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# Distribution Coefficients for Radionuclides in Aquatic Environments

Adsorption and Desorption Studies of  $^{85}\text{Sr}$

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- 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. 38 pp.
- Distribution Coefficients for Radionuclides in Aquatic Environments. II. Studies in Marine and Freshwater Sediments Systems Including the Radionuclides  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$ , and  $^{241}\text{Am}$ . Annual Report: August 1977 - July 1978. W. R. Schell, T. H. Sibley, A. Nevissi, and A. Sanchez. NUREG/CR-0802. 70 pp.
- Distribution Coefficients for Radionuclides in Aquatic Environments. III. Adsorption and Desorption Studies of  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ ,  $^{85}\text{Sr}$ , and  $^{237}\text{Pu}$  in Marine and Freshwater Systems. Annual Report: August 1978 - July 1979. W. R. Schell, A. L. Sanchez, T. H. Sibley and J. R. Clayton, Jr. NUREG/CR-0803. 76 pp.
- Distribution Coefficients for Radionuclides in Aquatic Environments. Methodology. W. R. Schell, A. Sanchez, T. H. Sibley, A. E. Nevissi, J. R. Clayton, Jr., and E. A. Wurtz. Laboratory of Radiation Ecology, Univ. Washington Topical Report NUREG/CR-1852, Vol. 1. 27 pp.
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- Distribution Coefficients for Radionuclides in Aquatic Environments. Adsorption and Desorption Studies of  $^{106}\text{Ru}$ . T. H. Sibley, A. L. Sanchez, and W. R. Schell. Laboratory of Radiation Ecology, Univ. Washington Topical Report NUREG/CR-1852, Vol. 3.

## ABSTRACT

Measurements have been made of the distribution coefficients for  $^{85}\text{Sr}$  in different sediment-water systems. Strontium has the lowest distribution coefficient of the radionuclides normally expected to be released downstream from nuclear power reactors. Estuarine and marine environments limit the sorption of  $^{85}\text{Sr}$  onto sediments. However,  $^{85}\text{Sr}$  does adsorb onto organic detrital matter in marine waters. There was no apparent pH effect on the distribution coefficient values for  $^{85}\text{Sr}$  measured between pH 4 and pH 10 for marine and estuarine waters. Higher values were found in fresh waters above the estuary in the Hudson River and these values increased gradually from pH 4 to pH 10. At the Cattaraugus Creek, the distribution coefficients increased gradually with pH from pH 4 to pH 8 and then a sudden increase in  $K_d$  values was measured at pH 9.2 indicating possible precipitation in this sediment-water system. A large increase in distribution coefficients was found with decrease in sediment concentration.

## TABLE OF CONTENTS

	Page
1.0 INTRODUCTION . . . . .	1
1.1 Background . . . . .	2
1.1.1 Previous Information . . . . .	2
1.1.2 Present Studies . . . . .	3
1.2 Objectives . . . . .	5
2.0 MATERIALS AND METHODS . . . . .	6
2.1 Sources and Types of Samples . . . . .	6
2.2 Constant Shaking Experiments . . . . .	11
2.3 Dialysis Experiments . . . . .	13
2.4 Measurement of Radioactivity . . . . .	15
3.0 RESULTS AND DISCUSSION . . . . .	16
3.1 $K_d$ Values by Constant Shaking Experiments . . . . .	16
3.2 Effect of pH on $K_d$ Values . . . . .	19
3.3 Effect of Sediment Concentration . . . . .	25
3.4 Dialysis Experiments . . . . .	27
4.0 CONCLUSIONS AND RECOMMENDATIONS . . . . .	32
5.0 SUMMARY - $^{85}\text{Sr}$ . . . . .	33
6.0 REFERENCES . . . . .	34

## LIST OF FIGURES

Figure	Page
1. Sequential steps involved in obtaining adsorption and desorption distribution coefficients with the constant shaking technique . . . . .	12
2. Schematic diagram of apparatus used for dialysis experiments . . . . .	14
3. Effect of pH on adsorption distribution coefficient of $^{90}\text{Sr}$ at Hudson River and Cattaraugus Creek stations . . . . .	20
4. Concentration of soluble $^{85}\text{Sr}$ as a function of time in the various compartments in a dialysis experiment for the Lake Michigan sediment-water system . . . . .	30

## LIST OF TABLES

Table	Page
1. Location of sediments, sewer and freshwater samples used in distribution coefficient studies . . . . .	7
2. Particle size, density, carbon and nitrogen content of sediments used in the experiments . . . . .	9
3. Chemical properties of water samples used in the early experiments . . . . .	10
4. Summary of adsorption-desorption distribution coefficients for $^{85}\text{Sr}$ for different sediment-water systems . . . . .	17
5. Effect of pH on $K_d$ values for Hudson River Stations and at Cattaraugus Creek, New York . . . . .	21
6. Adsorption distribution coefficients for $^{85}\text{Sr}$ as a function of sediment concentration . . . . .	26
7. Relative concentrations of $^{85}\text{Sr}$ measured during a 15-day dialysis experiment . . . . .	28
8. Comparisons of $K_d$ values for adsorption of $^{85}\text{Sr}$ by the constant shaking method in Lake Michigan sediment-water system . . . . .	31

# DISTRIBUTION COEFFICIENTS FOR RADIONUCLIDES IN AQUATIC ENVIRONMENTS

## Adsorption and Desorption Studies of $^{85}\text{Sr}$

### 1.0 INTRODUCTION

The problems associated with radioactive materials entering into the environment are with us today and will continue to be with us for some time in the future. The sources of these radioactive waste materials are nuclear-electric power generation and defense projects. Because of this "fait accompli" we must examine some of the critical natural processes and identify biogeochemical pathways which influence the introduction of radioactive materials into the biosphere.

The eventual fate of any pollutant in aquatic ecosystems is greatly affected by its uptake on inorganic and organic particulates. Chemical elements (stable or radioactive) which are introduced into aquatic environments are removed from the water by sorption onto organic and inorganic particulate matter and, to a lesser extent, by adsorption to and uptake by living organisms. The physical, chemical and biochemical factors which govern the removal processes are numerous and many are not well understood. An important key to understanding the behavior of any radionuclide in aquatic environments is knowledge of the physico-chemical state of the nuclide. Unfortunately, knowledge of the physico-chemical states of the radionuclides is difficult to obtain and may be altered by environmental factors. Therefore, the approach used in these studies is not to study physico-chemical states of radionuclides, per se, but to evaluate the distribution of radionuclides between the aqueous and particulate phases in laboratory systems using sediments and water from natural environments. This more applied approach can give specific information on radionuclide removal processes important to modeling the hydrological regimes at specific locations of interest.

During the past several years we have been evaluating several processes controlling the distribution coefficients  $K_d^*$  values, of radionuclides sorbed onto different natural matrices. Distribution coefficients provide a quantitative measure of the accumulation of radionuclides, or other pollutants, by suspended particulate matter. Therefore, they are an important parameter for predicting the fate of actual or potential aquatic pollutants. Although a great deal of research has been conducted to determine the  $K_d$  values of various radionuclides in specific environments, it is often difficult to utilize the results from one environment to make predictions about new environments. Furthermore,

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

there is remarkably little information about the transuranic elements. Therefore, this program was started to obtain  $K_d$  values for plutonium and americium in marine and freshwater environments. It was then expanded to include other radionuclides and selected environments that were of particular concern to Nuclear Regulatory Commission (NRC).

## 1.1 Background

### 1.1.1 Previous Information

As a result of nuclear weapons testing, fuel production and electrical power generating nuclear reactors, by-product radionuclides have entered the environment. Strontium-90 ( $t_{1/2}$  of 30 years) is foremost among these radionuclides due to its production from the fissioning of  $^{239}\text{Pu}$  and  $^{235}\text{U}$ . (Approximately 5% of the total products resulting from  $^{239}\text{Pu}$  and  $^{235}\text{U}$  fission produce  $^{90}\text{Sr}$ .) (1) Fallout  $^{90}\text{Sr}$  from nuclear weapons tests is present in the global environment and is used extensively in tracing ocean water masses since it is conservative, i.e., it is not removed appreciably by biogenic or detrital sediment particles in the deep ocean (2,3).

Environmental samples collected from the surface sediments and overlying water have been used in determining the partition or distribution coefficients. These measurements make the assumption that equilibrium is reached between the sediment and overlying waters and that there is no appreciable change in water concentration by advective mixing processes. This assumption may have merit at stagnant regions where currents are negligible and for radionuclides that have a short equilibration-exchange time.

Radiostrontium has been introduced into Perch Lake, Chalk River, Establishment, Canada, since 1955 through seepage from a nearby liquid waste disposal area. The distribution coefficient calculated for the upper one inch of sediment to the water has been measured by Ophel (4) to be about 180 ml/g. In the Guadalupe River, Texas, Reynolds and Gloyna (5) measured the distribution coefficient for  $^{90}\text{Sr}$  under static and flow conditions at 198 ml/g for a sediment with a cation exchange capacity of 35 meq/100 g and a suspended sediment concentration of 500 mg/l. The distribution coefficient for  $^{89}\text{Sr}$  in water of 16.5‰ salinity for these same sediments was 40.8 ml/g. The desorption distribution coefficient in bay water was measured at 2,520 ml/g. Huff and Kruger (6) determined that the fallout  $^{90}\text{Sr}$  distribution coefficient from six storms on the Saratoga Watershed, California, in 1965 and 1966 was  $302 \pm 30$  ml/g. In a study by Kulp and Schulert (7) 70-75% of the fallout  $^{90}\text{Sr}$  was found in the upper 2 inches of soil and 90-99% was in the upper six inches; there was no evidence for vertical movement of the  $^{90}\text{Sr}$  after initial deposition over a period of six years. In an experiment mentioned in this study where an equivalent of 300 inches (10 years worth) of rain was used, 90% of the  $^{85}\text{Sr}$  was retained on the top 1 1/2



inches of soil (7). Using these values the distribution coefficient for this soil-rainwater system is calculated to be 780 ml/g. Only 0.4 to 5% of the  $^{90}\text{Sr}$  is removed from soil by runoff. However, the soil carried by the runoff has 10 times higher concentrations of  $^{90}\text{Sr}$  than the average residual soil (7).

In the Clinch River system Parker et al. (8) measured  $^{90}\text{Sr}$  in water samples (total) and determined the percentages of  $^{90}\text{Sr}$  in the suspended solids together with the concentration in the water. From those data the following distribution coefficient values were determined:

<u>Station</u>	<u>Sediment Concentration</u>	<u>Distribution Coefficient</u>
CMR 41.5	185 mg/l	1701 ml/g
CMR 5.5	55 mg/l	1818 ml/g
CMR 14.5	25.3 mg/l	2553 ml/g
TRM 529.9	15 mg/l	6611 ml/g
TRM 471.0	9 mg/l	12345 ml/g

The pH for these river samples was from 7.6 to 7.8.

The uptake of  $^{90}\text{Sr}$  by open sea plankton gives concentration factors for  $^{90}\text{Sr}$  between 30 and 380 depending on the species (9). Adsorption was found in this study to play an important role in the accumulation of  $^{90}\text{Sr}$  by plankton. The  $^{85}\text{Sr}$  uptake, primarily by algae, in a fresh water flume has been measured by Gloyna et al. (10) and similar but higher sorption was found. From their data the distribution coefficient for  $^{85}\text{Sr}$  between water and suspended sediment has been calculated to be in the range of 2,465 to 21,764 ml/g at sediment concentrations of 54 to 6 mg/l respectively. These high distribution coefficient values indicate the importance of organic matter in the competition of  $^{85}\text{Sr}$  by particulate organic and inorganic matter in the suspended phase.

In a clay-water system cations are sorbed to the matrix through cation exchange equilibrium dominated by a) Coulombic interactions between counter ions (in various stages of hydration) and the fixed groups of the exchanger, and b) ion-dipole and ion-induced dipole interaction between counter ions and water molecules (ionic hydration) (11). Generally, (b) predominates and the "normal" Hoffmeister affinity series for cations is  $\text{Cs}^+ > \text{K}^+ > \text{Na}^+ > \text{Li}^+ > \text{Ba}^{+2} > \text{Sr}^{+2} > \text{Ca}^{+2} > \text{Mg}^{+2}$ . For example,  $\text{Cs}^+$  would be held more strongly to the clay than potassium and may therefore replace  $\text{K}^+$  in a clay-water system.

### 1.1.2 Present Studies

A great deal of research has been conducted to determine the  $K_d$  values for some radionuclides, but remarkably little information is available for the transuranic elements. Therefore, this program was 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  $^{85}\text{Sr}$ ,  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ , and  $^{244}\text{Cm}$ , in sediment-water systems that are of particular concern to NRC.

In August 1976 we began to investigate adsorption and desorption phenomena of  $^{237}\text{Pu}$  and  $^{241}\text{Am}$  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 (12). Results for preliminary experiments using  $^{65}\text{Zn}$  and  $^{137}\text{Cs}$  were compared to published results (13) 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  $^{237}\text{Pu}$  and  $^{241}\text{Am}$  in laboratory sediment-water systems. Dialysis experiments were used to study the distribution of  $^{60}\text{Co}$ ,  $^{65}\text{Zn}$ ,  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  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 uptake of soluble radionuclides by sediments and phytoplankton. Results from the first year of research are included in NUREG/CRO801 (14).

During FY 77-78 the project was extended to include new sediment types and additional radionuclides. Constant shaking experiments utilized  $^{85}\text{Sr}$ ,  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$  and  $^{241}\text{Am}$  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. We also evaluated the sedimentation technique (12) for determining  $K_d$ 's of  $^{237}\text{Pu}$  and  $^{241}\text{Am}$ , the effect of pH on the distribution coefficient of  $^{241}\text{Am}$  in sediment-water systems from Lake Nitinat and Lake Washington and particle formation by  $^{106}\text{Ru}$  and  $^{241}\text{Am}$  in the absence of suspended sediments. These results are available in NUREG/CRO802 (15). 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. Dialysis experiments were expanded in FY 77-78 to include the radionuclides  $^{59}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{65}\text{Zn}$ ,  $^{137}\text{Cs}$ ,  $^{204}\text{Bi}$ ,  $^{238}\text{Pu}$  and  $^{241}\text{Am}$ . Additional sacs were added containing a blank of filtered seawater and organic detritus as well as the sediment and plankton.

During FY 1978-79, adsorption distribution coefficients for  $^{85}\text{Sr}$ ,  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$  and  $^{237}\text{Pu}$  were determined in constant shaking experiments using filtered water and <63  $\mu$ -sized sediments from Lake Michigan, Clinch River, the mouth of Cattaraugus Creek where it flows into Lake Erie, Skagit Bay and Sinclair Inlet in Puget Sound, Washington and three stations in the Hudson River estuary. Desorption distribution coefficients for  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$  and  $^{241}\text{Am}$  were determined for sediment-water systems from Lake Michigan and the three stations in the Hudson River

estuary. For  $^{85}\text{Sr}$  and  $^{237}\text{Pu}$  desorption distribution coefficients were determined for Lake Michigan, the Hudson River estuary, Clinch River and Sinclair Inlet. The desorption  $K_d$  values were generally more than an order of magnitude higher than the adsorption  $K_d$  values. During FY 78-79 the dialysis experiments were continued using the radionuclides  $^{85}\text{Sr}$ ,  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$ ,  $^{237}\text{Pu}$  and  $^{241}\text{Am}$  with sediments and water from Lake Michigan. We also conducted preliminary experiments to investigate the effects of pH, suspended sediment concentration and selected organic ligands. Results from the FY 78-79 studies were published in NUREG/CRO803 (16).

The experimental program was extended into FY 79-80 to complete the experiments on adsorption and desorption distribution coefficients and to obtain additional information on the effects of organic ligands, pH and sediment concentration. Adsorption and desorption distribution coefficients for  $^{244}\text{Cm}$  were measured for the Hudson River estuary and the effect of pH and sediment concentration on the  $^{244}\text{Cm}$   $K_d$  value for Cattaraugus Creek was determined. Results from these studies will be presented in later reports in this series.

## 1.2 Objectives

The general objective for this program is to obtain 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. Such models can be used to predict the eventual environmental distribution of radionuclides following a release.

This report describes the experimental program and evaluates the results obtained for the distribution coefficients of  $^{85}\text{Sr}$  in different sediment-water systems.

## 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 (14,15,16) and will be discussed briefly below. The thin layer and suspended sediment techniques were investigated during the first year but reproducibility was not found sufficient so the constant shaking method was used in all the later studies.

### 2.1 Sources and Types of Samples

For the first two 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 in later years. Information for these samples is provided in Table 1.

During the third and fourth years of the program, water and sediment samples were collected at locations throughout the U.S.A in collaboration with other laboratories. In particular, Argonne National Laboratories (ANL) provided sediments and water from Lake Michigan and Lamont-Doherty Geological Observatory (LDGL) provided several samples from the Hudson River estuary. Samples from Cattaraugus and Buttermilk Creeks, New York State, 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), and samples from the Clinch River, Tennessee, were collected with the cooperation of Oak Ridge National Laboratories (ONL). Specific information for nonlocal samples is also provided in Table 1.

For the initial laboratory studies, large quantities of "standard" sediment and water samples were prepared from samples collected at three local stations--one of anoxic marine sediments from Lake Nitinat (a fjord), Vancouver Island, British Columbia; one of marine sediments collected from off the mouth of the Columbia River; and one of the freshwater sediments from the deepest part (62 m) of Lake Washington, Seattle. Both water and sediment samples were frozen after collection, and kept frozen until they were used in experiments. All water samples were filtered through 0.3  $\mu$ m Millipore filters. By having a large quantity of sediment and water available, replicate experiments could be conducted.

Knowledge of the physical and chemical characteristics of the sediments and water is necessary for proper interpretation of the distribution coefficient values for various water-sediment systems. Sediment characteristics that are of particular importance in determining the adsorptive properties of a sediment-water system are the surface area, ca-

Table 1. Location of sediments, seawater and freshwater samples used in distribution coefficient studies.

Sample type	Sediment depth (m)	pH	Salinity ‰	Hardness mg/l	Surface area m <sup>2</sup> /g (SD)	C mg/g	N mg/g	Treatment	Collection Site
Lake Michigan	67	7.90	<1.0	143	17.19(.48)	62.0	1.8	Frozen	43° 00'N, 83° 20' W - ANL Station 5
Clinch River	3.0	7.92	<1.0	158	13.37(.35)	14.0	1.8	Fresh (cooled)	16 mile upstream from Tennessee River
Hudson River									
MP 59.8 SLOSH VII	3.0	7.90	<1.0		12.24(.24)			Fresh (cooled)	15 miles upstream from Indian Point Reactor
MP 43.3 SLOSH III	3.0	7.90	<1.0	171	11.99(.34)	34.4	1.8	Freeze-dried	Adjacent to Indian Point Reactor
SLOSH VIII		--	3.2	--	9.24(.34)			Fresh (cooled)	Adjacent to Indian Point Reactor
MP 18.6 SLOSH II	3.0	7.82	2-3	378	8.98(.14)	23.0	1.1	Freeze-dried	18.6 miles upstream from southern tip of Manhattan Island
SLOSH IX		--	10.7	--	10.95(.12)			Fresh (cooled)	18.6 miles upstream from southern tip of Manhattan Island
MP 0.1 SLOSH V	6.0	7.70	15	2963	7.99(.18)	13.8	5.2	Freeze-dried	0.1 mile upstream from southern tip of Manhattan Island
SLOSH X		--	21.0	--	12.34(.29)			Fresh (cooled)	0.1 mile upstream from southern tip of Manhattan Island
Cattaraugus Creek	1.5		<1	202	10.75(.04)	16.8	0.8	Fresh (cooled)	PNL-station CC-11, mouth of creek
Skagit Bay-1	9	7.70	29.62	6662	2.03(.09)			Fresh (cooled)	48° 18.0'N, 122°29.0'W sandy sediment
" " 4	59	7.70	31.05	5325	7.01(.79)			Fresh (cooled)	48° 15.5'N, 122°32.5'W silt and clay sediment
Columbia River	100	7.80	32.56	6250	7.10				Outside bar on continental shelf, mouth of Columbia River
Saanich Inlet	225	7.80	31.24	5316					48°35.4'N, 123°30.2'W anoxic sediment and water
Lake Nitinat	203	7.80	31.22	6188	39.8			Fresh (cooled)	Deep anoxic lake on Vancouver Island, B.C.
Sinclair Inlet	15.1	7.75	28.89	5506				Fresh (cooled)	Oxic organic rich sediment in Puget Sound
Lake Washington	62	7.80	<1	42	31.4(6.2)			Fresh (cooled)	Deepest point in lake at Madison Park
Organic Detritus									
Lake Michigan	67	7.90	<1	143					
Skagit Bay	59	7.70		6662					
Montmorillonite	59	7.70	31.05	5325	87.82(2.3)				

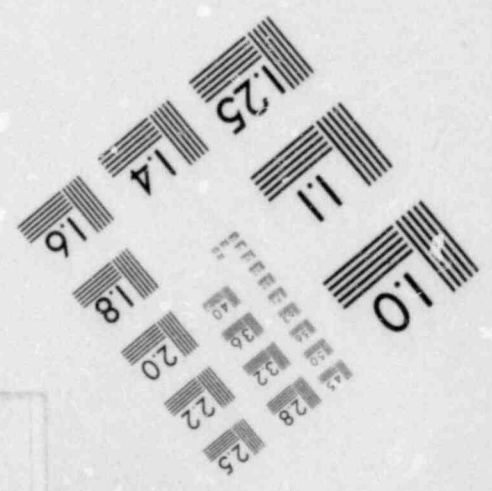
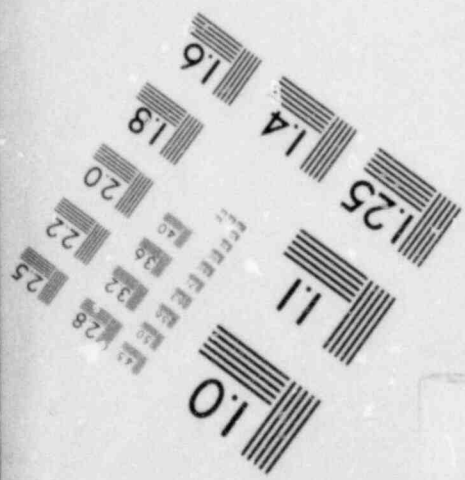
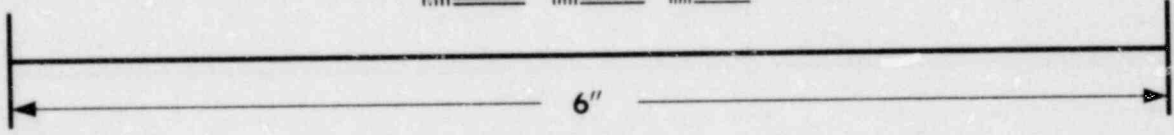
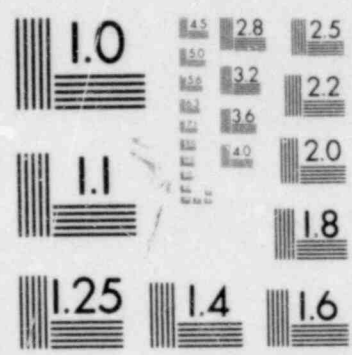
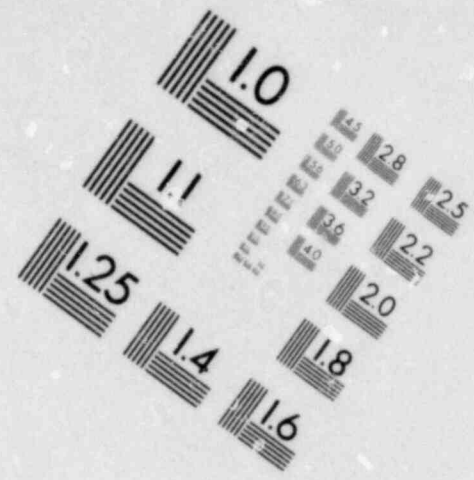
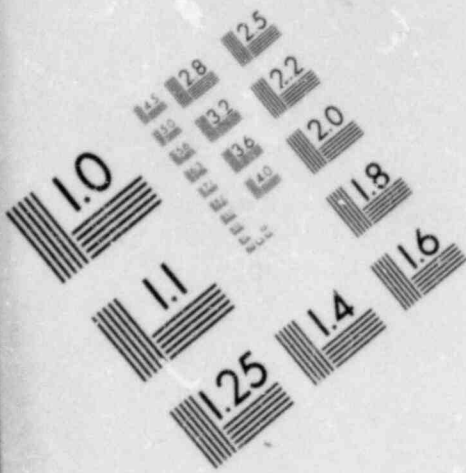
tion exchange capacity, the particle size distribution and the chemical composition of the sediments. An example of this type of information on particle size distribution and carbon and nitrogen composition of three particular sediments is shown in Table 2. It was observed that the clay fraction is predominant (75%-80%) in both the Lake Washington and Lake Nitinat (saltwater) samples. The marine sediment is composed of sand (40%), silt (21%), and clay (39%).

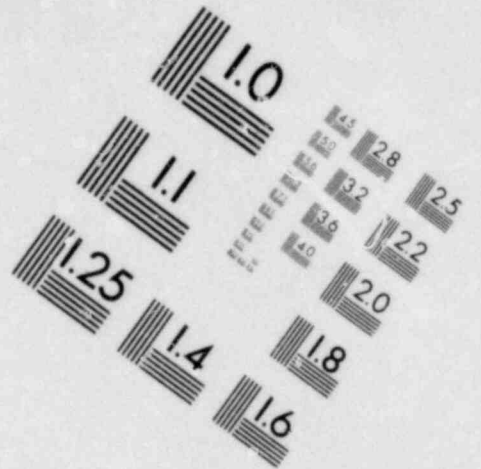
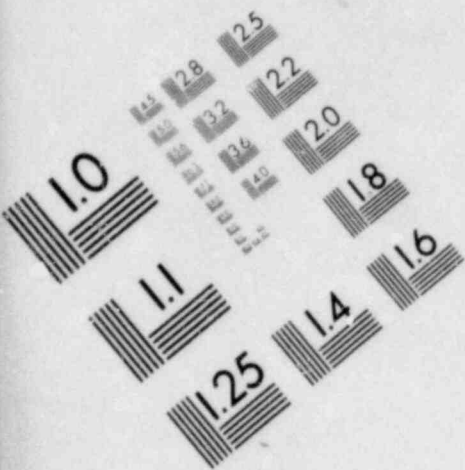
The water properties of a sediment-water system which are important in adsorption are the chemical composition, the solution pH and the oxidation-reduction potential of the solution. Some chemical and physical properties of three water samples used in our experiments are given in Table 3. Storage of water and sediment samples in a manner which minimizes changes in their physical and chemical properties presents a difficult problem. We examined several different methods including storage at 4°C, freezing and sterilization by heat or by irradiation with a <sup>60</sup>Co food irradiator. The chemical analyses of the marine water samples were made shortly after collection and then again after freezing and storage for about 1 year. The freshwater samples (Lake Washington) were frozen and stored for 1 month after collection before they were used in the experiments and analyzed for nutrients. The chemical analysis of anoxic marine water (Lake Nitinat) showed large differences in the nutrients following filtration and/or sterilization. Similar treatment produced only small differences for most nutrients in the marine and freshwater samples. These differences are due to both removal of particulates (filtration) and to irradiation. The effects of the irradiation alone on the water have not been identified. However, the changes in chemical properties of the water illustrate that the laboratory experimental conditions do not necessarily represent chemical and biological conditions found in the field.

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

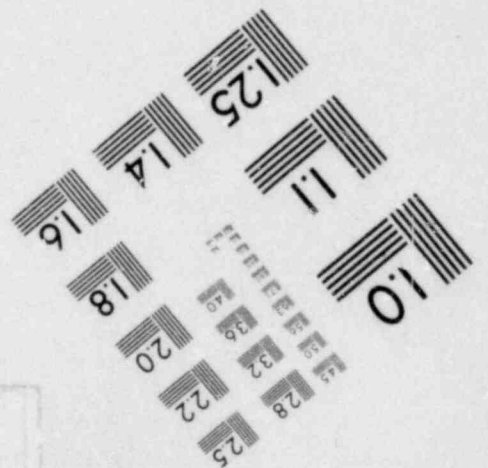
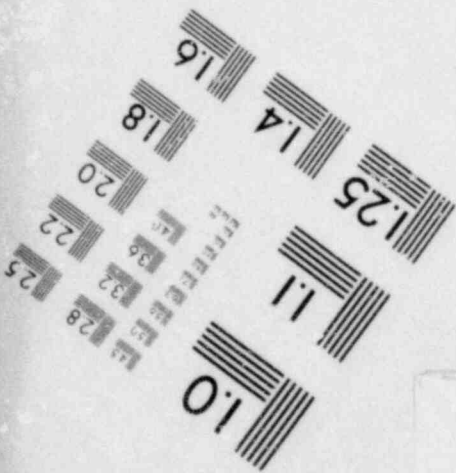
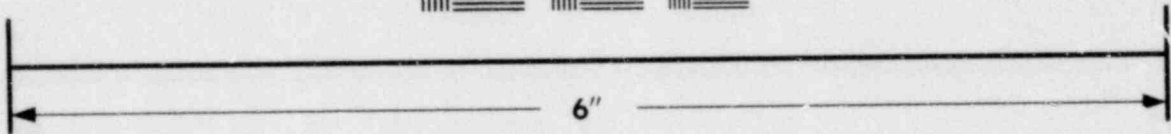
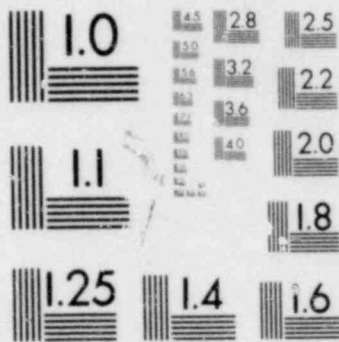
A question has arisen regarding the representative nature of  $K_d$  values obtained from samples which were frozen or freeze-dried before use. These values also must be compared to what is occurring naturally in the field. Ideally, measurements should be made under natural conditions but this is often 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 4°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 elements present in pore water with the sediment and would

IMAGE EVALUATION  
TEST TARGET (MT-3)





**IMAGE EVALUATION  
TEST TARGET (MT-3)**





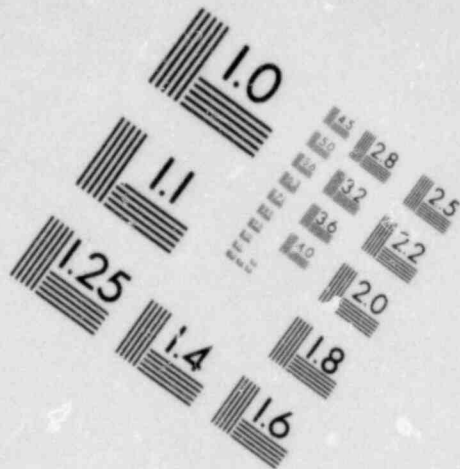
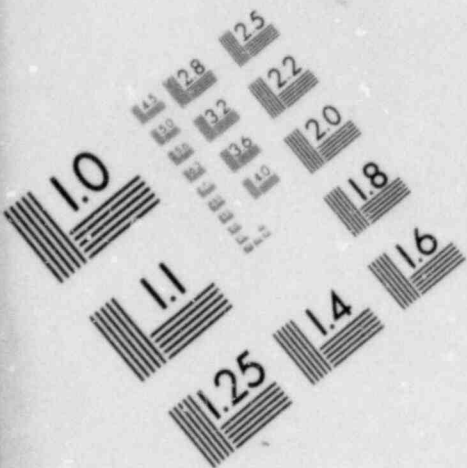


IMAGE EVALUATION  
TEST TARGET (MT-3)

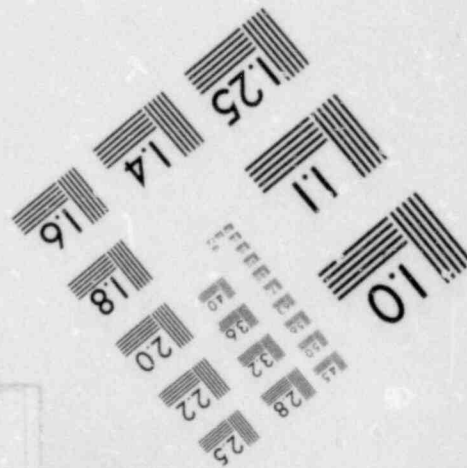
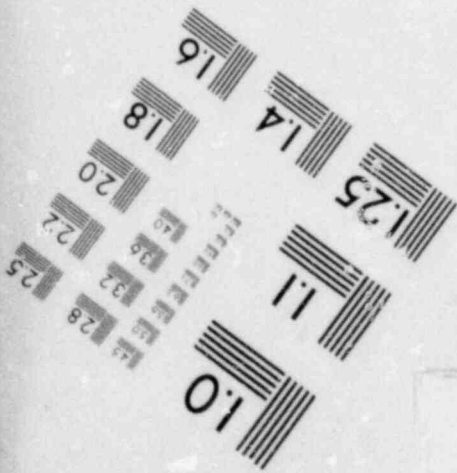
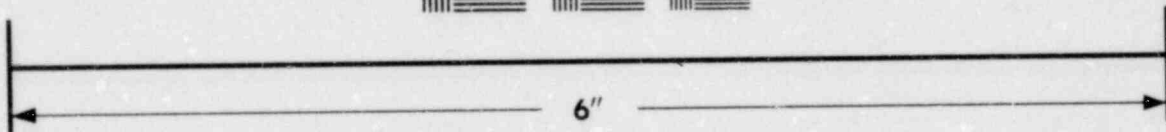
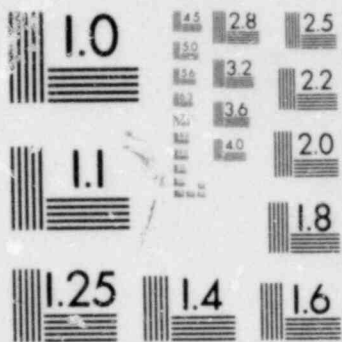


Table 2. Particle size, density, carbon and nitrogen content of sediments used in the experiments.

Particle			Percentage in sediments		
Ø Size	Diameter mm		Lake Washington	Lake Nitinat	Marine
-1	2		0	0.17	0.17
0	1		0.17	0.42	0.33
1	0.5		0.43	1.18	0.67
2	0.25		1.20	1.10	1.84
3	0.125		0.47	0.48	13.40
4	0.0625		0.21	0.49	24.00
Sand	(0-4)		2.48	3.67	40.24
6	0.0156		3.31	3.50	12.58
7	0.008		3.31	5.61	2.58
9	0.002		8.97	11.88	5.67
Silt	(5-9)		15.99	21.39	20.83
Clays	(>9)		81.93	74.93	38.7
Density	g/cm <sup>3</sup>		2.38	2.55*	2.56*
<u>Carbon and nitrogen contents</u>					
Total carbon %			5.25	2.77	1.81
Total nitrogen %			0.41	0.26	0.08
C/N ratio			11.9	10.5	21.7

\*Corrected for 4% salt content.

Table 3. Chemical properties of water samples used in the early experiments.

Sample Type	Temp °C	Salinity ‰	Reactive Phosphate μM	Reactive Silicate μM	Reactive Nitrate μM	Reactive Nitrite μM	Reactive Ammonia μM	Hydrogen Sulfide μM	Remarks
Lake Nitinat 48° 44.6'N 124° 45.3'W	10.11	31.267	3.30	104.43	0.00	0.06	76.86	166	Unfiltered
			2.14	43.2	26.7	0.23	2.0	-	Filtered
			3.58	67.1	14.9	0.72	49.3	-	Filtered Sterilized
Marine Water 48° 38.0'N 124° 53.0'W	8.29	32.566	2.37	44.96	26.83	0.16	0.61	-	Unfiltered
			2.24	45.9	28.5	0.23	0.4	-	Filtered
			1.37	24.2	23.6	1.01	3.4	-	Filtered Sterilized
Lake Washington 47° 37.5'N 122° 16' W			0.06	14.2	3.4	0.00	2.0	-	Filtered
			0.09	15.3	5.7	0.00	2.2	-	Filtered Sterilized

10

\* Filtered samples were passed through 0.3μM Millipore filters.

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. The results of the tests show that the freeze-dried samples are a factor of 0 to 4 times greater in  $K_d$  values than the fresh samples collected in the Hudson River in 1979.

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 (13,16,17). Briefly, the method consists of:

1. Adding the radionuclides to a known volume of filtered (0.45  $\mu$ m or 0.2  $\mu$ m) water sample and adjusting the pH to the initial pH of the water sample with 0.2 M NaOH.
2. Adding sediments previously sieved to less than 63  $\mu$ m 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. Figure 1 shows the sequential steps that are involved in obtaining adsorption and desorption distribution coefficients with the constant shaking technique.

### 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

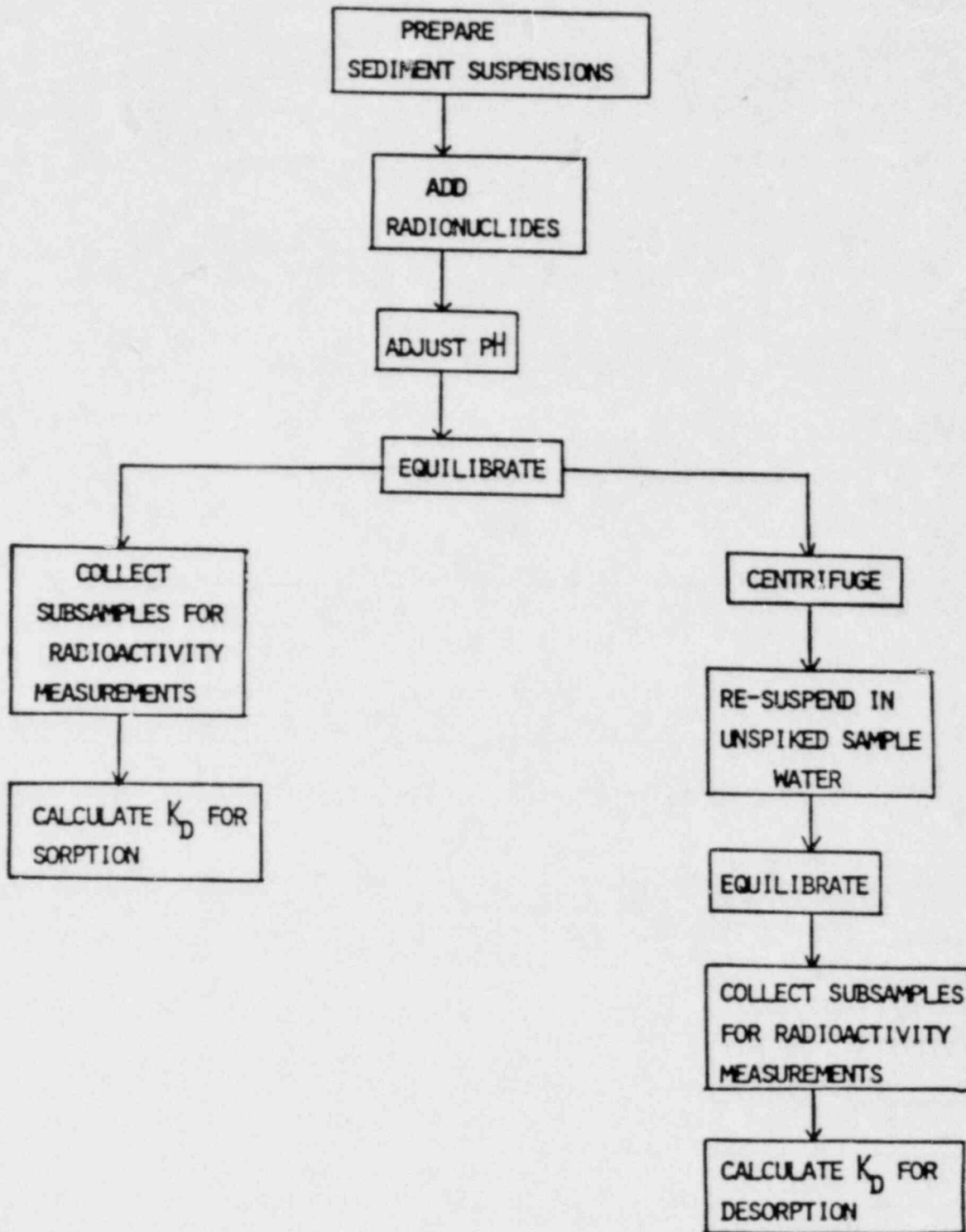


Figure 1. Sequential steps involved in obtaining adsorption and desorption distribution coefficients with the constant shaking technique.

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

The dialysis apparatus (Fig. 2) used for these experiments was adapted from similar equipment described by Barsdate (18) and Dawson and Duursma (19). In all our experiments, the outside compartment contains filtered (0.22  $\mu\text{m}$  Millipore filters) water that has been spiked with radionuclides and allowed to equilibrate for at least 10 days before the experiments begin. It is important to attain equilibrium between stable and radioisotopes and among different physico-chemical species since the experiments are designed to show how the physico-chemical species found in natural environments may behave. After the equilibration period, dialysis sacs containing filtered, or distilled water were submerged in the radioactive solution and the suspended particulates were then added to the dialysis sacs. Initially only two dialysis sacs were used, one containing phytoplankton and the other containing suspended sediments. In later experiments, two additional dialysis sacs were added in order to consider uptake of radionuclides by organic detritus and to have a control sac to study diffusion across the dialysis membrane.

Initial experiments investigated systems with different volumes of water. However, the present system consists of a 1000-ml beaker containing 700 ml of spiked, filtered water. Dialysis sacs, containing 50-70 ml of the same filtered water without the radionuclides, are then submerged in the outside compartment. During the experiment, 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 are intended to exclude species above a specified molecular weight. For these experiments dialysis membranes with a 6000-8000 molecular weight cut-off were used. Thus, only ionic species, low molecular weight complexes, and small colloids can diffuse across the membrane. Samples were removed from the dialysis sacs and the outside chamber at predetermined times and filtered through Millipore or Nucleopore membrane filters in order to separate the particulate and soluble phases. Both the filter and filtrate are then measured to determine the concentration of particulate and dissolved radionuclides in each chamber of the experiment.

The phytoplankton species used in these experiments, Phaeodactylum tricornutum and Chlorella vulgaris were obtained from a culture maintained by Dr. Frieda Taub, College of Fisheries, University of Washington. Prior to use in the experiments, a stock algal suspension ( $8.6 \times 10^6$  cells/ml) was centrifuged at 6000 rpm for 15 min and re-suspended in 32 ‰ NaCl solution. This process was repeated four times to remove the

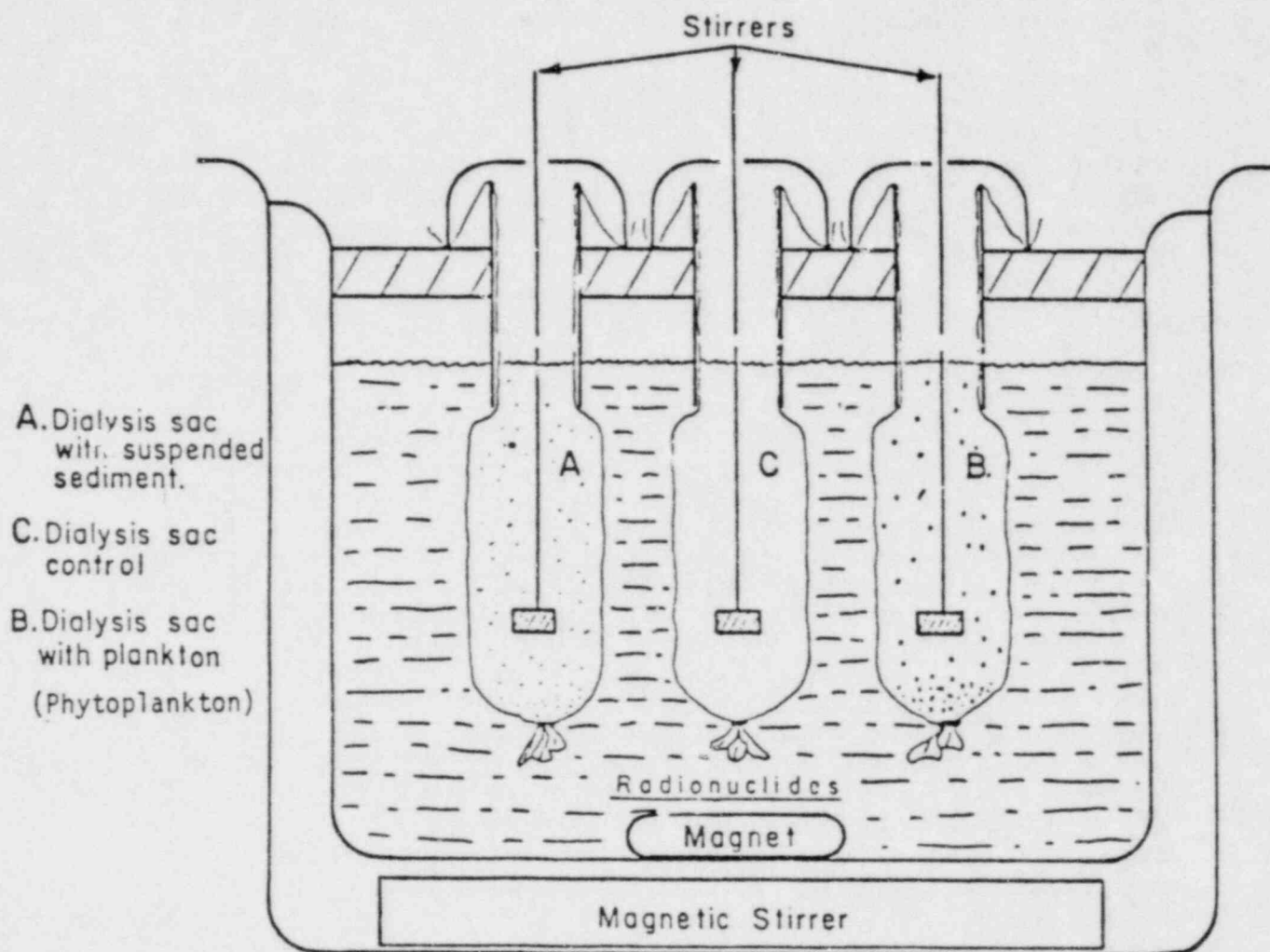


Figure 2. Schematic diagram of apparatus used for dialysis experiments.

nutrient media and the final rinse was in 32 ‰ filtered seawater. The phytoplankton were then re-suspended in filtered seawater before being added to the dialysis sac.

The suspended sediments were either natural sediments from Lake Nitinat, an anoxic fjord located on the western coast of Vancouver Island, British Columbia, or the reference clay, calcium montmorillonite, obtained from the Georgia Kaolin Company. In each case, a concentrated stock solution was prepared with a known sediment concentration (mg/l) and specified volume of the stock solution was added to the sediment dialysis sac.

Organic detritus was prepared from *Zostera marina*, a common marine macrophyte in Puget Sound. After washing and drying at 80° C, the *Zostera* was ground in a ball mill and sieved. For the experiments, a weighed amount of the <63 µm size fraction was suspended in filtered seawater to obtain a stock suspension. A known volume of this suspension was then added to a dialysis sac so that the weight of detritus in the experiment could be calculated.

#### 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,  $^{57}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$ , and  $^{241}\text{Am}$ , 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  $^{85}\text{Sr}$  and  $^{237}\text{Pu}$ , 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.



### 3.0 RESULTS AND DISCUSSION

#### 3.1 K<sub>d</sub> 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.

#### Adsorption of <sup>85</sup>Sr

Constant shaking experiments were conducted to determine the adsorption K<sub>d</sub> values of <sup>85</sup>Sr, the gamma emitting tracer of the radio strontium isotopes, 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 <sup>85</sup>Sr are shown in Table 4.

The highest K<sub>d</sub> values for <sup>85</sup>Sr are found consistently in freshwater systems. At MP 18.6, salinity of 2 - 3‰, the K<sub>d</sub> value is a factor of approximately 2 or more lower than for the freshwater systems, MP 43.3 and MP 59.8. At higher salinities (15‰ - 31‰), <sup>85</sup>Sr was not significantly sorbed to the sediments. Two explanations can be proposed for the low K<sub>d</sub> values for <sup>85</sup>Sr in marine systems: a) isotopic equilibrium between tracer <sup>85</sup>Sr and stable strontium may not be reached during the experiment and/or, b) ion exchange on the sediment surface favors sodium over strontium ions as shown by the "normal" Hoffmeister affinity series for cations (see Section 1.1.1). 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<sub>d</sub> values in Lake Michigan and Skagit Estuary systems are similar. This suggests that the low K<sub>d</sub> for <sup>85</sup>Sr is not due to competition with the stable strontium, but rather to saturation of available exchange sites with a more active cation such as sodium. The higher K<sub>d</sub> value for <sup>85</sup>Sr on marine detritus relative to suspended sediments suggests that organic surfaces may be more important than inorganic sediments for scavenging and transporting radiostrontium in marine environments.

For all of the sediment-water systems, <sup>85</sup>Sr has the lowest K<sub>d</sub> value of the five radionuclides measured, generally less than 100, and <sup>241</sup>Am has the highest value, greater than 10<sup>5</sup>. The order of increasing K<sub>d</sub> values is <sup>85</sup>Sr < <sup>137</sup>Cs < (<sup>106</sup>Ru or <sup>237</sup>Pu) < <sup>241</sup>Am. The radionuclides

Table 4. Summary of adsorption-desorption distribution coefficients for  $^{85}\text{Sr}$  for different sediment-water systems.

Sample type	Average $K_d$ (SD) ml/g				Treatment
	n(a)	Adsorption	n(b)	Desorption	
Lake Michigan	7	82.2 (7.0)	10	1410 (510)	Frozen
Clinch River	6	124.4 (7.5)	8	455 (89)	Fresh
Hudson River					
MP 59.8	9	113.0 (13.5)		(b)	Fresh
MP 43.3	6	73.7 (10.5)	9	486 (135)	Freeze-dried
"		84.4 (23)		(b)	Freeze-dried
MP 18.6	6	34.8 (5.5)	9	(b)	Freeze-dried
"	4	38.9 (16.9)		(b)	Fresh
MP 0.1		(b)	12	(b)	Freeze-dried
"		(b)		(b)	Fresh
Cattaraugus Creek	8	62.2 (6.2)		(b)	Fresh
Skagit Bay -1	6	(b)		(b)	Fresh
" -4	6	(b)		(b)	Fresh
Columbia River					Fresh
Saanish Inlet					Fresh
Lake Niinat					Fresh
Sinclair Inlet	9	(b)	9	(b)	Fresh
Lake Washington					Fresh
Organic detritus					
Lake Michigan	4	75.2 (4.1)		-	Dried, ground
Skagit Bay	6	68.3 (4.4)		-	Dried, ground
Montmorillonite					

(a) n is the total number of  $K_d$  values determined for a given sediment-water system.  
 (b) Concentrations in particulate phase was below detection limits.

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

#### Desorption of $^{85}\text{Sr}$

Experiments have been completed to determine the desorption  $K_d$  values of  $^{85}\text{Sr}$  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/l sediment in unspiked water from the same sampling location. The suspensions were kept under constant shaking in a cold room at 4°C.

The results of the desorption distribution coefficient experiments also are shown in Table 4. The desorption  $K_d$  values for this isotope in the freshwater systems varied as follows:

Clinch River < Hudson River < Lake Michigan

The estuarine (Hudson River MP 0.1) 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 in the high salt environment.

#### Summary of Sorption and 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 4. For  $^{85}\text{Sr}$ , the  $K_d$  values for desorption were higher than the adsorption  $K_d$  values by factors of 4 to 17. These results suggest that under these experimental conditions, sorption is not completely reversible. Because of the low adsorption  $K_d$  of  $^{85}\text{Sr}$  to the sediment, the desorption  $K_d$  values often were not measurable because of the low counting rates of the particulate and soluble fractions. 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,  $^{85}\text{Sr}$  showed the lowest desorption  $K_d$  values, and  $^{241}\text{Am}$  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  $^{241}\text{Am}$  (14). The results presented below are from experiments with water and sediments from the Hudson River, New York shown in Fig. 3(a-d) and the Cattaraugus Creek shown in Fig. 3e, and Table 5.

At the lower Hudson river stations MP 0.1 and MP 18.6, the  $K_d$  value of 66 ml/g was almost unchanged for the pH range of 4 to 10. At MP 43.3 and at MP 59.6 there appeared to be an increase in  $K_d$  values with increasing pH;  $K_d$  values at MP 43.3 ranged from 56 ml/g at pH 4.6 to 100 ml/g at pH 10.0 whereas  $K_d$  values at MP 59.8 ranged from 86 ml/g at pH 4 to 170 ml/g at pH 9.8. It appears that salt water intrusion influences the  $K_d$  values for  $^{85}\text{Sr}$  as far as MP 18.6. The  $K_d$  values for the MP 0.1 and MP 18.6 required 4 to 10 days to achieve equilibrium. This probably results from the exchange equilibrium of sorbed Sr with Ca and Na ions in the brackish estuarine water-sediment system. At 10 days and pH 7.68, the sorption of  $^{85}\text{Sr}$  on the particulate fraction is not measurable, as shown in Table 5. The upper river stations, where marine water is not present, exhibit the typical sediment behavior of ion exchange and substitution of strontium on the exchangeable lattice sites of the clays.

At the Cattaraugus Creek Station CC-11, the  $K_d$  value increased gradually from 70 ml/g at pH 4.3 to 90 ml/g at pH 8. However, between pH 8 and 9.2 the  $K_d$  value increased by a factor of 24 at pH 9.2 and reached the highest  $K_d$  value measured on any sediment of 4570 ml/g at pH 9.9. The isoelectric pH for strontium in this soil-clay-water system must be about pH 9.0. At this point the strontium sorbs strongly to the sediment, possibly as  $\text{SrCO}_3$  and is removed from solution giving the high  $K_d$  value. The high bicarbonate content of this water could contribute to the precipitation of  $\text{SrCO}_3$ .

During the equilibrium time of the experiment (i.e., 1, 4, 10 days), the pH varied as much as 2 pH units for some sediment-water systems. After a sample was taken, the pH was then readjusted to the original pH with NaOH or HCl and the sample was replaced on the shaker. The average pH was determined from the initial reading and final reading, assuming a linear change with time. For the equilibrium time period of 10 days for samples from the Hudson River stations MP 0.1 and MP 18.6, non-significant  $K_d$  values were found. This finding must be the result of the exchange of sodium and/or hydrogen ions for the  $^{85}\text{Sr}$  initially adsorbed onto the sediment. The  $^{85}\text{Sr}$  was found mostly in the aqueous fraction and at values below the instrumental detection limits for the particulate fraction. Another possible reason for these low values at later times may be the result of coagulation of particles resulting in the non-homogeneous sampling of the suspended sediment fraction. Since the concentration of sediment is so low, a 5 ml aliquot of the total volume would

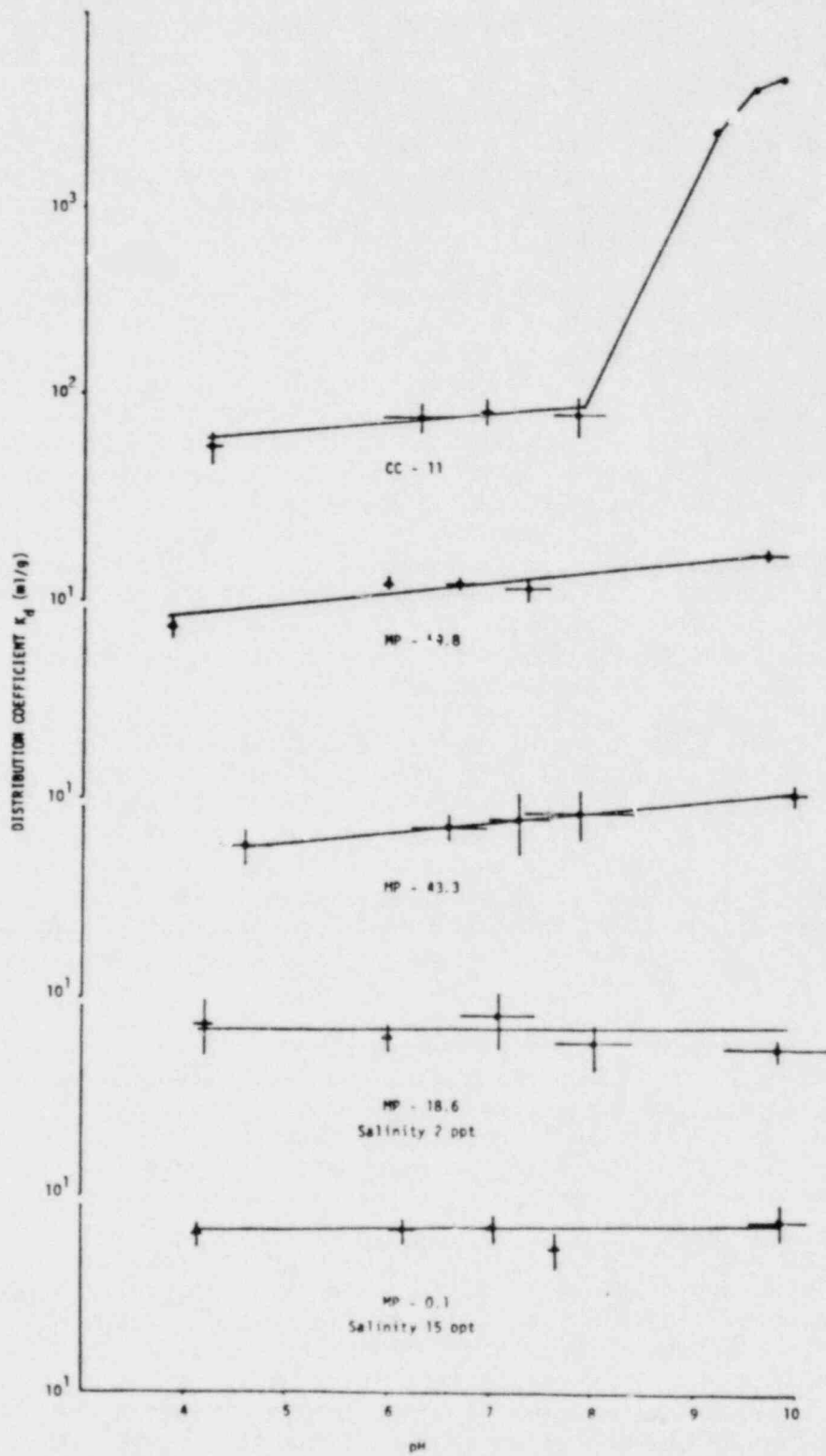


Figure 3. Effect of pH on adsorption distribution coefficient of  $^{90}\text{Sr}$  at Hudson River and Cattaraugus Creek stations.

Table 5. Effect of pH on  $K_d$  values for Hudson River Stations and at Cattaraugus Creek, New York.

Station	Time (days)	pH	$K_d$ (ml/g)
Hudson River MP 0	1	~4	74.9 + 8.7
	4		53.8 + 7.8
	10		NS
	Ave	4.11 + .02	64.3 +11
	1	~6	68.4 + 8.4
	4		62.7 + 8.1
	10		NS
	Ave	6.18 + .13	65.5 +11
	1	~7	80.8 + 8.9
	4		56.5 + 8.9
	10		NS
	Ave	7.04 + .02	68.6 8.5
	1	~8	53.9 + 22
	1		76.1 + 8.5
	1		54.6 + 7.7
	4		53.4 + 7.9
	4		49.8 + 7.4
	4		52.5 + 8.4
	10		NS
	10		NS
10		NS	
Ave	7.68 + .09	53.6 +11	
1	~10	89.3 + 3.4	
4		57.3 + 8.8	
10		NS	
Ave	9.87 + .33	73.2 +15	
MP 18.6	1	~4	97.1 + 9.3
	4		49.6 + 7.5
	10		NS
	Ave	4.20 + .09	73.3 + 23
	1	~6	72.3 + 7.6
	4		53.1 + 8.6
	10		NS
	Ave	6.00	62.7 + 9.6
	1	~7	103 +10.2
	4		54.6 + 8
	10		NS
	Ave	7.09 + .26	78.8 +24
	1	~8	53.8 + 7.9
1		64.4 + 8.3	
1		81.6 + 9.6	
4		49.7 + 8.0	

Table 5. Effect of pH on  $K_d$  values for Hudson River Stations and at Cattaraugus Creek, New York - continued.

Station	Time (days)	pH	$K_d$ (ml/g)
MP 18.6	4		51.1 + 7.1
	4		40.2 + 7.6
	10		NS
	10		NS
	10		14.6 + 9
	Ave	8.02 + .38	56.8 + 14.4
	1	~10	61.3 + 7.7
	4		48.0 + 8.1
	10		22.7 + 9.5
	Ave	9.84 + .54	54.6 + 6.7
	MP 43.3	1	~4
4			50.0 + 10.4
10			15.0 + 9.1
Ave		4.60 + .23	57.8 + 11.0
1		~6	77.6 + 9.6
4			64.3 + 11.0
10			16.9 + 9.3
Ave		6.60 + .38	70.9 + 9.4
1		~7	104 + 9.3
4			50.3 + 8.0
10			29.9 + 10.0
Ave		7.30 + .30	77.1 + 26
1		~8	85.0 + 9.8
1			126.0 + 11.4
1			89.9 + 10.0
4			56.1 + 8.8
4			82.0 + 11.3
4			70.1 + 10.9
10			36.1 + 10.3
10			35.3 + 10.2
10		30.7 + 8.1	
Ave	7.90 + .56	84.8 + 23	
1	~10	115 + 10.5	
4		93.5 + 12.2	
10		49.5 + 10.7	
Ave	10.0 + .11	104 + 11	
MP 59.8	1	~4	76.2 + 7.4
	4		83.1 + 10.1
	10		64.3 + 11.3
	Ave	3.89 + .01	74.5 + 9.5
	1	~6	119 + 9.5

Table 5. Effect of pH on  $K_d$  values for Hudson River Stations and at Cattaraugus Creek, New York - continued.

Station	Time (days)	pH	$K_d$ (ml/g)
MP 59.8	4		132 + 12.7
	10		116 + 13.5
	Ave	6.0 +0.0	122 + 8.5
	1	-7	120 + 12.5
	4		123 + 11.8
	10		125 + 13.4
	Ave	6.70 + .13	122 + 3
	1	-8	108 + 11.5
	1		119 + 9.9
	1		86.7 + 10.5
	4		135 + 13.7
	4		123 + 11.2
	4		108 + 10.8
	10		120 + 13.5
	10		113 + 13.2
	10		106 + 13.7
	Ave	7.38 + .22	113 + 13.5
	1	-10	165 +
	4		176 + 14.4
	10		172 + 15.4
Ave	9.75 + .18	171 + 6.0	
<u>Cattaraugus Creek</u>			
CC-11	1	-4	138 + 9.4
	4		60.9 + 16.5
	10		60.2 + 11.1
	Ave	4.28 + .08	60.5 + 11
	1	-6	88.9 + 9.1
	4		97.1 + 12.1
	10		69.6 + 11.8
	Ave	6.31 + .36	85.2 + 14
	1	-7	91.8 + 8.7
	4		101 + 12.5
	10		77.4 + 0
	Ave	6.98 + .07	90.0 + 12
	1	-8	126 + 13.0
	1		89.8 + 6.2
	1		77.5 + 10.3
	4		78.2 + 11.6
	4		95.5 + 12.5
	4		74.4 + 11.4
	10		68.8 + 12.3
	10		76.3 + 12.1



Table 5. Effect of pH on  $K_d$  values for Hudson River Stations and at Cattaraugus Creek, New York - continued.

Station	Time (days)	pH	$K_d$ (ml/g)
CC-11	10		89.9 + 12.1
	Ave	7.88 + .26	86.3 + 17
	1	9.23	2430 +108
	4	9.62	4090 +156
	10	9.90	4570 +189

not give sufficient sample to weigh accurately. Thus, volume measurements have been required. As the suspended sediment concentration becomes smaller a volume measurement is the most practical method for sampling. If the coagulation of particles leads to non-homogeneity in the vessel at sampling time, the low  $K_d$  values at later time (10 days) could be explained.

The clay colloid stability decreases with increasing ionic strength. Thus, at stations MP 18.6 and MP 0.1, where sea water is present, coagulation of the particles is more important than at the upstream stations MP 43.3 and MP 59.8. The possibility of adding  $\text{Na}^+$  or  $\text{H}^+$  ions to adjust the pH may also desorb  $^{85}\text{Sr}$  from the particles at the later times during the experiment and competition for precipitation and ion exchange occurs. Thus, an average value for  $K_d$  has been used to determine the pH effect for the experimental samples.

Experiments were also made to determine the desorption  $K_d$  values for  $^{85}\text{Sr}$ . However, because of the low  $K_d$  values for sorption and the ion exchange and coagulation problems, no desorption  $K_d$  values were measurable, except for Lake Michigan, Clinch River, and one experiment at MP 43.3 of the Hudson River.

### 3.3 Effect of Sediment Concentration

As for pH, some experiments have been reported previously on the effects on the  $K_d$  values of sediment concentration ranging from 100 to 1000 mg/l of  $^{241}\text{Am}$  in sterile and unsterilized systems from Lake Washington (14). We have completed and analyzed an experiment on the effects of sediment concentrations ranging from 16 mg/l to 340 mg/l in the Lake Michigan sediment-water system (16). For the radionuclides  $^{241}\text{Am}$ ,  $^{237}\text{Pu}$ ,  $^{106}\text{Ru}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  there is a significant increase in  $K_d$  value at lower sediment concentrations.

Sediment-water systems from the Hudson River and Cattaraugus Creek were used to determine the effect of sediment concentration on the  $K_d$  values of  $^{85}\text{Sr}$  Table 6. The distribution coefficient  $^{85}\text{Sr}$  increases at lower sediment concentrations by factors of 2 to 40 in those river samples at experimental sediment concentrations of 1000 and 200 mg/l. These results agree with results reported by Aston and Duursma (20); they showed a rapid increase in  $K_d$  values of  $^{65}\text{Zn}$ ,  $^{106}\text{Ru}$ , and  $^{137}\text{Cs}$  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. For example, as shown in Section 1.1.1, the distribution coefficients for environmental  $^{90}\text{Sr}$  in the Clinch river sediment-water system increases by a factor of 7 at sediment concentrations of from 55 to 9 mg/l (8).

Table 6. Adsorption distribution coefficients for  $^{85}\text{Sr}$  as a function of sediment concentration.

Station/concentration	Average $K_d$ (SD) ml/g			
	n*	200 mg/l	n*	1000 mg/l
<u>Hudson River</u>				
MP 0.1		---	9	17.2 (1.5)
MP 18.6	6	34.8 (5.3)	9	16.7 (1.6)
MP 43.3	6	73.7 (10.5)	6	41.9 (2.9)
MP 59.8			9	107.7 (4.8)
<u>Cattaraugus Creek</u>				
CC 11	8	62.2 (6.2)	9	16.6 (1.0)

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

### 3.4 Dialysis Experiments

Two dialysis experiments have been made 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, (14,15).

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  $\mu$ m 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  $\mu$ m size fraction.

In each experiment, 700 ml of filtered (<0.22  $\mu$ m) 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. 2).

At each sampling time, a 5 ml sample was removed from each chamber and filtered through a 0.22  $\mu$ m 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<sub>3</sub>.

A dialysis experiment was set up to investigate the behavior of <sup>85</sup>Sr and <sup>237</sup>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 Table 7.

Table 7. Relative concentrations of  $^{85}\text{Sr}$  measured during a 15-day dialysis experiment.

Time		Spiked chamber cpm/ml + 2SD	Dialysis chambers			
			Blank cpm/ml + 2SD	Plankton cpm/ml + 2SD	Sediment cpm/ml + 2SD	Detritus cpm/ml + 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)

Strontium-85 (Table 7) occurs almost entirely in the soluble phase. The concentration of particulate  $^{85}\text{Sr}$  never exceeds 3% of the total  $^{85}\text{Sr}$  concentration. In most cases, the concentration listed as particulate probably corresponds to  $^{85}\text{Sr}$  adsorbed on the filter rather than the actual particulate phase. The concentration of soluble  $^{85}\text{Sr}$  is plotted in Fig. 4. 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  $^{60}\text{Co}$  and  $^{137}\text{Cs}$ .

#### 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 8. Both sediments and detritus show greater  $K_d$  values in the constant shaking experiments than in the dialysis experiments for  $^{85}\text{Sr}$ ; the  $K_d$  measurements in the dialysis experiments is at least four times less than the adsorption  $K_d$  values.

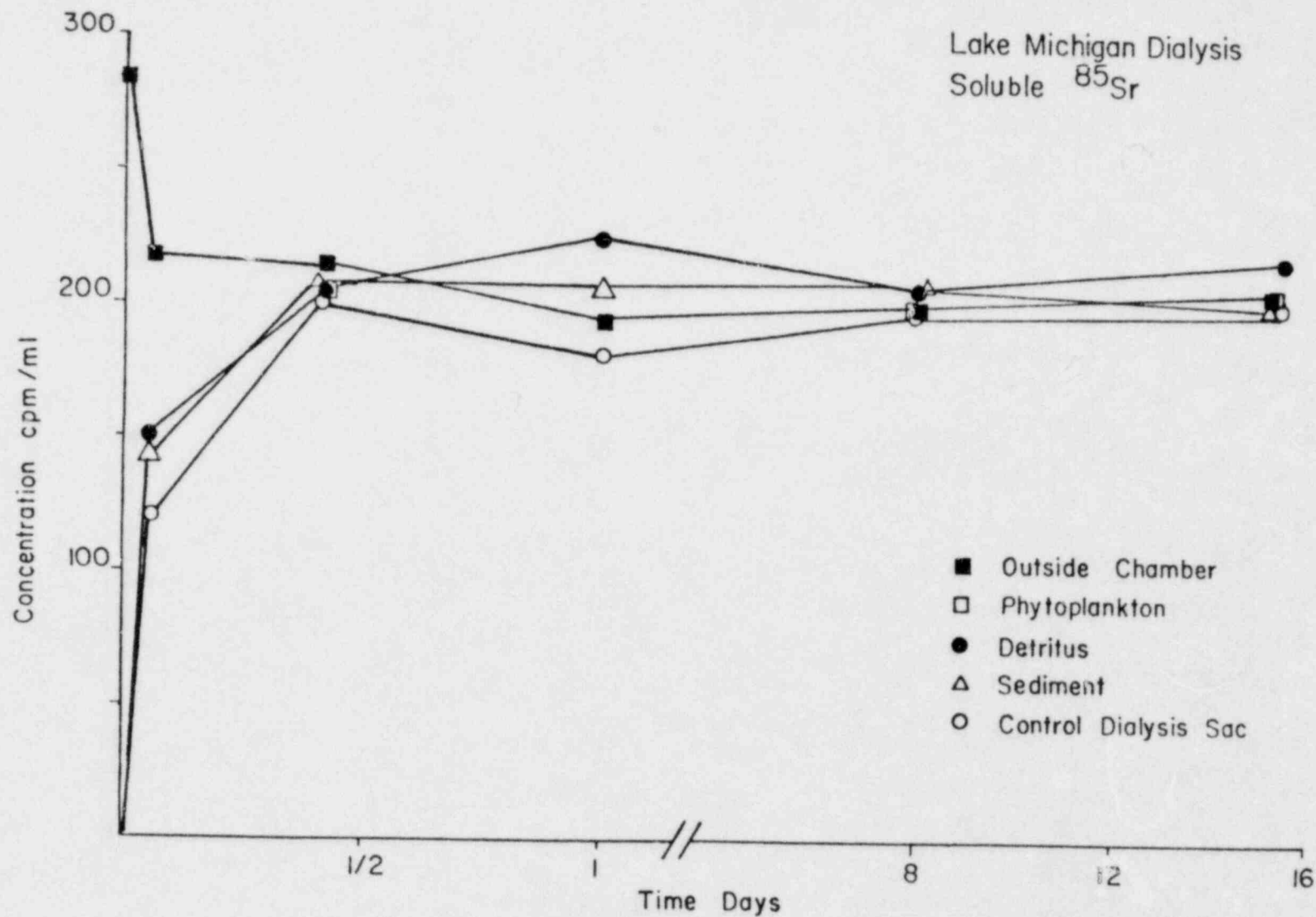


Figure 4. Concentration of soluble  $^{85}\text{Sr}$  as a function of time in the various compartments in a dialysis experiment for the Lake Michigan sediment-water system. The particulate fraction was below detection limits.

Table 8. Comparisons of  $K_d$  values for adsorption of  $^{85}\text{Sr}$  by the constant shaking method with  $K_d$  values for adsorption by the dialysis method in Lake Michigan sediment-water system.

Time (days)	Sediment		Detritus	
	Dialysis $K_d$ (ml/g)	Adsorp. $K_d$ (ml/g)	Dialysis $K_d$ (ml/g)	Adsorp. $K_d$ (ml/g)
1	83.3 (7.5)*	---	213 (13)	---
8	21.8 (4.9)	82.2 (7.0)	200 (6)	75.2 (4.1)
15	24.7 (7.4)	---	18.2 (4.6)	---

\* Average values of distribution coefficients  $K_d$  (SD).



## 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  $^{85}\text{Sr}$  to greater than  $10^5$  for  $^{241}\text{Am}$ . The values for  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  are greater in freshwater systems than in the marine sediment-water systems.
2. The desorption  $K_d$  values range from 4 to 17 times greater than the adsorption  $K_d$  values for  $^{85}\text{Sr}$  in the freshwater-sediment and marine water-sediment systems. The lowest desorption  $K_d$  value is found for  $^{85}\text{Sr}$  and the highest is found for  $^{241}\text{Am}$ ; 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  $^{85}\text{Sr}$ . At Hudson river stations MP 0.1 and MP 18.6 there was no pH effect. These stations were effected by marine water. At the upper river stations a slow increase in  $K_d$  values was found to pH 10. At Cattaraugus Creek, the  $K_d$  value increased gradually to pH 8.0 and then a factor of 24 increase occurred to pH 9.2. The  $K_d$  value of 4570 ml/g was found at pH 9.9.
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. 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 mechanisms 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.

5.0 SUMMARY -  $^{85}\text{Sr}$ 

A summary of the adsorption and desorption  $K_d$  values for  $^{85}\text{Sr}$  is shown in Table 4 for the several stations measured. The values range from non-significant in the estuarine areas where the sodium concentration is high, Hudson River, MP 0.1, to 124 for the Clinch River. The desorption  $K_d$  value is a factor of 17 greater than the adsorption  $K_d$  value for Lake Michigan and a factor of 6 to 7 times greater for the river stations. The pH range of 4 to 10 has no effect on the  $K_d$  value at the estuarine stations of the Hudson River; however, the  $K_d$  value increases with pH at the upper river stations. At Cattaraugus Creek, the  $K_d$  value increases gradually from pH 4 to 8 and then increases abruptly by a factor of 25 at pH 9.2. The  $K_d$  values increase with decreasing suspended sediment concentration. Laboratory measurements of  $K_d$  values at the normal suspended sediment concentration ( $\sim 200$  mg/l) may be a factor of 10 to 100 times lower than the environmental  $K_d$  values for river suspended sediment concentrations of 9-185 mg/l. The data of Parker et al. (8) were used and represent natural suspended particles including plankton, and organic detritus as well as the inorganic sediments generally used in laboratory  $K_d$  studies.

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