
Distribution Coefficients for Radionuclides in Aquatic Environments

Methodology

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

College of Fisheries
University of Washington
Seattle, WA 98195

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ABSTRACT

Methods used to determine distribution coefficients for ^{60}Co , ^{85}Sr , ^{106}Ru , ^{137}Cs , ^{237}Pu , ^{241}Am , and ^{244}Cm in natural sediment-water systems were tested and evaluated. The methods included the thin layer technique, sedimentation technique, suspension or constant shaking technique, and the dialysis technique (1,7). Problems associated with sample storage and treatment, the effect of bacteria, equilibration time and particle formation of ^{106}Ru and ^{241}Am in control experiments were investigated. The counting methods have utilized high sensitivity Ge(Li) detectors with multi-channel analyzers and computer data collection and reduction, NaI(Tl) crystal detectors with single-channel analyzers, and liquid scintillation counting. A comparison of the techniques for different sediment-water systems indicated that the most reproducible data was collected by the suspension or constant shaking technique.

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DISTRIBUTION COEFFICIENTS FOR RADIONUCLIDES IN AQUATIC ENVIRONMENTS

Methodology

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

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

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

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 (1). Results for preliminary experiments using ^{65}Zn and ^{137}Cs were compared to published results (2) in order to evaluate our experimental techniques. Sediments and water collected from Lake Washington (freshwater), Lake Nitinat (anoxic marine) and the mouth of the Columbia River (oxic marine) were used to determine distribution coefficients for ^{237}Pu and ^{241}Am in laboratory sediment-water systems. Dialysis experiments were used to study the distribution of ^{60}Co , ^{65}Zn , ^{137}Cs , ^{239}Pu and ^{241}Am among suspended sediments, phytoplankton and filtered seawater. Since diffusion across the dialysis membranes is limited to chemical species of sizes less than 6000-8000 nominal molecular weight, these experiments provide information on the uptake of soluble radionuclides by sediments and phytoplankton. Results from the first year of research are included in NUREG/CRO801 (3).

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 (1) for determining K_d 's of ^{237}Pu and ^{241}Am , the effect of pH on the distribution coefficient of ^{241}Am in sediment-water systems from Lake Nitinat and Lake Washington and particle formation by ^{106}Ru and ^{241}Am in the absence of suspended sediments. These results are available in NUREG/CRO802 (4). 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 $<63\mu\text{m}$ -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/CRO803 (8).

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 and evaluate the various methods used for determining distribution coefficients. In addition, recommendations will be made on the design and appropriate methodology to be used for future distribution coefficient studies.

2.0 MATERIALS AND METHODS

The experimental procedures used in this research were adapted from published reports of other workers and modified as needed for our experiments. In particular, the methods developed by E.K. Duursma and his colleagues at International Atomic Energy Agency (IAEA) Marine Laboratory, Monaco, have been used extensively. Our application of these techniques has been described previously (3,4,8) and will be discussed below.

2.1 Sources and Types of Samples

For the first two years of this program, experiments were conducted with sediments and water that were collected 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 μ 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, cation exchange capacity, the particle size distribution and the chemical composition of the sediments. An example of this type of information on

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	Remarks
Lake Michigan	67	7.90	<1.0	143	17.19(.48)	62.0	1.8	Frozen	Collected near east shore
Clinch River	3.0	7.92	<1.0	158	13.37(.35)	14.0	1.8	Fresh (cooled)	Collected 1.5 miles downstream from breeder reactor
Hudson River									
MP 59.8	3.0	7.90	<1.0		12.24(.24)				Collected 15 miles upstream from Indian Point Reactor
MP 43.3	3.0	7.90	<1	171	11.99(.34)	34.4	1.8	Freeze-dried	" adjacent to " " "
"	"	"	"	"	9.24(.34)	"	"	Fresh (cooled)	" " " "
MP 18.6	3.0	7.82	2-3	378	8.98(.14)	23.0	1.1	Freeze-dried	Collected 18.6 miles upstream
"	"	"	"	"	10.95(.12)	"	"	Fresh (cooled)	" " " "
MP 0.1	6.0	7.70	15	2963	7.99(.18)	13.8	5.2	Freeze-dried	" 0.1 mile upstream from tip of Manhattan Island
"	"	"	"	"	12.34(.29)	"	"	Fresh (cooled)	" " " " " " " "
Cattaraugus Creek	1.5		<1	202	10.75(.04)	16.8	0.8	Fresh (cooled)	" at mouth of delta into Lake Erie - STA CC-11
Skagit Bay-1	9	7.70	29.62	6662	2.03(.09)			Fresh (cooled)	" 48° 18.0' N, 122° 29.0' 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
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)	Collected at deep anoxic lake on Vancouver Island, B.C.
Sinclair Inlet	15.1	7.75	28.89	5506				Fresh (cooled)	" anoxic organic rich sediment Sta. 2.
Lake Washington	62	7.70	<1	42	31.4(6.2)			Fresh (cooled)	" at 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)				

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.

2.2 Determination of Distribution Coefficients in Laboratory Water-Sediment Systems

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

Thin-layer technique was used for the initial experiments. This technique consists of depositing sediment on filter paper, submerging the sediment and filter into a solution of radionuclides and determining the accumulation of radionuclides by the sediments. In our experiments, six duplicate sediment samples of 10 mg were deposited on filter papers and with six blank filter papers were submerged into the radioactive solution. The sorption of the radionuclide to the sediment was calculated

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*
<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

* Filtered samples were passed through 0.3μM millipore filters.

as the difference in the concentration of the radionuclides in the sediment and the blank filters after various exposure periods. In 1976-1977, we conducted a few experiments to evaluate this technique. The results of these experiments indicated that during sorption, agitation of the water while sampling may cause significant variations in the data due to loss of sediment from the filter. We have found that the sediments will not adhere well to the filters unless the size of the particles is similar to the particle size of silty-clay. It was found by Duursma and Eisma (2) and subsequently verified in our experiments that the rate of sorption to sediments in thin-layer experiments is rapid for the first 48 hours and decreases after that time. We believe this technique provides less reliable results than other techniques.

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

The sedimentation technique was used to determine the K_d value for ^{237}Pu and ^{241}Am in a sterile marine system and the K_d value for ^{241}Am in a non-sterile marine system. The concentration of ^{241}Am in the nonsterile marine system is plotted against time in Figure 1, to indicate the type of results that are obtained with this technique. Results from the first seven days (before the addition of sediments) provide an estimate of the loss of radionuclides due to adsorption on the cylinder walls. Approximately 4-6% of ^{241}Am is removed by adsorption. In the sedimentation technique the distribution coefficient is determined by the extent of removal of radionuclide for each sediment addition; that is, from the slope of ^{241}Am concentration versus time for days 8 through 14. Higher distribution coefficients have steeper slopes.

Constant Shaking Technique - Adsorption K_d Values

The vast majority of our experiments to determine distribution coefficients in laboratory sediment-water systems used the constant shaking technique (1, 5). Briefly, the method consists of:

1. Adding the radionuclides to a known volume of filtered ($0.45\ \mu\text{m}$ or $0.22\ \mu\text{m}$) water sample and adjusting the pH to the initial pH of the water sample with 0.2 M NaOH.
2. Adding sediments from a stock sediment suspension to make a final predetermined sediment concentration, generally 200 mg/l.

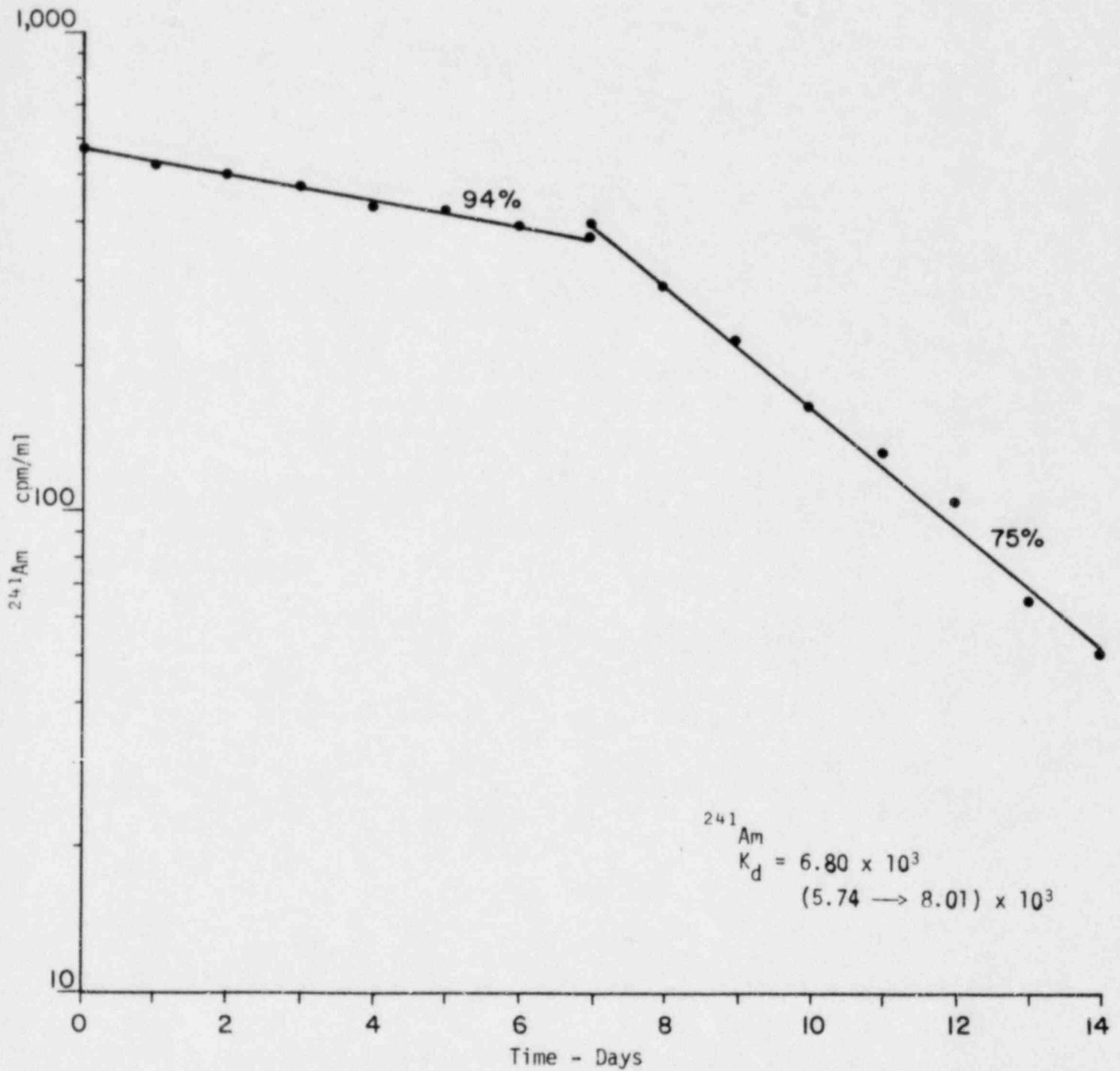


Figure 1. Results of sedimentation technique experiment for determining the distribution coefficient for ^{241}Am in a marine sediment from the mouth of the Columbia River.

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 2 shows the sequential steps that are involved in obtaining adsorption and desorption distribution coefficients with the constant shaking technique.

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. 3) used for these experiments was adapted from similar equipment described by Barsdate (6) and Dawson and Duursma (7). In all our experiments, the outside compartment contains filtered (0.22 μ 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

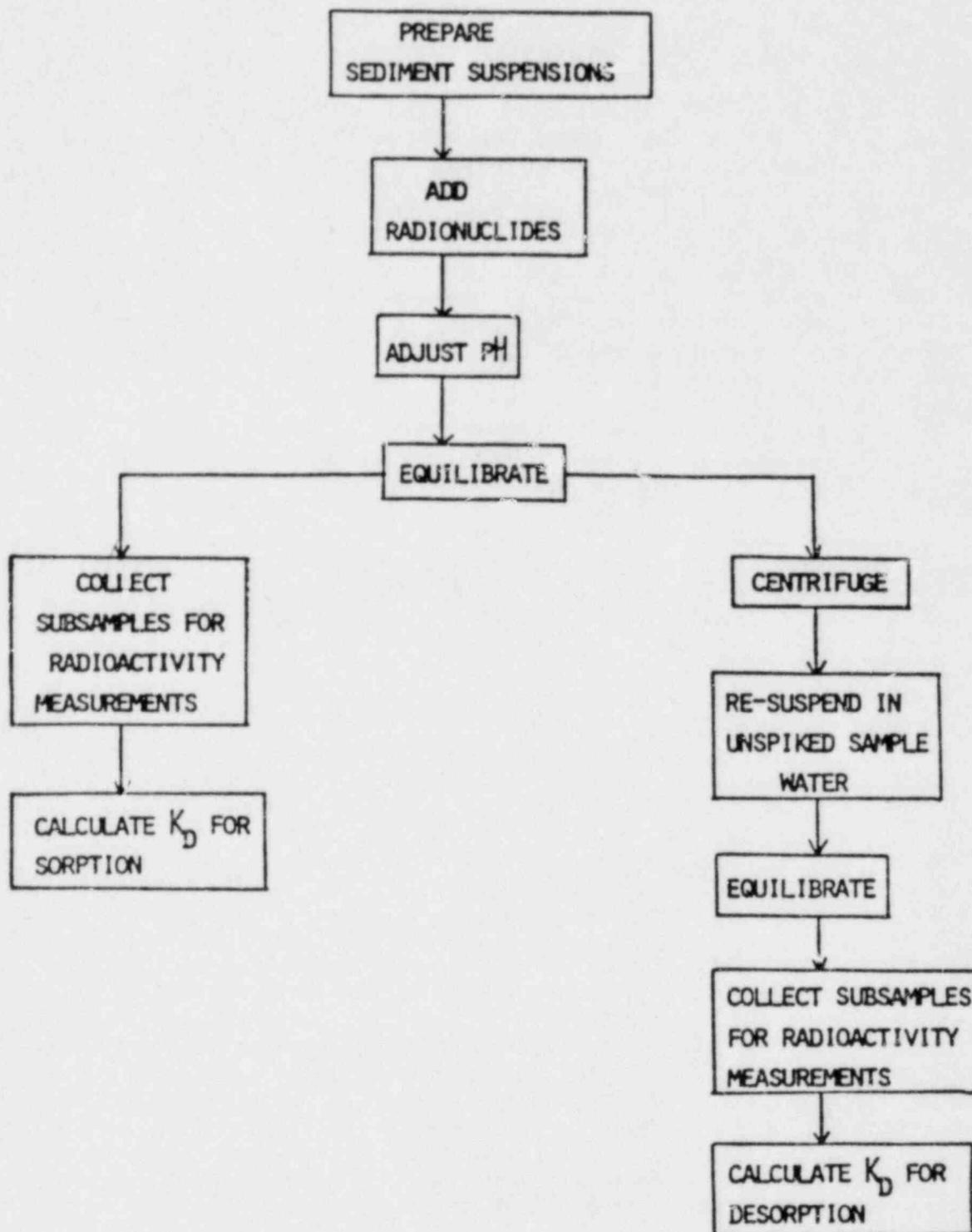


Figure 2. Sequential steps involved in obtaining adsorption and desorption distribution coefficients with the constant shaking technique.

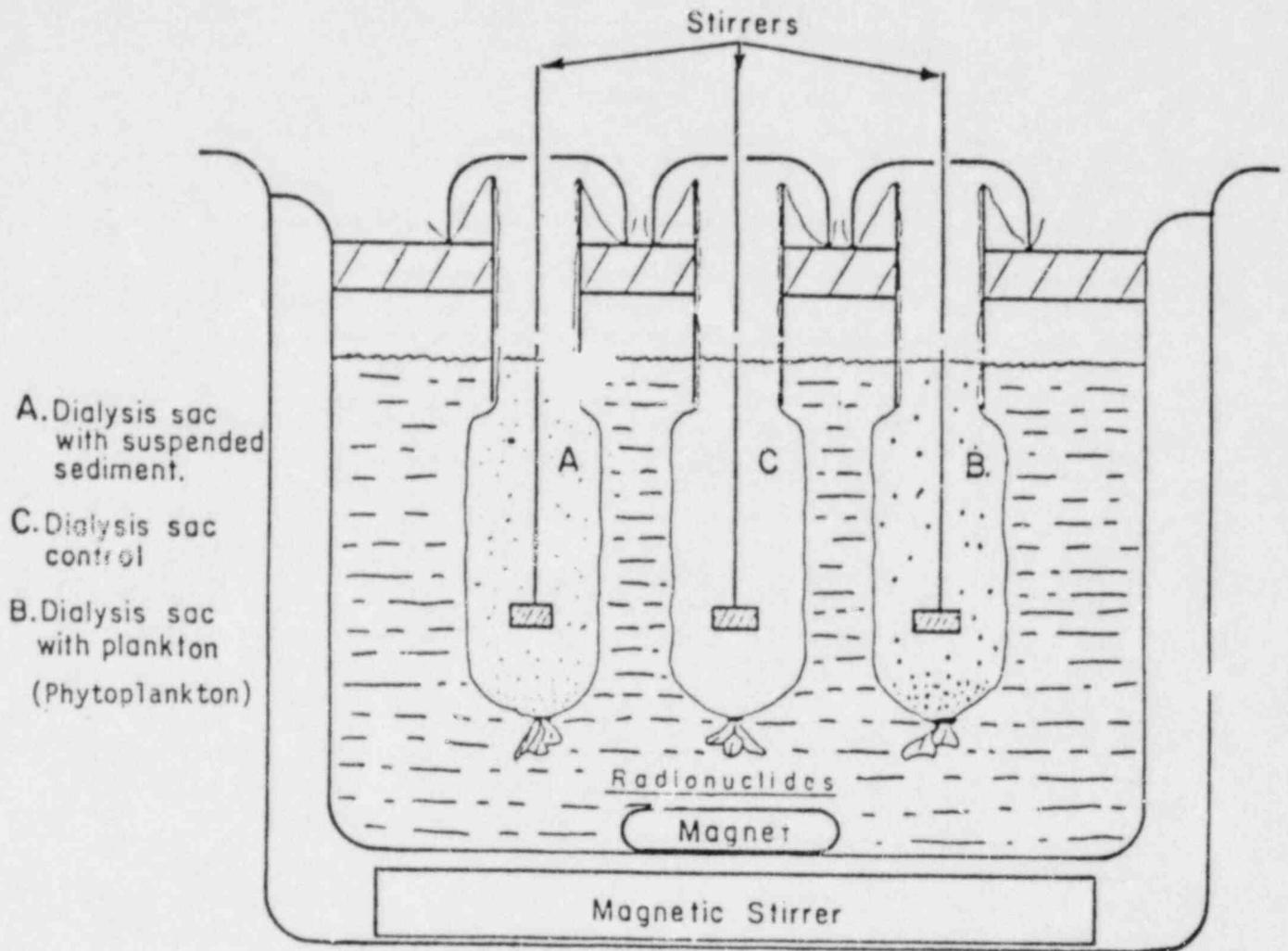


Figure 3. Schematic diagram of apparatus used for dialysis experiments.

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 (8.6×10^6 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 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 μ m 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 Sterilization of Samples

To evaluate the role of bacteria as a factor in the distribution of the radionuclides in the water-sediment system, experiments were conducted during the first year to compare the results in sterilized and unsterilized systems. Water and sediment samples were sterilized to remove bacteria with the expectation that the sterilization process would

not significantly change the physical and/or chemical characteristics of the system (3). There are three practical methods for deactivating bacteria in sediments: heat, addition of an anti-bacterial agent, or irradiation. Although each of these treatments may cause changes in the sediment and water properties, it was decided that sterilization by irradiation would be less damaging than the other methods. Sediment and water samples were sterilized with a dose of 10^6 rads of gamma radiation from the ^{60}Co food irradiator at the College of Fisheries.

Experiments were conducted to test for the presence of bacteria in the sterilized and unsterilized samples using both aerobic and anaerobic culture media incubated at 22°C . The results in Table 4 show that no bacterial growth was detected in either the filtered water or the unfiltered samples. Unsterilized samples of the three types of sediments showed positive test results for both aerobic and anaerobic bacteria after 4 days of incubation. Anaerobic bacteria were not detected in either marine or Lake Nitinat sediments after 90 days. The sterilized freshwater sediments (Lake Washington) showed positive indications of anaerobic and aerobic bacteria after only 13 days of incubation. Thus, the indications are that 10^6 rads were effective in sterilizing sediment from Lake Nitinat, partly effective in sterilizing marine sediments (off the Columbia River), and much less effective in sterilizing freshwater sediments. Considering that the distribution coefficient experiments were conducted at a lower temperature ($5-8^\circ\text{C}$) than the temperature for the bacteria growth experiments (22°C), less effect of bacteria would be expected. Therefore, the experiments with sterilized sediment may indicate the role of bacteria even if the system was not completely sterilized.

2.5 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.

In tracer experiments using only ^{244}Cm , a simple analytical method is to measure the gamma or X-ray radiation emitted in the decay. This method does not require the complex and lengthy radiochemical separation procedures of alpha spectroscopy. However, gamma or X-ray emitted radiation is of low abundance in the decay of curium. This fact, coupled

Table 4. Results of bacterial culture experiments on water and sediment collected from marine and freshwater environments.

	Growth	Days of incubation	Remarks
I. Water			
Seawater, sterilized (1)	-	90	
" " (2)	-	"	
" unsterilized (1)	-	"	
" " (2)	-	"	
Lake Nitinat, sterilized (1)	-	"	
" " (2)	-	"	
" unsterilized (1)	-	"	
" " (2)	-	"	
Lake Washington, sterilized (1)	-	"	
" " (2)	-	"	
" unsterilized (1)	-	"	
" " (2)	-	"	
II. Sediments			
Marine, sterilized (1)	+	39	slight turbidity change
" " (2)	-	90	
" unsterilized (1)	+	4	{meat turned black after 20 days
" " (2)	+	4	
Lake Nitinat, sterilized (1)	-	90	
" " (2)	-	90	
" unsterilized (1)	+	4	{meat turned black after 4 days of incubation indi- cating high anaerobic content of H ₂ S-producing bacteria
" " (2)	+	4	
Lake Washington, sterilized (1)	+	13	{meat turned slightly black after 40 days of incubation
" " (2)	+	13	
" unsterilized (1)	+	4	{meat medium black after 20 days of incubation
" " (2)	+	4	
(1) aerobic culture			
(2) anaerobic culture			

with the low detection efficiency using Ge(Li) or NaI(Tl) detectors, limits the utility of the gamma and X-rays for analysis. We attempted to develop a procedure for measuring the 18.3 KeV L, X-ray decay with a germanium intrinsic detector and pulse height analyzer and found it necessary to use ^{244}Cm alone rather than with several other radionuclides in these experiments. In this procedure, particulate ^{244}Cm is counted directly while soluble ^{244}Cm is coprecipitated with ferric hydroxide and ^{244}Cm is measured in the precipitate. This procedure requires an unacceptably long counting time and we finally adopted a method based on the more sensitive method of liquid scintillation counting.

Samples were prepared for liquid scintillation spectrometry according to a procedure developed by Alberts and Sibley as follows:

i) Sediments

Filters containing sediments were placed in 22 ml glass scintillation vials. The vials were capped after the addition of 1 ml of concentrated H_2SO_4 . Following acid digestion for 1 hour at room temperature, 10 ml of distilled, deionized H_2O was added to the sediment and dissolved filter. After thorough shaking, 4 ml of sample was added to 10 ml of scintillation cocktail (3a70B, Research Products International Corp.) in a 22 ml glass scintillation vial. This mixture was immediately shaken to insure uniform distribution of the radionuclides in the resulting gel.

ii) Solution

The filtrate from each sample was acidified with 1/10 volume concentrated H_2SO_4 ; 4 ml of this solution was added to 10 ml scintillation cocktail in a glass scintillation vial and shaken. The addition of H_2SO_4 to the filtrate sample was to insure equal counting efficiencies of the filter and filtrate samples. The ^{244}Cm alpha particle activity was measured using a Packard Tri-Carb Liquid Scintillation Spectrometer at 5°C. A 2% gain was used with lower and upper discriminator settings of 200 and 450, respectively. The lowest detectable limit of activity during a 200 min counting period was approximately 1 dpm.

3.0 RESULTS AND DISCUSSION

3.1 Sample Treatment and Storage

A question has arisen regarding the representative nature of K_d values obtained from samples which were frozen or freeze-dried before use. 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 stored at $\sim 4^\circ\text{C}$ to minimize alterations by chemical or microbial activity. Samples from Lake Michigan were frozen when we obtained them from Argonne National Laboratories and were kept frozen until used in the experiments to limit changes in sediment or water properties and to decrease microbial activity. The original 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 rehydrate 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 are from 0 to 4 times lower for freeze-dried compared to untreated samples, depending on the radionuclide.

3.2 Particle Formation During Constant-Shaking Experiments

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

Controls consisting of spiked seawater without sediments have been analyzed with our samples. Figure 4 shows the activity of ^{106}Ru on the filters and in solution for a control used in the Skagit River estuary sediment-water experiment. There was a continuous increase in ^{106}Ru on the filter for the first 70-90 hours with a corresponding decrease of ^{106}Ru in solution. After about 80 hours there was a small simultaneous

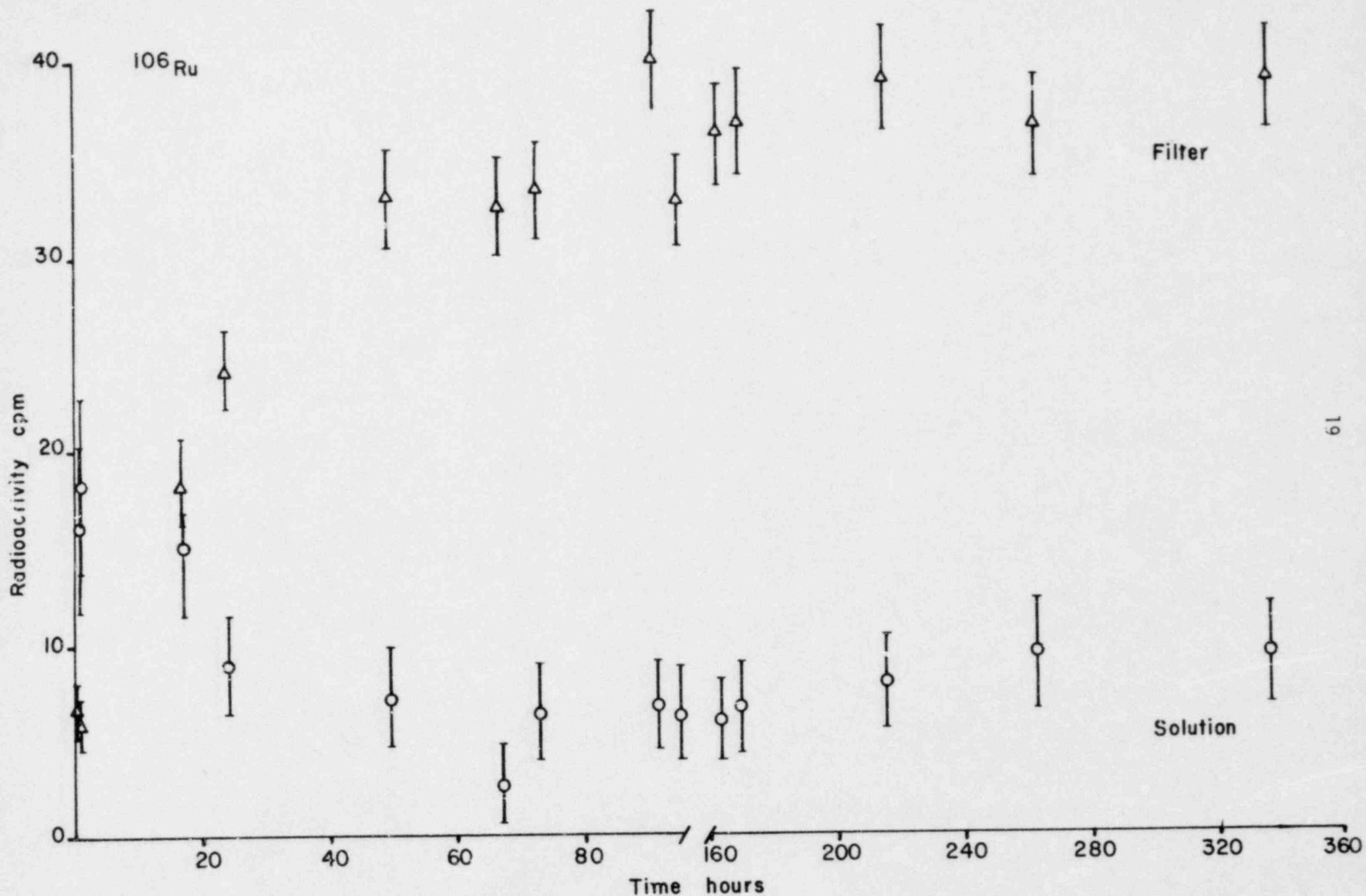


Figure 4. Concentration of ^{106}Ru on filters and in solution as a function of time for a control vessel in a constant shaking experiment using Skagit River estuary collections.

increase in ^{106}Ru for both the soluble and particulate phases. The increase in activity after 80 hours probably results from desorption of ^{106}Ru from the walls of the experiment vessel. At the beginning of an experiment when the concentration of soluble ^{106}Ru is high, the radioisotope could adsorb to the container walls. The formation of particulate ^{106}Ru , however, will reduce significantly the concentration of dissolved species. Therefore, ^{106}Ru which had adsorbed to the container walls may subsequently desorb and distribute into both the soluble and particulate phases. In a second control from the Skagit River estuary ^{106}Ru in the particulate phase increased and the soluble phase decreased after 320 hours (15 days). This indicates that for some radionuclides the time necessary to reach steady state conditions may be significantly greater than the 24-40 hours that we originally assumed to be sufficient for ^{241}Am and ^{237}Pu . After the first 80 hours, however, the small changes in the concentration of soluble and particulate ^{106}Ru probably do not alter the K_d values significantly.

For ^{241}Am in salt water approximately 90% of the total radioactivity in the controls is retained on the filter. Furthermore, the conversion of soluble ^{241}Am in the spike to particulate ^{241}Am occurs quite rapidly. After only one hour 75-80% of the total ^{241}Am activity is present in the particulate phase. There is, however, a slight increase in the concentration of particulate ^{241}Am throughout the experiment. Figure 5 shows the percent of the ^{241}Am in the particulate phase as a function of time for two controls from the Skagit River estuary.

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

3.3 Comparisons of Different Experimental Techniques

Distribution coefficients for ^{241}Am and ^{237}Pu determined by the sedimentation technique are one or two orders of magnitude lower than the distribution coefficients determined by the constant shaking technique. Duursma and Bosch (1) pointed out that the K_d value is dependent upon the analytical technique used to determine the K_d . However, the results for a given technique are generally consistent. One reason for the lower K_d values using the sedimentation technique compared to the constant shaking technique may be the shorter contact time between the sediments and the radionuclides in solution. We would expect that K_d values from the constant shaking and sedimentation techniques would be more similar for radionuclides with rapid sorption rates such as ^{137}Cs .

Replicate experiments using the sedimentation technique gave consistent results for K_d values as shown in Table 5. This may be a preferred method of determining distribution coefficients for some sediment types;

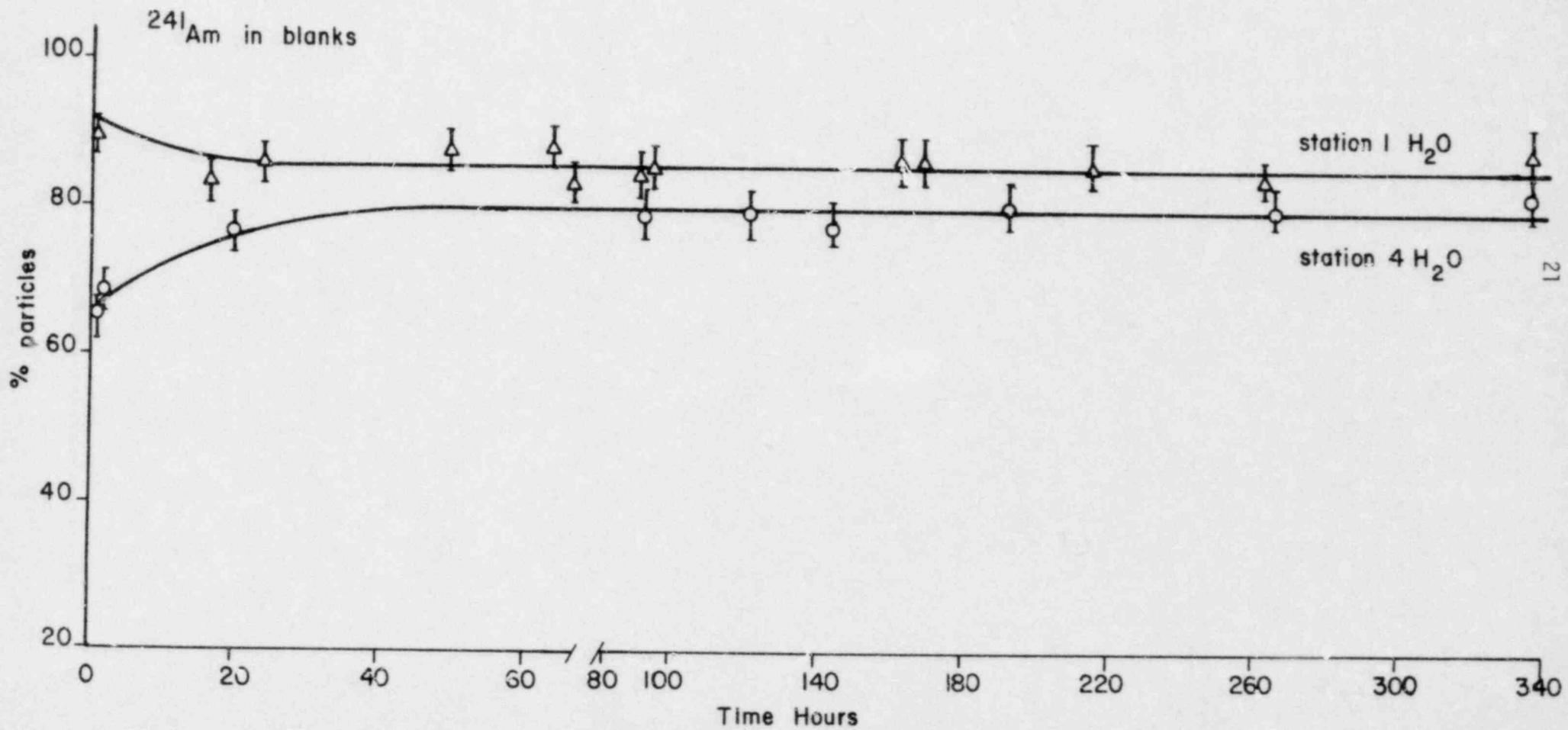


Figure 5. Fraction of ²⁴¹Am in the particulate state as a function of time for control vessels in a constant shaking experiment using Skagit River estuary samples.

Table 5. Distribution coefficients for ^{241}Am and ^{237}Pu by the sedimentation technique for sterile and nonsterile sediments collected at the mouth of the Columbia River.

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

for example, coarse sediments which, in the natural environment, are only briefly suspended in the water column.

The K_d value for ^{241}Am in the sterile marine system is approximately seven times higher than the K_d value in the non-sterile system. A similar trend between sterile and non-sterile systems was observed using the constant shaking technique (NUREG/CR-0801), (3).

4.0 CONCLUSIONS AND RECOMMENDATIONS

Measurements of distribution coefficients have been made using the methods established by Duursma and Bosch (1) and adapted in this work. The question of whether laboratory measurements of K_d values are equivalent to K_d values in the field has been addressed but no simple answer can be given.

Both sorption and desorption occurs in a dynamic natural environment and must be addressed separately. When radionuclides are introduced into the environment, they first interact with the suspended particulate matter and are sorbed. The initial removal processes is the sorption distribution coefficient. However, as time progresses, the radionuclide-particle experiences a natural environment which may alter this initial bonding. Organic compounds, pH changes, salinity, carbonate concentration, bacteria, etc. can alter this bonding. In the natural environment then, possible desorption processes influence the availability of the radionuclide for uptake by organisms, bottom sediment and suspended detrital material. Thus the desorption distribution coefficients may dominate the radionuclide-particle interaction. We have found that desorption K_d values are generally a factor of 10 or more greater than sorption K_d values.

In reality, the K_d value is an equilibrium value and thus sorption and desorption should be similar. However, in real world systems kinetic rather than equilibrium processes may control the eventual partitioning between soluble and particulate phases. This factor complicates the evaluation of laboratory values compared to real world values. As this study continues more information can be synthesized into a better model of the important parameters which influence the radionuclide-particle interaction. The methodology developed so far should be sufficient to give values accurate to within a factor of 5.

The method utilized to gain the most reproducible data for a wide range of sediment types was the batch or constant shaking technique. This technique is recommended for both sorption and desorption K_d value measurements. The thin-layer technique was used in the initial experimentation but the values obtained were not reproducible. We attributed the lack of reproducibility to agitation of the water while sampling which caused losses of the sediment from the filters. The sedimentation technique was also evaluated but found only to be useful for coarse sediments which, in the natural environment, are only briefly suspended in the water column.

The treatment of sediments prior to use included freezing, fresh or storage at $\sim 4^{\circ}\text{C}$ and freeze-drying. Our limited evaluation of these treatments indicated that the values obtained for freeze-dried were 0 to 4 times lower than fresh samples were comparable for the Hudson Canyon sediments. Freezing limits chemical and bacterial activity so would be

recommended for long-time storage. Use of the sediment immediately after collection, is, of course, our best recommendation.

Irradiation of the sediments with 10^6 Rads from a ^{60}Co irradiator was sufficient to minimize bacterial growth for marine and estuarine sediments but the irradiated Lake Washington sediments showed bacterial growth after 13 days incubation. Because of the low temperature storage of sediment and because of the 4-8°C temperature used for the experiments we believe that the sterilization treatment with 10^6 Rads would minimize the effect of bacterial growth on K_d values.

5.0 SUMMARY - METHODOLOGY

Methods have been developed and tested to measure distribution coefficient values for radionuclides in sediment-water systems. A series of samples of sediment and water from Saanich Inlet, Skagit River estuary, Hudson River, Clinch River, Cattaraugus and Buttermilk Creeks, Lake Michigan and Lake Washington were collected and stored, either frozen, cooled at 4°C or freeze-dried before being used in the experiments. The K_d values have been determined on sediment-water, organic detritus-water, and plankton-water systems. Techniques evaluated and utilized were the thin layer method, suspended sediment method and constant shaking method. The most reliable and reproducible method was the constant shaking method which was used for most of the experiments. Both sorption and desorption K_d values were determined using these methods. In addition, dialyses membranes were introduced into the system to measure K_d values for radionuclide-molecule complexes which diffused through 6000-8000 molecular weight pore sizes. These K_d values have been compared to the values found by the constant shaking method. The measurements of the radionuclides were by high sensitivity Ge(Li) detectors with computer data collection and reduction, by NaI(Tl) well crystal detectors with pulse height analyzers, by alpha radionuclide separation chemical procedures, and by liquid scintillation counting.

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