NUREG/CR-1852
Vol. 2
RE

Distribution Coefficients for Radionuclides in Aquatic Environments

Adsorption and Desorption Studies of ^{137}Cs

Manuscript Completed: December 1980
Date Published: May 1981

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

College of Fisheries
University of Washington
Seattle, WA 98195

**Prepared for
Division of Health, Siting and Waste Management
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555
NRC FIN B5749**

8106010536

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. 38 pp.

Distribution Coefficients for Radionuclides in Aquatic Environments. II. Studies in 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. 70 pp.

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. 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. of Washington, Topical Report NUREG/CR1852, Vol. 1. 27 pp.

NRC FORM 335
(7-77)

U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

1. REPORT NUMBER *(Assigned by DDC)*
NUREG-CR-1852, Vol. 2

4. TITLE AND SUBTITLE *(Add Volume No., if appropriate)*
Distribution Coefficients for Radionuclides in Aquatic Environments
Subtitle: Adsorption and Desorption Studies of ^{137}Cs

2. *(Leave blank)*

3. RECIPIENT'S ACCESSION NO.

7. AUTHOR(S)
W.R. Schell, A.L. Sanchez, and T.H. Sibley

5. DATE REPORT COMPLETED
MONTH December YEAR 1980

9. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS *(Include Zip Code)*
College of Fisheries
University of Washington
Seattle, WA 98195

DATE REPORT ISSUED
MONTH May YEAR 1981

6. *(Leave blank)*

8. *(Leave blank)*

12. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS *(Include Zip Code)*
U.S. Nuclear Regulatory Commission
Division of Health, Siting and Waste Management
Office of Nuclear Regulatory Research
Washington, DC 20555

10. PROJECT/TASK/WORK UNIT NO.

11. CONTRACT NO.
FIN B5749

13. TYPE OF REPORT
Topical

PERIOD COVERED *(Inclusive dates)*

15. SUPPLEMENTARY NOTES

14. *(Leave blank)*

16. ABSTRACT *(200 words or less)*

This report investigates the several parameters important to measuring the distribution coefficients for ^{137}Cs in natural sediment-water systems. The samples of water and sediment for the experimental measurements were collected from several sites in the U.S.A. including the Hudson River, Clinch River, Cattaraugus Creek, Lake Michigan, Skagit Bay, Columbia River, and Lake Washington. The results of the experiments have given sorption distribution coefficients for ^{137}Cs of from 17 to 1700 ml/g; the desorption distribution coefficient is a factor of 10 greater. The lowest values were found in marine systems and the highest values in freshwater. Organic compounds of different complexing functional groups added to the experimental system, and the change in pH between 4 and 10 had no effect on the distribution coefficients measured. Sediment concentration and salinity changes influenced strongly the experimental distribution coefficients measured.

17. KEY WORDS AND DOCUMENT ANALYSIS

Distribution Coefficients, Dialysis Experiments, ^{137}Cs, Sediment Concentration, pH

17a. DESCRIPTORS

17b. IDENTIFIERS/OPEN-ENDED TERMS

18. AVAILABILITY STATEMENT
Unlimited

19. SECURITY CLASS *(This report)*
Unclassified

21. NO. OF PAGES

20. SECURITY CLASS *(This page)*
Unclassified

22. PRICE
$

ABSTRACT

As a result of the nuclear weapons production and the nuclear industry, radionuclides have entered the environment and thus have participated in the biogeochemical cycle of elements. Because of this radioactivity they pose a potential hazard as well as serve as tracers within the ecosystem. An important part of the biogeochemical cycle is the uptake of the radionuclides on particulate matter and this uptake can be measured quantitatively by the distribution coefficients between soluble and particulate fractions.

This report investigates the several parameters important to measuring the distribution coefficients for ^{137}Cs in natural sediment-water systems. The samples of water and sediment for the experimental measurements were collected from several sites in the U.S.A. including the Hudson River, Clinch River, Cattaraugus Creek, Lake Michigan, Skagit Bay, Columbia River and Lake Washington. The results of the experiments have given sorption distribution coefficients for ^{137}Cs of from 17 to 1700 ml/g; the desorption distribution coefficient is a factor of 10 greater. The lowest values were found in marine systems and the highest values in freshwater. Organic compounds of different complexing functional groups added to the experimental system, and the change in pH between 4 and 10 had no effect on the distribution coefficients measured. Sediment concentration and salinity changes influenced strongly the experimental distribution coefficients measured.

TABLE OF CONTENTS

Page

LIST OF FIGURES

LIST OF TABLES

DISTRIBUTION COEFFICIENTS FOR RADIONUCLIDES IN AQUATIC ENVIRONMENTS

Adsorption and Desorption Studies of ^{137}Cs.

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 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

---

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

one environment to make predictions about new environments. Furthermore, 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. Cesium-137 ($t_{1/2}$ of 30 years) is foremost among these radionuclides due to its production from the fissioning of ^{239}Pu and ^{235}U. (Approximately 6% of the total products resulting from ^{239}Pu and ^{235}U fission produce ^{137}Cs.) Fallout ^{137}Cs from nuclear weapons tests is used as a tracer in determining sedimentation rates in lakes and reservoirs since it adheres strongly to clay particulate matter in fresh water (1). It 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. Recent measurements of sediment and overlying water at the German coast revealed distribution coefficients for ^{137}Cs of (70-760) ml/g with a mean value of 330 ml/g (4). Heatherington and Jefferies (5) show that the uptake of ^{137}Cs in estuarine sediments decreases with increasing particle size of silt and fine sand supporting the view that uptake is a surface adsorption process. Patel et al. (6) also show that the surface processes dominate in ^{137}Cs uptake by sediments. They find that the ^{137}Cs desorption (i.e., the loss from sediment) increases with increasing salinity. They relate this process to the available binding sites on the clay mineral and the replacement of Cs^+ by K^+, Na^+, and Mg^{++} in the increasingly saline water.

In Clinch River sediments, Pickering (7) shows that ^{137}Cs is not removed by leaching after it becomes associated with sediments. The sediment types used were the clay-silt fraction which had 35% mica and clay minerals and 65% quartz; the cation exchange capacity was 33-37 meq/100 g. El Prince et al. (8) studied ^{137}Cs near water cooled reactors on the Savannah River and found a distribution coefficient range of 18-130 ml/g at 27°C. The adsorption of ^{137}Cs by sediments follows a 1/T°K relationship (i.e., the adsorption K_d value decreases with increasing temperature). The mechanisms for the desorption of ^{137}Cs with increasing temperature is thought to be due to the competition of H_3O^+ for the Cs^+ at the alumino-silica matrix sites of adsorption.

In the cooling pond system at the Denora Station of the Savannah River Nuclear Plant, South Carolina, high distribution coefficient values for ^{137}Cs of 2 to 4 x 10^4 ml/g were measured on the small suspended particle (5-80 μm)-water system (9). Huff and Kruger (10) also found high distribution coefficient values ($2.5 \pm 1.0 \times 10^4$ ml/g) for fallout ^{137}Cs on suspended sediment-water in the Saratoga watershed of California during six cyclonic storms in 1965 and 1966. This study was designed to investigate hydrological transport of fallout particles through overland flow and erosion in a semi-arid region. Because of the high erosion rate, suspended sediment concentrations were 295 to 5,297 mg/l and equilibrium between sediment and water probably was not reached. No information is reported on the particle sizes or composition of the sediments but one would expect that fine clay particles were being transported by the precipitation in this erosion study.

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 $Cs^+ > K^+ > Na^+ > Li^+ > Ba^{+2} > Sr^{+2} > Ca^{2+} > Mg^{2+}$. For example, Cs^+ would be held more strongly to the clay than potassium and may therefore replace K^+ in a clay-water system.

In the biological system, a review by Davis (12) of cesium relationships to potassium shows that biological organisms may discriminate between the two elements. In potassium deficient water, marine algae cells readily take up cesium. However, when potassium is in sufficient concentrations cesium is not preferentially taken up. In dead cells, the differential permeability of potassium and cesium does not occur and concentration factors of 420 have been found in Chlorella cells (12). When trace amounts of ^{137}Cs are introduced into freshwater environments, they are readily accumulated and transferred through the food web.

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}Sr, ^{106}Ru, ^{137}Cs, ^{60}Co, and ^{244}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}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 (13). Results for preliminary experiments using ^{65}Zn and ^{137}Cs were compared to published results (14) 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 uptake of soluble radionuclides by sediments and phytoplankton. Results from the first year of research are included in NUREG/CR0801 (15).

During FY 77-78 the project was extended to include new sediment types and additional radionuclides. 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. We also evaluated the sedimentation technique (13) 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 (16). 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}Fe, ^{60}Co, ^{65}Zn, ^{137}Cs, ^{204}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 FY 1978-79, adsorption distribution coefficients for ^{85}Sr, ^{106}Ru, ^{137}Cs, ^{241}Am and ^{237}Pu were determined in constant shaking experiments using filtered water and <63 m-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}Ru, ^{137}Cs and ^{241}Am were determined for sediment-water systems from Lake Michigan and the three stations in the Hudson River estuary. For ^{85}Sr and ^{237}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}Sr, ^{106}Ru,

^{137}Cs, ^{237}Pu and ^{241}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/CR0803 (17).

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}Cm were measured for the Hudson River estuary and the effect of pH and sediment concentration on the ^{244}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 will describe the data obtained from distribution coefficient measurements of ^{137}Cs in 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 (15,16,17) and will be discussed briefly below. The thin layer and suspended sediment techniques were investigated during the first year but reproducability was not found sufficient so the constant shaking method was used in all the later studies. See NRC 1852, Volume 1 for the complete methodology used in these studies (18).

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 during the past year. 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 μm 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

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^2/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)				

characteristics that are of particular importance in determining the adsorptive properties of a sediment-water system are the surface area, cation 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 ^{60}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 $\sim$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 original Hudson River sediment samples

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

Particle		Percentage in sediments		
ϕ Size	Diameter mm	Lake Washington	Lake Nitinat	Marine
-1	2	0	0.11	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	3.90	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^3		2.38	2.55*	2.56*
Carbon and nitrogen contents				
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	8.30	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

*Filtered samples were passed through 0.3μM Millipore filters.

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 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.

To compare the results obtained with freeze-dried samples to those from untreated samples, a second series of sediment and water samples were collected from the Hudson River estuary in August and September 1979. These samples were stored under ice or at 4°C until used in the experiments. Results from adsorption experiments, to be presented in future reports, indicate that the K_d values for ^{137}Cs are 2 to 4 times lower for freeze-dried compared to untreated samples.

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 (13,18,19,20). Briefly, the method consists of:

1. Adding the radionuclides to a known volume of filtered (0.45 μm or 0.22 μm) water sample and adjusting the pH to the initial pH of the water sample with 0.2 M NaOH.

2. Adding sediments previously seived to less than 63 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.

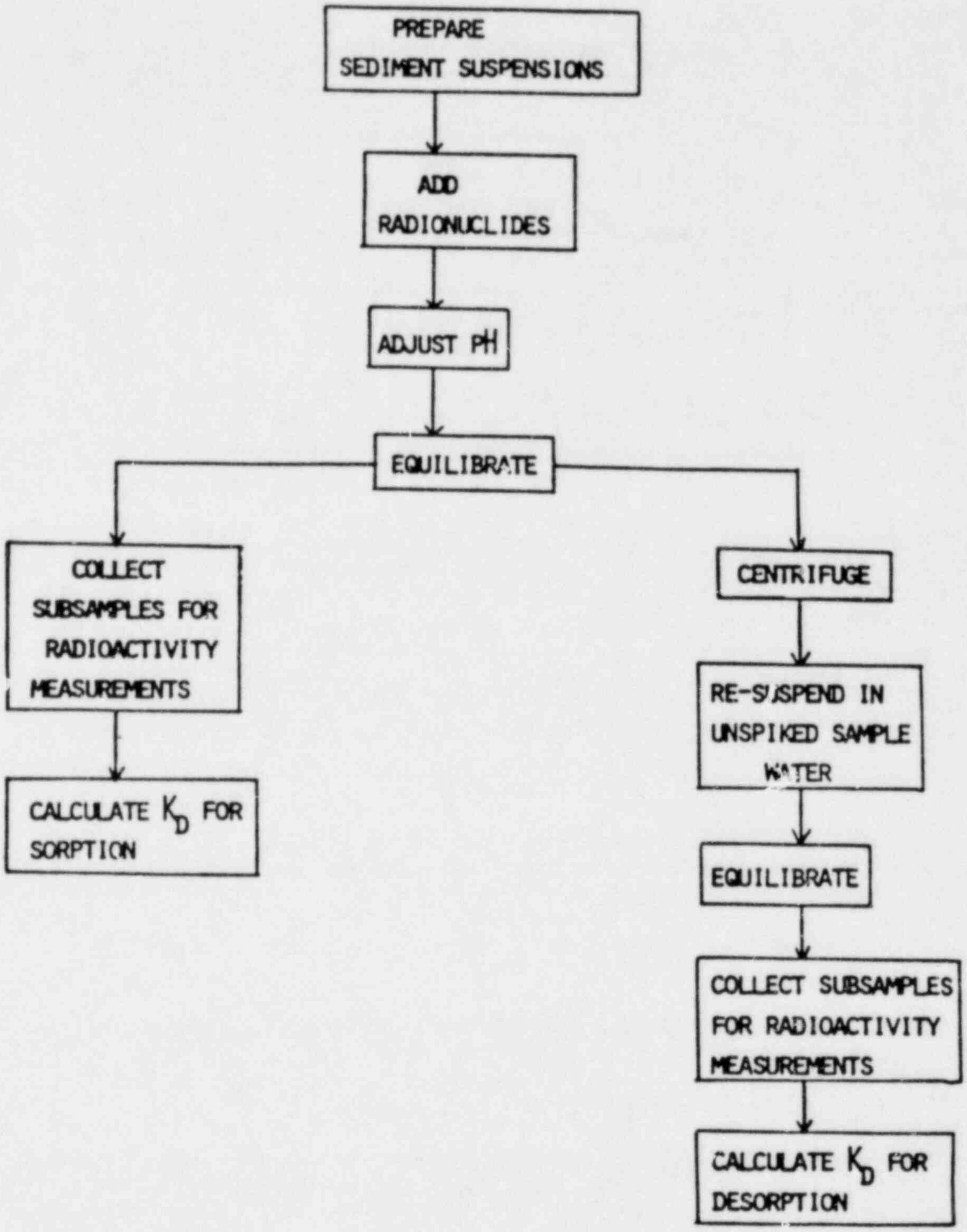


Figure 1. Sequential steps involved in obtaining adsorption and desorption distribution coefficients with the constant shaking technique.

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 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 (2) and Dawson and Duursma (22). In all our experiments, the outside compartment contains filtered (0.22 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.

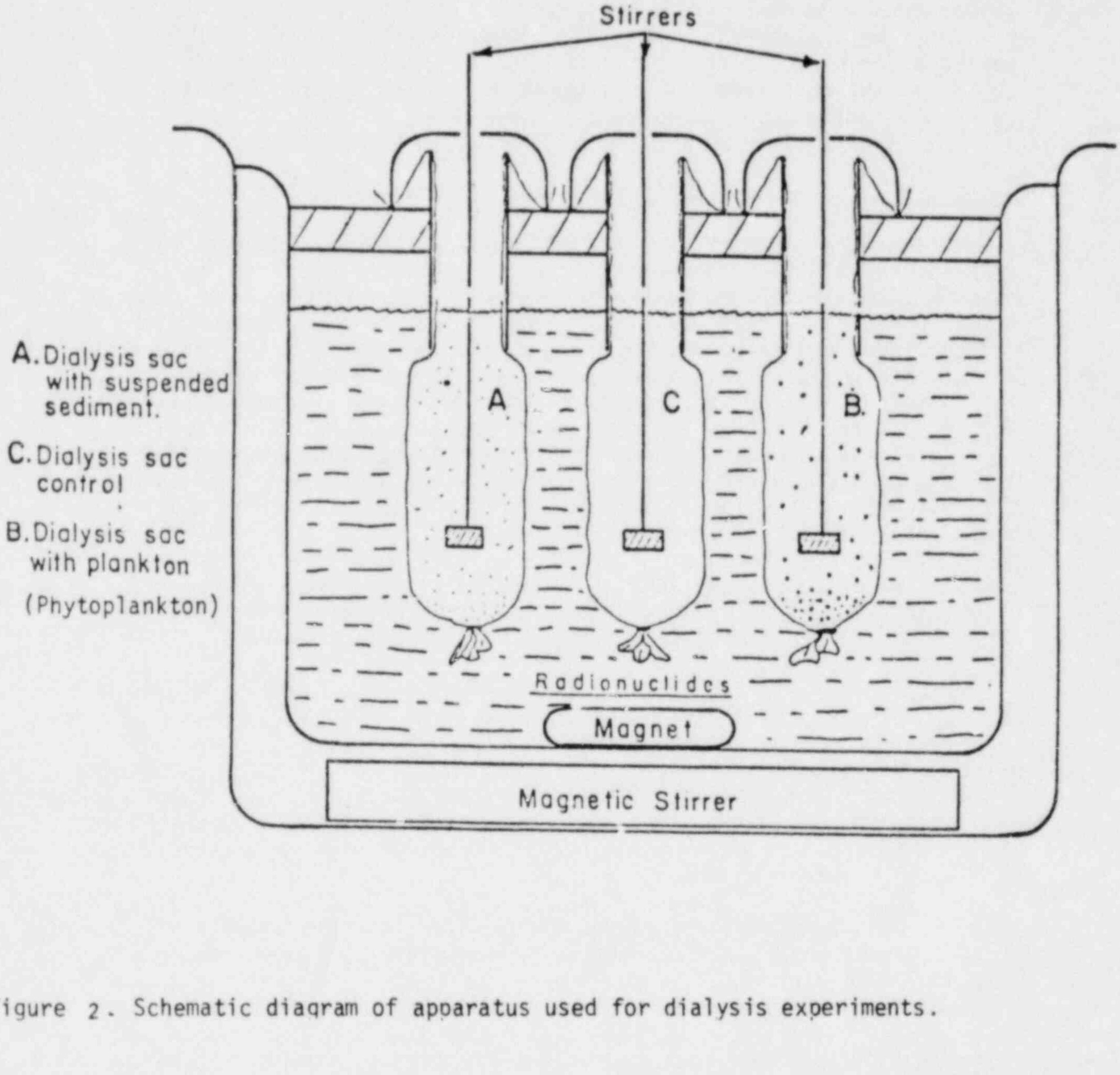


Figure 2. Schematic diagram of apparatus used for dialysis experiments.

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 x 10^6 cells/ml) was centrifuged at 6000 rpm for 15 min and re-suspended in 32 ^{0}/oo NaCl solution. This process was repeated four times to remove the nutrient media and the final rinse was in 32 ^{0}/oo 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 montmorillinite, 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 80^0 C, the Zostera was ground in a ball mill and sieved. For the experiments, a weighed amount of the <63 m size fraction was suspended in filtered seawater to obtain a stock suspension. A known volume of this suspension was then added to a dilaysis 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}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 analysis.

3.0 RESULTS AND DISCUSSION

3.1 Sorption and Desorption 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 m sediment fraction for cores from the Skagit River Estuary and Saanich Inlet.

Adsorption of ^{137}Cs

Distribution coefficients have been calculated for ^{137}Cs for sediment-water systems from the Skagit River Estuary, Saanich Inlet, Lake Michigan, and for calcium montmorillonite. An example of the approach to equilibrium of ^{137}Cs between the particulate and soluble fractions is shown by the concentration of ^{137}Cs in suspended sediments as a function of time for the sediment-water system of the Skagit River estuary, Fig. 3. These results are given in Table 4. The distribution coefficients for ^{137}Cs are highest for the Lake Michigan sediment-water system and lowest for calcium montmorillonite-Skagit Estuary water system. The high K_d values for ^{137}Cs 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 (100 meq/ 100 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 ^{137}Cs.

The data for ^{137}Cs from Cattaraugus Creek (Fig. 4) 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 from Saanich Inlet but not from Skagit River estuary. Similar results are found for ^{106}Ru during experiments with the Lake Michigan sediment-water systems.

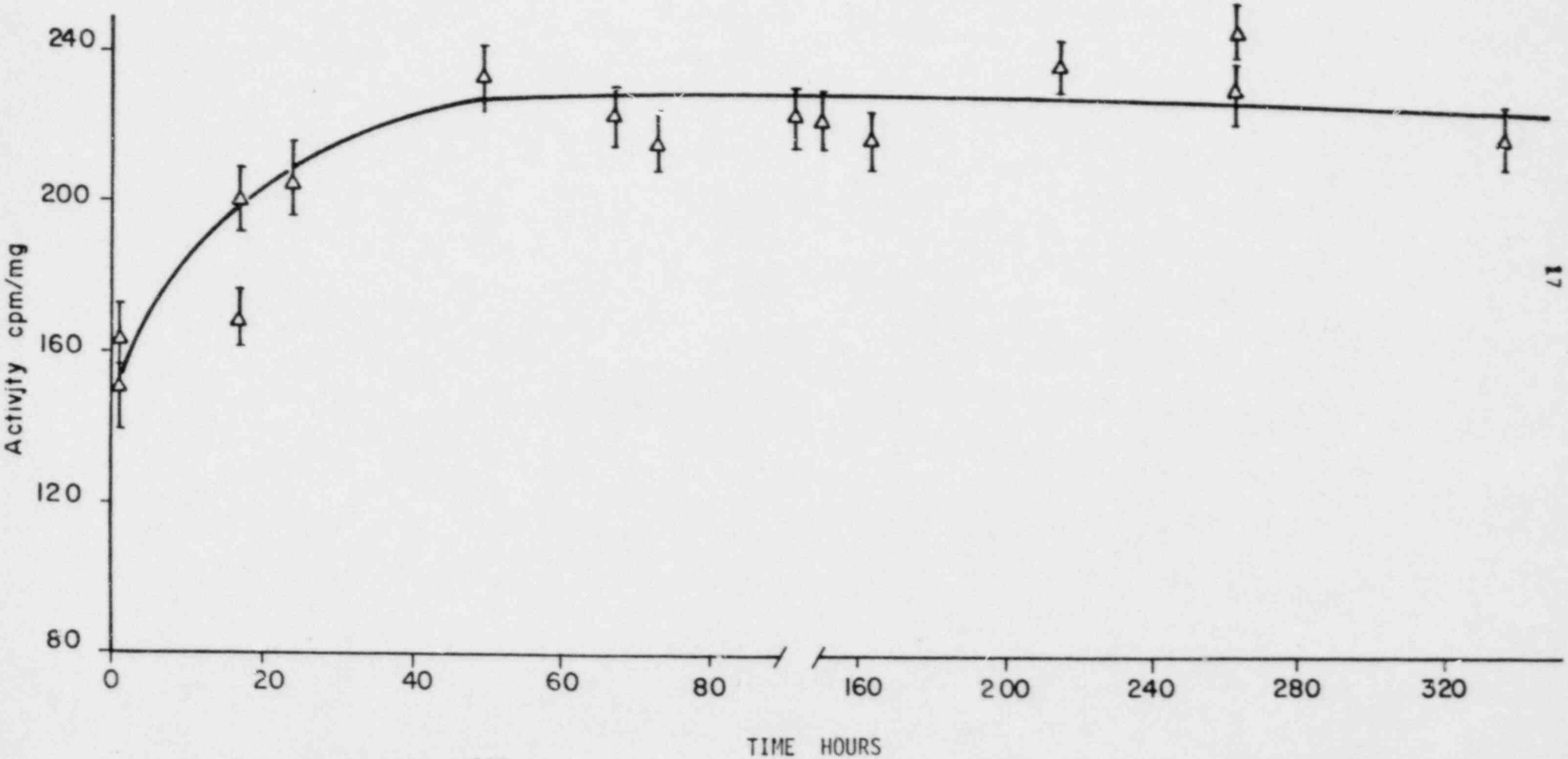


Figure 3. Concentration of ^{137}Cs in suspended sediments as a function of time for Skagit River estuary sediment-water systems. Errors are 2SD of propagated counting errors.

Table 4. Summary of adsorption-desorption distribution coefficients for ^{137}Cs for different sediment-water systems.

	Average K_d (SD) ml/g x 10^{-2}				
Sample type	n$^{(a)}$	Adsorption	n$^{(a)}$	Desorption	Treatment
Lake Michigan	6	5.09 (0.31)	6	55.7 (0.06)	Frozen
Clinch River	9	13.6 (0.40)			Fresh
Hudson River					
MP 59.8	5	16.6 (1.30)			Fresh
MP 43.3	9	4.01 (0.20)	9	36.5 (5.2)	Freeze-dried
"	6	17.40 (0.80)			Fresh
MP 18.6	9	8.78 (0.71)	9	33.4 (6.9)	Freeze-dried
"	7	5.82 (0.70)			Fresh
MP 0.1	9	3.56 (0.21)	9	17.9 (4.7)	Freeze-dried
"	6	6.36 (0.63)			Fresh
Cattaraugus Creek	3	6.0			Fresh
Skagit Bay -1	9	1.90 (0.15)			Fresh
" -4	5	3.53 (0.07)			Fresh
Columbia River	3	0.17 (.02)$^{(b)}$			Fresh
Saanish Inlet	3	0.61 (0.24)			Fresh
Lake Nitinat	6	0.52 (0.03)$^{(b)}$			Fresh
Sinclair Inlet	9	1.24 (0.09)			Fresh
Lake Washington	4	0.50 (.04)$^{(b)}$			Fresh
Organic detritus					
Lake Michigan	1	20.6$^{(c)}$			Dried, ground
Skagit Bay	1	<1.0$^{(c)}$			Dried, ground
Montmorillonite	6	0.65 (0.07)			

(a) n is the number of experimental vessels from which K_d was calculated.
(b) Values obtained by thin layer technique.
(c) Values obtained by dialysis technique.

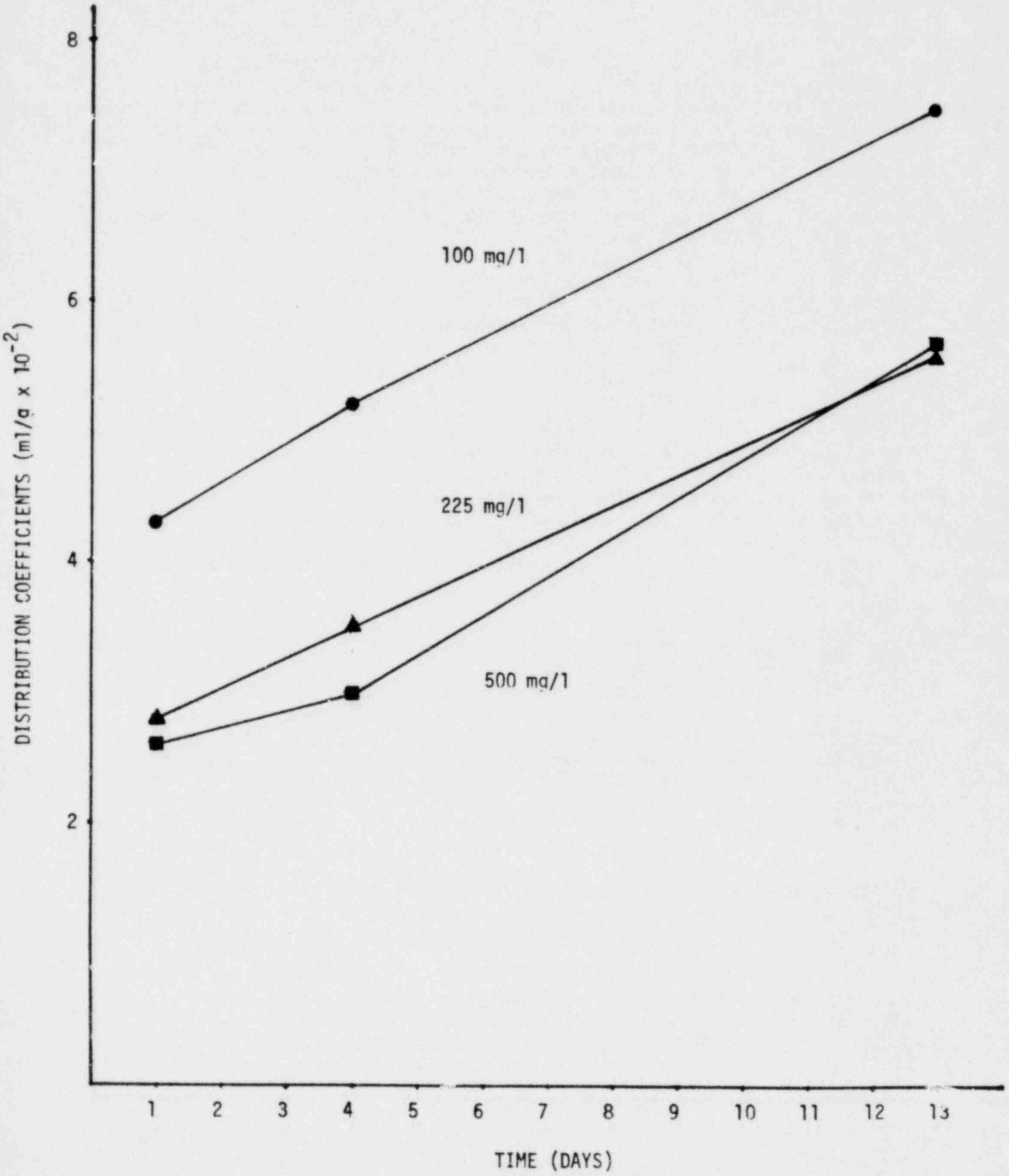


Figure 4. Effect of "equilibrium" time and suspended sediment concentration on the distribution coefficient values for ^{137}Cs in the Cattaraugus Creek sediment-water system.

Table 4 shows the K_d values of ^{137}Cs for Clinch River, Sinclair Inlet, and the Hudson River Estuary. 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 ^{137}Cs 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 sediments from MP 18.6 may have a higher than usual affinity for ^{137}Cs which could mask any salinity effects.

Desorption of ^{137}Cs

Experiments have been completed to determine the desorption K_d values of ^{137}Cs 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 $\sim 4^{o}C$.

For evaluating the desorption of ^{137}Cs from Lake Michigan sediments, a different approach was taken: Three sets of samples corresponding to three different sorption times for the radionuclides were 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-sampled for radioactivity measurements were collected after 1 hour, 24 hours, and 96 hours of desorption.

The results of the desorption distribution coefficient experiments also are shown in Table 4. The desorption K_d for ^{137}Cs 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.

Summary of Adsorption 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 ^{137}Cs the K_d values for desorption were higher than the adsorption K_d values by at least a factor of 10. These results suggest that under these experimental conditions, sorption is not completely reversible. For an estuarine sediment-water system, measurements were made of the approach to equilibrium K_d values as a function of time. Figure 2 shows the concentration of ^{137}Cs accumulated by sediments as a function of time for the Skagit River Estuary Core. Equilibrium appears to be reached after about 40 hours. 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 radionuclides from suspended or bed sediments, but should be appropriate for the initial removal from the water.

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: $^{85}\text{Sr} < {}^{137}\text{Cs} < ({}^{106}\text{Ru or } {}^{237}\text{Pu}) < {}^{241}\text{Am}$ (18).

3.2 Effect of Sediment Concentration on K_d Values

The data for ^{137}Cs from Cattaraugus Creek (Fig. 4) indicate that the K_d values incease 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 from Saanich Inlet.

An experiment on the effects of sediment concentration on the K_d values for ^{137}Cs has been completed in the Lake Michigan sediment-water system. The sediment concentrations ranged from 16 mg/l to 340 mg/l. Results of this experiment are shown in Figure 5. For the radionuclide there is a significant increase in K_d value at lower sediment concentrations. The apparently sharp increase in K_d values of ^{137}Cs at sediment concentraton below $\sim$50 mg/l may be an experimental artifact resulting from sorption of these radionuclides to the filters.

Sediment-water systems from Catarraugus Creek were used to determine the effect of sediment concentrations on the K_d values of ^{137}Cs (Figure 4). The distribution coefficients of ^{137}Cs incrase at lower sediment concentrations, especially between 100 and 250 mg/l. These results agree with results reported by Aston and Duursma (20); they showed a rapid incrase 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

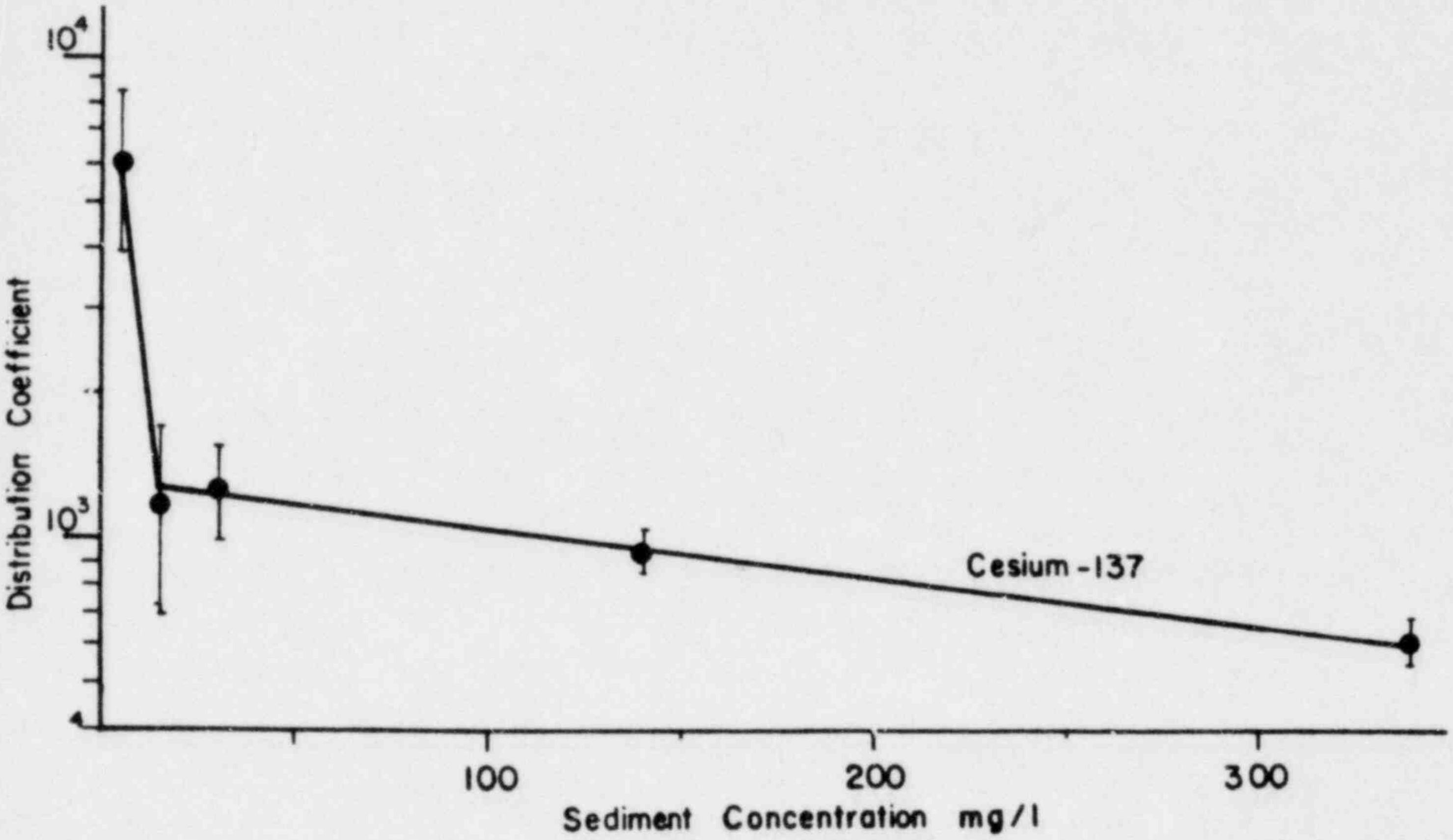


Figure 5. Effect of sediment concentrations on the distribution coefficients for ^{137}Cs in Lake Michigan sediment-water system. Errors are 2SD of propagated counting error.

extended to even lower sediment concentrations in order to correspond to the concentration of suspended sediments in most natural waters.

A later experiment was run on the effect of sediment concentration on the distribution coefficient values using 1000 mg/l instead of 200 mg/l for the Hudson River stations. This larger sediment concentration was prepared using fresh rather than the freeze-dried sediments as before. The fresh 200 mg/l sediments have greater K_d values than the freeze-dried sediments as shown in Table 5. The estuarine or brackish water K_d values MP 0.1 and MP 18.6 have similar K_d values for both the 200 and 1000 mg/l. However, the freshwater stations, MP 43.3 and MP 59.8 have significantly higher values for the 1000 mg/l concentration. This finding was not expected since K_d values have been shown previously to increase with decreasing sediment concentration. These experiments should be repeated to verify the "anomalous" findings and to determine the possible reasons for the differences.

3.3 Effect of pH of 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 (16). The results presented below are from experiments with water and sediments from Clinch River, Tennessee.

The K_d values obtained, for ^{137}Cs in the Clinch River sediment-water system are shown in Figure 6. 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 for ^{137}Cs are not affected by the pH changes that are commonly observed in natural waters.

The effect of pH on distribution coefficient values for the Hudson River stations is shown in Table 6. Again the pH has little effect on the K_d values determined at the four river-estuary stations MP 0.1, 18.6, 43.3 and 59.8. The value at pH of 4.0 was slightly lower than the average values found for the range, pH 4.0 to pH 10.0.

3.4 Effect of Organic Ligands

The effect of organic ligands on the values of sediment-water distribution coefficients for ^{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

Table 5. Effect of sediment concentration on distribution coefficients for ^{137}Cs for Hudson River sediment water systems.

Station/Concentration	Average K_d (SD) ml/g					
	200 mg/ℓ				1000 mg/ℓ	
	n*		n*		n*	
Hudson River						
MP 0.1	9	356 (21)	6	635 (63)**	9	700 (80)**
MP 18.6	9	878 (71)	7	582 (70)	9	740 (230)
MP 43.3	9	401 (20)	6	1740 (80)	9	3490 (180)
MP 59.8	-	-	5	1660 (130)	6	7380 (1000)

[*] n is the number of experimental vessels from which the K_d was calculated.

[**] These K_d values were measured on fresh samples and show 2 to 4 times greater K_d value than the freeze-dried samples in column 4.

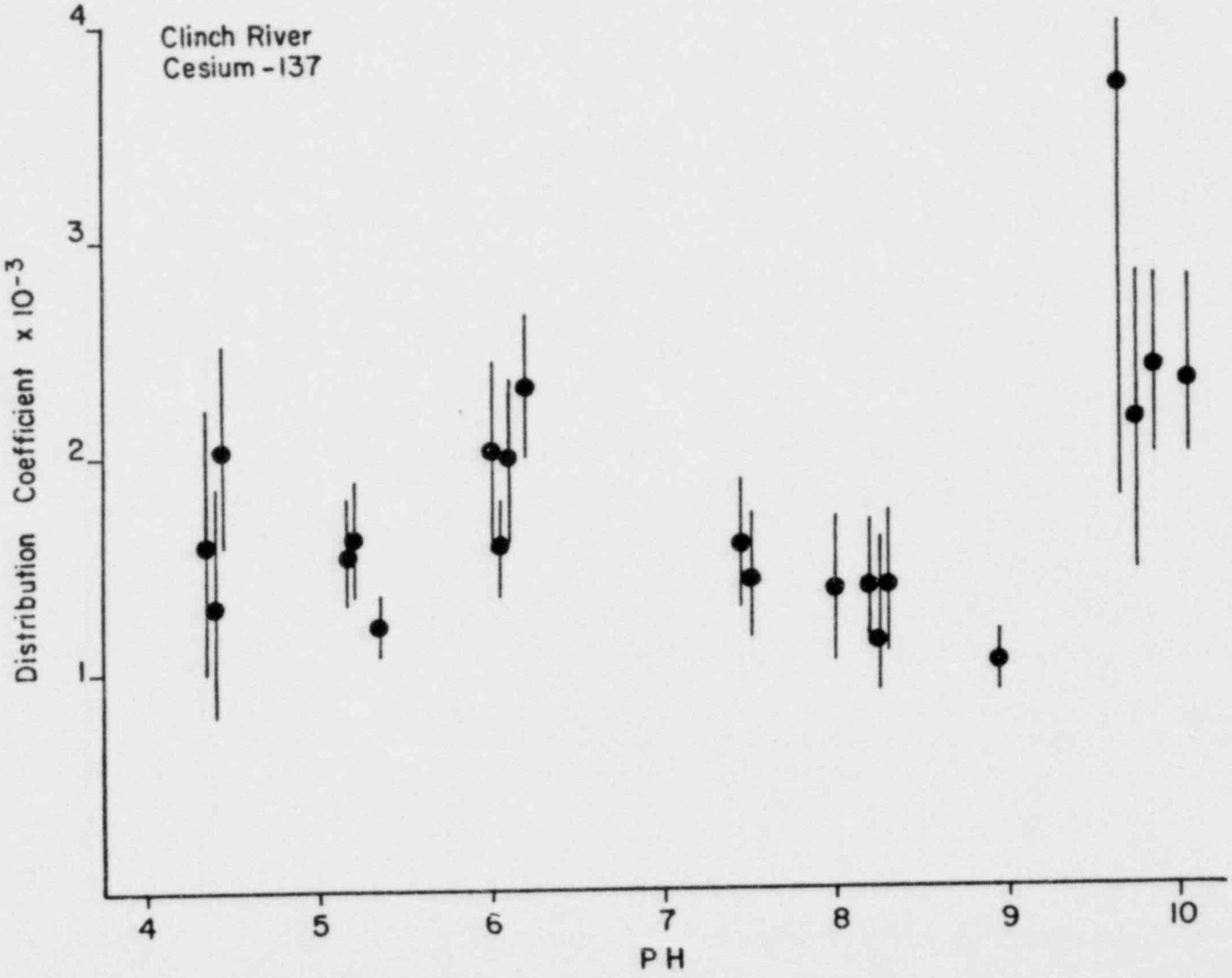


Figure 6. Effect of pH on the distribution coefficients for ^{137}Cs in the Clinch River sediment-water system.

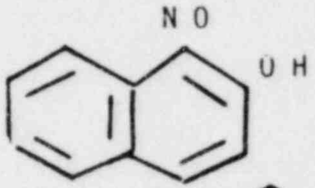
Table 6. Effect of pH on distribution coefficient for ^{137}Cs for Hudson River stations.

Station	pH	n*	K_d (ml/g)
Hudson River	4.0	3	540 (30)
MP 0.1	6.0	3	520 (60)
	7.0	3	440 (80)
	8.0	3	530 (80)
	8.0	3	430 (25)
	10.0	3	370 (10)
MP 18.6	4.0	3	459 (62)
	6.0	3	500 (20)
	7.0	3	440 (130)
	8.0	3	480 (110)
	8.0	3	518 (37)
	10.0	3	590 (80)
MP 43.3	4.0	3	1334 (56)
	6.0	3	1570 (110)
	7.0	3	1670 (220)
	8.0	3	1500 (140)
	8.0	3	1472 (73)
	10.0	3	1480 (100)
MP 59.8	4.0	3	1025 (74)
	6.0	2	1520 (100)
	7.0	2	1670 (100)
	8.0	3	1350 (330)
	10.0	3	1723 (234)

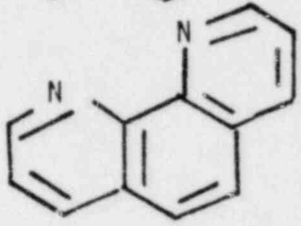
---

* n is the number of experimental vessels from which K_d was calculated.

1 - Nitroso-2-Naphthol

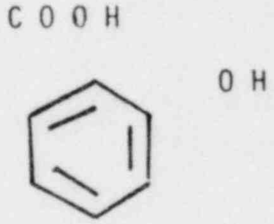


1,10-Phenanthroline



Acetic Acid $CH_3 - COOH$

Salicylic Acid



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

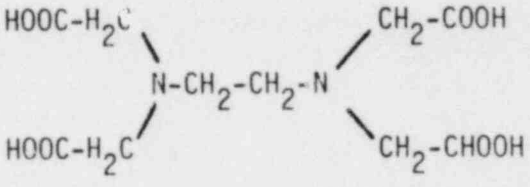


Figure 7. Organic compounds used to determine effects on distribution coefficient for sediment-water system.

Fig. 7) 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 0.22 μm Nuclepore 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 procedure may have produced some chemical alterations in the <u>in situ</u> organic matter and sediments. The sediment size fraction passing through a 63 μm 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 ^{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 7. For both concentrations of EDTA no significant differences were observed between K_d values at 4 and 480 hours (student "t" test, $p = 0.05$). 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. 7 on sediment-water K_d values for ^{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. A final experiment was conducted using Hudson River samples from stations MP 0.1, 18.6 and 43.3. Humic acids and glycolic acid was used in addition to the organic compounds tested above.

Table 7. Effect of the addition of two concentrations of EDTA on the distribution coefficients for ^{137}Cs for Cattaraugus Creek sediment-water system.

Time (hr)	Average K_d (SD) ml/g			
	n*	10^{-6}M EDTA	n*	10^{-10} M EDTA
4	3	1400 ± 100	3	1800 ± 400
480	3	1900 ± 800	3	2400 ± 2100

---

* n is the number of experimental vessels from which the average K_d was calculated.

The results of the experiments to determine K_d values as a function of various organic compounds are summarized in Table 8 and in Fig. 8. For EDTA, the individual values in Table 8 represent only the means of triplicate measurements on individual sample bottles. In Fig. 8, however, the EDTA values are presented as mean values with one standard deviation unit for the three sample bottles. This information gives the intrasample variability to be expected in the measurements from an individual sample bottle. Using the data in Table 8, Kruskal-Wallis statistical tests (23) 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 statistically significant differences ($p = 0.05$ level) were not observed for ^{137}Cs.

3.5 Effect of Salinity

A series of samples from the Hudson River and estuary were measured for ^{137}Cs K_d values. The salinity changes from a high value of 15-25 ^{o}/oo at station MP 0.1 to 2-10^o/oo MP 18.6 to $<2^o$/oo at MP 43.3 and 59.8. Salinity would be expected to have the most profound effect, by decreasing the K_d values, at the high salinity stations. The ^{137}Cs once adsorbed to the sediment, on subsequent transport down stream to the estuary could be expected to exchange the Cs atoms for the excess concentrations of Na atoms at the exchangeable sites of the sediment. For minerals with lattice bound Cs, the exchange of Na^+ for Cs^+ would be much slower and the experimental K_d value would be expected to remain almost constant. Figure 9 shows the K_d values for the upriver-freshwater and estuary-brackish water stations. The K_d values for ^{137}Cs appears to decrease by a factor of about 27 times from freshwater to brackish water; this means that ^{137}Cs is solublized in the brackish water. There are no apparent differences in the K_d values between the two estuarine stations which have salinities greater than 2 ^{o}/oo.

3.6 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 physicochemical 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, (15 and 16).

Table 8. Distribution coefficient values for ^{137}Cs in the Lake Washington and Hudson River sediment-water systems with the addition of specific organic compounds.

	Average K_d (SD) ml/g							
			Hudson River					
Compound Added	n*	Lake Washington	n*	MP 0.1	n*	MP 18.6	n*	MP 43.3
None (control)	3	415 (100)	6	635 (63)	7	582 (70)	6	1740 (80)
1-nitroso-2-naphthol $10^{-5}M$	3	504 (69)	4	724 (54)	4	603 (40)	4	1770 (50)
1, 10 phenanthroline $10^{-4}M$	3	320 (39)	4	746 (56)	4	647 (22)		1600 (90)
Acetic Acid $10^{-4}M$	2	270 (77)		-		-		-
Salicylic Acid $10^{-4}M$	3	371 (61)		-		-		-
EDTA $10^{-4}M$	3	347 (76)	4	656 (53)	4	624 (45)	4	1770 (90)
Humic Acid		-	4	740 (61)	4	696 (51)	4	1850 (90)
Glycolic Acid		-	4	742 (49)	4	618 (56)	4	1700 (90)

---

*n is the number of experimental vessels from which the average K_d was calculated.

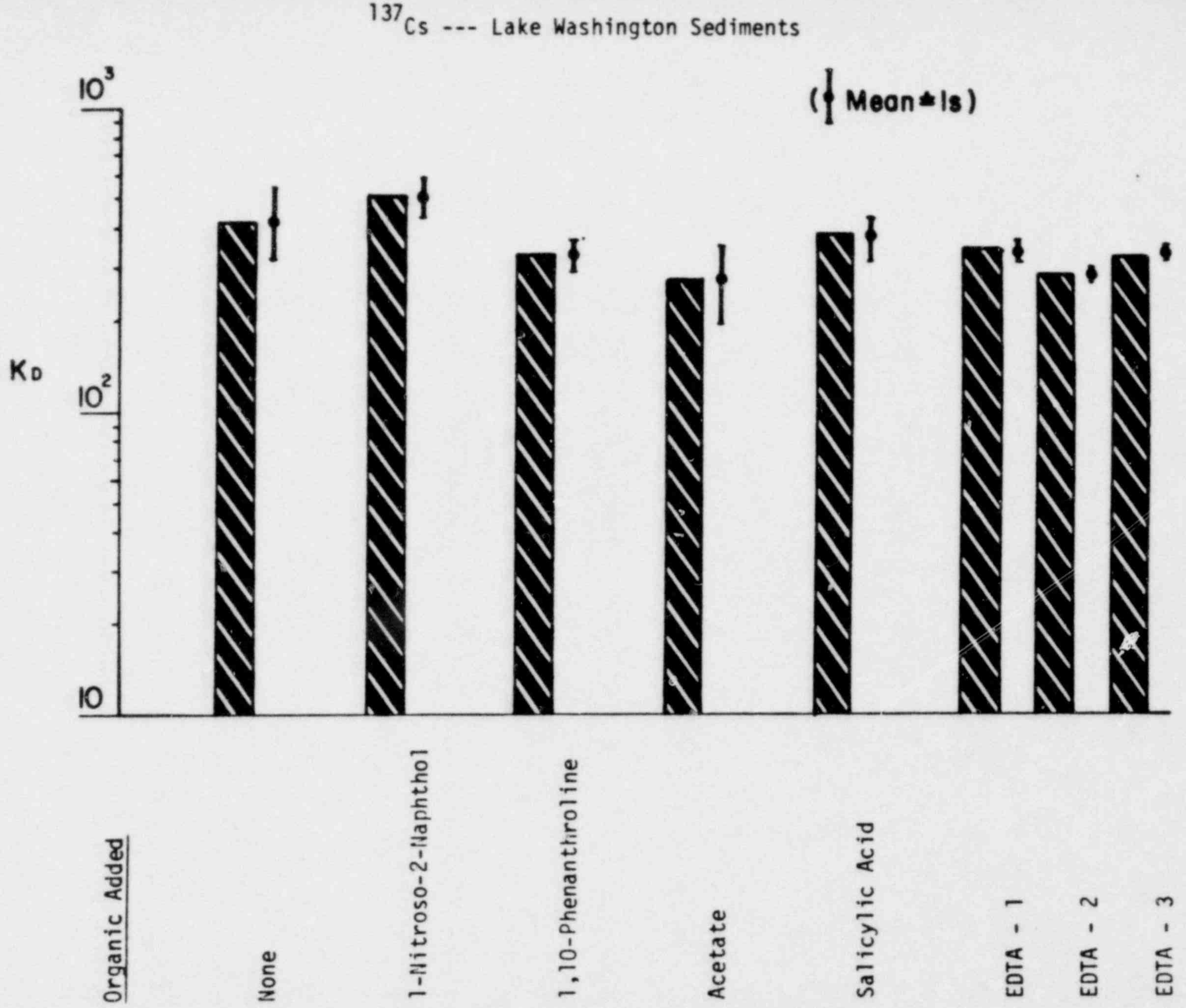


Figure 8. Effects of organic compounds on the distribution coefficients for ^{137}Cs in the Lake Washington sediment-water system

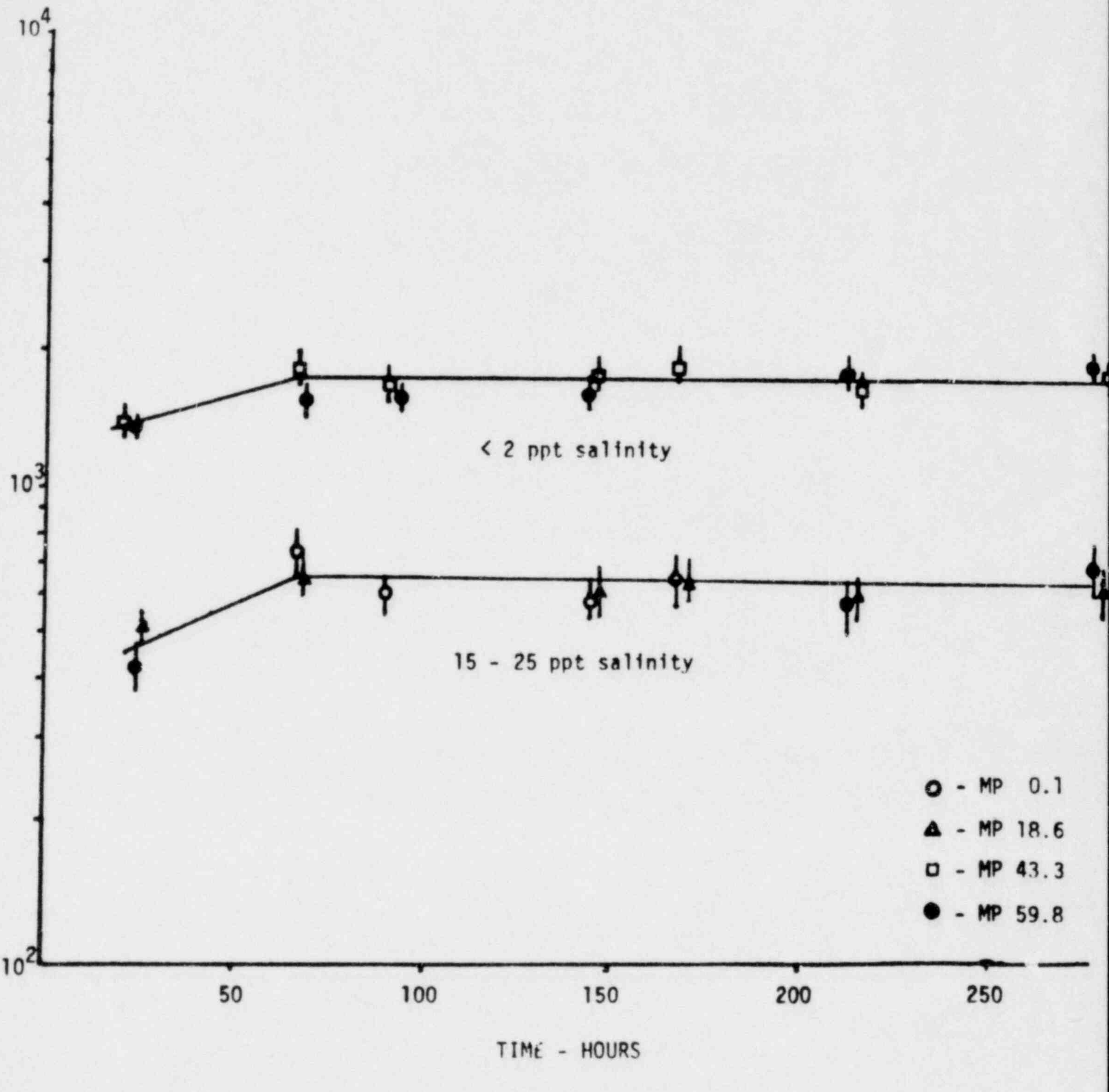


Figure 9. Effects of salinity on the distribution coefficients for ^{137}Cs in the Hudson River sediment-water system.

Table 9. Relative concentrations measured in dialysis experiments for ^{137}Cs in the Lake Michigan sediment-water system.

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

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 procelain 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. 2).

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.

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.

The results are presented in Table 9 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 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.

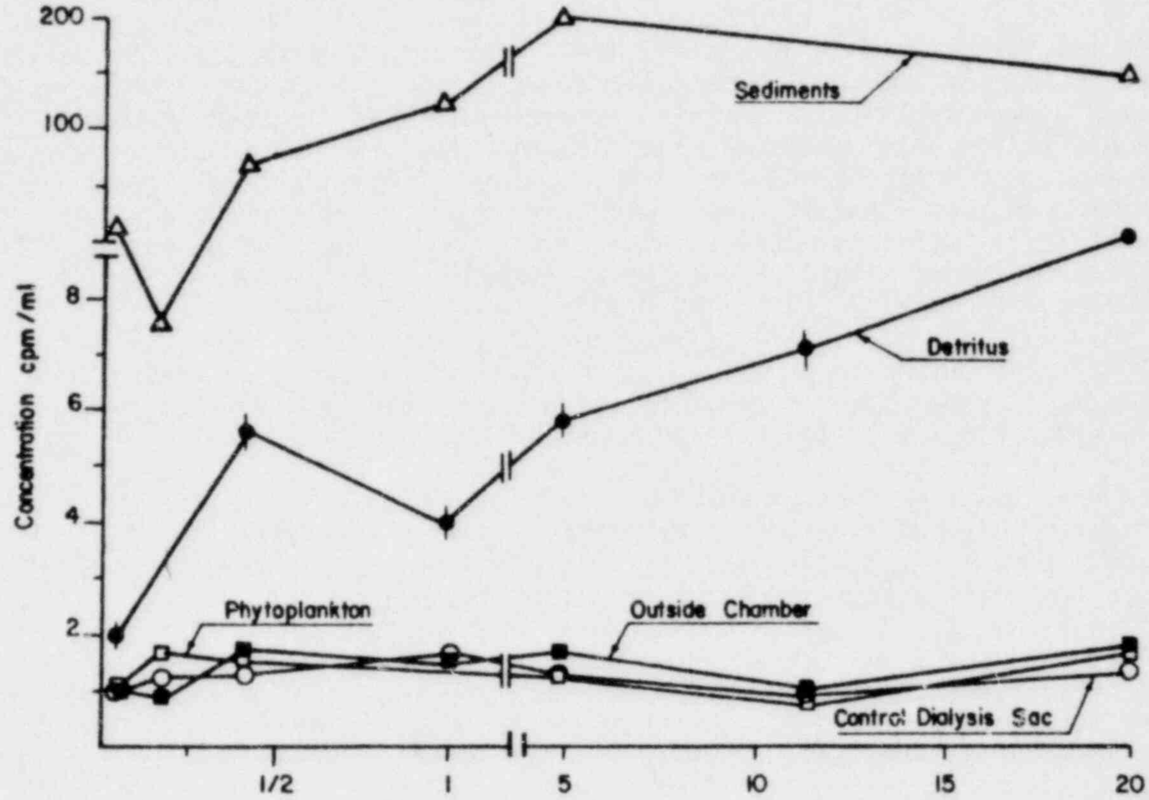
Cesium-137 occurs primarily as an ionic species and behaves very differently from ^{106}Ru or ^{241}Am which exist in several different physico-chemical states including radiocolloids. Figure 10a shows the concentration of soluble ^{137}Cs in each of the chambers throughout the experiment. There is a 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. 10b. 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 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.

Dialysis experiments have been conducted using filtered water from the Eastern Pacific Ocean and the Skagit River Estuary. For one experiment, filtered Eastern Pacific seawater (32 $^{0}/oo$, pH = 8.0) collected at 150 m depth off Port Angeles, Washington was spiked with radionuclides and aged. Approximately 3,000 ml of spiked seawater was placed in the outer compartment and three dialysis sacs with 100 ml of distilled water each were submerged in the seawater. Five ml of a Phaeodactylum tricornutum culture (2 x 10^5 cells/ml) were added to the phytoplankton sac and 5 ml of filtrate from the algal culture were added to the control and sediment sacs; the sediment sac contained 765 mg of sediment from Lake Nitinat. The experiment was conducted at pH = 7.5 and the equilibrium salinity was 29.1 $^{0}/oo$. Samples were collected from all four compartments at the beginning of the experiment and after 1, 2, 4, and 11 days and analyzed for radioactivity in the soluble (<0.22 μm) and particulate phase. The radioactivity of ^{137}Cs was determined by counting using the Ge(Li) gamma ray spectroscopy system.

Data are presented in terms of radionuclide concentration in the soluble and particulate phases of each compartment as a function of time. Figure 11a shows the changes in concentration with time measured for ^{137}Cs in the outer, spiked seawater compartment. During the first two days there is rapid decline in the concentration of ^{137}Cs in the soluble fraction. The decrease in concentration results primarily from losses in the outer compartment as the soluble species diffuse across the dialysis membranes and into the dialysis sacs. After the fourth day there are no significant changes, i.e., within counting errors, in the concentration of the radionuclide in either the soluble or the particulate phase. It is important to note the scale used for the particulate and soluble data for ^{137}Cs. The radionuclide concentration in the particulate phase is generally less than 1% of that in the soluble phase.

(a) Particulate



(b) Soluble

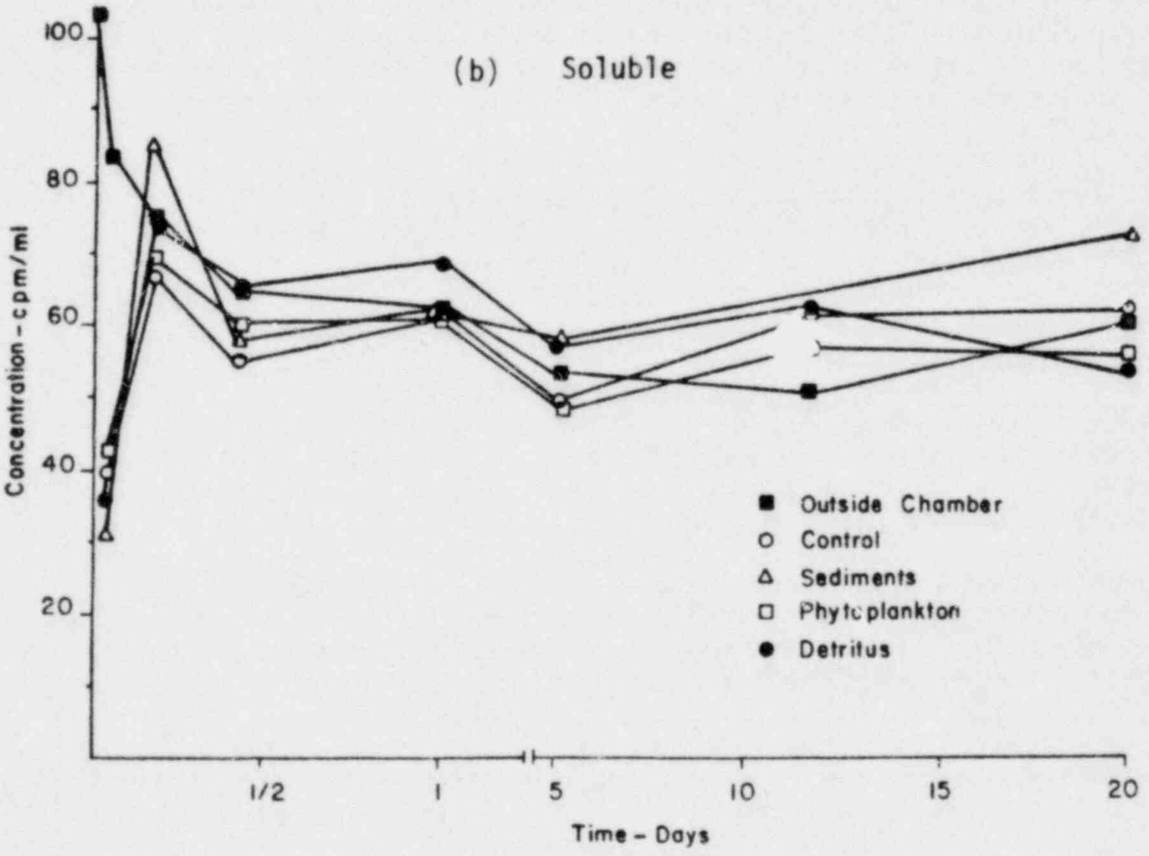


Figure 10. Concentrations of ^{137}Cs in the soluble and particulate fractions as a function of time in a dialysis experiment for the Lake Michigan sediment-water system.

Inside the control sac there was a significant increase in the soluble concentration of ^{137}Cs during the first day (Figure 11b) due to the soluble radionuclides diffusing across the membrane. For ^{137}Cs the soluble concentrations were unchanged from day 1 until the end of the experiment. The concentrations of particulate ^{137}Cs increased significantly between days 4 and 11, indicating that equilibrium may not have been reached. As with the outside compartment, the control sac contained only a small fraction (<5%) of the total concentration of ^{137}Cs in the particulate phase.

The distribution of ^{137}Cs in the plankton and sediment chambers is shown in Figures 11c and 11d. In the sediment sac approximately 40% of ^{137}Cs is associated with the particulates.

The concentration of ^{137}Cs per unit weight of sediment and phytoplankton is shown in Figure 11e. After 2 days the radionuclides in solution and on the sediments and phytoplankton have reached equilibrium, except for ^{137}Cs on phytoplankton. Significant quantities of ^{137}Cs are accumulated by sediments, which appears at equilibrium.

Mass balance calculations were made for ^{137}Cs in this experiment at day 11 (shown in Table 10). These calculations include the activity in the particulate and soluble phase for each of the compartments at day 11, the activity removed by sampling during the experiment and the activity adsorbed to miscellaneous apparatus. The latter includes activity adsorbed to the dialysis membranes and that removed from the glassware by three washings with hot HNO_3. Approximately 100% of the ^{137}Cs was recovered. In terms of the percent recovered, the major portion of the ^{137}Cs occurs in the soluble fraction of the outer compartment.

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

Samples were collected from each chamber at 2, 10 24, 48 and 96 hours and the activity (cpm/ml) was determined for the soluble (<0.45µm) and particulate (<0.45 µm) fractions. By comparing the concentration of soluble radionuclides in each compartment we can estimate the time required to reach equilibrium between the outside chamber and the inside of the dialysis sacs for each radionuclide, Cesium-137 reaches equilibrium after 10 hours.

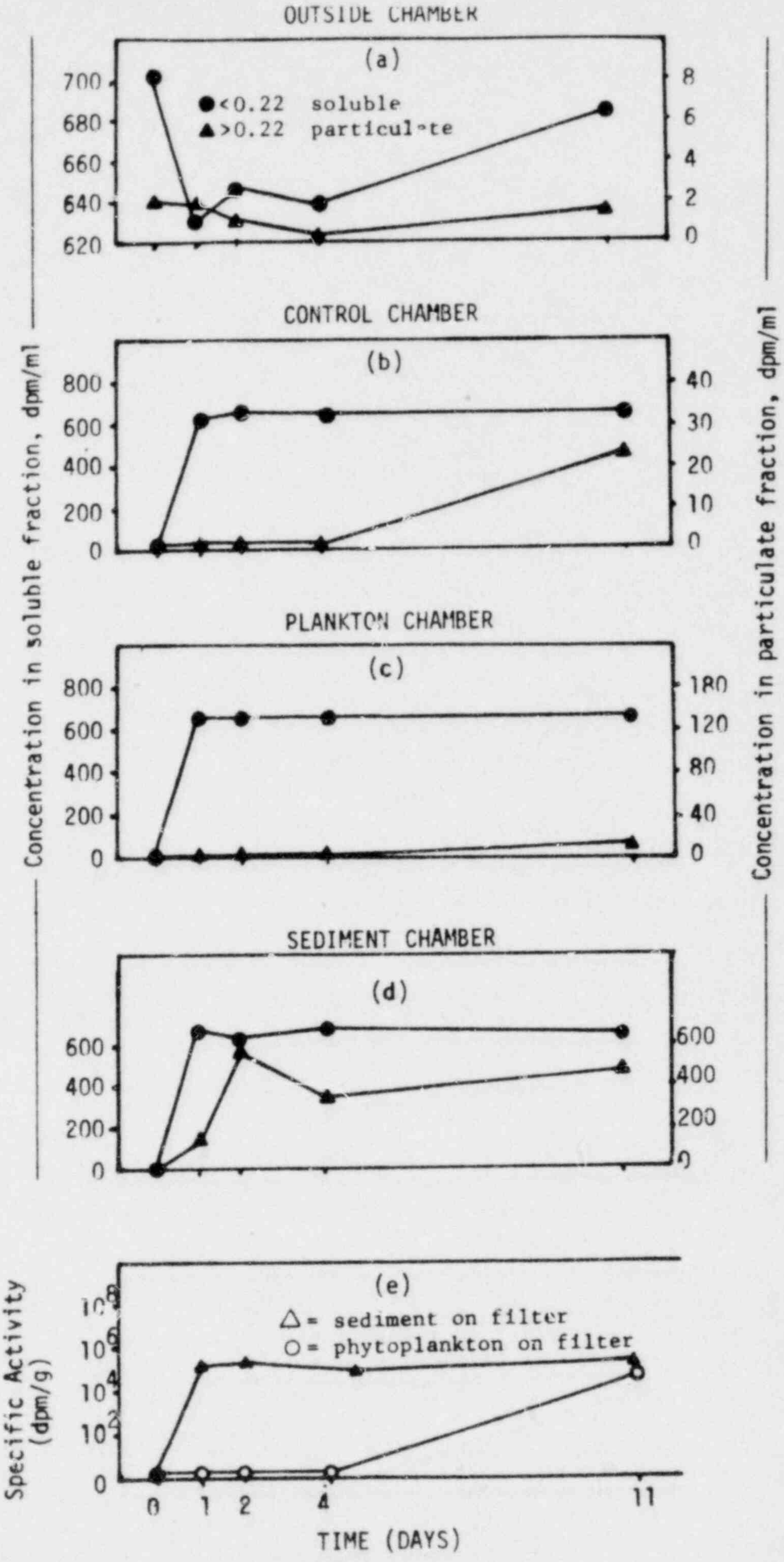


Figure 11. Effect of time on the distribution of ^{137}Cs in the dialysis chambers for a marine sediment-water system.

Table 10. Mass balance calculations for ^{137}Cs after 11 days of dialysis experiment for a marine sediment-water system (Lake Nitinat).

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

---

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

†Amount added = spiked water at day 0.

Figure 12 shows the concentration of ^{137}Cs in the soluble phase for each compartment throughout the experiment. All the dialysis sacs appear to be in equilibrium with the outer compartment after 10 hours and there are only small changes in concentration after that time. Table 11 shows that the concentration of particulate ^{137}Cs never exceeded detection limits and less than 1% of the total ^{137}Cs is in the particulate phase.

Comparison 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 12. For ^{137}Cs, 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.

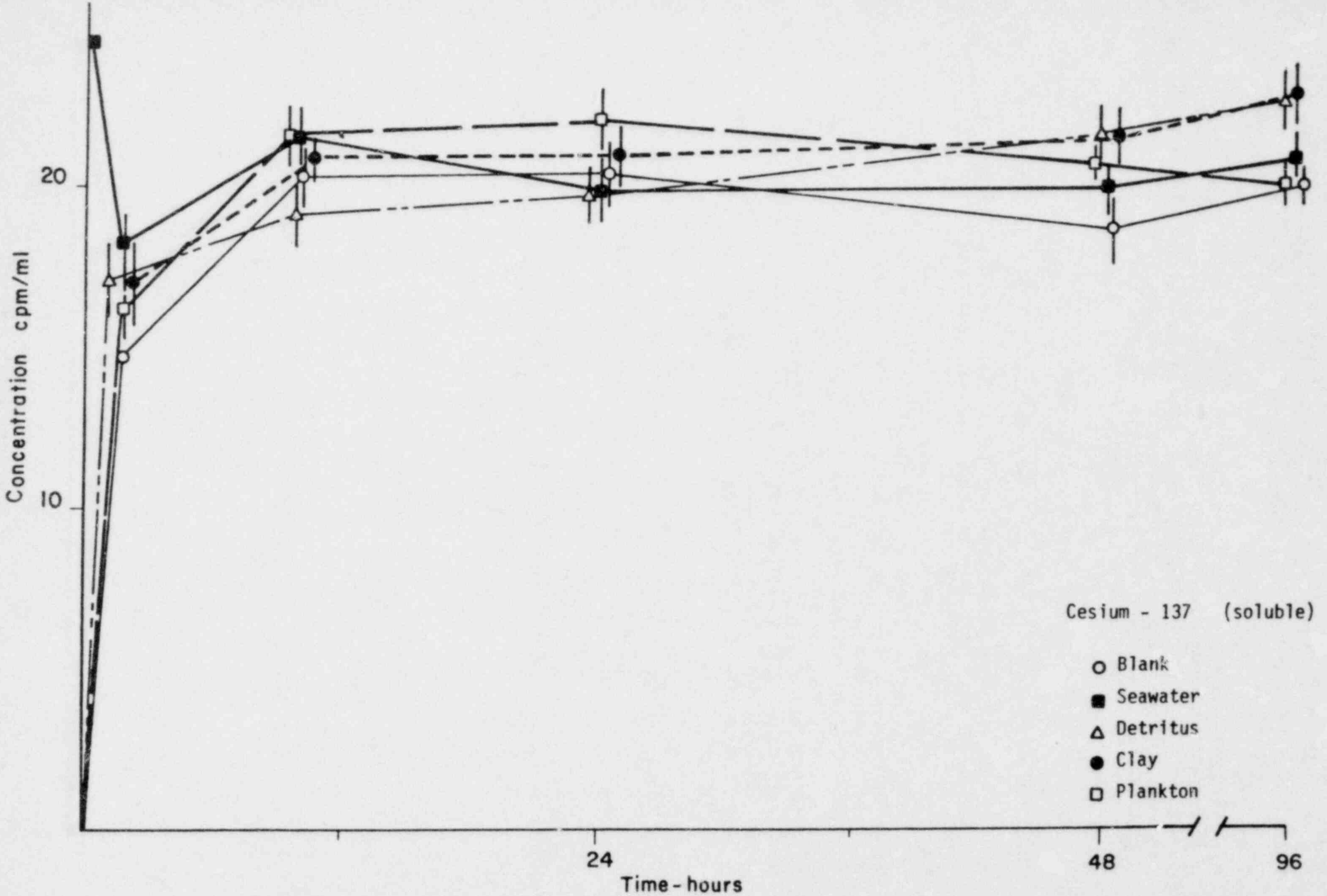


Figure 12. Concentration of ^{137}Cs in the soluble fraction as a function of time in a dialysis experiment for a marine sediment-water system. The particulate concentration in the fraction was below detection limits.

Table 11. Distribution of ^{137}Cs into soluble and particulate fractions during a 96-hour dialysis experiment for a marine sediment-water system (Skagit Estuary).

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

Table 12. Comparisons of K_d values for adsorption by the constant shaking method with K_d values calculated from 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)
^{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	$(1.04 \pm 0.02) \times 10^4$		$(2.06 \pm 0.09) \times 10^3$	

4.0 CONCLUSIONS AND RECOMMENDATIONS

The average sorption distribution coefficient, K_d, values measured for ^{137}Cs in experimental fresh, estuarine and marine sediment-water systems ranged from 17 to 1740 ml/g; organic detritus in dialysis experiments from freshwater was 10^4 ml/g. The average desorption distribution coefficients were often a factor of 10 times greater than the sorption K_d values. This illustrates that cesium, once sorbed or exchanged, is not easily replaced on certain sedimentary minerals. The lowest values were found in the marine sediment-water system and the highest values were found at the Hudson and Clinch river sediment-water systems. Organic detritus accumulates ^{137}Cs in freshwater systems but does not accumulate ^{137}Cs in marine or estuarine systems. In terms of chemical reactivity, cesium is higher on the replacement series of cations and cannot be readily replaced by Na, K, or Ca which may be present in the solution.

There is an apparent increase in K_d values for ^{137}Cs as the sediment concentration decreases. There is also an increase in K_d values as a function of time for all sediment concentrations. These two observations may be related to the surface and lattice properties of the sedimentary material. A rapid initial adsorption may occur first on the sediment surface followed by an ion exchange for potassium in the mineral lattice.

There appears to be no effect on the K_d values for ^{137}Cs at pH values of 4 to 9, which are typical of most natural waters. Organic compounds added to the sediment-water mixtures had no significant effect on the K_d values for ^{137}Cs.

In the dialysis studies, ^{137}Cs occurs primarily as ionic species and diffuses easily through the membrane. As expected, ^{137}Cs concentrates strongly in the sediment chamber with the organic detritus also concentrating ^{137}Cs. Phytoplankton did not concentrate ^{137}Cs extensively, possibly because of the cellular discrimination factor of potassium over cesium. The K_d values calculated from the dialysis experiments appear to be 20 times greater than the equilibrium K_d values for adsorption (constant shaking method). The results of this study suggest the complex reactions which occur in an experimental system can be influenced by detritus water, phytoplankton, and sediment and that these same reactions occur in natural water systems. It is recommended that in using data on K_d values in hydrological modeling of radionuclides released from nuclear reactors, the effects of the parameters investigated in these studies be evaluated.

5.0 SUMMARY - ^{137}Cs

A summary of the adsorption and desorption K_d values for ^{137}Cs is shown in Table 4 for the several stations measured. The values range from a low of 17 ml/g in Columbia River Sediments placed in contact with Eastern Pacific Ocean water (thin-layer technique) to a high of 1740 ml/g for Hudson River sediments (<63μm size) suspended in Hudson River (fresh) water above Station MP 43.3. The desorption distribution coefficient generally is a factor of 10 times greater than the adsorption distribution coefficient. This difference between adsorption and desorption is observed by most investigators and indicates that the equilibrium of cesium between the aqueous and particulate phases has not occurred within the time period of the experiments. Substitution of K^+ for Cs^+ on exchangeable sites, substitution of ^{137}Cs for Cs^+ bound in the clay crystalline lattice and other environmental factors may influence this equilibrium. It appears that the liquid effluent containing radionuclides from a nuclear reactor may initially experience adsorption to environmental particles giving a much lower K_d value than that achieved at later times. Fallout or other sources of radionuclides present in the environment for long time periods probably have reached this equilibrium and thus laboratory and field measurements may differ. Regulatory requirements for nuclear reactor effluents must take both time dependent effects into consideration. To evaluate the effective distribution coefficient for a given system, information on the particle size, sediment concentration, mineral type, salinity and/or hardness of the water are required. Additional information is required on mechanisms responsible for: a) the differences measured between the adsorption and desorption K_d; b) the kinetic reactions which control the ion exchange/lattice substitution equilibrium; and c) the environmental effects peculiar to a given locati

6.0 REFERENCES

(1) Schell, W. R., and A. E. Nevissi. 1980. Detrital sedimentation in lakes and reservoirs. In Guidebook on Nuclear Techniques in Hydrology - 1980. International Atomic Energy Agency, Vienna. In press.

(2) Bowen, V. T., V. E. Noshkin, H. D. Livingston, and H. L. Volchok. 1980. Fallout radionuclides in the Pacific Ocean; vertical and horizontal distributions, largely from GEOSECS stations. Woods Hole Oceanographic Institute Report No. COO-3563-88.

(3) Goldberg, E. D.., U. S. Broecker, M. G. Gross, and K. K. Turekian. 1971. Marine Chemistry. In Radioactivity in the marine environment. National Academy of Sciencies, Washington, D.C. 272 pp.

(4) Marray, C. N., and H. F. Eicke. 1977. German coastal survey, a comparison of the distribution and behavior of cesium and some transuranic isotopes. Deutsche Hydrographisches Inst. Zeitschrift 30:1-8.

(5) Heatherington, I. A., and D. F. Jefferies. 1974. Distribution of some fission product radionuclides in sea and estuarine sediments. Neth. J. Sea Res. 8:319-338.

(6) Patel, B. S., S. Patel, and S. Pavar. 1978. Desorption of radioactivity from nearshore sediment. Estuarine and Coastal Marine Science 7:49-58.

(7) Pickering, R. J. 1969. Distribution of radionuclides in bottom sediment of the Clinch River, Eastern Tennessee. United States Geological Survey. Prof. Paper 433-H. 25 pp.

(8) Elprince, A. M., C. I. Rich, and D. C. Martens. 1977. Effect of temperature and hydroxyaluminum interlayers on the adsorption of trace radioactive cesium by sediments near water-cooled nuclear reactors. Water Resource Res. 13:375-381.

(9) Sure, D. J., and M. K. Gottshalk. 1976. Cesium-137 dynamics within a reactor effluent stream in South Carlina. In Radioecology and Energy Resources, Proc. Symp. on Radioecology 4th. The Ecological Soc. America. Spec. Publ. No. 1. 234-241 pp.

(10) Huff, D. D., and D. Kruger. 1970. Simulation of the hydrologic transport of radioactive aerosols. In R. F. Gould, ed. Radionuclides in the Environment. Advances in Chemistry Series 93. Am. Chem. Soc. Publ., Washington D.C. 487-505 pp.

(11) Stumm, W., and J. J. Morgan. 1970. Aquatic Chemistry. Wiley Interscience. New York. 583 pp.

(12) Davis, J. J. 1963. Cesium and its relationship to potassium in ecology. *In* E. V. Schultz and A. W. Klement, Jr., eds. Radioecology. Proc. Symp. on Radioecology 1st. Reinhold Publ. Corp. New York. 539-556 pp.

(13) Duursma, E. K., and C. J. Bosch. 1970. Theoretical, experimental, and field studies concerning diffusion of radioisotopes in sediments and suspended particles of the sea. Part B. Methods and experiments. *Neth. J. Sea Res.* 4:395-469.

(14) Duursma, E. K., and D. Eisma. 1973. Theoretical, experimental and field studies concerning reactions of radioisotopes with sediments and suspended particles of the sea. Part C. Applications to field studies. *Neth. J. Sea Res.* 6:265-324.

(15) Seymour, A. H., A. Nevissi, W. R. Schell, and A. Sanchez. 1979. 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 Progress Report NUREG/CR-0801, Univ. Washington, Laboratory of Radiation Ecology for Nuclear Regulatory Commission, Washington, D.C. 38 pp.*

(16) Schell, W. R., T. H. Sibley, A. Nevissi, and A. Sanchez. 1979. Distribution coefficients for transuranic elements in aquatic environments. II. Studies on marine and freshwater sediment systems including the radionuclides 106-Ru, 137-Cs, and 241-Am. Annual Progress Report NUREG/CR-0802, Univ. Washington, Laboratory of Radiation Ecology for Nuclear Regulatory Commission, Washington, D.C. 70 pp.*

(17) Schell, W. R., A. L. Sanchez, T. H. Sibley, and J. R. Clayton. 1980. Distribution coefficients for transuranic elements 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 Progress Report NUREG/CR-0803, Univ. Washington, Laboratory of Radiation Ecology for Nuclear Regulatory Commission, Washington, D.C. 76 pp.*

(18) Schell, W. R., A. Sanchez, T. H. Sibley, A. Nevissi, J. Clayton, Jr., and E. A. Wurtz. 1981. Distribution coefficients for radionuclides in aquatic environments. Methodology. Topical Report NUREG/CR-1852, Vol. 1, Univ. Washington, Laboratory of Radiation Ecology for Nuclear Regulatory Commission, Washington, D.C. 27 pp.*

(19) Murray, C. N., and L. Murray. 1973. Adsorption-desorption equilibrium of some radionuclides in sediment-fresh water and sediment-seawater systems. *In* Radiactive contamination of the marine environment, Proc. Symp., Seattle 10-14, July 1972. IAEA, Vienna.

(20) Aston, S. R., and E. K. Duursma. 1973. Concentration effects of cesium-137, zinc-65, cobalt-60, and ruthenium-106 sorption by marine sediments with geochemical implications. Neth. J. Sea Res. 6:225-240.

(21) Bartsdate, R. J. 1970. Transition metal binding by large molecules in high latitudinal waters. Pages 425-493 *in* P. W. Hood, ed. Organic Matter in Natural Waters. Occ. Publ. No. 1, Inst. Mar. Sci., Univ. Alaska, Fairbanks, AK.

(22) Dawson, R., and E. K. Duursma. 1974. Distribution of radioisotopes between plankton, sediment and seawater in a dialysis compartment system. Neth. J. Sea Res. 8:339-353.

(23) Sokal, R. R., and J. F. Rohlf. 1969. Biometry. W. H. Freeman and Co., San Francisco. 776 pp.

---

*Available for purchase from the NRC/GPO Sales Program, U.S. Nuclear Regulatory Commission, Washington, DC 20555, and/or the National Technical Information Service, Springfield, VA 22161.

NRC FORM 335
(7-77)

U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

1. REPORT NUMBER (Assigned by DDC)
NUREG-CR-1852, Vol. 2

2. (Leave blank)

3. RECIPIENT'S ACCESSION NO.

4. TITLE AND SUBTITLE (Add Volume No., if appropriate)
Distribution Coefficients for Radionuclides in Aquatic Environments
Subtitle: Adsorption and Desorption Studies of ^{137}Cs

5. DATE REPORT COMPLETED
MONTH December YEAR 1980

6. (Leave blank)

7. AUTHOR(S)
W.R. Schell, A.L. Sanchez, and T.H. Sibley

8. (Leave blank)

9. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)

College of Fisheries
University of Washington
Seattle, WA 98195

DATE REPORT ISSUED
MONTH May YEAR 1981

10. PROJECT/TASK/WORK UNIT NO.

11. CONTRACT NO.
FIN B5749

12. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)
U.S. Nuclear Regulatory Commission
Division of Health, Siting and Waste Management
Office of Nuclear Regulatory Research
Washington, DC 20555

13. TYPE OF REPORT
Topical

PERIOD COVERED (Inclusive dates)

14. (Leave blank)

15. SUPPLEMENTARY NOTES

16. ABSTRACT (200 words or less)

This report investigates the several parameters important to measuring the distribution coefficients for ^{137}Cs in natural sediment-water systems. The samples of water and sediment for the experimental measurements were collected from several sites in the U.S.A. including the Hudson River, Clinch River, Cattaraugus Creek, Lake Michigan, Skagit Bay, Columbia River, and Lake Washington. The results of the experiments have given sorption distribution coefficients for ^{137}Cs of from 17 to 1700 ml/g; the desorption distribution coefficient is a factor of 10 greater. The lowest values were found in marine systems and the highest values in freshwater. Organic compounds of different complexing functional groups added to the experimental system, and the change in pH between 4 and 10 had no effect on the distribution coefficients measured. Sediment concentration and salinity changes influenced strongly the experimental distribution coefficients measured.

17. KEY WORDS AND DOCUMENT ANALYSIS
Distribution Coefficients, Dialysis Experiments, ^{137}Cs, Sediment Concentration, pH

17a. DESCRIPTORS

17b. IDENTIFIERS/OPEN-ENDED TERMS

18. AVAILABILITY STATEMENT
Unlimited

19. SECURITY CLASS (This report)
Unclassified

20. SECURITY CLASS (This page)
Unclassified

21. NO. OF PAGES

22. PRICE
$

NRC FORM 335 (7-77)