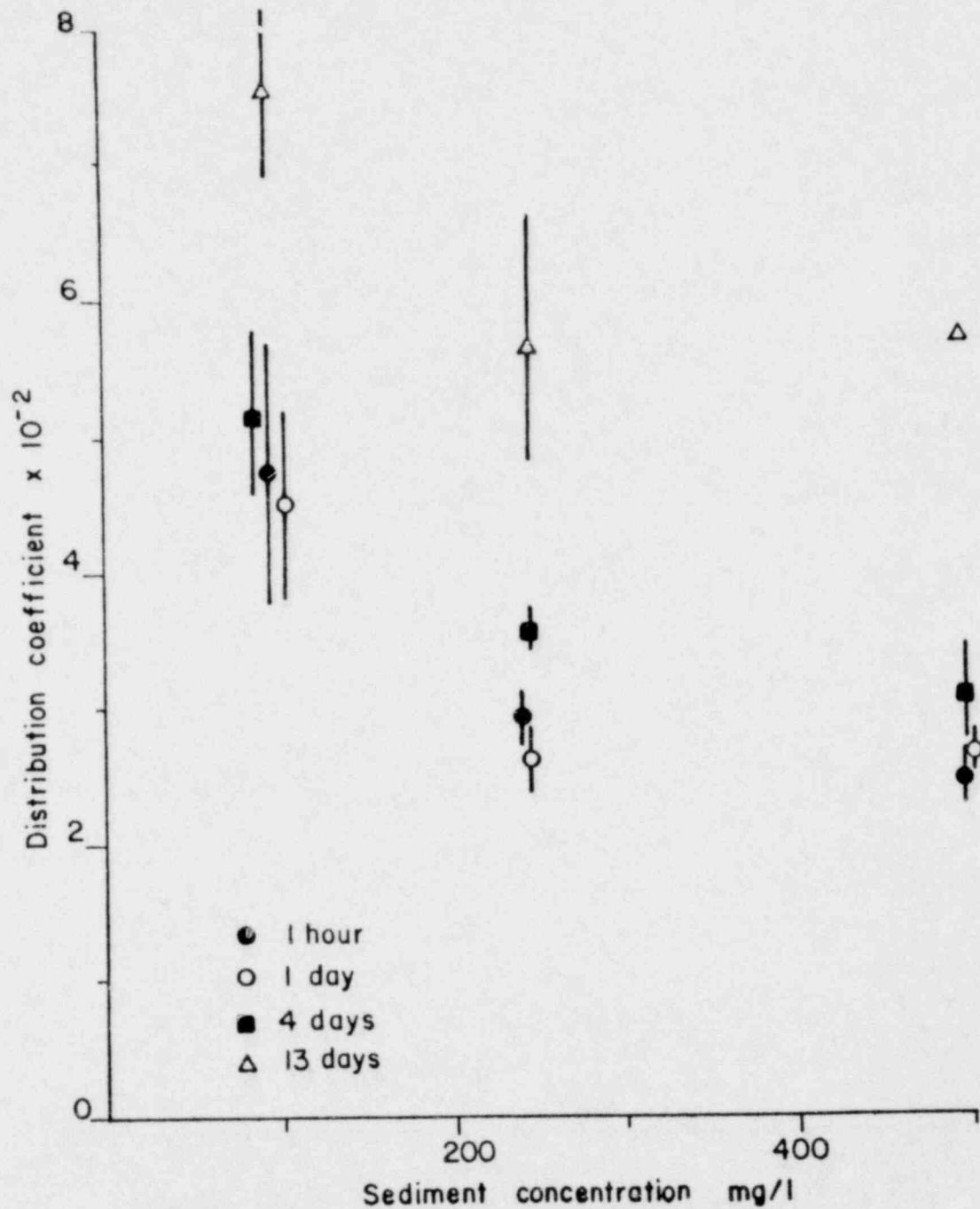


Figure 2. Adsorption distribution coefficients for ^{137}Cs in the Cattaraugus Creek sediment-water system as a function of sediment concentration. Error bars are 1σ difference between means of replicate samples. Lack of error bars indicates that only one sample was analyzed.

from Saanich Inlet. Similar results are found for ^{106}Ru during experiments with the Lake Michigan sediment-water systems.

Tables 6-8 show the K_d values of ^{106}Ru , ^{137}Cs , and ^{241}Am for Clinch River, Sinclair Inlet, and the Hudson River Estuary. For all three radionuclides, as well as for ^{85}Sr (see next section), the K_d value in the Clinch River sediment-water system is the highest value we have found in any oxic experimental system. These results suggest that the ion exchange capacity of Clinch River sediments may be significantly higher than for other sediments we have used. The differences between Clinch River and other freshwater systems indicate the necessity of conducting experiments with specific sediment-water systems before modeling radionuclide transport in a given system.

The K_d values of ^{106}Ru , ^{137}Cs , and ^{241}Am for Sinclair Inlet and the Hudson River Estuary agree with the values obtained for other experimental systems. Comparison of the results from the Hudson River Estuary and Sinclair Inlet suggest that ^{241}Am , like ^{237}Pu , has higher K_d values at higher salinity. No salinity effect is noted for ^{137}Cs , or ^{106}Ru . However, it appears that the sediments from SLOSH II may have a higher than usual affinity for ^{137}Cs and ^{106}Ru which could mask any salinity effects. New experiments are planned with sediments from SLOSH II over a range of salinity values.

Adsorption of ^{85}Sr and ^{237}Pu : Constant shaking experiments were conducted to determine the adsorption K_d values of ^{85}Sr and ^{237}Pu for a variety of sediment-water systems and for organic detritus in Lake Michigan and the Skagit River Estuary. Replicate experimental vessels were used for each sediment-water system and samples were withdrawn after various time intervals to assure that equilibrium had been reached. Radioactivity measurements were made with a 2" NaI(Tl) well crystal. Distribution coefficients for ^{85}Sr and ^{237}Pu are shown in Tables 9 and 10, respectively.

The highest K_d values for ^{85}Sr are found consistently in freshwater systems. At SLOSH II, salinity of 2 - 30/00, the K_d value is a factor of approximately 2 lower than for freshwater systems. At higher salinities (150/00 - 310/00), ^{85}Sr was not significantly sorbed to the sediments. Two explanations can be proposed for the low K_d values for ^{85}Sr in marine systems: a) isotopic equilibrium between tracer ^{85}Sr and stable strontium may not be reached during the experiment and/or, b) the physico-chemical state of strontium may be different between the marine and freshwater systems. Since the concentration of stable strontium is much higher in marine systems it may be that the available sites for sorbing strontium are predominantly filled by the stable isotope. However, in experiments with organic detritus, the K_d values in Lake Michigan and Skagit Estuary systems are similar. This suggests that the low K_d for ^{85}Sr is not due to competition with the stable strontium, but rather to changes in the physico-chemical state of the ^{85}Sr tracer. The higher K_d value for ^{85}Sr on marine detritus

Table 6. Adsorption distribution coefficients for ruthenium-106.

Sediment-Water System	Salinity ppt	pH	n*	Average K_d^{**} ml/g x 10^{-4}	Range*** ml/g x 10^{-4}
Clinch River	<1	7.80 - 8.00	12	6.75 ± 0.52	$(6.12 \pm 0.26) - (7.79 \pm 0.94)$
Hudson River	<1	7.85 - 8.00	12	3.92 ± 0.40	$(3.29 \pm 0.36) - (4.84 \pm 1.04)$
SLOSH III					
SLOSH II	2 - 3	7.70 - 7.85	11	5.15 ± 1.03	$(3.96 \pm 0.85) - (6.89 \pm 0.95)$
SLOSH V	15	7.70 - 7.85	11	4.53 ± 0.37	$(3.98 \pm 0.35) - (5.18 \pm 1.12)$
Sinclair Inlet	28.9	7.70 - 7.80	12	5.42 ± 0.43	$(4.86 \pm 0.14) - (6.11 \pm 1.12)$

*n = total number of K_d values determined for a radionuclide in a given sediment-water system.

**Average K_d is the mean for all determinations. Error terms are one standard deviation from the mean of replicate determinations.

***Range in K_d is the lowest and highest K_d values obtained in each sediment-water system. Error terms are two standard deviations propagated counting errors.

Table 7. Adsorption distribution coefficients for cesium-137.

Sediment-Water System	Salinity ppt	pH	n*	Average K_d^{**} ml/g	Range*** ml/g
Clinch River	<1	7.80 - 8.00	9	1355 ± 40	(1302 ± 50) - (1425 ± 50)
Hudson River					
SLOSH III	<1	7.85 - 8.00	9	401 ± 20	(378 ± 26) - (437 ± 29)
SLOSH II	2 - 3	7.70 - 7.85	9	878 ± 71	(795 ± 41) - (1018 ± 72)
SLOSH V	15	7.70 - 7.85	8	356 ± 21	(326 ± 25) - (134 ± 18)
Sinclair Inlet	28.9	7.70 - 7.80	9	124 ± 9	(108 ± 19) - (134 ± 18)

*n = the total number of K_d values determined for a radionuclide in a given sediment-water system.

**Average K_d is the mean for all determinations. Error terms are one standard deviation from the mean of replicate determinations.

***Range in K_d is the lowest and highest K_d values obtained in each sediment-water system. Error terms are two standard deviations propagated counting errors.

Table 8. Adsorption distribution coefficients for americium-241.

Sediment-Water System	Salinity ppt	pH	n*	Average K_d^{**} ml/g x 10^{-5}	Range*** ml/g x 10^{-5}
Clinch River	<1	7.80 - 8.00	8	7.33 ± 3.64	$(3.52 \pm 2.11) - (13.17 \pm 11.81)$
Hudson River					
SLOSH III	<1	7.85 - 8.00	9	1.26 ± 0.13	$(1.11 \pm 0.33) - (1.50 \pm 0.42)$
SLOSH II	2 - 5	7.70 - 7.85	9	2.37 ± 0.88	$(1.37 \pm 0.46) - (3.97 \pm 2.72)$
SLOSH V	15	7.70 - 7.85	6	2.96 ± 0.88	$(2.58 \pm 1.09) - (4.62 \pm 3.62)$
Sinclair Inlet	28.9	7.70 - 7.80	9	4.54 ± 1.20	$(2.71 \pm 0.64) - (5.97 \pm 2.38)$

*n = the total number of K_d values determined for a radionuclide in a given sediment-water system.

**Average K_d is the mean for all determinations. Error terms are one standard deviation from the mean of replicate determinations.

***Range in K_d is the lowest and highest K_d values obtained in each sediment-water system. Error terms are two standard deviations propagated counting errors.

Table 9. Adsorption distribution coefficients for strontium-85.

Sediment-Water System	Salinity ppt	pH	n*	Average K_d^{**} (ml/g)	Range*** (ml/g)
Clinch River	<1	7.8 - 8.0	6	124.4 ± 7.5	(118.0 ± 23.5) - (137.2 ± 23.1)
Cattaragus Creek	<1	8.0 - 8.2	8	62.2 ± 6.2	(52.9 ± 3.3) - (70.2 ± 3.0)
Lake Michigan	<1	7.8 - 8.0	7	82.2 ± 7.0	(73.5 ± 22.9) - (93.6 ± 20.7)
Hudson River					
SLOSH III	<1	7.8 - 8.0	6	73.7 ± 10.5	(58.3 ± 19.9) - (90.5 ± 16.2)
SLOSH II	2 - 3	7.7 - 7.9	6	34.8 ± 5.3	(28.9 ± 11.8) - (42.9 ± 15.5)
SLOSH V	15	7.7 - 7.9	}	}	^{85}Sr was not sorbed to sediments $K_d = 0.$
Skagit Estuary	31.3	7.6 - 7.8			
Sinclair Inlet	28.9	7.7 - 7.8			
Detritus					
Lake Michigan	<1	8.2 - 8.5	4	75.2 ± 4.1	(70.6 ± 4.1) - (79.1 ± 6.6)
Skagit Estuary	29.6	7.9 - 8.0	6	68.3 ± 4.4	(62.2 ± 5.0) - (72.5 ± 3.1)

*n = the total number of K_d values determined for a radionuclide in a given sediment-water system.

**Average K_d is the mean for all determinations. Error terms are one standard deviation from the mean of replicate determinations.

***Range in K_d is the lowest and highest K_d values obtained in each sediment-water system. Error terms are two standard deviations propagated counting errors.

Table 10. Adsorption distribution coefficients for plutonium-237.

Sediment-Water System	Salinity ppt	pH	n*	Average K_d^{**} ml/g x 10^{-4}	Range*** ml/g x 10^{-4}
Clinch River	<1	7.8 - 8.0	6	4.71 ± 0.40	(4.21 ± 0.26) - (5.17 ± 0.37)
Cattaraugus Creek	<1	8.0 - 8.2	7	2.09 ± 0.65	(1.19 ± 0.07) - (2.73 ± 0.32)
Lake Michigan	<1	7.8 - 8.0	7	14.1 ± 1.8	(12.4 ± 2.4) - (16.8 ± 4.1)
Hudson River					
SLOSH III	<1	7.8 - 8.0	9	0.93 ± 1.4	(0.83 ± 0.03) - (1.14 ± 0.04)
SLOSH II	2 - 3	7.7 - 7.9	6	3.12 ± 0.22	(2.83 ± 0.16) - (3.37 ± 0.07)
SLOSH V	15	7.7 - 7.9	4	3.87 ± 0.11	(3.71 ± 0.23) - (3.97 ± 0.25)
Skagit Estuary	31.3	7.6 - 7.8	6	10.3 ± 0.8	(9.18 ± 1.80) - (11.2 ± 2.2)
Sinclair Inlet	28.9	7.7 - 7.8	9	7.40 ± 1.21	(4.79 ± 0.31) - (8.67 ± 1.19)
Detritus					
Lake Michigan	<1	8.2 - 8.5	6	2.59 ± 0.94	(1.54 ± 0.18) - (4.06 ± 1.09)
Skagit Estuary	29.6	7.9 - 8.0	9	$1.34 \pm 0.94\#$	(0.34 ± 0.15) - (2.81 ± 2.71)

*n = the total number of K_d values determined for a radionuclide in a given sediment-water system.

**Average K_d is the mean for all determinations. Error terms are one standard deviation from the mean of replicate determinations.

***Range K_d is the lowest and highest K_d values obtained in each sediment-water system. Error terms are two standard deviations propagated counting errors.

#Three replicate experimental vessels had significantly different K_d 's: 2.20 ± 0.63 , 0.93 ± 0.62 , 0.34 ± 0.15 .

relative to suspended sediments suggests that organic surfaces may be more important than inorganic sediments for scavenging and transporting ^{85}Sr in marine environments.

For both suspended sediments and detritus, the highest K_d values for ^{237}Pu are found for the Lake Michigan system. For freshwater-sediment systems K_d values for ^{237}Pu range over more than an order of magnitude. This suggests that differences among sediment-water systems result from different water quality parameters or different sediment characteristics among the various systems.

The data for the Hudson River Estuary and the marine systems suggest that the K_d value of ^{237}Pu may increase with an increase in salinity. That is, as plutonium is transported from freshwater through estuarine to marine environments, a greater percentage is found in the particulate phase.

Summary of Adsorption K_d Values. Constant shaking experiments have been completed to determine the adsorption K_d values of ^{85}Sr , ^{106}Ru , ^{137}Cs , ^{237}Pu , and ^{241}Am in sediment-water systems from Lake Michigan, Clinch River, Cattaraugus Creek, and the Hudson River Estuary. A summary of the adsorption K_d values is presented in Table 11. Many of these values have been presented previously but they are collected here in a single table. The data are presented such that K_d values of different radionuclides can be compared within a given system and K_d values of selected radionuclides can also be compared among the different sediment-water systems.

For all of the sediment-water systems, ^{85}Sr has the lowest K_d value, generally less than 100, and ^{241}Am has the highest value, greater than 10^5 . The order of increasing K_d values is $^{85}\text{Sr} < ^{137}\text{Cs} < (^{106}\text{Ru} \text{ or } ^{237}\text{Pu}) < ^{241}\text{Am}$. The radionuclides ^{106}Ru and ^{237}Pu are coupled in this sequence because the K_d value of ^{106}Ru is higher than ^{237}Pu for sediment-water systems from the Clinch River and Hudson River Estuary systems but in the other systems K_d values for ^{237}Pu are higher than for ^{106}Ru . The K_d values for ^{85}Sr and ^{137}Cs are greater in the freshwater systems than in the marine sediment-water systems but no similar trends were observed for ^{106}Ru , ^{237}Pu , or ^{241}Am . For all three of these latter radionuclides both the highest and lowest K_d values were found in sediment-freshwater systems.

Desorption of ^{106}Ru , ^{137}Cs , ^{241}Am , ^{85}Sr , and ^{237}Pu . Experiments have been completed to determine the desorption K_d values of ^{106}Ru , ^{137}Cs , ^{241}Am , ^{85}Sr , and ^{237}Pu in sediment-water systems from Lake Michigan, Clinch River, three locations in the Hudson River Estuary and Sinclair Inlet in Puget Sound. Desorption studies were conducted by first sorbing radionuclides to sediments, centrifuging and then resuspending the sediments at a concentration of 200 mg sediment/l in unspiked water from the same sampling location. The suspensions were kept under constant shaking in a cold room at $\sim 4^\circ\text{C}$.

For evaluating the desorption of ^{106}Ru , ^{137}Cs , and ^{241}Am from Lake Michigan sediments, a different approach was taken: Three sets of samples corresponding to three different sorption times for the radionuclides were

Table 11. Adsorption K_d values for ^{85}Sr , ^{106}Ru , ^{137}Cs , ^{237}Pu and ^{241}Am in selected sediment-water systems.

Sediment-Water System	n*	^{85}Sr K_d^{**}	n*	^{106}Ru $K_d \times 10^{-4}$	n*	^{137}Cs $K_d \times 10^{-2}$	n*	^{237}Pu $K_d \times 10^{-4}$	n*	^{241}Am $K_d \times 10^{-5}$
Lake Michigan	7	82.2 (7.0)	12	5.23 (1.68)	17	5.09 (0.37)	7	14.1 (1.8)	12	5.48 (3.73)
Clinch River	6	124.4 (7.5)	12	6.72 (0.52)	9	13.6 (0.40)	6	4.71(0.40)	8	7.33 (3.64)
Cattaraugus Creek	7	62.3 (6.2)	9	1.40	#		7	2.09(0.65)	12	2.26 (1.47)
Hudson River										
mp 43	6	73.7 (10.5)	12	3.92 (0.40)	9	4.07 (0.20)	6	0.93(0.14)	9	1.26 (0.13)
mp 18	6	34.8 (5.3)	11	5.15 (1.03)	9	8.78 (0.71)	6	3.12(0.22)	9	2.37 (0.88)
mp 0		##	11	4.53 (0.37)	8	3.56 (0.21)	4	3.87(0.11)	6	2.96 (0.88)
Skagit Estuary										
Core No. 1		##	21	3.61 (0.63)	9	1.87 (0.15)	9	10.3 (0.8)	15	2.68 (0.74)
Core No. 4		-	12	4.51 (0.76)	5	2.42 (0.07)		-	10	3.80 (1.06)
Saanich Inlet		-	17	4.42 (0.52)	#			-	14	3.71 (0.84)
Sinclair Inlet		##	12	5.42 (0.43)	9	1.24 (0.09)	9	7.40(1.21)	9	4.54 (1.20)

*n = the total number of K_d calculations made for a radionuclide in a given sediment-water system.

** K_d is average value for all determinations. Number in parentheses is one standard deviation from mean of replicate determinations

#Did not reach equilibrium.

##Concentration in particulate phase was below detection limits.

prepared. A first set consisted of sediment samples which were allowed to sorb the radionuclides for one hour only before the desorption experiments were started. A second set was prepared from sediments which were allowed to sorb the radionuclides for 24 hours, and a third set of sediments had a sorption time of 96 hours. Immediately after each sorption period, the sediments were centrifuged and then resuspended in new sample water. The suspensions were then kept under constant shaking, and sub-samples for radioactivity measurements were collected after 1 hour, 24 hours, and 96 hours of desorption. The experimental conditions in each sediment-water system are presented in Table 12.

The results of the desorption distribution coefficient experiments are shown in Table 13. The average desorption K_d and the range in K_d values obtained for each radionuclide in different sediment-water systems are presented in this table. The following is a brief discussion of the desorption values for the individual radionuclides:

^{85}Sr . Desorption K_d values for this isotope in the freshwater systems varied as follows:

Clinch River < Hudson River < Lake Michigan
(SLOSH III)

The estuarine (Hudson River SLOSH II and V) and marine (Sinclair Inlet) sediment-water systems did not show measurable concentrations of this isotope in the sediments, suggesting that it may have been desorbed completely.

^{237}Pu . This isotope showed high desorption K_d values (greater than 10^5 ml/g) in all the sediment-water systems studied. There is considerable overlap in K_d values for some systems, and the overall trend in average K_d values is:

Clinch River < Hudson River (SLOSH II) < Hudson River (SLOSH V) < Sinclair Inlet < Hudson River (SLOSH III) < Lake Michigan

^{137}Cs . The desorption K_d for this radionuclide was determined only for the three Hudson River samples and for Lake Michigan. For the Lake Michigan experiment, the equilibrium K_d values corresponding to sorption periods of one hour, 24 hours, and 96 hours, are 3.67×10^3 , 5.50×10^3 , and 5.62×10^3 ml/g, respectively. These results indicate that lesser amounts of ^{137}Cs were sorbed by the sediments in one hour than in 24 hours or 96 hours, and that equilibrium distribution was attained in 24 hours, as shown by the same desorption K_d values obtained for the 24-hour and 96-hour experiments.

Table 12. Summary of experimental conditions for desorption K_d measurements. All experiments were done under constant shaking at 200 rpm in a cold room (5 - 8°C). Sediment concentration = 0.20 mg/ml in each system.

Sediment-Water System	Salinity	pH
Hudson River		
mp 43 (SLOSH III)	<1	7.6 - 8.0
mp 18 (SLOSH II)	2 - 3	7.5 - 7.7
mp 0 (SLOSH V)	15	7.7 - 7.9
Clinch River	<1	7.6 - 8.1
Lake Michigan	<1	7.9 - 8.2
Sinclair Inlet	28.9	7.8 - 8.0

Table 13. Desorption distribution coefficients for ^{85}Sr , ^{237}Pu , ^{137}Cs , ^{241}Am and ^{106}Ru in different sediment-water systems.

Sediment-water System	Radionuclides	n*	Average K_d^{**}	Distribution Coefficient, ml/g Range in K_d^{***}	
Lake Michigan	^{85}Sr	10	$(1.41 \pm 0.51) \times 10^3$	$(0.57 \pm 0.13) \times 10^3$ to $(2.33 \pm 0.46) \times 10^3$	
	^{237}Pu	10	$(4.79 \pm 1.19) \times 10^5$	$(3.10 \pm 0.35) \times 10^5$ to $(6.47 \pm 1.95) \times 10^5$	
	^{137}Cs (A)#	6	$(3.67 \pm 0.53) \times 10^3$	$(3.03 \pm 0.48) \times 10^3$ to $(4.27 \pm 0.38) \times 10^3$	
		(B)#	4	$(5.50 \pm 0.51) \times 10^3$	$(4.98 \pm 0.75) \times 10^3$ to $(6.17 \pm 0.81) \times 10^3$
		(C)#	6	$(5.62 \pm 0.73) \times 10^3$	$(4.56 \pm 0.61) \times 10^3$ to $(6.73 \pm 0.91) \times 10^3$
	^{241}Am (A)#	6	$(5.62 \pm 0.73) \times 10^3$	$(2.81 \pm 0.77) \times 10^5$ to $(12.20 \pm 1.93) \times 10^5$	
		(B)#		See Text	$(6.66 \pm 2.23) \times 10^5$ to $(15.34 \pm 4.64) \times 10^5$
		(C)#			$(7.94 \pm 5.22) \times 10^5$ to $(22.34 \pm 12.00) \times 10^5$
	^{106}Ru (A)#	8	$(3.51 \pm 0.88) \times 10^4$	$(2.57 \pm 0.32) \times 10^4$ to $(4.97 \pm 0.83) \times 10^4$	
		(B)#	9	$(1.91 \pm 0.34) \times 10^5$	$(1.61 \pm 0.47) \times 10^5$ to $(2.69 \pm 1.35) \times 10^5$
(C)#		9	$(2.79 \pm 0.31) \times 10^5$	$(2.31 \pm 1.41) \times 10^5$ to $(3.34 \pm 2.11) \times 10^5$	
Clinch River	^{85}Sr	8	$(4.55 \pm 0.89) \times 10^2$	$(3.57 \pm 3.34) \times 10^2$ to $(6.38 \pm 3.30) \times 10^2$	
	^{237}Pu	9	$(1.54 \pm 0.09) \times 10^5$	$(1.38 \pm 0.11) \times 10^5$ to $(1.66 \pm 0.15) \times 10^5$	
Hudson River SLOSH III mp 43	^{85}Sr	9	$(4.86 \pm 1.35) \times 10^2$	$(3.07 \pm 2.61) \times 10^2$ to $(7.02 \pm 2.89) \times 10^2$	
	^{237}Pu	6	$(3.60 \pm 0.37) \times 10^5$	$(3.10 \pm 0.56) \times 10^5$ to $(3.91 \pm 0.99) \times 10^5$	
	^{137}Cs	5	$(3.65 \pm 0.52) \times 10^3$	$(3.08 \pm 0.37) \times 10^3$ to $(4.43 \pm 0.78) \times 10^3$	
	^{241}Am	4	$(5.39 \pm 0.84) \times 10^5$	$(4.36 \pm 1.48) \times 10^5$ to $(6.21 \pm 2.41) \times 10^5$	
	^{106}Ru	5	$(2.52 \pm 0.71) \times 10^5$	$(1.73 \pm 0.13) \times 10^5$ to $(3.49 \pm 1.62) \times 10^5$	
Hudson River SLOSH II mp 18	^{85}Sr	-	-	-	
	^{237}Pu	9	$(2.98 \pm 0.22) \times 10^5$	$(2.69 \pm 0.35) \times 10^5$ to $(3.39 \pm 0.51) \times 10^5$	
	^{137}Cs	2	$(3.34 \pm 0.69) \times 10^3$	$(2.85 \pm 0.19) \times 10^3$ to $(3.82 \pm 0.41) \times 10^3$	
	^{241}Am	5	$(8.47 \pm 1.52) \times 10^5$	$(6.52 \pm 0.61) \times 10^5$ to $(10.20 \pm 2.46) \times 10^5$	
	^{106}Ru	5	$(3.35 \pm 0.37) \times 10^5$	$(2.93 \pm 0.48) \times 10^5$ to $(3.94 \pm 0.71) \times 10^5$	
Hudson River SLOSH V mp 0	^{85}Sr	-	-	-	
	^{237}Pu	12	$(3.21 \pm 1.35) \times 10^5$	$(1.09 \pm 0.09) \times 10^5$ to $(5.26 \pm 0.97) \times 10^5$	
	^{137}Cs	5	$(1.79 \pm 0.47) \times 10^3$	$(1.29 \pm 0.19) \times 10^3$ to $(2.48 \pm 0.39) \times 10^3$	
	^{241}Am	4	$(21.73 \pm 5.12) \times 10^5$	$(16.78 \pm 5.71) \times 10^5$ to $(26.46 \pm 12.83) \times 10^5$	
	^{106}Ru	6	$(3.25 \pm 0.46) \times 10^5$	$(2.72 \pm 0.51) \times 10^5$ to $(4.06 \pm 0.91) \times 10^5$	
Sinclair Inlet	^{85}Sr	-	-	-	
	^{237}Pu	9	$(3.59 \pm 1.32) \times 10^5$	$(1.87 \pm 0.19) \times 10^5$ to $(5.63 \pm 0.48) \times 10^5$	

*n = the total number of K_d values obtained for each radionuclide.

**Average K_d represents the average value and one standard deviation from this average for the n number of K_d measurements.

***Range in K_d represents the lowest and highest values of the distribution coefficient obtained in the experiments. The error term represents 2σ propagated counting error for each isotope.

#For ^{137}Cs and ^{241}Am and ^{106}Ru in Lake Michigan, A, B, C correspond to initial sorption periods of 1, 24 and 96 hours, respectively.

For the Hudson River experiments, the desorption K_d values increased as follows:

$$\text{SLOSH V} < \text{SLOSH II} \sim \text{SLOSH III.}$$

mp 0 mp 18 mp 43

There was notable overlap in the K_d values for the SLOSH II and III samples (Table 13), and the average K_d values were approximately twice that of the SLOSH V sediment-water system. The results suggest possible effects of salinity (or ionic strength) on the desorption K_d for ^{137}Cs .

^{106}Ru . As for ^{137}Cs , the desorption K_d for this radionuclide was determined only for Hudson River and Lake Michigan samples. For Lake Michigan, desorption K_d values appear to increase with increasing sorption time, again indicating that lesser amounts of ^{106}Ru were sorbed by the sediments in one hour than in 24 hours or 96 hours of sorption. The desorption K_d value for the third set (sediments allowed to sorb the radionuclide for 96 hours) probably represents an equilibrium K_d value, since earlier sorption experiments for this isotope showed that equilibrium distribution in Lake Michigan samples is attained after 48 hours.

For the Hudson River Estuary sediment-water systems, the desorption K_d values varied as follows:

$$\text{SLOSH III} < \text{SLOSH II} \sim \text{SLOSH V}$$

Mile Post:	43	18	0
Salinity:	00/00	20/00	150/00

However, the lower value for desorption K_d in the freshwater (SLOSH III) system compared to those for the 2 ppt salinity (SLOSH II) and 15 ppt salinity (SLOSH V) systems does not necessarily indicate that ^{106}Ru is more readily desorbed in freshwater because the total amount of ^{106}Ru sorbed by the freshwater sediments was less than the amounts sorbed in the 2 ppt and 15 ppt salinity sediment samples.

^{241}Am . The highest desorption K_d values obtained in the experiments were for this isotope. The K_d values appear to increase with increasing salinity for the Hudson River Estuary samples. For Lake Michigan samples, equilibrium K_d values were not attained in the three different experimental systems within the 4-day duration of the experiments. A large variation in the K_d values was observed for this isotope particularly due to the very low levels of soluble ^{241}Am in the systems (near minimum detection levels of 0.40 dpm/ml). The range in desorption K_d values for the Lake Michigan sediment-water system are given in Table 13.

Summary of Desorption K_d Values. The desorption distribution coefficients for the different radionuclides are compared to adsorption K_d values for the same sediment-water systems in Table 14. For all radionuclides, the K_d values for desorption were higher than the adsorption K_d values by at

Table 14. Summary of adsorption-desorption distribution coefficients with properties of the sediment-water systems.

Sample	pH	Hardness mg/l	Salinity 0/00	Surface Area m ² /g	C mg/g dry	N mg/g dry	K _d ml/g										Sample Treatment		
							137Cs		85Sr		106Ru		237Pu		241Am				
							ADS K _d x 10 ⁻²	DES K _d x 10 ⁻³	ADS K _d x 10 ⁰	DES K _d x 10 ⁻³	ADS K _d x 10 ⁻⁴	DES K _d x 10 ⁻⁵	ADS K _d x 10 ⁻⁴	DES K _d x 10 ⁻⁵	ADS K _d x 10 ⁻⁵	DES K _d x 10 ⁻⁵			
Lake Michigan	7.90	143	<1		62.0	1.8	5.09 (0.31)	5.57 (0.62)	82.2 (7.0)	1.41 (0.51)	5.23 (1.68)	2.79 (0.31)	14.1 (1.7)	1.79 (1.19)	5.48 (3.73)	No equil.	Frozen		
Lake Washington	7.8	42	<1	31.4±6.2									12.0 (1.4)	3.00 (0.12)	1.63 (0.24)	5.22 (2.11)	Fresh (cooled)		
Clinch River	7.92	158	<1		14.0	1.8	13.6 (.40)		124.4 (7.5)	.46 (0.09)	6.75 (0.52)		4.71 (0.40)	1.54 (0.09)	7.33 (3.64)		Fresh (cooled)		
Cattaraugus Creek		202	<1		16.8	0.8	No equil.		62.3 (6.2)		1.39 (0.12)		2.09 (0.65)		2.26 (1.47)		Fresh (cooled)		
Hudson River mp 43	7.90	171	<1		34.4	1.8	4.01 (.20)	3.65 (0.52)	73.7 (10.5)	0.49 (0.14)	3.92 (0.40)	2.52 (0.71)	.93 (0.14)	3.60 (0.37)	1.26 (0.13)	5.29 (0.84)		Freeze dried	
mp 18	7.82	378	2-3		23.0	1.1	8.78 (0.71)	3.34 (0.69)	34.8 (5.3)	MDL (~ 0)	5.15 (1.03)	3.35 (0.37)	3.12 (0.22)	2.98 (0.22)	2.37 (0.88)	8.47 (1.52)		Freeze dried	
mp 0	7.70	2963	15		31.8	5.2	3.56 (0.21)	1.79 (0.47)	MDL (~ 0)	MDL (~ 0)	4.53 (0. .)	1.25 (0.46)	3.87 (0.11)	3.21 (1.35)	2.96 (0.88)	21.73 (5.12)		Freeze dried	
Skagit Bay #1	7.70	6662	29.62	2.6±0.6			1.87 (0.15)				3.61 (0.63)				2.68 (0.74)			Fresh (cooled)	
#4	7.70	5325	31.05	7.0±1.6	13.7	1.6	3.53 (0.07)		MDL (~ 0)		4.51 (0.76)		10.3 (0.8)		2.80 (1.06)			Fresh (cooled)	
Columbia River Mouth	7.8	6250	32.566	8.7±2									1.62 (0.19)	0.25 (0.04)	1.03 (0.18)	0.70 (0.08)		Fresh (cooled)	
Sinclair Inlet	7.82	5606	28.89				1.24 (0.09)		MDL (~ 0)	MDL (~ 0)	5.42 (0.43)		7.40 (1.21)	3.59 (1.32)	4.54 (1.20)			Fresh (cooled)	
Saanish Inlet	7.80	5316	31.243								4.42 (0.52)				3.71 (0.84)			Frozen	
Lake Nitinat	7.8	6188	31.217	40.3±3									4.40 (0.7)	1.74 (0.17)	2.54 (0.31)	>10 (MDL)		Frozen anoxic	
Montmorillonite	7.70	5325	.05				.65 (0.07)								1.91 (0.41)				
Organic Detritus Lake Michigan		141	<1						75.2 (4.1)				2.59 (0.94)						Dried, ground < 63 μm
Skagit Bay		666	29.6						68.3 (4.4)				1.34 (0.94)						Dried, ground < 63 μm

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least a factor of 3. More than an order of magnitude increase in K_d values for desorption compared to adsorption was observed for some radionuclides. These results suggest that under these experimental conditions, sorption is not completely reversible. Radionuclides are strongly bound to the sediments and may be unavailable for release. This indicates that K_d values obtained in adsorption experiments may not be applicable to modeling the release of sediments from suspended or bed sediments.

As in the adsorption K_d measurements, ^{85}Sr showed the lowest desorption K_d values, and ^{241}Am has the highest desorption K_d value. The same increasing trend in desorption K_d values for the radionuclides as in the adsorption experiments is obtained: $^{85}\text{Sr} < ^{137}\text{Cs} < (^{106}\text{Ru} \text{ or } ^{237}\text{Pu}) < ^{241}\text{Am}$.

3.2 Effect of pH on K_d Values

The adsorption of radionuclides to suspended particulates is dependent upon the physico-chemical species of radionuclides and the surface characteristics of the sediments. Both of these may change as a function of pH. Thus, the K_d value of some radionuclides may be affected by changes in pH. Previous experiments in our laboratory have shown some effects of pH variation in both freshwater and anoxic marine systems for ^{241}Am (4). The results presented below are from experiments with water and sediments from Clinch River, Tennessee.

The K_d values obtained, for ^{106}Ru and ^{137}Cs in the Clinch River sediment-water system are shown in Figs. 3 and 4, respectively. The K_d value of ^{106}Ru increases by a factor of approximately 5 between pH 4 and 6. At pH values above 6 there is considerable scatter in the data with no obvious trends. The apparent decrease in K_d values between pH 8 and 10 is believed to be an experimental artifact but will be evaluated in future experiments. Unlike ^{106}Ru which exhibits an increase in K_d values at relatively low pH, there appears to be no effect on the K_d values of ^{137}Cs at pH < 9. There is, however, a slight increase in K_d between pH 9 and 10. It appears that in the Clinch River sediment-water system the K_d values of ^{106}Ru and ^{137}Cs are not affected by the pH changes that are commonly observed in natural waters.

Another radionuclide that was included in the Clinch River experiments was ^{60}Co . Although we do not generally report on ^{60}Co , these results are presented here (Fig. 5) to indicate the effect that changes in pH may have on some radionuclides. Between pH 6.0 and 7.5 the K_d value of ^{60}Co increases more than two orders of magnitude. Not only is this a significantly larger effect than was noted for ^{106}Ru or ^{137}Cs , it also occurs within the pH range of most natural waters. Thus, hydrologic models for ^{60}Co must be more sensitive to pH than models for ^{106}Ru or ^{137}Cs .

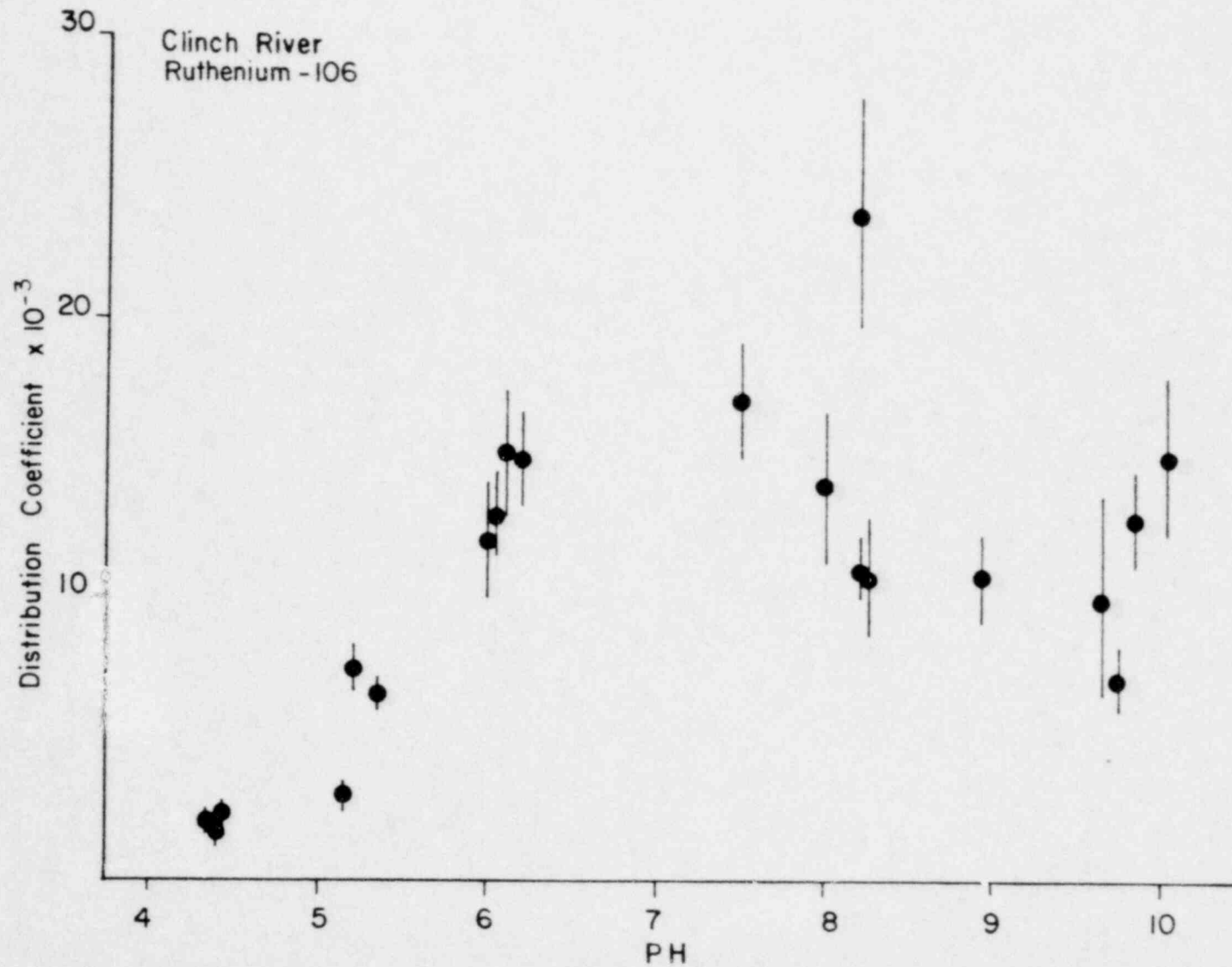


Figure 3. Distribution coefficients as a function of pH for ^{106}Ru in sediment-water systems from Clinch River. Error bars represent 2σ propagated counting error for a single sample.

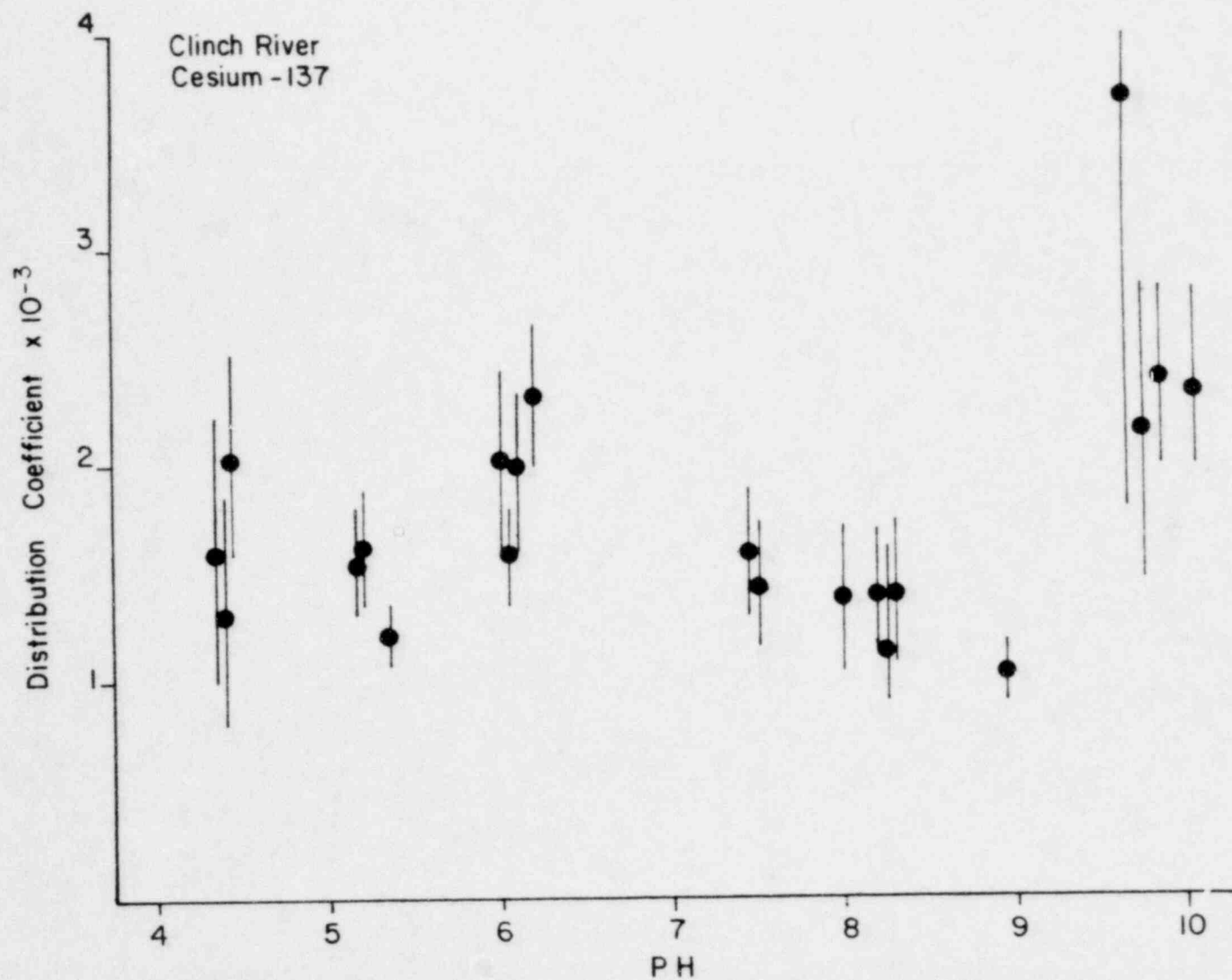


Figure 4. Distribution coefficients as a function of pH for ^{137}Cs in sediment-water systems from Clinch River. Error bars represent 2σ propagated counting error for a single sample.

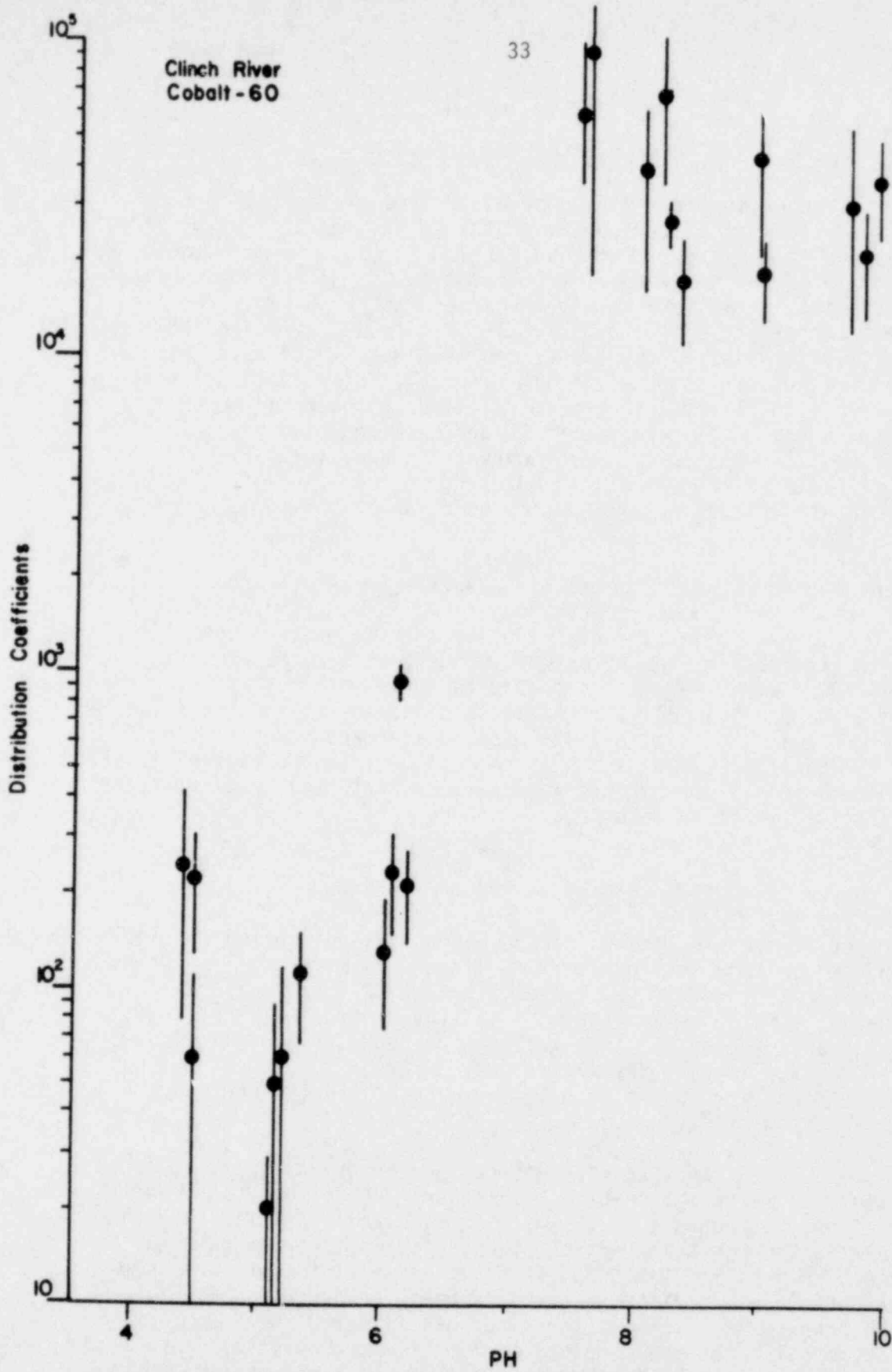


Figure 5. Distribution Coefficients as a function of pH for ^{60}Co in sediment-water systems from Clinch River. Error bars represent 2σ propagated counting error for a single sample.

3.3 Effect of Sediment Concentration

As for pH, some experiments have been reported previously on the effects of sediment concentration on the K_d values of ^{241}Am in sterile and unsterilized systems from Lake Washington (3). More recently, we have completed and analyzed an experiment on the effects of sediment concentration in the Lake Michigan sediment-water system. The sediment concentrations ranged from 16 mg/l to 340 mg/l and the radionuclides ^{237}Pu , ^{137}Cs , ^{106}Ru , and ^{60}Co were included. Results of this experiment are shown in Fig. 6. For all these radionuclides there is a significant increase in K_d value at lower sediment concentrations. This corresponds to the data reported previously for ^{241}Am (3). The apparently sharp increase in K_d values of ^{106}Ru and ^{137}Cs at sediment concentrations below 50 mg/l is thought to be an experimental artifact resulting from sorption of these radionuclides to the filters.

Sediment-water systems from Cattaraugus Creek were used to determine the effect of sediment concentration on the K_d values of ^{137}Cs and ^{241}Am (Figs. 7 and 8). The distribution coefficients of both ^{137}Cs and ^{241}Am increase at lower sediment concentrations, especially between 100 and 250 mg/l. These results agree with results reported by Aston and Duursma (8); they showed a rapid increase in K_d values of ^{65}Zn , ^{106}Ru , and ^{137}Cs with a decrease in sediment concentrations below 400 mg/l. These experiments need to be extended to even lower sediment concentrations in order to correspond to the concentration of suspended sediments in most natural waters.

3.4 Effect of Organic Ligands

The effect of organic ligands on the values of sediment-water distribution coefficients for four radionuclides (^{241}Am , ^{57}Co , ^{106}Ru , and ^{137}Cs) was determined in the presence of one of five organic compounds: 1-nitroso-2-naphthol, 1, 10-phenanthroline, acetic acid, salicylic acid, or EDTA. These organic compounds (shown in Fig. 9) were selected primarily for the variety of their functional groups available for co-ordination complex bonding of the radionuclides and/or sorption by sediments.

Experiments were conducted with natural sediments and water collected from two freshwater sites--Lake Washington (Washington State) and the mouth of the Cattaraugus Creek entering Lake Erie (New York State). To minimize the potential effect of bacteria on experimental results all reaction vessels (i.e., 250 ml screw cap plastic bottles) were either autoclaved or rinsed with 95% ethanol before use. The water from both environmental sampling sites was filtered through a 0.20 μm Nucleopore polycarbonate membrane to remove microorganisms. The sediments from Lake Washington (but not Cattaraugus Creek) were subjected to ^{60}Co irradiation for sterilization purposes, although this

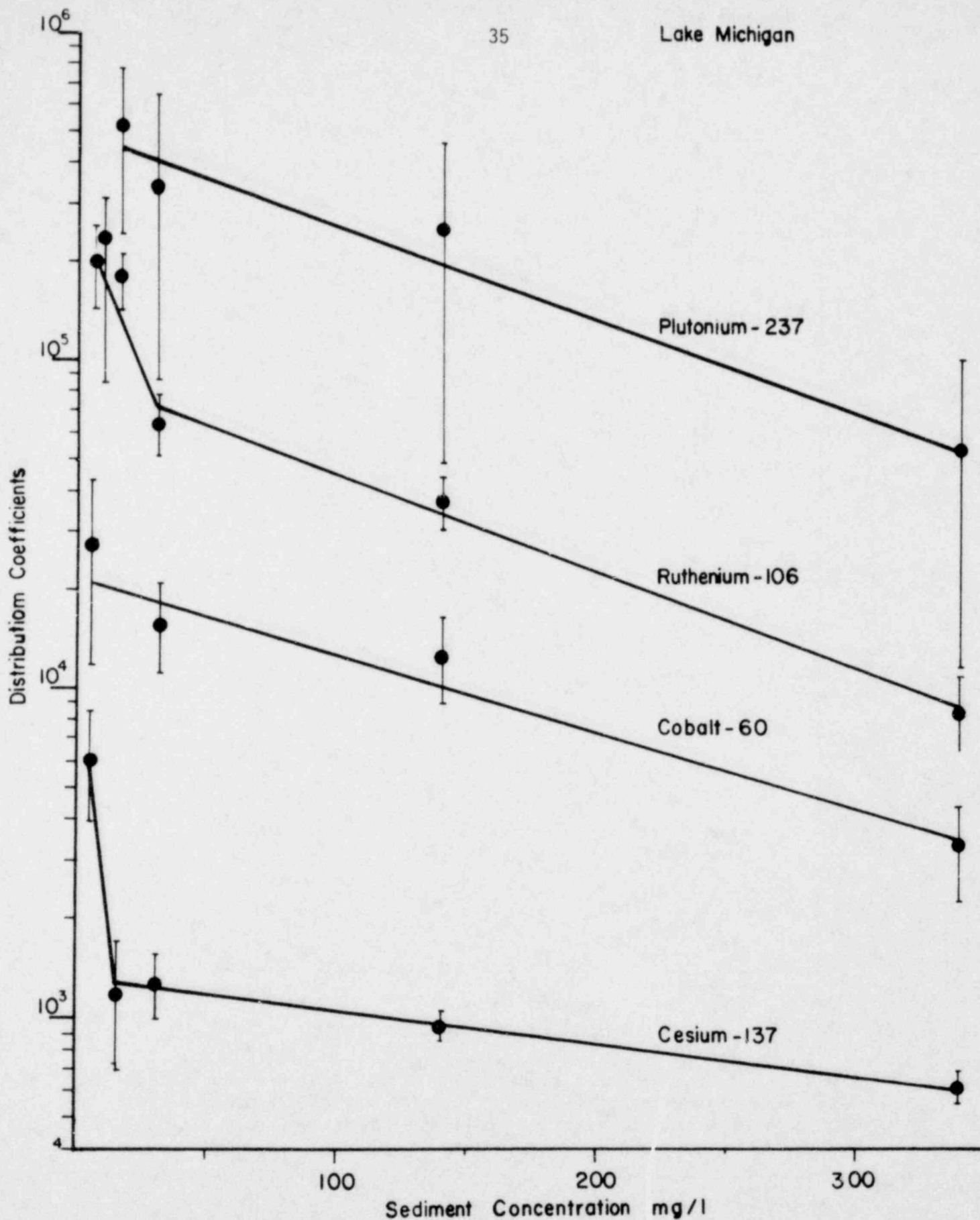


Figure 6. Distribution coefficients as a function of sediment concentration for ⁶⁰Co, ¹⁰⁶Ru, ¹³⁷Cs and ²³⁷Pu in sediment-water systems from Lake Michigan. Error bars are 2σ propagated counting error for a single sample.

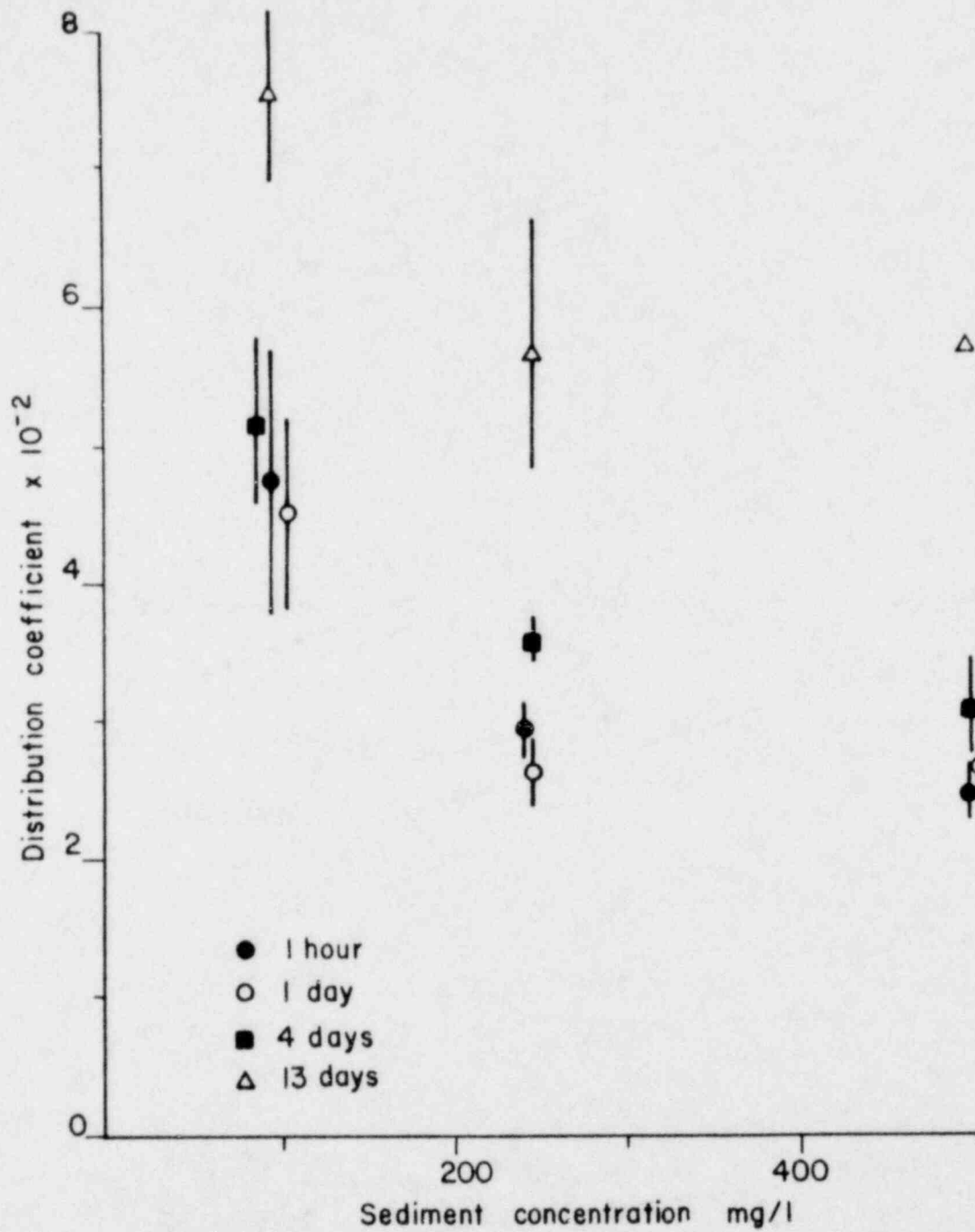


Figure 7. Distribution coefficients for ^{137}Cs in the Cattaraugus Creek sediment-water system as a function of sediment concentration. Error bars are 1σ difference between means of replicate samples. Lack of error bars indicates that only one sample was analyzed.

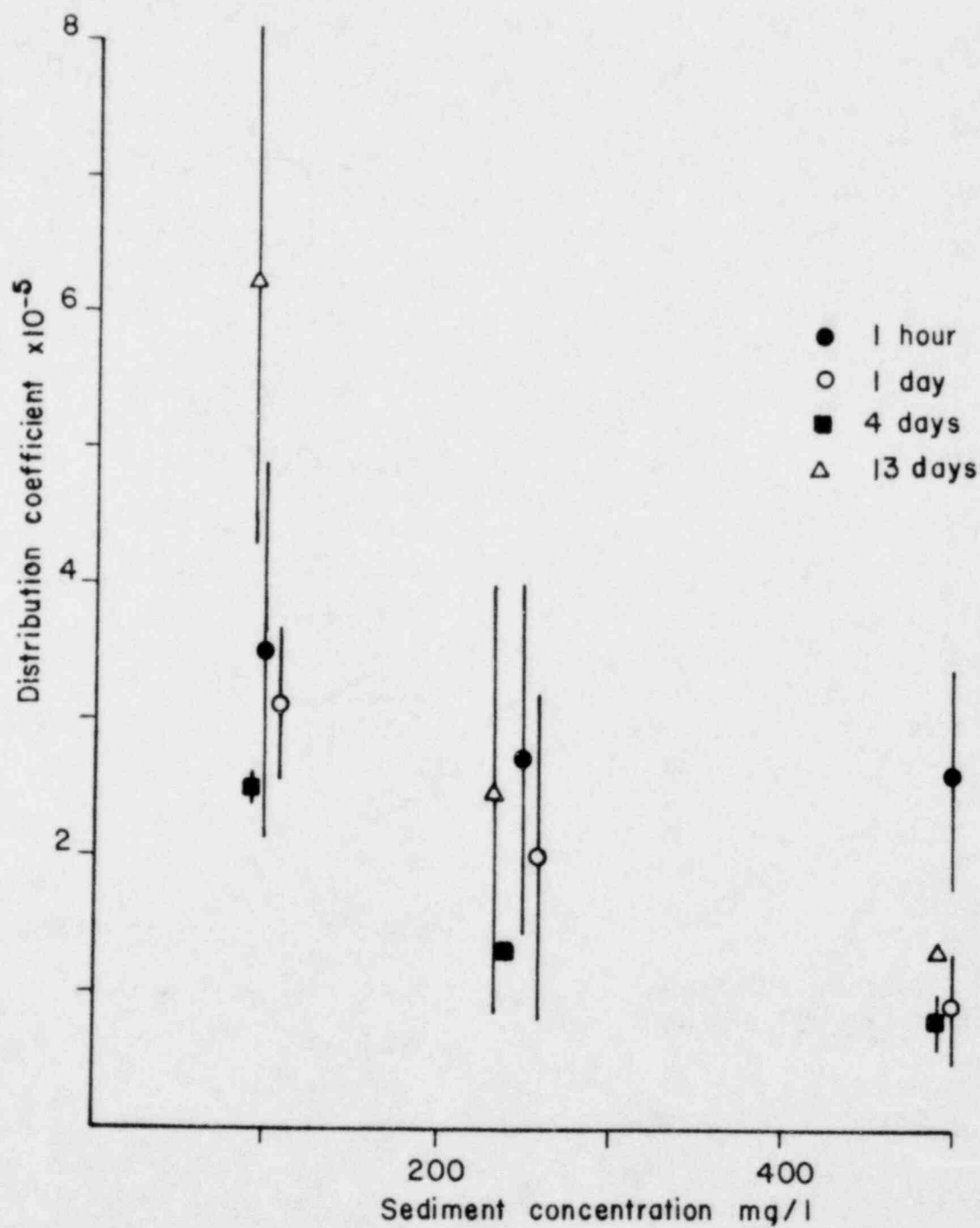
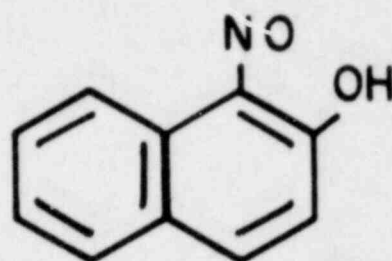
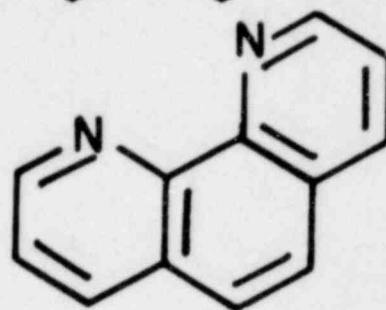


Figure 8. Distribution coefficients for ^{241}Am in the Cattaraugus Creek sediment-water system as a function of sediment concentration. Error bars are 1σ difference between means of replicate samples. Lack of error bars indicates that only one sample was analyzed.

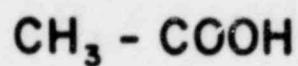
1-Nitroso-2-Naphthol



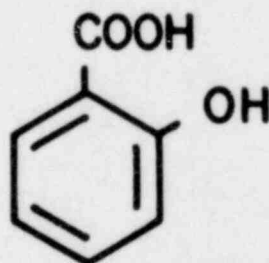
1,10-Phenanthroline



Acetic Acid



Salicylic Acid



EDTA (N,N'-1,2-Ethanediylobis[N-(carboxymethyl)glycine])

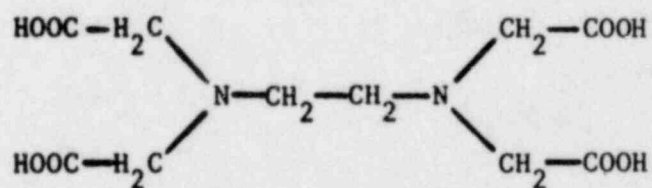


Figure 9. Organic compounds tested for effects upon sediment-water K_d values.

procedure may have produced some chemical alterations in the in situ organic matter and sediments. The sediment size fraction passing through a 63 μ m sieve was used for the experiments.

To estimate the length of time required for sediment-water-organic systems to achieve equilibrium, an initial experiment was conducted in which K_d values for ^{241}Am , ^{106}Ru , and ^{137}Cs were determined as a function of time. For this work the system chosen included sediments and water from Cattaraugus Creek and EDTA as the organic compound. Two concentrations of EDTA-- 10^{-6} M and 10^{-10} M--were tested. Although data were obtained for five time periods (i.e., 4 hr, 52 hr, 101 hr, 169 hr, and 480 hr), only the results for the minimum and maximum time periods are summarized in Table 15. For all three radionuclides and for both concentrations of EDTA no significant differences were observed between K_d values at 4 and 480 hours (student "t" test, $\alpha = 0.05$). Consequently, for subsequent experiments, it was assumed that a time period of approximately 150 hours would be sufficient to achieve stable values of K_d .

The next experiment, using Lake Washington sediments and water, involved an evaluation of the effects of the five organic compounds shown in Fig. 9 on sediment-water K_d values for ^{241}Am , ^{57}Co , ^{106}Ru , and ^{137}Cs . To determine whether any of the organics would, in fact, affect K_d values, relatively high concentrations of the organic compounds were tested (i.e., 10^{-4} M for 1,10-phenanthroline, acetic acid, salicylic acid, and EDTA and 10^{-5} M for 1-nitroso 2-naphthol). For the purpose of statistical analyses, three sample bottles were run for each organic compound tested. Furthermore, to estimate intrasample variability, three sample aliquots were withdrawn and analyzed separately from each of the three EDTA sample bottles. All sample aliquots were collected for radioactivity measurements after 165 hours of shaking. The ranges of pH values in samples at the beginning and the end of the experiment were 7.05-7.15 and 7.9-9.1, respectively. The distribution of the final pH values showed no apparent correlation with the different organic compound types tested.

The results of the experiment to determine K_d values as a function of various organic compounds are summarized in Table 16 and in Figs. 10 through 13. For EDTA, the individual values in Table 16 represent only the means of triplicate measurements on individual sample bottles. In Figs. 10 through 13, however, the EDTA values are presented as mean values with one standard deviation unit for each of the three sample bottles (i.e., EDTA-1, EDTA-2, and EDTA-3). The latter information gives the intrasample variability to be expected in the measurements from an individual sample bottle. Using the data in Table 16, Kruskal-Wallis statistical tests (9) were performed for each of the radionuclides to determine if the associated K_d values for the organic compounds and the control (i.e., no organic compound added) were different. The results of these analyses indicated that

Table 15. Sediment-water K_d values for Cattaraugus Creek system at two different time periods and two concentrations of EDTA

Time (hr)		K_d (ml/g)													
		10^{-6} M EDTA						10^{-10} M EDTA							
		n	$^{241}\text{Am}(x10^5)$		$^{106}\text{Ru}(x10^5)$		$^{137}\text{Cs}(x10^3)$		n	$^{241}\text{Am}(x10^5)$		$^{106}\text{Ru}(x10^5)$		$^{137}\text{Cs}(x10^3)$	
\bar{x}	s		\bar{x}	s	\bar{x}	s	\bar{x}	s		\bar{x}	s	\bar{x}	s		
4	3	5.9	2.8	1.6	0.4	1.4	0.1	3	11.3	12.0	2.2	1.0	1.8	0.4	
480	3	11.2	4.9	1.2	0.3	1.9	0.8	3	30.7	20.5	2.2	1.6	2.4	2.1	

n = number of measurements

\bar{x} = mean

s = one standard deviation unit

Table 16. Sediment-water K_d values for Lake Washington system as a function of different organic compounds. Triplicate samples done for each organic compound (except acetic acid).

Organic Compound Added (Concentration)	K_d			
	$^{241}\text{Am}(\times 10^4)^*$	$^{57}\text{Co}(\times 10^4)^*$	$^{106}\text{Ru}(\times 10^4)^*$	$^{137}\text{Cs}(\times 10^2)$
None (i.e., control) $\bar{x} \pm s^{**}$	3.11 1.78 1.96 2.28 \pm 0.71	0.31 0.36 0.30 0.32 \pm 0.03	1.54 0.83 0.90 1.09 \pm 0.39	3.81 3.36 5.28 4.15 \pm 1.00
1-nitroso-2-naphthol (10^{-5} M) $\bar{x} \pm s^{**}$	11.24 6.07 5.98 7.76 \pm 3.01	54.41 18.31 34.73 35.82 \pm 18.07	3.79 1.99 2.04 2.61 \pm 1.03	5.83 4.71 4.57 5.04 \pm 0.69
1,10-phenanthroline (10^{-4} M) $\bar{x} \pm s^{**}$	23.64 28.10 24.17 25.30 \pm 2.44	20.19 12.14 18.10 16.81 \pm 4.18	3.76 3.81 3.76 3.78 \pm 0.03	3.58 2.81 3.26 3.22 \pm 0.39
acetic acid (10^{-4} M) $\bar{x} \pm s^{**}$	3.88 7.32 5.60 \pm 2.43	0.42 0.48 0.44 \pm 0.04	1.70 2.89 2.30 \pm 0.84	2.15 3.24 2.70 \pm 0.77
Salicylic acid (10^{-4} M) $\bar{x} \pm s^{**}$	3.87 3.13 1.82 2.94 \pm 1.04	0.38 0.26 0.26 0.30 \pm 0.07	1.72 1.20 0.76 1.23 \pm 0.48	3.17 4.39 3.76 3.77 \pm 0.61
EDTA (10^{-4} M) $\bar{x} \pm s^{**}$	0.05 0.04 0.04 0.05 \pm 0.01	0.02 0.00 0.01 0.01 \pm 0.01	4.04 2.97 3.35 3.45 \pm 0.54	3.34 2.78 4.29 3.47 \pm 0.76

* significant differences between groups at $\alpha = 0.05$ level by Kruskal-Wallis statistical test

** \bar{x} and s correspond to the mean and one standard deviation unit, respectively, for the triplicate values listed

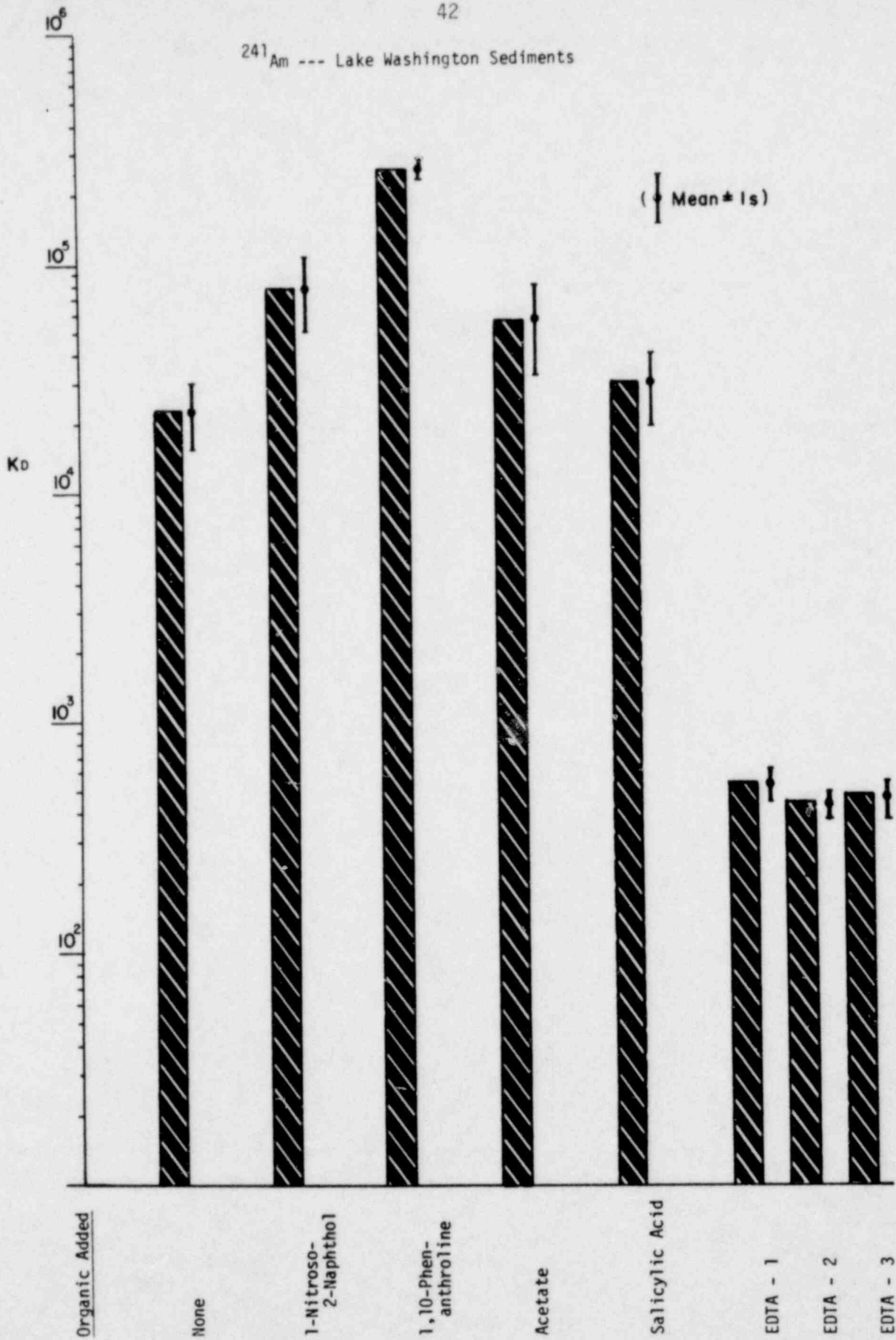
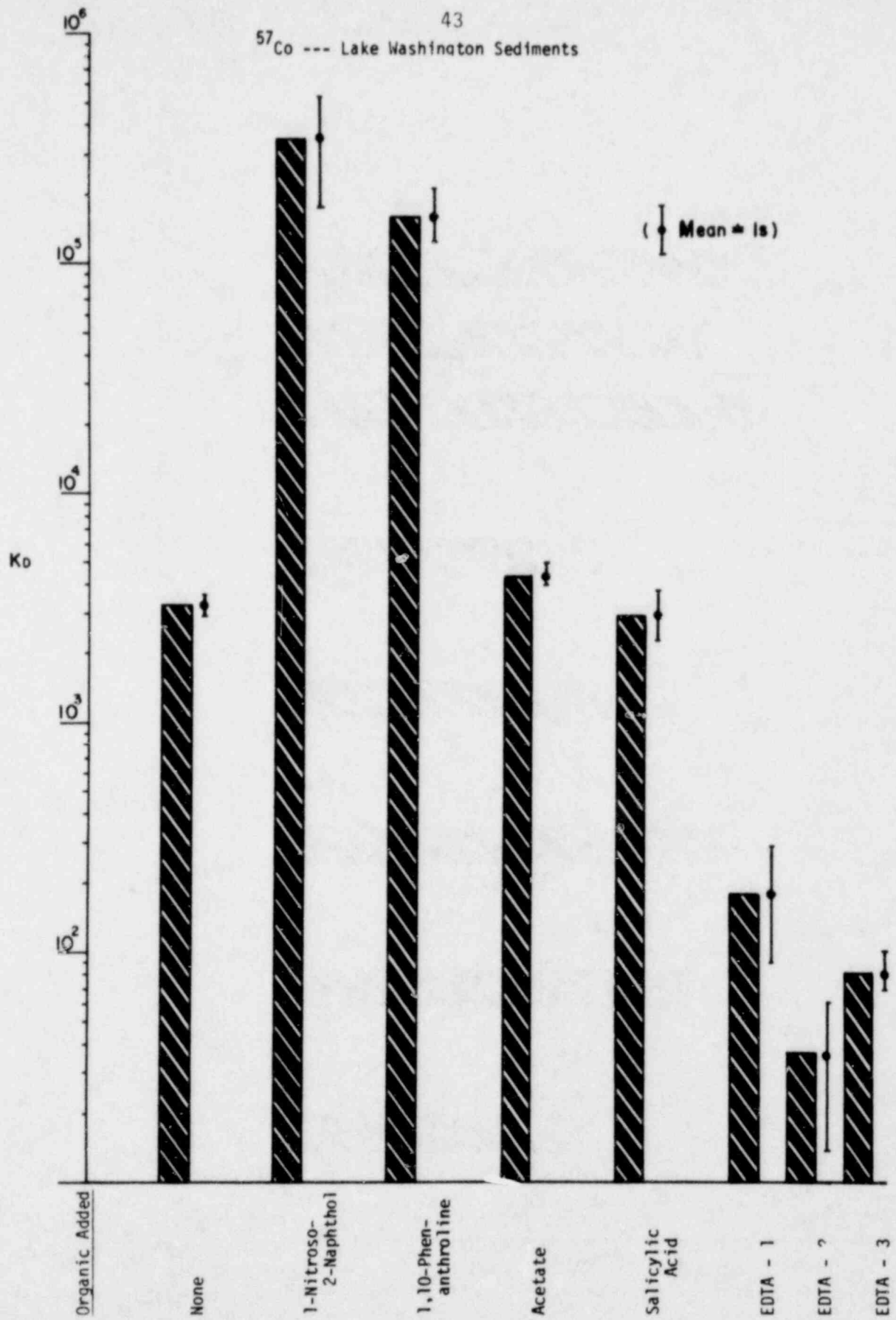
^{241}Am --- Lake Washington Sediments

Figure 10. Effects of organic compounds on K_d values of ^{241}Am .

^{57}Co --- Lake Washington SedimentsFigure 11. Effects of organic compounds on K_d values of ^{57}Co .

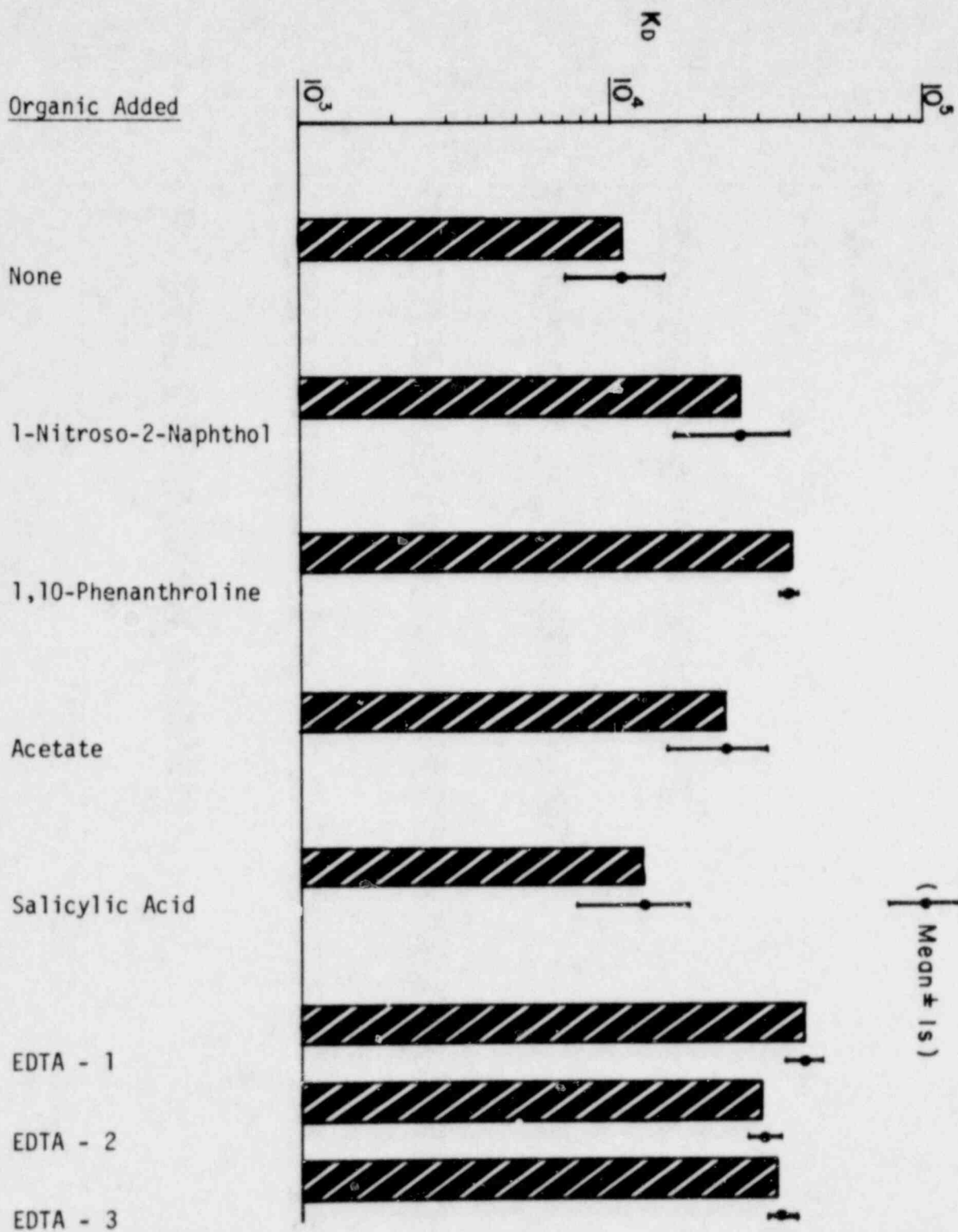


Figure 12. Effects of organic compounds on K_d values of ^{106}Ru .

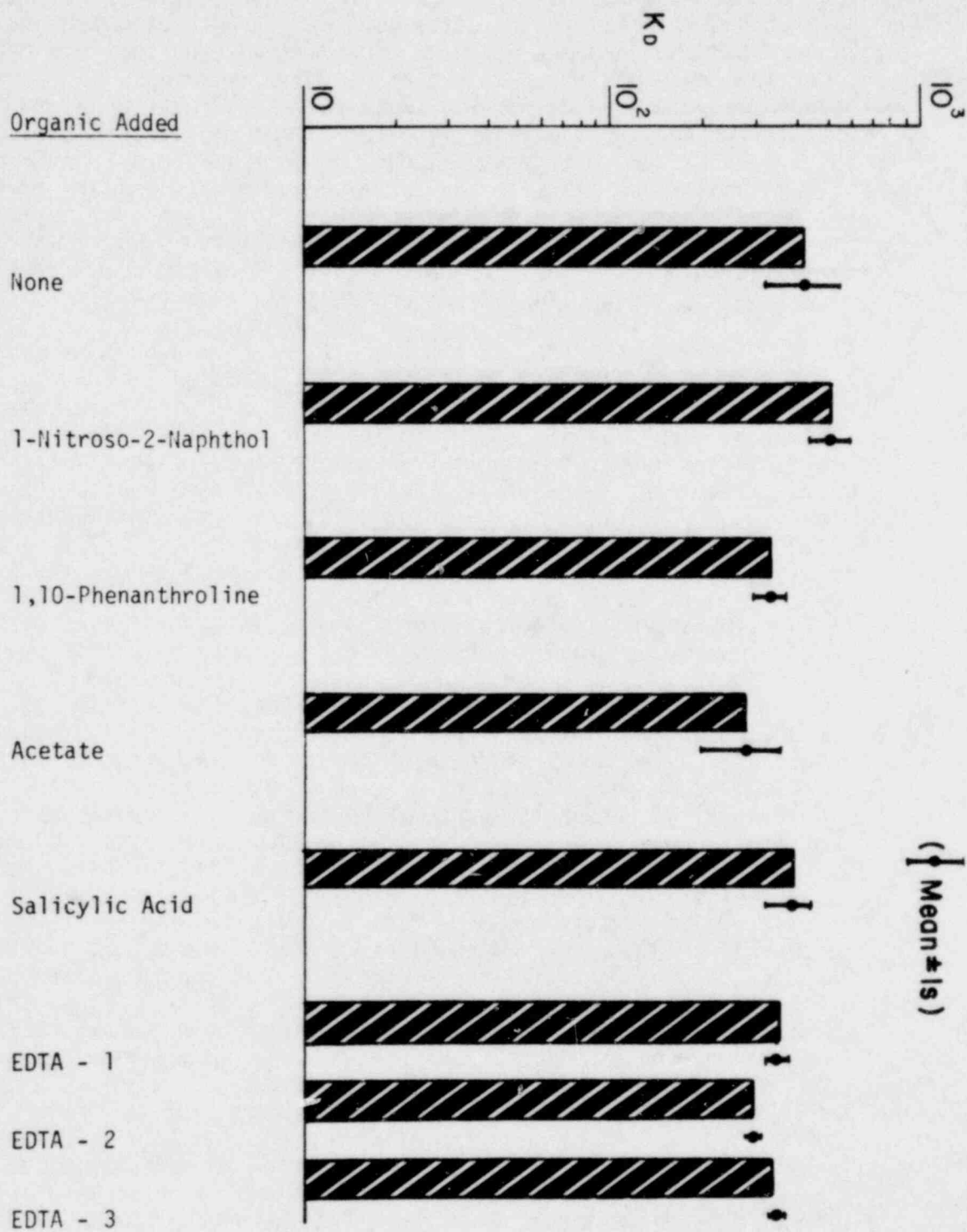


Figure 13. Effects of organic compounds on K_d values of ^{137}Cs .

statistically significant differences ($\alpha = 0.05$ level) were observed for the K_d values of ^{241}Am , ^{57}Co , and ^{106}Ru , but not for ^{137}Cs . For both ^{241}Am and ^{57}Co , values of K_d relative to the controls were higher with 1-nitroso-2-naphthol, 1,10-phenanthroline, and possibly, acetic acid, whereas much lower values were observed with EDTA. For ^{106}Ru 1-nitroso-2-naphthol, 1,10-phenanthroline, acetic acid and EDTA all appeared to produce higher K_d values.

During the studies summarized in Table 16, concurrent control samples were run with identical amounts of water, organic compounds and radionuclides but no sediment. Information from these controls was intended to give upper limit estimates of the amounts of each radionuclide that might be retained on filters independent of the sediment particles. The results of these measurements, presented in Table 17, are reported as the ratios of the filter radionuclide concentrations of systems without sediment to those with sediment. Since these ratios are low in general, the magnitude of the differences indicated between K_d values in Table 16 and Figs. 10 through 13 cannot be explained simply by retention of the radionuclides on filters independent of sediment. Hence, the observed trends in the K_d values of Table 16 appear to be real rather than experimental artifacts that might have resulted from retention of non-sediment associated organo-radionuclide complexes.

A next step in evaluating the effects of organic compounds on sediment-water associations of radionuclides is to determine K_d values as a function of varying concentrations of organic compounds. For this work those organics which affected K_d values at relatively high concentrations (see Table 16) will be tested. One such preliminary experiment with EDTA and the combined radionuclides ^{241}Am , ^{106}Ru , and ^{137}Cs has been conducted. Figure 14 illustrates the results of this experiment for ^{241}Am in a Cattaraugus Creek sediment-water system. A marked decrease in the K_d value for ^{241}Am occurs between EDTA concentrations of 10^{-4} M and 10^{-6} M. The accompanying data for ^{106}Ru and ^{137}Cs are not presented since no obvious concentration-dependent trends were observed for either of these radionuclides.

The above preliminary results confirm the hypothesis that certain organic compounds (e.g., 1-nitroso-2-naphthol, 1,10-phenanthroline, EDTA and acetic acid) have the potential to affect sediment-water distribution coefficients for some of the radionuclides tested (i.e., ^{241}Am , ^{57}Co , and ^{106}Ru). However, the magnitude and direction (i.e., higher or lower K_d values) of these effects are functions of not only the physico-chemical properties but also the environmental (i.e., system) reactions of the radionuclides and the organic compounds. For example, the dramatic decrease in the solubilizing effect of EDTA upon ^{241}Am occurred only between 10^{-4} M and 10^{-6} M concentrations of this ligand (see Fig. 14). One hypothesis that might explain such concentration dependent behavior is that at concentrations lower than 10^{-6} M some other element(s) may be competing with ^{241}Am for this

Table 17. Ratios (R) of radionuclide activities retained on filters from systems without sediment to systems with sediment.

Organic Compound Added (concentration)	R			
	^{241}Am	^{57}Co	^{106}Ru	^{137}Cs
None (i.e., control)	0.08	0.02	0.08	0.20
1-nitroso-2-naphthol (10^{-5} M)	0.03	0.28	0.15	0.14
1,10-phenanthroline (10^{-4} M)	0.03	0.01	0.03	0.20
Acetic Acid (10^{-4} M)	0.07	0.03	0.05	0.29
Salicylic Acid (10^{-4} M)	0.10	0.02	0.07	0.14
EDTA (10^{-4} M)	0.15	0.64	0.02	0.19

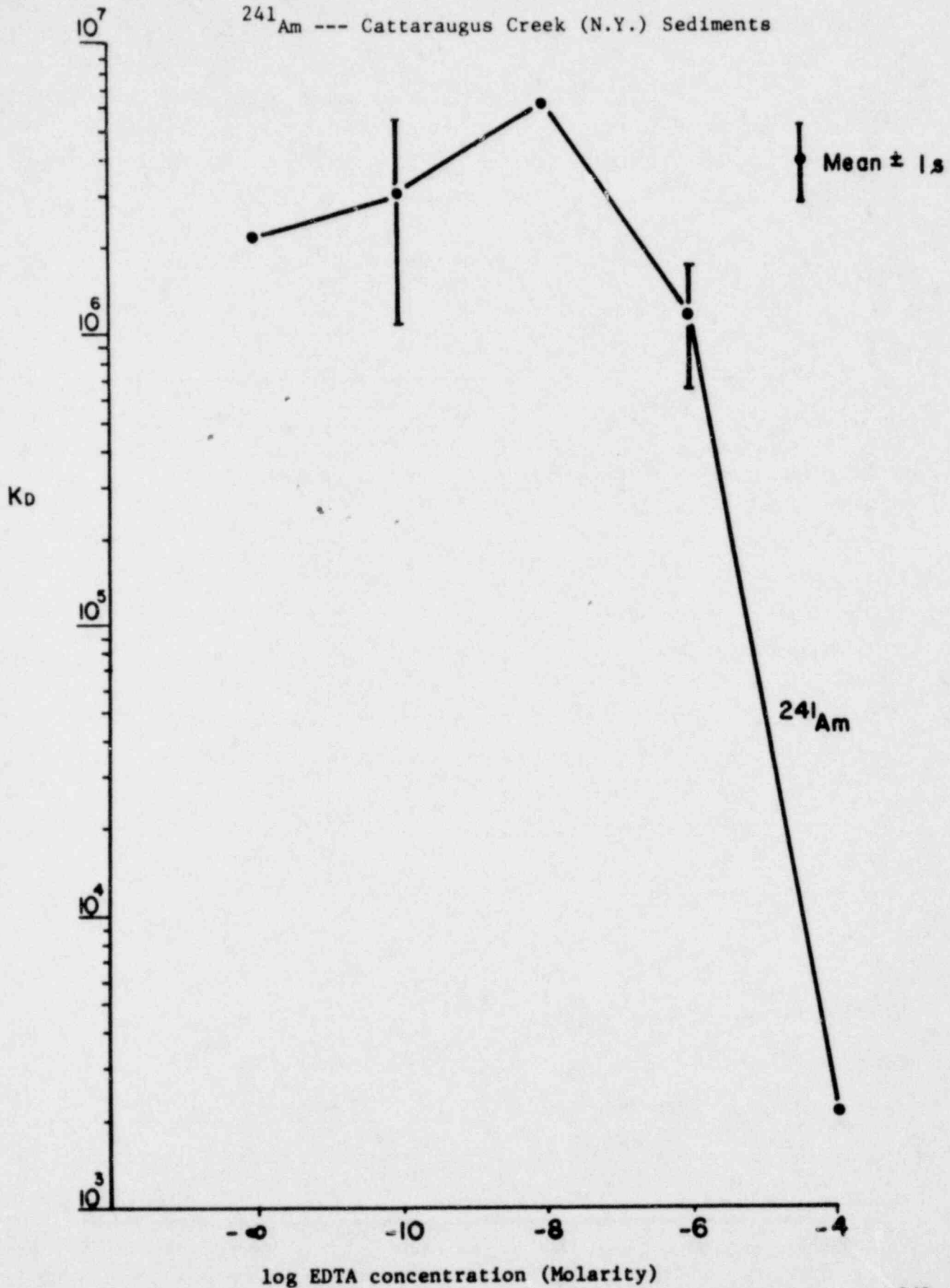


Figure 14. Effect of varying EDTA concentration of K_d values of ^{241}Am . Values at 0 M, 10^{-8} and 10^{-4} M EDTA are single point measurements. Values at 10^{-10} and 10^{-6} M EDTA are the mean \pm one standard deviation unit for triplicate sample measurements.

organic ligand. Such an explanation emphasizes the need to chemically characterize each environmental system before the distribution coefficient behavior of a given radionuclide can be fully understood and predicted.

Of equal interest in the above work is the observation that certain organic ligands (e.g., 1-nitroso-2-naphthol and 1,10-phenanthroline) can increase the associations of certain radionuclides (e.g., ^{241}Am and ^{57}Co) with a sediment phase. This implies that such organic ligands are involved in two reaction processes: a) they are complexing the radionuclide and b) they are associating with the sediment particles. Similar phenomena have been observed for certain trace metal-organic combinations by Davis and Leckie (10), (11). Depending upon the relative affinities of such sediment-associating ligands for the radionuclide and for the sediment particles, a lowering of the concentration of such a ligand may result in a K_d value lower than that in a control system (i.e., no ligand added). This "solubilizing" effect, which is opposite to that observed at higher concentrations of the ligand, would result from the continued complexing of the radionuclide by the ligand but a concentration-dependent dissociation of the majority of the ligand molecules from the sediment particles (i.e., a phenomenon analogous to that observed for EDTA in the above work). It is our intention to look for such ligand concentration-dependent effects on K_d values for both 1-nitroso-2-naphthol and 1,10-phenanthroline in the subsequent work of this research project.

In summary, organic compounds certainly have the capacity to affect the distributions of certain radionuclides in natural sediment-water systems. For this reason studies of organic ligand-radionuclide-sediment associations warrant attention. However, it must be noted that a full understanding of these interactions requires information on the physical and chemical properties of: a) the given radionuclide, b) the given organic ligand, c) the given solid phases, and d) the aqueous environment.

3.5 Dialysis Experiments

During the past year we have completed two dialysis experiments to compare the uptake of radionuclides by suspended sediments, phytoplankton and organic detritus in filtered water from Lake Michigan. These experiments complement the constant shaking experiments for the determination of K_d values. In addition, they indicate which biogeochemical pathways are most important for determining the transport and eventual deposition of individual radionuclides. Since the presence of a dialysis membrane prohibits the transfer of physico-chemical species larger than 6000-8000 nominal molecular weight, the data from these experiments may indicate which physico-chemical species predominate in the Lake Michigan system.

Finally, these results can be compared to dialysis experiments that have been completed and reported previously for marine waters from the Eastern Pacific Ocean and the Skagit River Estuary, (3 and 4).

For the Lake Michigan experiments, water and sediment were provided by Dr. David Edgington of Argonne National Laboratory. Both water and sediment were collected from the eastern side of Lake Michigan at approximately 43° 00' N., 86° 20' W. A sediment core was collected from a depth of 63 m in September 1976, sectioned into 2 cm slices and frozen. The clay and silt fraction (< 63 μm particle size) from the top 4 cm was used for these experiments. The water sample was collected from 60 m in December 1977. The phytoplankton species, *Chlorella vulgaris*, was obtained from a stock culture maintained by Dr. Frieda Taub of the College of Fisheries, University of Washington. Detritus was prepared from aquatic macrophytes collected from Lake Washington. Samples of *Myriophyllum* sp. were washed with distilled water, dried at 103°C, ground with a porcelain mortar and pestle and sieved to obtain the < 63 μm size fraction.

In each experiment, 700 ml of filtered (<0.22 μm) Lake Michigan water was spiked and allowed to equilibrate for at least ten days. At the beginning of the experiment ($t = 0$), dialysis sacs containing 60 ml of filtered, unspiked Lake Michigan water and predetermined amounts of particulates were suspended in the spiked outer chamber. To facilitate transfer across the dialysis membrane and maintain homogeneity within each of the chambers, glass rods with vanes were used to stir within the dialysis sacs and a magnetic stirrer was used to circulate water in the outer chamber (see Fig. 1).

At each sampling time, a 5 ml sample was removed from each chamber and filtered through a 0.22 μm filter to collect the particulate fraction; 3 ml of the filtrate were then used to measure the soluble concentration. In order to assure identical counting geometries, the filter with the particulates was dissolved in the counting vial with 3 ml of 8 N HNO_3 .

One experiment was spiked with ^{241}Am , ^{137}Cs , ^{106}Ru , ^{60}Co , ^{90}Sr , and ^{244}Cm . During the first day samples were collected from each chamber after 2, 4, 10, and 24 hrs to assess the rate of diffusion across the dialysis membrane. Subsequent samples were taken after 5, 12, and 20 days to determine if equilibrium among the different chambers was attained and maintained throughout the experiment.

Radiochemical analyses for ^{90}Sr and ^{244}Cm have not been completed; however, the results for the γ -emitting radionuclides ^{241}Am , ^{137}Cs , ^{106}Ru , and ^{60}Co are presented in Tables 18-21. These results are presented as relative concentrations (cpm/ml) of soluble and particulate radionuclides within each chamber at each sampling time. By comparing the concentration of soluble radionuclides in each compartment, we can estimate the time required for each radionuclide

Table 18. Relative concentrations of ^{241}Am during a 20 day dialysis experiment.

Time		Spiked Chamber cpm/ml \pm 2SD	Dialysis Chambers			
			Blank cpm/ml \pm 2SD	Plankton cpm/ml \pm 2SD	Sediment cpm/ml \pm 2SD	Detritus cpm/ml \pm 2SD
T=0	soluble	161.4 (0.5)				
	particulate	66.7 (0.4)				
1 hr	soluble	247.0 (2.8)	< 0.14	< 0.44	< 0.45	< 0.38
	particulate	3.7 (0.3)	< 0.08	< 0.11	< 0.18	< 0.18
4 hrs	soluble	83.4 (0.5)	< 0.32	< 0.25	7.3 (0.8)	0.9 (0.6)
	particulate	139.4 (0.5)	0.2 (0.1)	< 0.12	0.2 (0.1)	< 0.13
10 hrs	soluble	92.0 (1.8)	0.6 (0.2)	0.9 (0.7)	0.4 (0.2)	2.2 (0.9)
	particulate	134.9 (1.5)	0.3 (0.2)	0.4 (0.1)	1.3 (0.2)	1.5 (0.3)
24 hrs	soluble	124.7 (2.1)	2.9 (1.3)	4.7 (1.0)	< 1.3	8.9 (1.1)
	particulate	101.6 (0.5)	1.1 (0.2)	Lost	4.9 (0.4)	2.7 (0.4)
5 days	soluble	97.0 (1.9)	8.8 (1.0)	12.7 (1.1)	6.3 (1.0)	10.4 (1.4)
	particulate	36.9 (0.9)	5.2 (0.3)	17.9 (0.6)	73.7 (1.8)	41.9 (0.9)
12 days	soluble	30.8 (0.4)	12.7 (1.1)	20.1 (0.7)	113.9 (3.0)	77.9 (1.7)
	particulate	51.9 (0.4)	1.6 (0.4)	13.3 (0.6)	8.5 (0.5)	38.0 (0.9)
20 days	soluble	7.8 (1.0)	4.8 (1.0)	17.4 (1.6)	4.9 (0.9)	10.3 (1.0)
	particulate	52.4 (1.0)	4.4 (0.4)	21.0 (0.7)	144.3 (1.0)	126.5 (1.5)

Table 19. Relative concentrations of ^{137}Cs during a 20 day dialysis experiment.

Time	Spiked Chamber cpm/ml \pm 2SD	Dialysis Chambers				
		Blank cpm/ml \pm 2SD	Plankton cpm/ml \pm 2SD	Sediment cpm/ml \pm 2SD	Detritus cpm/ml \pm 2SD	
T=0	soluble particulate	103.6 (0.4) 1.2 (0.6)				
1 hr	soluble particulate	83.4 (1.5) 1.1 (0.1)	39.7 (0.3) 1.0 (0.1)	42.5 (1.0) 1.1 (0.1)	31.3 (0.9) 12.5 (0.4)	35.9 (0.9) 2.0 (0.2)
4 hrs	soluble particulate	75.2 (0.4) 0.8 (0.1)	66.9 (1.3) 1.2 (0.1)	69.2 (0.5) 1.6 (0.2)	85.1 (1.4) 7.5 (0.1)	74.5 (1.4) 1.0 (0.1)
10 hrs	soluble particulate	64.7 (1.3) 1.7 (0.2)	54.8 (0.4) 1.3 (0.1)	59.7 (1.2) 1.5 (0.1)	58.1 (0.4) 73.0 (0.4)	64.6 (1.3) 5.6 (0.3)
24 hrs	soluble particulate	62.2 (1.3) 1.6 (0.1)	61.9 (1.8) 1.6 (0.1)	60.7 (1.3) Lost	61.0 (1.8) 125.8 (0.8)	69.0 (1.4) 4.0 (0.3)
5 days	soluble particulate	53.1 (1.2) 1.7 (0.2)	49.1 (1.2) 1.2 (0.1)	48.8 (1.2) 2.4 (0.2)	57.9 (1.2) 206.2 (2.4)	57.3 (1.8) 5.8 (0.3)
12 days	soluble particulate	51.3 (0.4) 1.0 (0.1)	61.6 (1.3) 0.7 (0.2)	57.1 (0.8) 0.8 (0.2)	283.2 (3.7) 16.7 (0.5)	62.6 (1.3) 7.1 (0.3)
20 days	soluble particulate	60.6 (1.3) 1.8 (0.2)	62.8 (1.3) 1.3 (0.2)	56.1 (1.7) 1.7 (0.2)	72.6 (1.4) 150.7 (0.9)	54.8 (1.2) 11.3 (0.4)

Table 20. Relative concentrations of ^{106}Ru during a 20 day dialysis experiment.

Time		Spiked Chamber cpm/ml \pm 2SD	Dialysis Chambers			
			Blank cpm/ml \pm 2SD	Plankton cpm/ml \pm 2SD	Sediment cpm/ml \pm 2SD	Detritus cpm/ml \pm 2SD
T=0	soluble	10.5 (0.2)				
	particulate	4.0 (0.1)				
1 hr	soluble	16.0 (0.8)	0.31 (0.11)	<0.35	0.41 (0.31)	0.42 (0.32)
	particulate	0.4 (0.1)	<0.08	<0.07	0.23 (0.12)	0.18 (0.08)
4 hrs	soluble	6.2 (0.2)	0.65 (0.49)	0.95 (0.20)	0.67 (0.52)	0.90 (0.52)
	particulate	7.6 (0.1)	0.17 (0.08)	<0.09	0.08 (0.04)	0.16 (0.08)
10 hrs	soluble	7.5 (0.7)	0.96 (0.17)	1.07 (0.53)	0.85 (0.17)	1.64 (0.60)
	particulate	7.6 (0.3)	0.35 (0.07)	0.31 (0.05)	0.24 (0.11)	0.35 (0.16)
24 hrs	soluble	9.1 (0.7)	1.20 (0.85)	0.94 (0.59)	1.44 (0.82)	2.11 (0.61)
	particulate	6.0 (0.1)	0.35 (0.10)	Lost	0.94 (0.25)	0.41 (0.20)
5 days	soluble	7.1 (2.7)	1.96 (0.57)	1.95 (0.59)	1.97 (0.59)	1.89 (0.85)
	particulate	2.2 (0.2)	0.27 (0.09)	0.42 (0.18)	0.86 (0.74)	0.71 (0.21)
12 days	soluble	2.6 (0.2)	2.18 (0.58)	1.61 (0.37)	3.60 (1.22)	2.23 (0.62)
	particulate	3.8 (0.1)	0.26 (0.16)	0.66 (0.17)	0.36 (0.19)	1.15 (0.19)
20 days	soluble	1.7 (0.6)	1.34 (0.59)	1.60 (0.81)	1.64 (0.58)	2.21 (0.59)
	particulate	3.8 (0.3)	0.37 (0.15)	0.99 (0.20)	2.35 (0.28)	2.04 (0.26)

Table 21. Relative concentrations of ^{60}Co during a 20 day dialysis experiment.

Time		Spiked Chambers cpm/ml \pm 2SD	Dialysis Chambers			
			Blank cpm/ml \pm 2SD	Plankton cpm/ml \pm 2SD	Sediment cpm/ml \pm 2SD	Detritus cpm/ml \pm 2SD
T=0	soluble	69.4 (0.3)				
	particulate	1.6 (0.1)				
1 hr	soluble	60.8 (1.2)	12.3 (0.2)	11.8 (0.5)	12.5 (0.6)	10.9 (0.5)
	particulate	0.7 (0.1)	0.3 (0.1)	0.3 (0.1)	0.6 (0.1)	1.3 (0.1)
4 hrs	soluble	56.1 (0.4)	33.6 (0.9)	37.6 (0.4)	35.0 (0.9)	34.2 (0.9)
	particulate	1.1 (0.1)	0.7 (0.1)	0.7 (0.1)	0.7 (0.1)	0.6 (0.1)
10 hrs	soluble	49.4 (1.1)	41.9 (0.3)	46.1 (1.1)	41.5 (0.3)	48.1 (1.1)
	particulate	1.6 (0.2)	1.1 (0.1)	1.1 (0.1)	6.7 (0.1)	8.1 (0.4)
24 hrs	soluble	48.3 (1.1)	49.7 (1.6)	46.9 (1.1)	53.2 (1.6)	56.4 (1.2)
	particulate	1.2 (0.1)	1.4 (0.1)	Lost	8.6 (0.2)	6.1 (0.3)
5 days	soluble	47.0 (1.1)	48.8 (1.1)	48.3 (1.1)	50.1 (1.1)	53.0 (1.6)
	particulate	1.5 (0.2)	1.2 (0.1)	1.5 (0.2)	14.1 (0.7)	2.9 (0.2)
12 days	soluble	44.9 (0.4)	51.4 (1.1)	49.8 (0.7)	60.2 (1.7)	56.2 (1.2)
	particulate	1.0 (0.1)	0.8 (0.1)	0.7 (0.1)	1.2 (0.2)	1.9 (0.2)
20 days	soluble	50.2 (1.1)	50.2 (1.1)	48.7 (1.6)	50.6 (1.1)	51.6 (1.1)
	particulate	1.6 (0.2)	1.4 (0.2)	1.7 (0.2)	11.0 (0.3)	6.0 (0.3)

to reach equilibrium between the outside chamber and the inside of the dialysis sacs. After radionuclides have entered the dialysis sacs, comparison of the concentration of particulate radionuclides in the different chambers indicates the relative affinity of individual radionuclides for sediments, phytoplankton, or organic detritus.

The concentrations of soluble and particulate ^{241}Am are plotted in Figs. 15 and 16. The soluble ^{241}Am in the outer chamber shows a rapid decline during the first 4 hrs followed by a slower decline for the duration of the experiment. The particulate ^{241}Am in the outer chamber declines gradually during the first 5 days but remains relatively constant from day 5 until day 20. Although there are detectable concentrations of both soluble and particulate ^{241}Am in all four dialysis sacs after 10 hrs, equilibrium among the different chambers does not occur during the first 12 days. After 20 days, the control dialysis sac, sediment and detritus chamber all appear to be in equilibrium with the outside chamber. That is, all show comparable decreases in activity as shown in Fig. 15. The phytoplankton chamber, however, has a significantly higher concentration of soluble ^{241}Am . This suggests that a soluble, probably organic, compound of ^{241}Am is formed within the phytoplankton chamber. This compound is apparently too large to pass through the membrane and come into equilibrium with the rest of the system so the concentration of soluble ^{241}Am remains higher in the phytoplankton chamber than in other compartments. The source of an organic ligand to form this compound is not currently known. It may be a phytoplankton exometabolite or may result from the decomposition of some phytoplankton cells during the experiment.

A significant portion of ^{241}Am from the outside chamber is retained on the filter even though no particulates were introduced into this chamber. The formation of ^{241}Am radiocolloids has been noted previously for both dialysis and constant shaking experiments. The most likely explanation for this is the presence in the outer chamber of iron oxide solids to which the ^{241}Am adsorbs. It is interesting that in the outer chamber as much as 87% of the ^{241}Am can be retained on the filters. In the control dialysis sac, however, the concentration of particulate ^{241}Am never exceeds 50%. This difference probably results from the lower total concentration of ^{241}Am in the control dialysis sac.

All three particulates adsorb significant amounts of ^{241}Am with the sediments adsorbing the largest quantities and phytoplankton the least. The concentration of particulate ^{241}Am in the phytoplankton chamber appears to be constant from day 5 through 20 and probably represents an equilibrium between soluble and particulate species. Both the detritus and the sediment chambers show increased concentrations of particulates on day 20. Therefore, we cannot be certain that equilibrium has been reached in these chambers even after 20 days. In both the detritus and sediments sacs, greater than 90% of the total ^{241}Am occurs in the particulate phase. The transfer of ^{241}Am

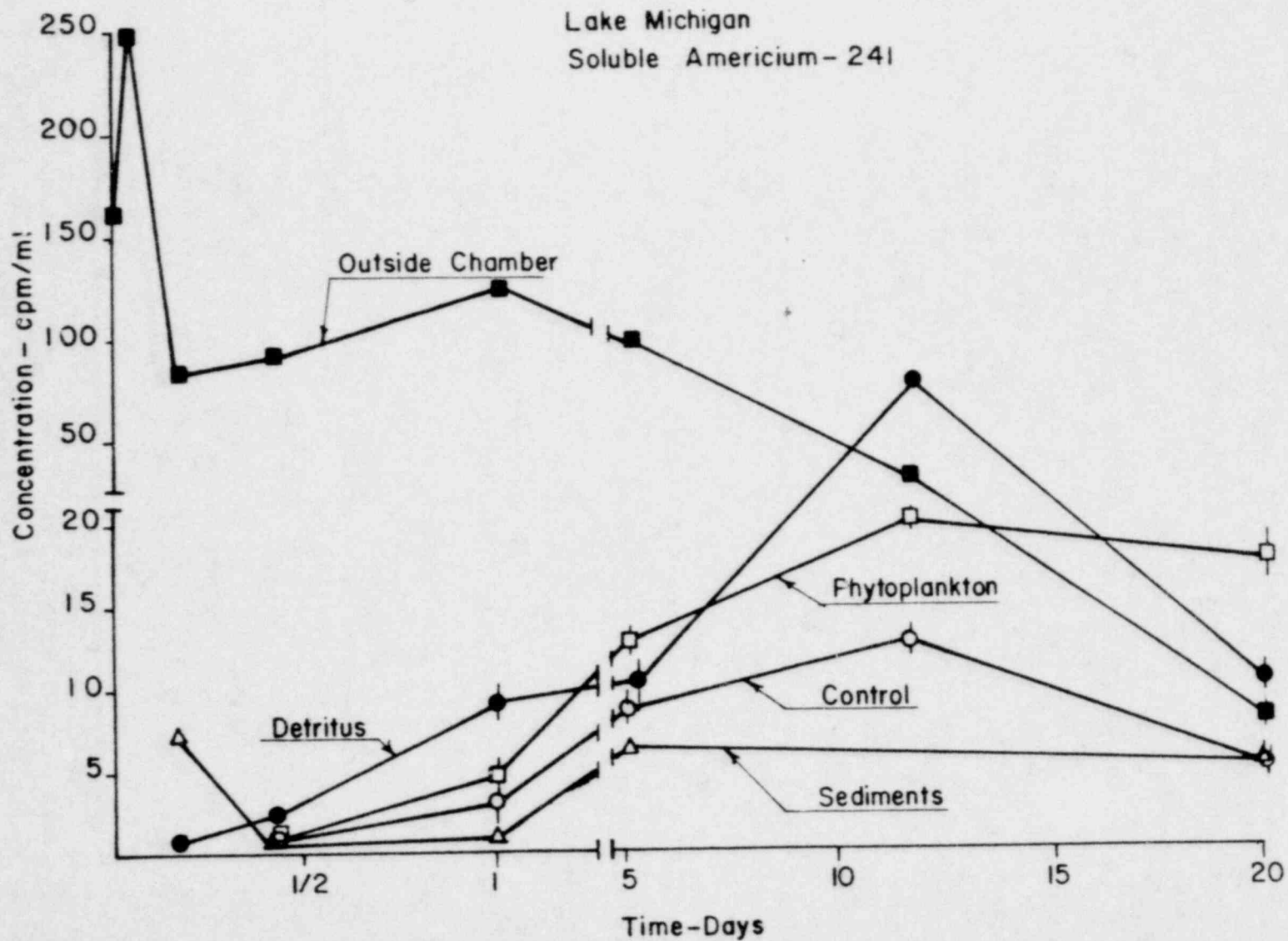


Figure 15. Relative concentrations (cpm/ml) of soluble ^{241}Am in each compartment as a function of time for a 20 day dialysis experiment. Sediments and water are from Lake Michigan. Phytoplankton is *Chlorella vulgaris*. Detritus is dried *Myriophyllum* sp. (a macrophyte) from Lake Washington.

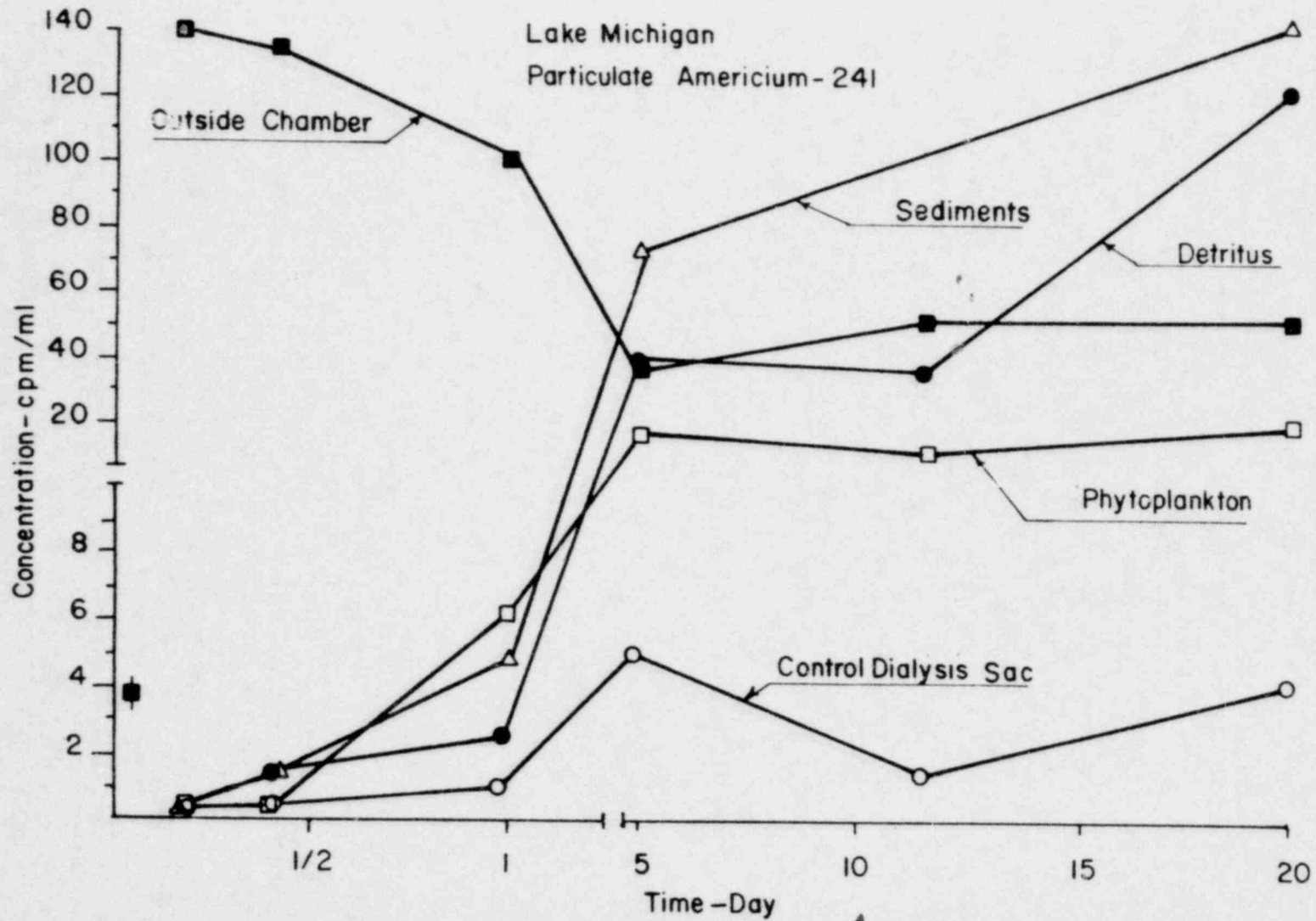


Figure 16. Relative concentrations (cpm/ml) of particulate ^{241}Am in each compartment as a function of time for a 20 day dialysis experiment. See Figure 15 for a description of components.

across the dialysis membranes is much slower in the Lake Michigan experiment than in previous experiments with seawater. Therefore, the time required to reach equilibrium is much longer and may not occur within 20 days in our dialysis apparatus.

The distributions of soluble and particulate ^{106}Ru are plotted in Figs. 17 and 18. Like ^{241}Am there is a rapid initial decline in the outer chamber, followed by a continuing gradual decline throughout the experiment. Each of the dialysis sacs behaves similarly with regard to the concentration of soluble ^{106}Ru . There are detectable concentrations of soluble ^{106}Ru in all the dialysis sacs after 4 hrs and a continual increase in each sac during the first 24 hrs. However, equilibrium among the dialysis sacs and the outside chamber is not reached during the first 12 days.

The highest concentration of particulate ^{106}Ru is found in the outside chamber throughout the experiment. It is most likely that this ^{106}Ru is adsorbed to iron oxides or other colloids that form in the outside chamber, while the spike is equilibrating. The control sac has very low concentrations of particulate ^{106}Ru although more than 20% of the total ^{106}Ru may occur in the particulate phase. Phytoplankton, detritus, and sediments all accumulate ^{106}Ru with phytoplankton accumulating significantly less than either detritus or sediment. It appears from Fig. 18 that sediments adsorb more ^{106}Ru than detritus. However, the differences between sediments and detritus do not exceed two standard deviations of propagated counting error. For all three types of particulates the majority of the adsorption occurs between days 5 and 20. In fact, there appears to be a linear increase with time during this period. It is not clear why adsorption occurs so slowly in this system. Furthermore, it is not certain that the adsorption sites have been saturated after 20 days.

The adsorption patterns for both ^{241}Am and ^{106}Ru in the Lake Michigan system are quite unexpected. They certainly require further evaluation and perhaps additional experimental confirmation.

Cesium-137 occurs primarily as an ionic species and behaves very differently from ^{106}Ru or ^{241}Am which exist in several different physico-chemical states including radiocolloids. Figure 19 shows the concentration of soluble ^{137}Cs in each of the chambers throughout the experiment. There is a rapid transfer of ^{137}Cs across the dialysis membranes during the 1st hr and after 4 hrs all the dialysis sacs are in equilibrium with the outside chamber. While there was a slight decrease in the concentration of soluble ^{137}Cs in all chambers, equilibrium among the different chambers was maintained.

The concentration of particulate ^{137}Cs is shown in Fig. 20. The major adsorption of ^{137}Cs occurs on sediments within the first day. The concentration of ^{137}Cs on sediments increases between days 1 and 5 and then declines slightly by day 20. Detritus accumulates ^{137}Cs although

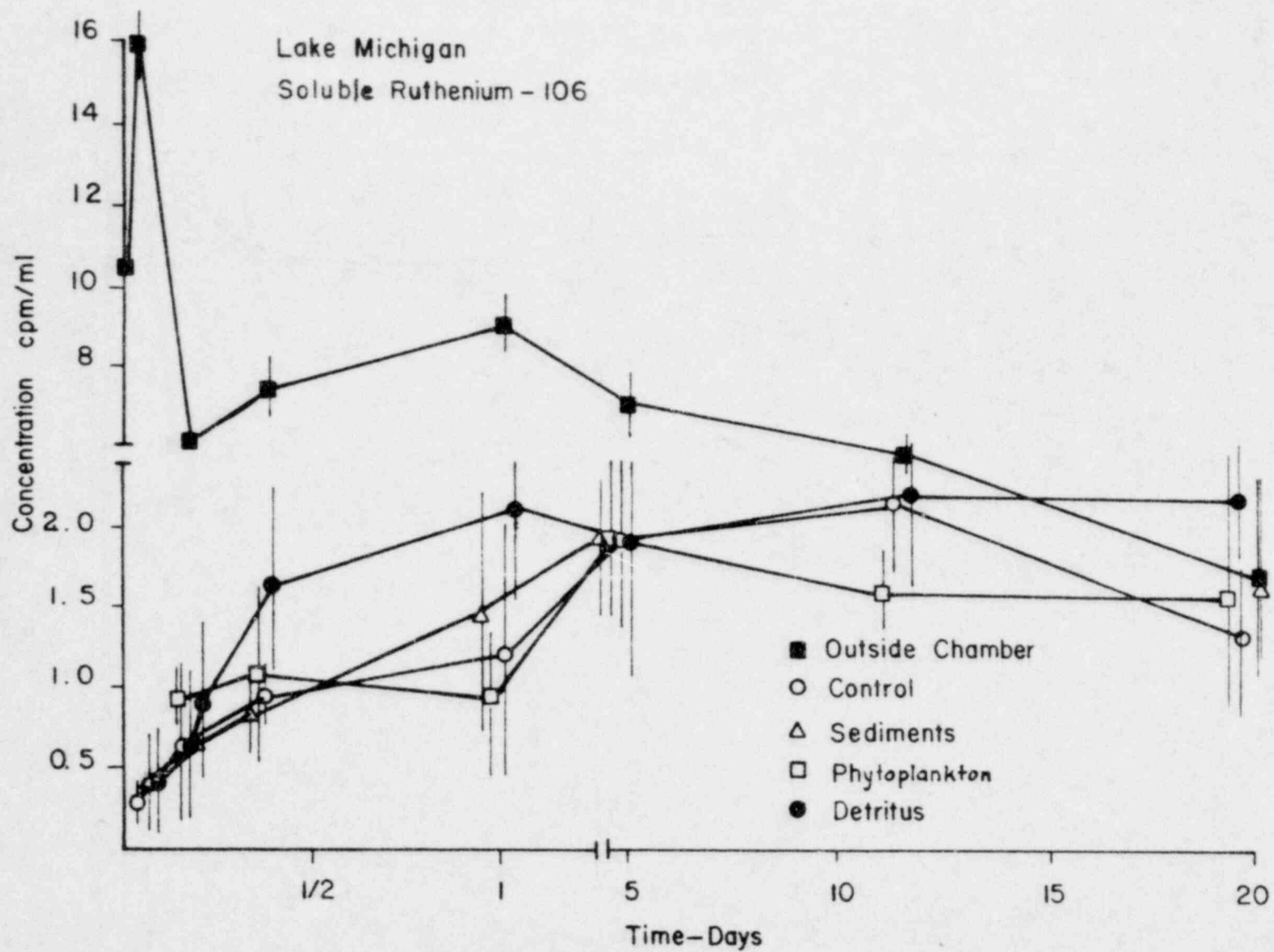


Figure 17. Relative concentrations (cpm/ml) of soluble ^{106}Ru in each compartment as a function of time for a 20 day dialysis experiment. See Figure 15 for a description of components.

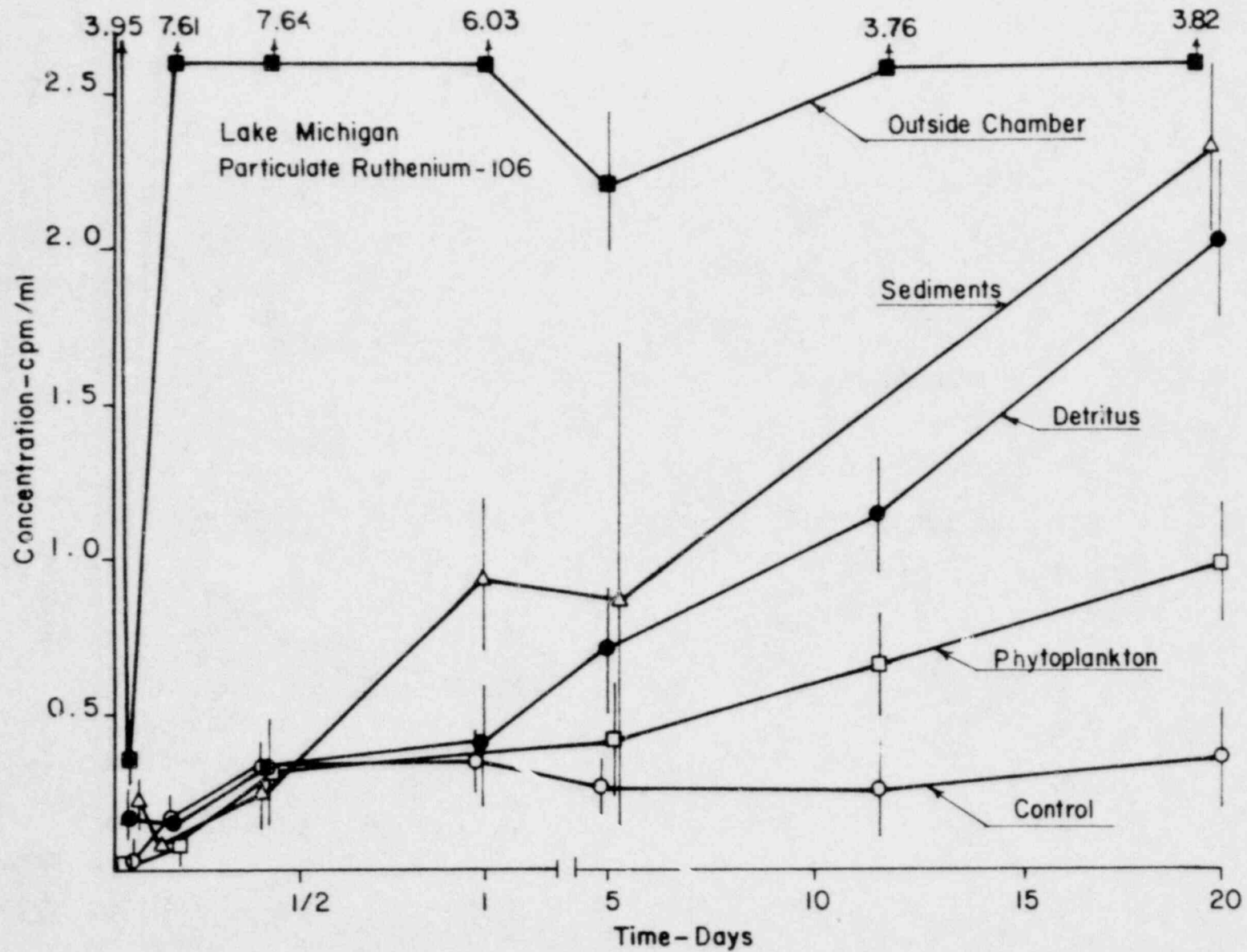


Table 13. Relative concentrations (cpm/ml) of particulate ^{106}Ru in each compartment as a function of time for a 20 day dialysis experiment. See Figure 15 for a description of components.

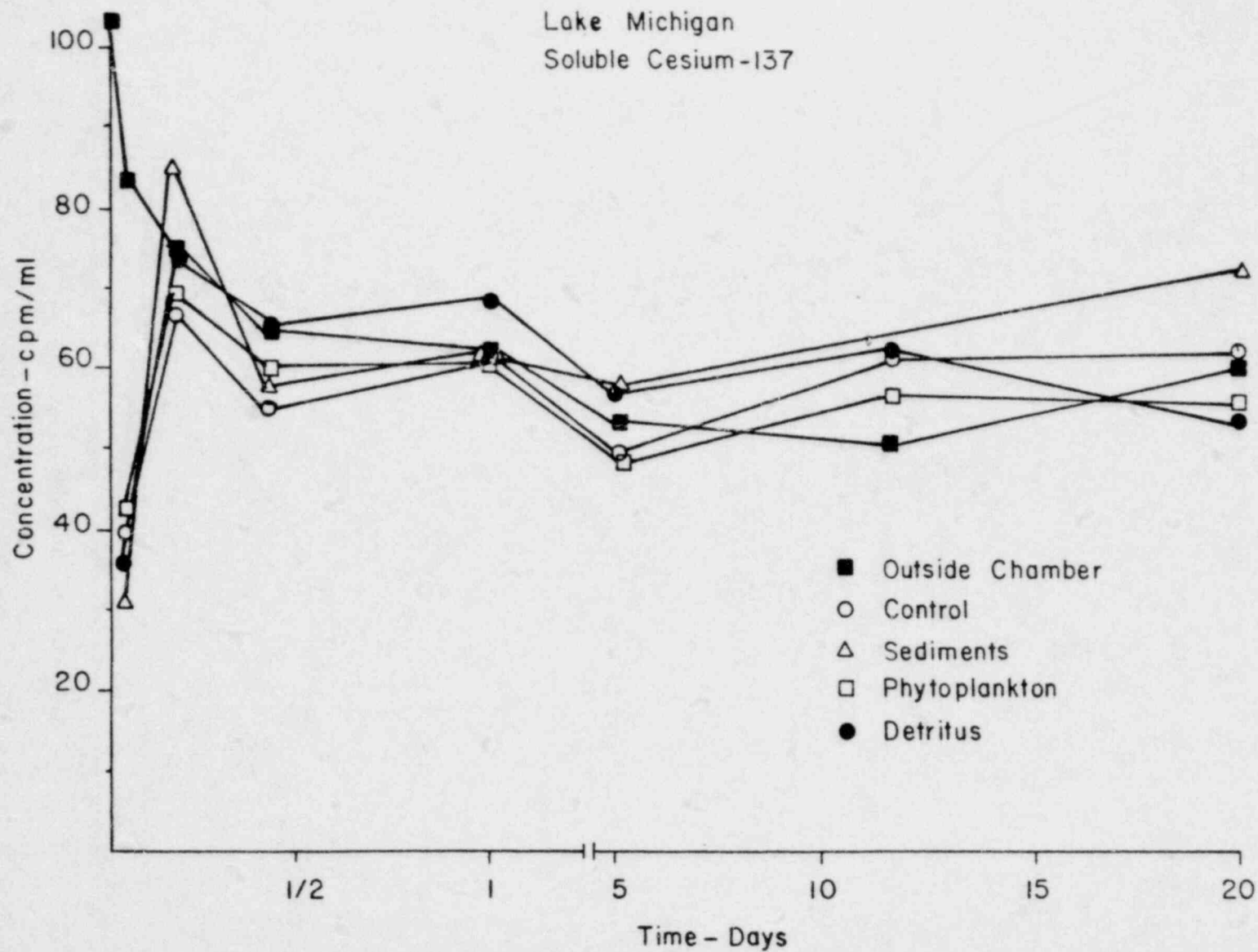


Figure 19. Relative concentrations (cpm/ml) of soluble ^{137}Cs in each compartment as a function of time for a 20 day dialysis experiment. See Figure 15 for a description of components.

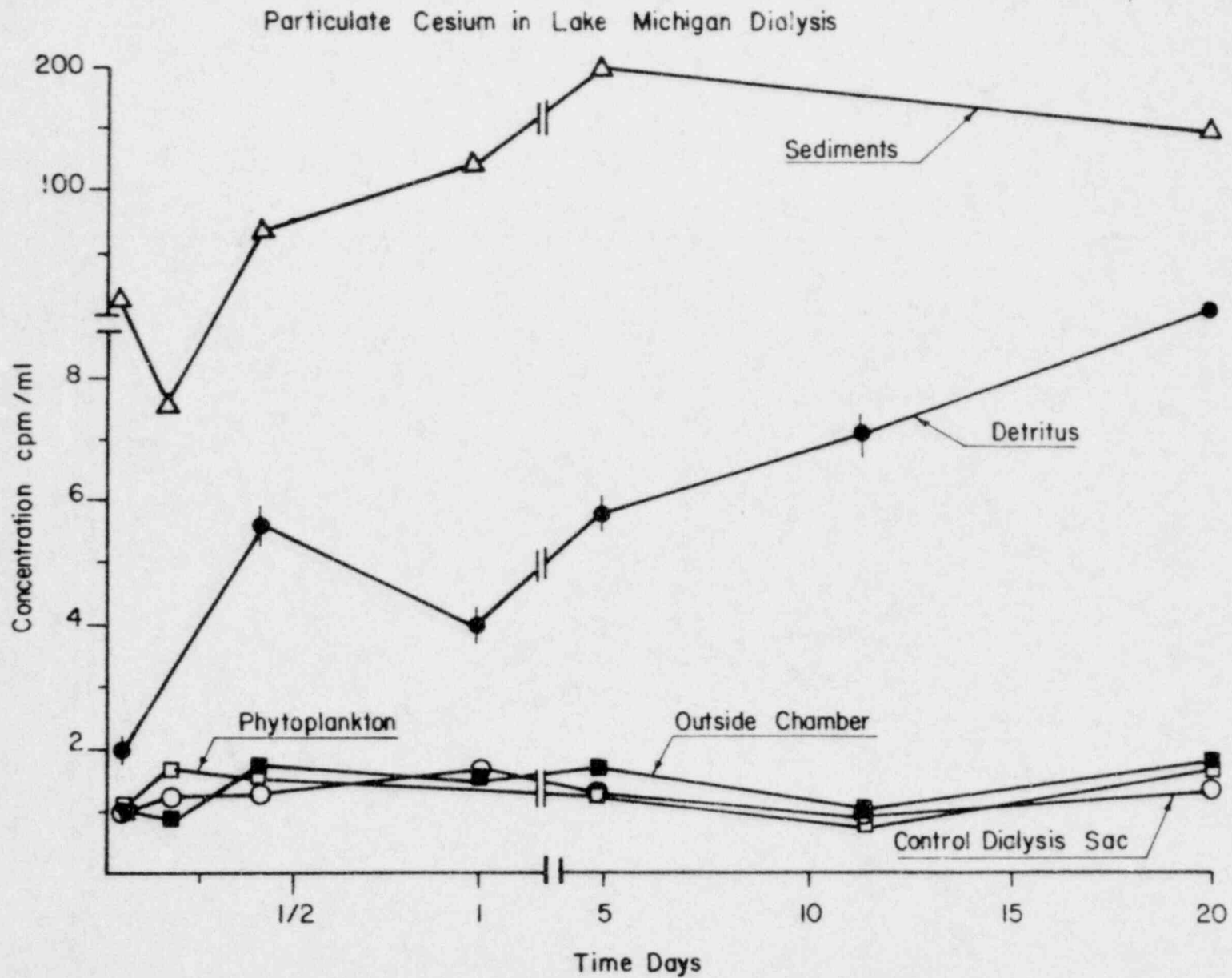


Figure 20. Relative concentrations (cpm/ml) of particulate ^{137}Cs in each compartment as a function of time for a 20 day dialysis experiment. See Figure 15 for a description of components.

at much lower concentrations than sediments. On detritus the major adsorption occurs during the first 10 hrs although there is a linear increase in concentration between days 5 and 20. Although some ^{137}Cs is retained on the filter papers, there is very little particulate ^{137}Cs in either the outside chamber or the control dialysis sac. Also, the phytoplankton do not accumulate any significant quantity of ^{137}Cs .

The final radionuclide included in this experiment, ^{60}Co , behaves similarly to ^{137}Cs . The concentrations of soluble ^{60}Co are shown in Fig. 21. There is a rapid decrease in the concentration of ^{60}Co in the outside compartment and a concomitant increase in each of the dialysis sacs. Equilibrium among soluble species of ^{60}Co in all the compartments is reached within the first day and maintained rather well throughout the remainder of the experiment.

The concentrations of particulate ^{60}Co are shown in Fig. 22. Like ^{137}Cs , there is very little, if any, particulate ^{60}Co in the outside chamber or control dialysis sac and there is no apparent accumulation in phytoplankton. Both detritus and sediments do accumulate significant amounts of ^{60}Co with the concentration in sediments being about three times as great as in detritus. The uptake of ^{60}Co by sediments and detritus is quite rapid during the first 10 hrs with relatively minor fluctuations during the remainder of the experiment.

The second dialysis experiment was set up to investigate the behavior of ^{85}Sr and ^{237}Pu during a 15-day dialysis experiment. Samples were collected from each chamber after 3, 10, and 24 hrs, 8 and 15 days. Results collected from this experiment are presented in Tables 22 and 23.

Strontium-85 (Table 22) occurs almost entirely in the soluble phase. The concentration of particulate ^{85}Sr never exceeds 3% of the total ^{85}Sr concentration. In most cases, the concentration listed as particulate ^{85}Sr probably corresponds to ^{85}Sr adsorbed on the filter rather than the actual particulate phase. The concentration of soluble ^{85}Sr is plotted in Fig. 23. Equilibrium among all the chambers is reached within 10 hrs and maintained for the duration of the experiment. This rapid approach to equilibrium is similar to the results obtained for ^{60}Co and ^{137}Cs in the previous experiment.

The distribution of soluble ^{237}Pu is shown in Fig. 24. There is a very rapid decrease in the concentration of soluble ^{237}Pu in the outside chamber during the initial 3 hrs, followed by a more gradual decline for the remainder of the experiment. There appears to be differences among the dialysis sacs regarding both the rate of diffusion across the membrane and the total concentration of soluble ^{237}Pu in the different chambers. During the first day the concentration of soluble ^{237}Pu is lower in the sediment sac than in

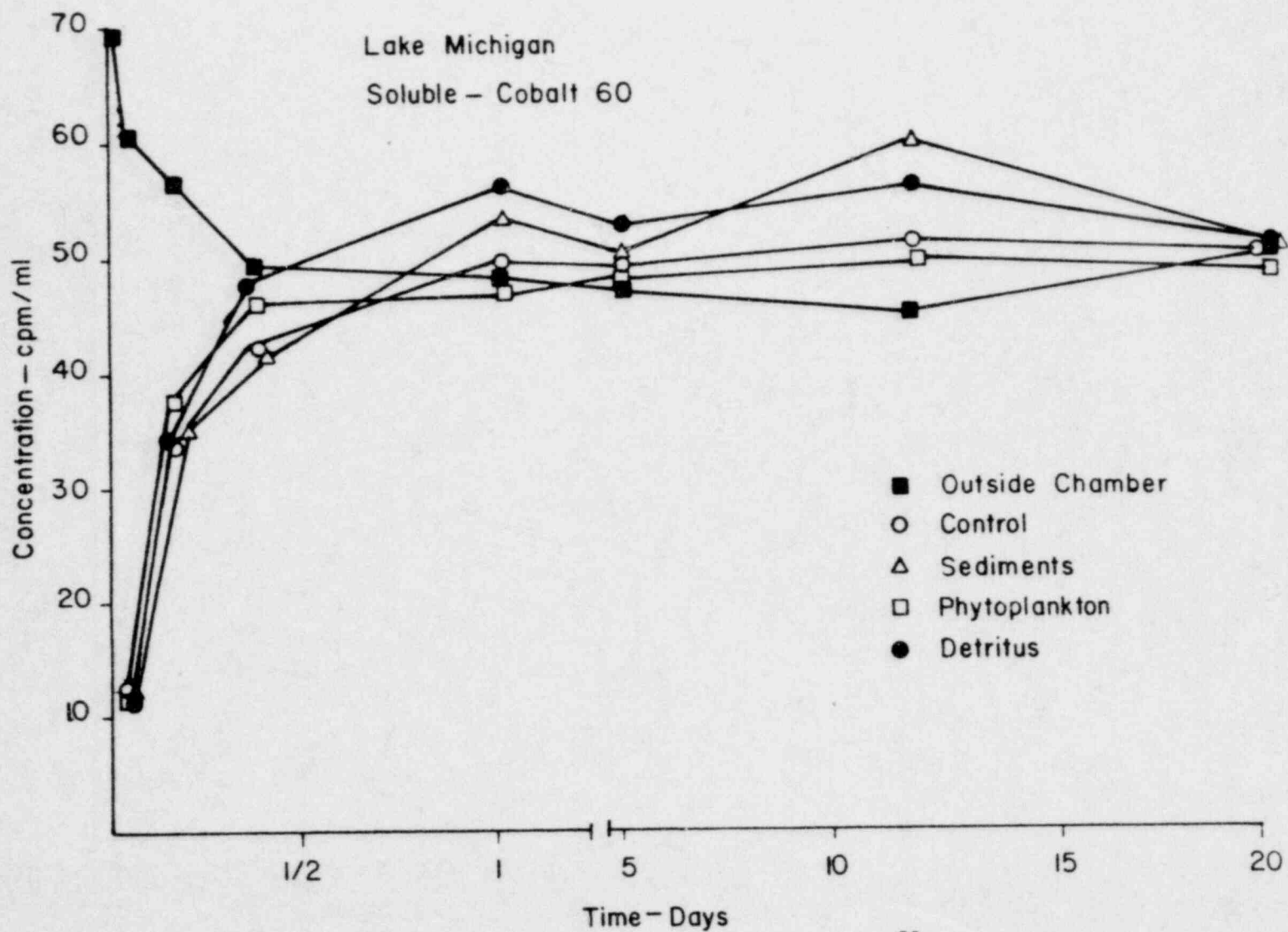


Figure 21. Relative concentrations (cpm/ml) of soluble ^{60}Co in each compartment as a function of time for a 20 day dialysis experiment. See Figure 15 for a description of components.

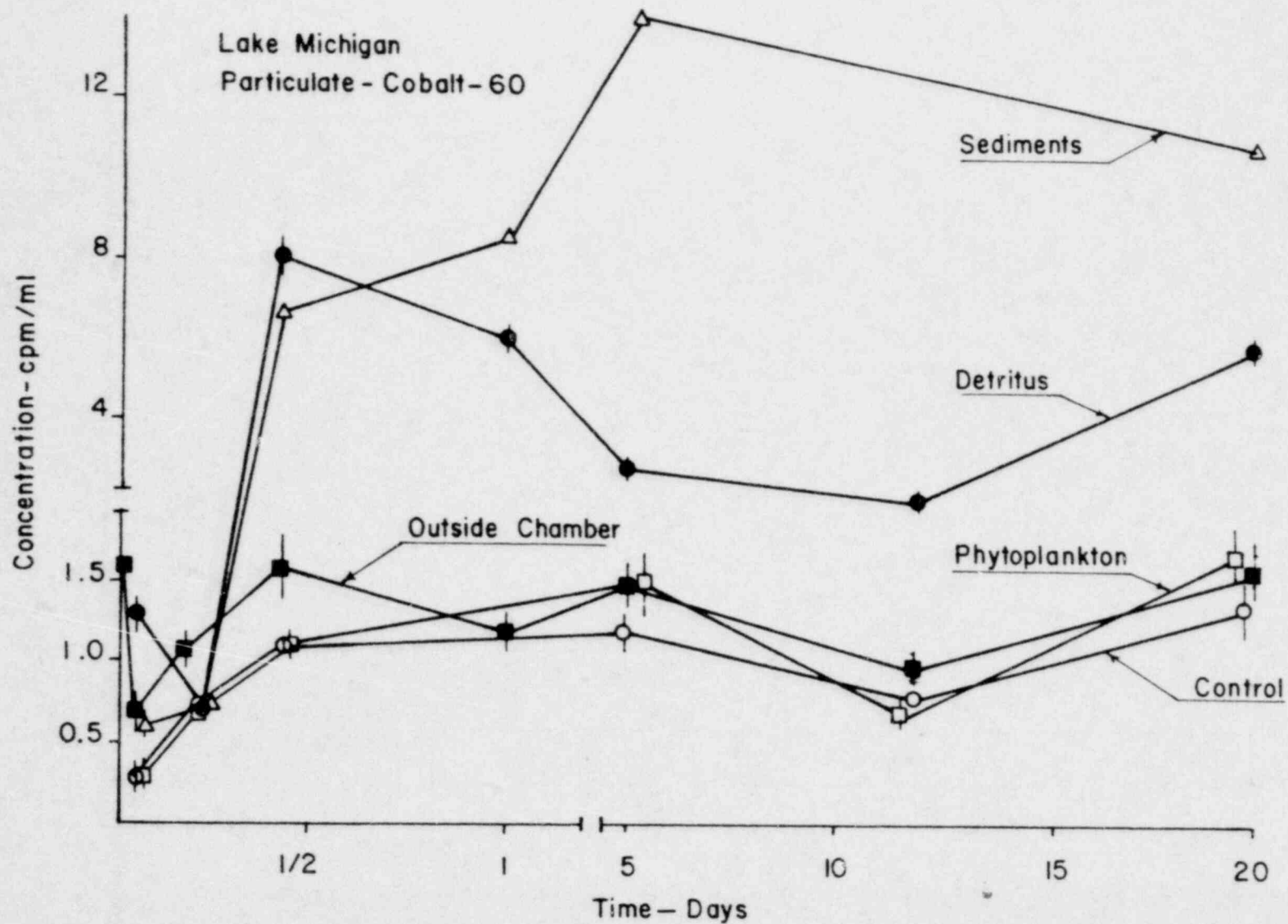


Figure 22. Relative concentrations (cpm/ml) of soluble ^{60}Co in each compartment as a function of time in a 20 day dialysis experiment. See Figure 15 for a description of components.

Table 22. Relative concentrations of ^{85}Sr during a 15 day dialysis experiment.

Time		Spiked Chamber cpm/ml \pm 2SD	Dialysis Chambers			
			Blank cpm/ml \pm 2SD	Plankton cpm/ml \pm 2SD	Sediment cpm/ml \pm 2SD	Detritus cpm/ml \pm 2SD
T=0	soluble	284.9 (4.4)				
	particulate	2.9 (0.3)				
3 hrs	soluble	219.0 (3.9)	124.4 (1.7)	Lost	143.2 (3.2)	140.6 (2.8)
	particulate	2.2 (0.3)	1.3 (0.3)	0.4 (0.1)	3.7 (0.3)	1.9 (0.3)
10 hrs	soluble	213.5 (3.8)	203.1 (3.2)	202.3 (2.2)	208.2 (3.8)	202.9 (3.7)
	particulate	2.9 (0.3)	2.3 (0.3)	2.6 (0.3)	5.5 (0.5)	5.8 (0.4)
24 hrs	soluble	195.7 (3.1)	178.1 (2.5)	411.0 (4.3)	204.0 (3.8)	234.7 (3.3)
	particulate	0.6 (0.1)	2.3 (0.3)	0.4 (0.2)	3.4 (0.3)	5.0 (0.3)
8 days	soluble	198.8 (2.2)	195.9 (3.2)	201.2 (5.5)	206.7 (3.2)	204.5 (3.1)
	particulate	0.7 (0.1)	0.4 (0.2)	1.7 (0.3)	0.9 (0.2)	4.1 (0.1)
15 days	soluble	205.4 (3.3)	198.5 (5.5)	204.9 (5.6)	202.8 (2.3)	219.9 (3.5)
	particulate	1.0 (0.3)	0.9 (0.2)	1.4 (0.3)	1.0 (0.3)	0.4 (0.1)

Table 23. Relative concentrations of ^{237}Pu during a 15 day dialysis experiment.

Time	Spiked Chamber cpm/ml \pm 2SD	Dialysis Chambers				
		Blank cpm/ml \pm 2SD	Plankton cpm/ml \pm 2SD	Sediment cpm/ml \pm 2SD	Detritus cpm/ml \pm 2SD	
T=0	soluble particulate	92.2 (3.8) 28.5 (0.7)				
3 hrs	soluble particulate	27.2 (2.9) 1.0 (0.3)	10.8 (1.2) 0.6 (0.2)	Lost 0.5 (0.1)	6.8 (2.2) 7.3 (0.4)	11.3 (2.0) 12.3 (0.5)
10 hrs	soluble particulate	23.6 (2.8) 2.1 (0.3)	17.9 (2.4) 0.7 (0.3)	18.3 (1.6) 1.4 (0.3)	9.8 (2.6) 19.2 (0.8)	15.8 (2.7) 20.2 (0.6)
24 hrs	soluble particulate	16.7 (2.2) 0.7 (0.2)	16.8 (1.8) 0.5 (0.2)	38.1 (3.2) 0.7 (0.2)	9.6 (2.6) 31.1 (0.7)	21.9 (2.4) 28.6 (0.7)
8 days	soluble particulate	13.8 (1.5) 1.4 (0.1)	12.1 (2.1) 0.5 (0.2)	21.8 (3.9) 0.7 (0.3)	16.0 (2.3) 5.1 (0.3)	18.5 (2.3) 21.2 (0.2)
15 days	soluble particulate	7.1 (2.3) 1.5 (0.3)	6.7 (3.9) 0.6 (0.2)	12.7 (4.1) 0.7 (0.3)	4.7 (1.6) 3.6 (0.3)	11.8 (2.5) 2.2 (0.2)

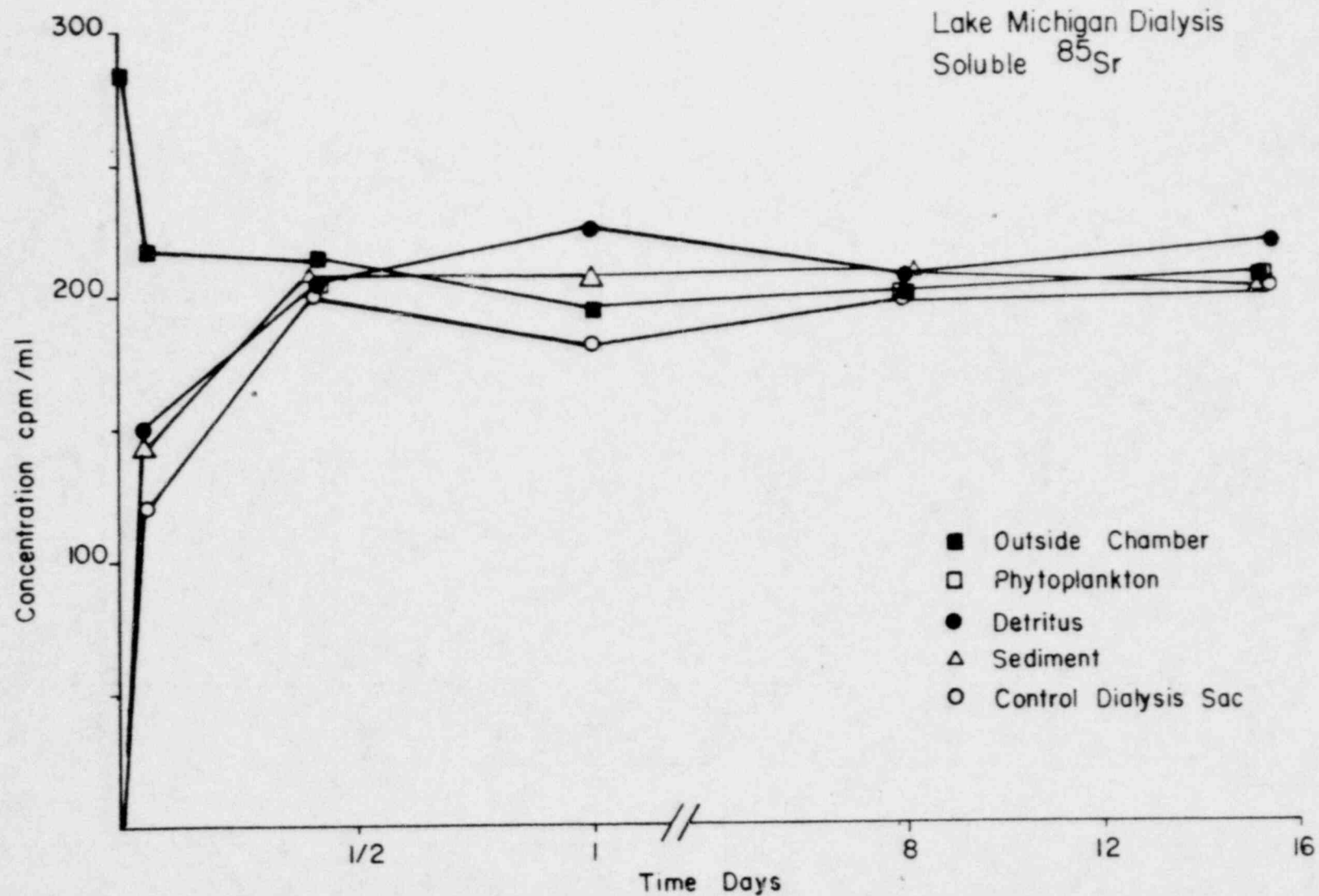


Figure 23. Relative concentrations (cpm/ml) of soluble ^{85}Sr in each compartment as a function of time in a 15 day dialysis experiment. See Figure 15 for a description of components.

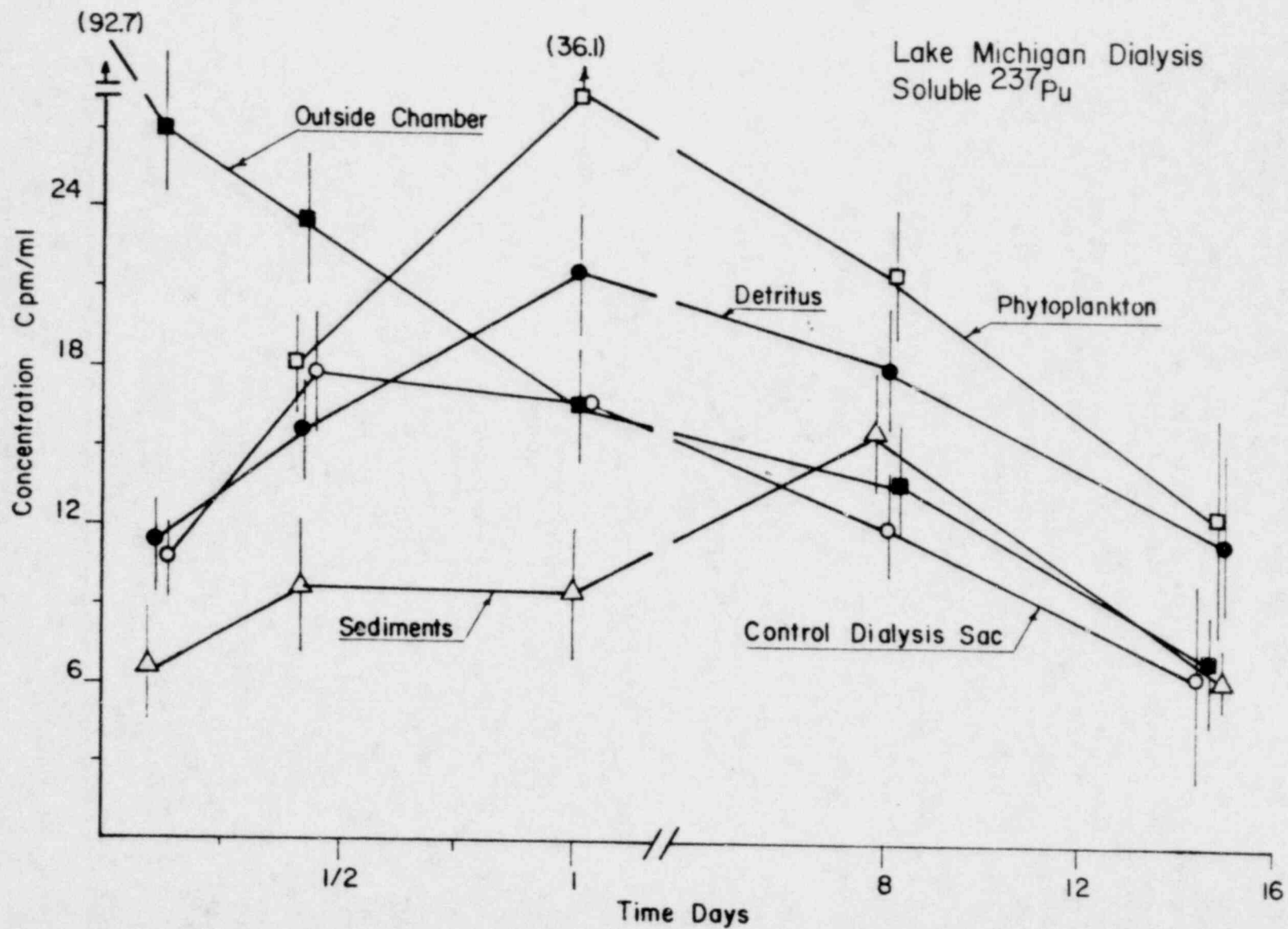


Figure 24. Relative concentrations (cpm/ml) of soluble ^{237}Pu in each compartment as a function of time in a 15 day dialysis experiment. See Figure 15 for a description of components.

crosses the dialysis membrane is rapidly adsorbed on the sediment and does not occur as soluble ^{237}Pu for very long. After 8 days the sediment sac is in equilibrium with both the control dialysis sac and the outside chamber. During most of the experiment, the concentration of soluble ^{237}Pu is higher in the phytoplankton and detritus sacs than in the other chambers. This suggests that organic molecules in these chambers may be complexing some ^{237}Pu and increasing the dissolved concentration. This is particularly interesting for phytoplankton since there is no accumulation of ^{237}Pu by phytoplankton (Fig. 25) during this experiment. The concentration of soluble ^{237}Pu in the phytoplankton dialysis sac after 1 day is extraordinarily high and cannot be explained at this time.

The concentrations of particulate ^{237}Pu are shown in Fig. 25. During the first three hours the concentration in the outside chamber drops more than an order of magnitude and remains low for the duration of the experiment. The concentrations of particulate ^{237}Pu in the phytoplankton and control dialysis sacs are comparable to the outside chamber at all sampling times. Both the sediments and detritus accumulate significant quantities of ^{237}Pu during the first day but also have significantly lower amounts on days 8 and 15. This is very unusual behavior and probably represents an experimental artifact which we would not expect in natural systems.

Comparisons Between Sorption and Dialysis K_d Values

In the Lake Michigan sediment-water system, the equilibrium K_d values for adsorption (constant shaking method) are compared with the K_d values obtained from the dialysis experiments in Table 24. Both sediments and detritus show greater K_d values in the constant shaking experiments than in the dialysis experiments for ^{237}Pu , ^{241}Am , ^{106}Ru , and ^{85}Sr ; a majority of the K_d measurements in the dialysis experiments is at least four times less than the adsorption K_d values. For ^{137}Cs , however, the reverse is observed: the equilibrium K_d value for adsorption is at least 20 times less than the dialysis K_d values. Dialysis membranes restrict the passage of molecules above certain molecular size ranges. Therefore, the differences in K_d values between constant shaking and dialysis experiments imply that chemical speciation of the radionuclides in the soluble phases can affect significantly particle-water distributions of the nuclides. The results suggest that interactions between radionuclides and sediments in a complex experimental system which includes detrital matter and phytoplankton are influenced by additional factors which can either increase or decrease the K_d values obtained for pure sediment-water systems.

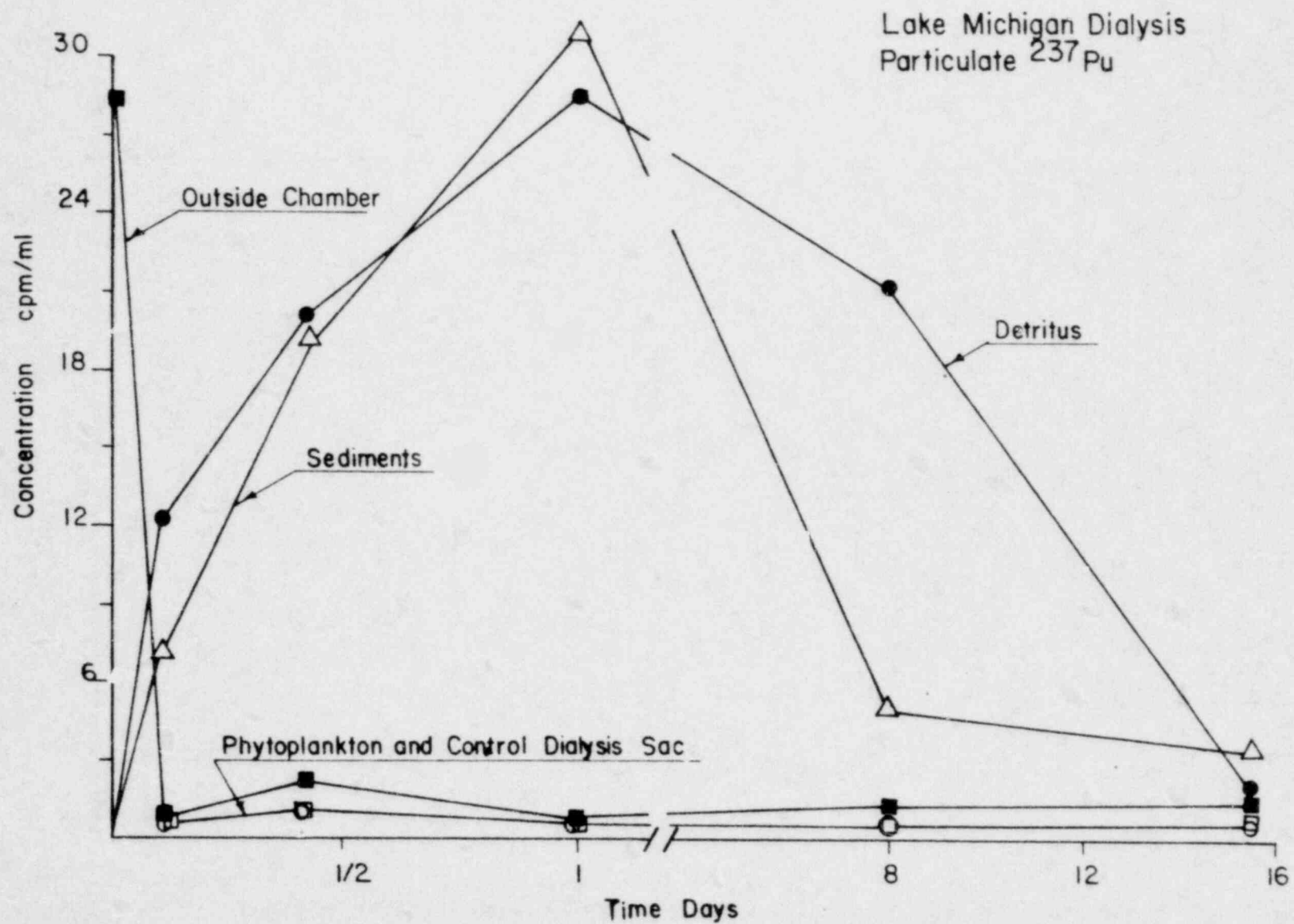


Figure 25. Relative concentrations (cpm/ml) of particulate ^{237}Pu in each compartment as a function of time in a 15 day dialysis experiment. See Figure 15 for a description of components.

Table 24. Comparisons of K_d values for adsorption by the constant shaking method with K_d values for adsorption by the dialysis method in Lake Michigan sediment-water system

Radionuclide	Days	Sediment		Detritus	
		Dialysis K_d values (ml/g)	Adsorption K_d values (ml/g)	Dialysis K_d values (ml/g)	Adsorption K_d values (ml/g)
^{237}Pu	1	$(1.63 \pm 0.44) \times 10^4$	$(1.41 \pm 0.18) \times 10^5$	$(1.31 \pm 0.15) \times 10^4$	$(2.59 \pm 0.94) \times 10^4$
	8	$(1.59 \pm 0.25) \times 10^3$		$(1.15 \pm 0.14) \times 10^4$	
	15	$(3.83 \pm 0.13) \times 10^3$		$(1.86 \pm 0.43) \times 10^3$	
^{241}Am	1	$> 1.88 \times 10^4$	$(5.48 \pm 3.73) \times 10^5$	$(3.03 \pm 0.59) \times 10^3$	72
	5	$(5.86 \pm 0.94) \times 10^4$		$(4.03 \pm 0.55) \times 10^4$	
	20	$(1.47 \pm 0.27) \times 10^5$		$(1.23 \pm 0.12) \times 10^5$	
^{106}Ru	1	$(3.26 \pm 2.05) \times 10^3$	$(5.23 \pm 1.68) \times 10^4$	$(1.94 \pm 1.10) \times 10^3$	
	5	$(2.18 \pm 1.99) \times 10^3$		$(3.76 \pm 2.02) \times 10^3$	
	20	$(7.16 \pm 0.27) \times 10^3$		$(9.23 \pm 2.73) \times 10^3$	
^{137}Cs	1	$(1.03 \pm 0.03) \times 10^4$	$(5.09 \pm 0.31) \times 10^2$	$(5.80 \pm 0.45) \times 10^2$	
	5	$(1.78 \pm 0.04) \times 10^4$		$(1.01 \pm 0.06) \times 10^3$	
	20	$(1.04 \pm 0.02) \times 10^4$		$(2.06 \pm 0.09) \times 10^3$	
^{85}Sr	1	83.3 ± 7.5	82.2 ± 7.0	$(2.13 \pm 0.13) \times 10^2$	75.2 ± 4.1
	8	21.8 ± 4.9		$(2.00 \pm 0.06) \times 10^2$	
	15	24.7 ± 7.4		18.2 ± 4.6	

4.0 CONCLUSIONS AND RECOMMENDATIONS

1. The order of increasing K_d values for adsorption is $^{85}\text{Sr} < ^{137}\text{Cs} < (^{106}\text{Ru} \text{ or } ^{237}\text{Pu}) < ^{241}\text{Am}$. The values range from less than 10^2 for ^{85}Sr to greater than 10^5 for ^{241}Am . The values for ^{85}Sr and ^{137}Cs are greater in freshwater systems than in the marine sediment-water systems. For ^{106}Ru , ^{237}Pu , or ^{241}Am , both the highest and lowest K_d values are found in sediment-freshwater systems.
2. The desorption K_d values range from 3 to 38 times greater than the adsorption K_d values for ^{137}Cs , ^{85}Sr , ^{106}Ru , ^{237}Pu , ^{241}Am in the freshwater-sediment and marine water-sediment systems. The lowest desorption K_d value is found for ^{85}Sr and the highest is found for ^{241}Am ; the desorption K_d values increase with the same order as for the adsorption K_d values.
3. The pH of specific sediment-water systems affects the K_d values for some radionuclides. For the sediment-water system from Clinch River, the K_d value for ^{106}Ru increased by a factor of 5 between pH 4 and 6. There appears to be no effect of pH on K_d values for ^{137}Cs for pH < 9. For ^{60}Co , the K_d value increased by two orders of magnitude between pH 6.0 and 7.5.
4. The effect of sediment concentration on the K_d values has been measured in the range of 16 to 340 mg/l for the Lake Michigan and Cattaraugus Creek sediment-water systems. Results show that large increases in the K_d values occur as the concentration of sediment decreases.
5. The effect of organic ligands on the K_d values has been measured using five organic compounds and two freshwater-sediment systems. Equilibrium apparently was reached rapidly (4 hrs) using EDTA as a test compound. Significant increases and/or decreases in K_d values of ^{241}Am , ^{60}Co , and ^{106}Ru relative to controls were found with several of the organic compounds. No differences in K_d values were found for the ^{137}Cs . Additional work is required to be able to further evaluate these findings.

These preliminary studies have utilized specific organic compounds which may not be representative of the types and complexities of organic molecules found in natural systems. Therefore, it would be valuable to determine sediment-water K_d values in the presence of natural organic compound assemblages (e.g., humic extracts from natural soils).

6. Different K_d values for ^{241}Am and ^{237}Pu between sterile and non-sterile systems have been found, indicating a possible microbial effect. It is unclear, however, whether these differences were due to microbes (e.g., bacteria) or chemical alterations produced

in the sediments by the sterilization technique. Hence, more definitive studies should be performed to define the potential of bacteria to affect K_d values. These experiments could include not only milder sterilization techniques but also additions of specific bacteria.

7. Although it is found in all our experiments that radionuclides are removed significantly by sediments, the mechanisms involved in this removal are not readily discernible from distribution coefficient studies. A major limitation in using the K_d data to elucidate such mechanisms is that natural sediment and water samples are very complex systems which are difficult to define. Some additional information on the mechanism responsible for the interactions is needed so that a generalized model can be prepared. It is therefore recommended that comparisons be made on the distribution of radionuclides between pure solids and water. Pure iron oxides, goethite, and some clay minerals which are present in natural systems and which have been well characterized should be used.

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16. ABSTRACT (200 words or less) Sediments potentially serve as important sinks and sources for radionuclides released into aquatic systems. In this regard, we have collected data over the past year to evaluate mechanisms affecting particle-water distributions of selected radionuclides (^{57}Co , ^{60}Co , ^{85}Sr , ^{106}Ru , ^{137}Cs , ^{237}Pu , ^{241}Am and ^{244}Cm). Samples used in these studies were obtained from several natural freshwater, estuarine and marine locations around the continental United States. Particle-water distribution coefficients (K_d) were found to vary from 10^2 to 10^6 depending on the specific radionuclide, the type of particle(s), and the overall physical and chemical properties of a given test system. In general, K_d values increased as follows: $^{85}\text{Sr} < ^{137}\text{Cs} < ^{106}\text{Ru}$ or $^{237}\text{Pu} < ^{241}\text{Am}$. However, both the absolute values of these K_d 's and their magnitudes relative to other radionuclides were found to be dramatically and nonuniformly affected by certain variables. The variables tested and discussed in this report include: the quantity and chemical composition of the particles, the degree of reversibility of radionuclide-sediment interactions, the pH of the water, the abundance and composition of organic ligands and the chemical speciation (i.e., molecular weight size associations) of the radionuclides. This data will contribute to the formulation of predictive models to describe distributions and eventual fates of radionuclides in aquatic environments.				10. PROJECT/TASK/WORK UNIT NO.	
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