Water quality

REPORT NO. 122

WRRI

TRANSPORT OF ANTHROPOGENIC RADIONUCLIDES IN THE UPPER SUB-BASIN OF THE SAVANNAH RIVER

by

Robert A. Fjeld
Alan W. Elzerman
Environmental Systems Engineering
Clemson University
Clemson, S. C. 29634-0919

June 1986

Technical Completion Report
G1043-7

TRANSPORT OF ANTHROPOGENIC RADIONUCLIDES IN THE UPPER SUB-BASIN OF THE SAVANNAH RIVER

by

Robert A. Fjeld, Alan W. Elzerman and James A. Vandeven
Environmental Systems Engineering
Clemson University
Clemson, SC 29631

Submitted to:
United States Department of Interior
U.S. Geological Survey
Reston, VA 22092

The research on which this report is based was financed in part by the United States Department of the Interior, Geological Survey, through the South Carolina Water Resources Research Institute

South Carolina Water Resources Research Institute
Clemson University
Clemson, SC 29634-2900
August, 1986

Project Agreement No. 14-08-0001-G1043
Period of Investigation: 6/15/85-6/30/86

ACKNOWLEDGEMENTS

The authors would like to acknowledge the efforts of graduate students Merri Anderson, Jay Vandeven, and Scott Reboul who assisted in this investigation. The diligence of Pamela Whiten in preparing the manuscript is appreciated as is the loan of water sampling equipment by Dr. David Hayes of the Savannah River Laboratory, the gift of mixed media gamma calibration standards by Duke Power Company, and the sediment cores provided by Frank Dunnivant of the Department of Environmental Systems Engineering and Steve Ashby of the U.S. Army Corps of Engineers. The research on which this report is based was funded in part by the United States Department of Interior as authorized under the Water Research and Development Act of 1978 as Project No. G1043-7 through the South Carolina Water Resources Research Institute, Clemson University, Clemson, S.C. Additional support was provided by the National Science Foundation under Grant No. ISP-8011451 and by the Department of Environmental Systems Engineering, Clemson University.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

I. INTRODUCTION

Man-made radionuclides are released to the environment as a result of nuclear fuel cycle activities, non-fuel cycle industrial applications of radioactive materials, and testing of nuclear weapons (Jacobs, 1983). The radionuclides become distributed throughout the environment as a result of transport in atmospheric, aquatic, and biological systems. Due to the potential health and environmental effects of ionizing radiation emitted in the radioactive decay of these radionuclides their transport, fate, and distribution in the environment are of considerable interest and importance.

Radionuclide transport in surface waters is of interest primarily because of radiation doses received through drinking water, fish, and irrigated food pathways; and secondarily due to doses by external exposure. Weapons related radionuclides enter the aquatic environment as fallout through deposition and subsequent transport in the watershed. These inputs are ubiquitous in the northern hemisphere (Russell and Bruce, 1969; Eisenbud, 1973) but have been declining since the Nuclear Test Ban Treaty was signed in 1963. Nuclear facilities, both federal and commercial, routinely release low levels of radioactivity in their liquid effluents, and nuclear reactors and associated fuel cycle industries are the predominant source of anthropogenic radioactivity in many adjacent aquatic systems.

In the United States, the effect on human health and the environment from nearly 30 years of releases in liquid effluents is generally considered to be inconsequential (Fabrikant, 1984).

However, if a reactor accident or a malfunction of a radwaste system were to result in a large, inadvertent release, the impact could be serious. Also, over extended periods of time radionuclides may accumulate in certain subcompartments of the aquatic environment, such as bottom sediments through adsorption to suspended particles and subsequent sedimentation (Eisenbud, 1973; Olsen et al., 1981) or aquatic organisms through biomagnification (Spigarelli, 1976). To evaluate the potential impact of these occurrences and to develop plans for mitigating any impacts that are of significance, it is necessary to understand and predict the transport and fate of radioactivity in the system.

This project is concerned with the transport of anthropogenic radionuclides in the upper sub-basin of the Savannah River. The upper sub-basin consists of a series of reservoirs (Jocassee, Keowee, Hartwell, Russell, and Clarks Hill), one of which, Hartwell Lake, has received low levels of radioactivity in effluents from the Oconee Nuclear Station for over ten years. The presence of trace levels of reactor related radionuclides in the system, coupled with the proximity of Clemson University, which is located on the shores of Hartwell Lake, to all of the reservoirs, provides a unique opportunity for the study of radionuclide transport. The ultimate goal of the research is to develop predictive models for transport in the system that are based on fundamental principles of environmental transport. Presented in this report are results of (1) field measurements of radionuclide concentrations in bottom sediment and in the water column, focusing on H-3 and Cs-137 and including Co-58, Co-60 and Cs-134; (2) laboratory and field investigations of the sorption of Co-60 and Cs-137 by particulate matter; and (3) an evaluation of a compartmental model for H-3 and Cs-137 transport in the system.

II. OBJECTIVES

The overall goal of an ongoing research effort on radionuclide transport in the upper sub-basin of the Savannah River is to develop and validate predictive models that have a basis in fundamental theory of environmental transport. This requires an understanding of the processes that control transport in the system. Toward this end, the objectives of the research presented here are as follows:

1. To determine the distribution of man-made, gamma emitting radionuclides in the system,
2. To quantify and determine the significance of adsorption on the transport of Co-60 and Cs-137,
3. To determine the distribution of H-3 in the system,
4. To evaluate a simple compartmental model for H-3 and Cs-137 in Hartwell Lake.

III. METHODOLOGY

Study Area

The upper sub-basin of the Savannah River extends from the Clarks Hill dam to the mountains of South Carolina, North Carolina, and Georgia (Figure 1.). The system consists of five reservoirs - Clarks Hill, Russell, Hartwell, Keowee, and Jocassee - and their respective watersheds. The Oconee Nuclear Station is located on Lake Keowee but releases its liquid effluents to the headwaters of the Seneca River arm of Hartwell Lake. The principal radionuclides released to Hartwell Lake from the plant are H-3, Co-58, Co-60, Cs-134, and Cs-137. Yearly releases from the plant are given in Table I. A peak in the releases is evident in the 1975-1978 time period, and releases of Cs-134, Cs-137, and Co-60 have been steadily decreasing in the intervening years, while H-3 and Co-58 reached a minimum around 1982 and have increased slightly in the past three years. Oconee is the sole source to the lake of Co-58, Co-60, and Cs-134, but it is not the sole source of H-3 and Cs-137, significant amounts of which are present due to atmospheric weapons testing. Lakes Jocassee and Keowee, being located above Hartwell Lake, are not affected by releases from Oconee. Thus, the only anthropogenic nuclides present are weapons related H-3 and Cs-137. Lakes Russell and Clarks Hill could potentially receive reactor related radionuclides from Hartwell. In fact, an unstated objective of this research was to determine if reactor related radionuclides could be detected in these two lower lakes.

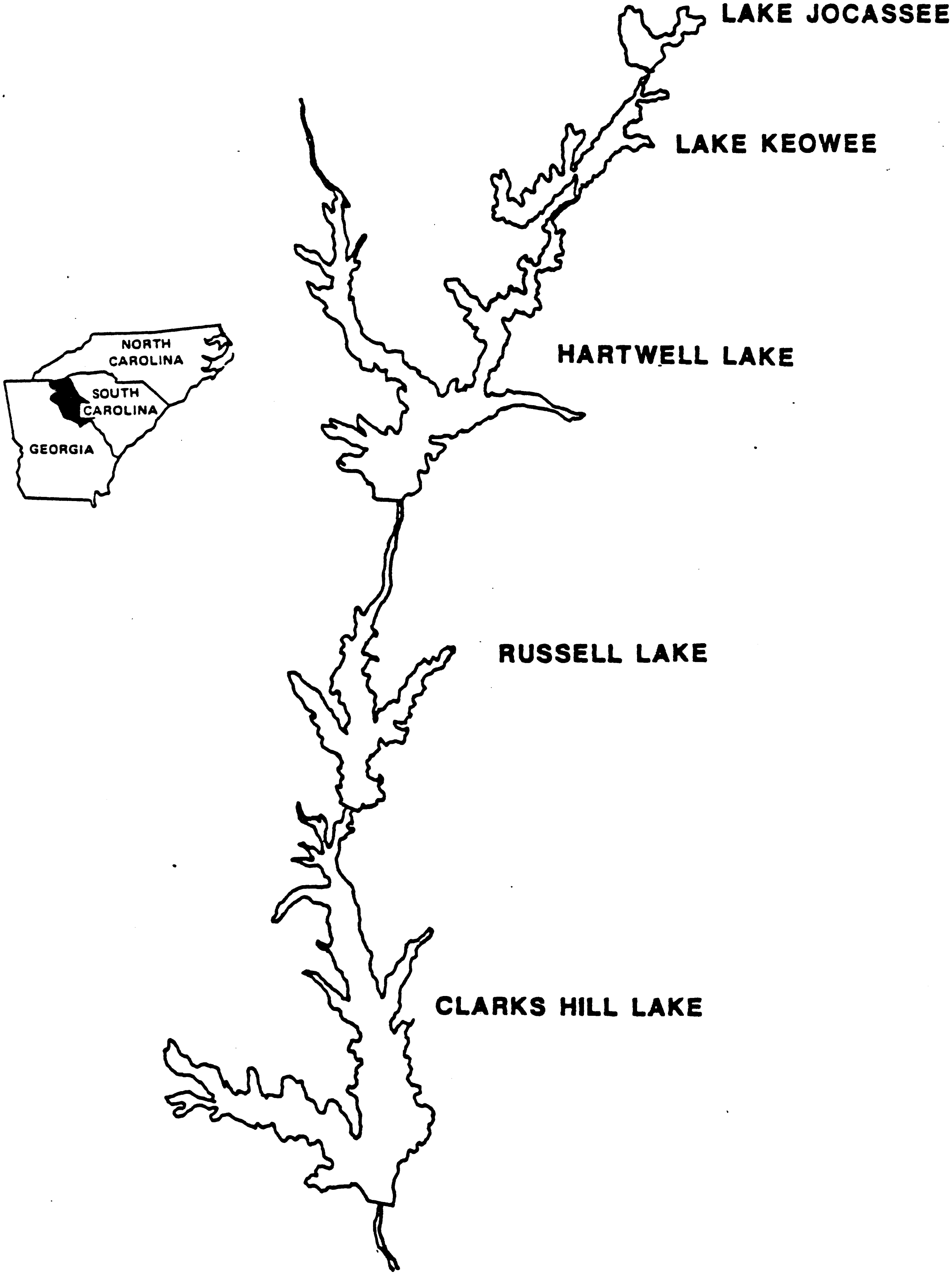


Figure 1. The upper sub-basin of the Savannah River

Table I. Yearly releases of major radionuclides from the Oconee Nuclear Station

YEAR	RELEASE RATE (Ci/y)				
	H-3	Co-58	Co-60	Cs-134	Cs-137
1973	71	0.40	0.04	-	0.01
1974	350	0.47	0.03	0.04	0.06
1975	3550	2.21	0.56	0.17	0.44
1976	2190	2.01	0.37	0.69	1.04
1977	1920	1.85	1.61	1.39	2.25
1978	1180	2.89	0.47	0.39	0.64
1979	894	0.18	0.05	0.12	0.24
1980	712	0.30	0.08	0.27	0.55
1981	507	0.20	0.03	0.49	0.67
1982	354	0.12	0.03	0.03	0.47
1983	1280	0.72	0.05	0.06	0.15
1984	1280	0.67	0.08	0.03	0.07
1985	1240				

Because Hartwell Lake receives direct inputs of reactor related radionuclides, much of the sampling effort was focused there. Presented in Figure 2 is a map of Hartwell Lake showing major highways, landmarks, and tributaries.

Sediment Sampling

The distribution of gamma emitting radionuclides in sediments was determined from over 80 bottom sediment grab samples. The majority of the samples were obtained from Hartwell Lake. These were supplemented with samples from Lake Keowee, Lake Issaqueena, and surrounding areas. Sampling was conducted using an Eckmann dredge. The depth of collection varied depending on the physical characteristics of the sampling location and the sediment, but generally was in the range of five to ten centimeters. Samples were transferred to plastic bags and, upon return to the laboratory, stored at 4°C until processing. Processing consisted of drying the sediment at 97°C for 48 hours followed by grinding either with a mortar and pestle or with a soil grinder. The sediment was then passed through a 2000 µm (#10) seive and stored for analysis. The typical sample dry weight following processing was 500 to 1000 g. Approximately 400 to 500g were put into a 1 L Marinelli beaker for analysis by gamma spectrometry.

Sections of a Hartwell Lake sediment core obtained near buoy S84 were provided by Frank Dunnivant of the Environmental Systems Engineering Department, Clemson University. Core sections from three locations in Clarks Hill Lake (the fore bay area near the dam, the mid-reservoir area near US 378, and the upper-reservoir

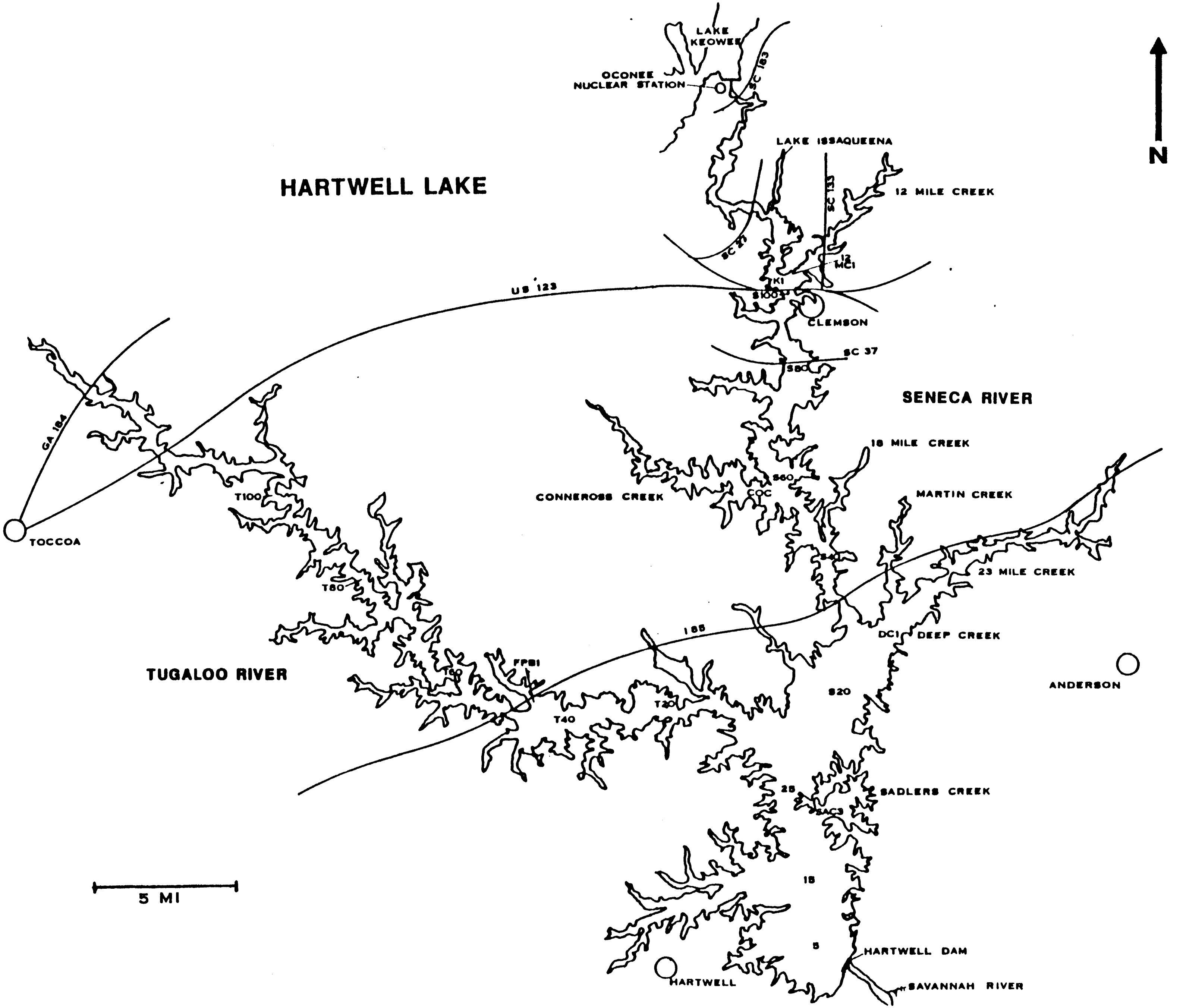


Figure 2. Map of Hartwell Lake

area north of Bobby Brown State Park) were provided by Steve Ashby of the U.S. Army Corps of Engineers. Cores from both lakes had been separated into four centimeter sections. They were dried at 98°C and transferred to plastic bottles for analysis by gamma spectrometry.

Water Column Sampling

Cesium and Cobalt

In situ separation of dissolved and particulate phase cesium and cobalt was performed. Fifteen samples were collected at eight locations on Hartwell Lake and a single sample was collected from Russell Lake. At locations on the Keowee River section of Hartwell Lake an effort was made to sample in the old riverbed. At all other locations, samples were taken at the center of the navigational channel. Water sampling was performed at a depth of at least three meters below the surface and at a height of at least two meters from the bottom to penetrate any existing thermocline and to prevent contamination from bottom sediments. Due to a mild autumn, the weather and lake conditions for all of the samples collected in 1984 were similar; with the weather conditions being warm with little or no precipitation for at least one week prior to sampling and the lake conditions being calm with suspended solids less than 5 mg/L.

The sampling methodology was based on suggestions by Dr. David Hayes of the Savannah River Laboratory. Dissolved radionuclides were extracted by ion-exchange resin and those associated with particulate matter were removed in 1 μm polypropylene cartridge filters. The system is shown in Figure 3 and is described in detail by Vandeven (1985). It consists of a 12V/7A DC pump (Model 12560-0001, Jabsco, Costa Mesa, CA), a 12V/60amp-hr deep cycle marine battery, a cartridge filter holder and 1 µm polypropylene cartridge filter (GX1-10, Hytrex, Hilliard, OH), an ion-exchange column with 16-50 mesh cation exchange resin (Research grade Rexyn 101H, Fischer Scientific, Springfield NJ), a flow meter, and polycarbonate tubing. The ion-exchange resin is similar to Dowex 50W-X8 (Dow Chemical Co., Midland, MI) which has been shown to be effective in removing ionic radiocesium and radiocobalt from freshwaters (Kreiger and Frishkorn, 1971; Luetzelschwab, 1976). The sampling system was capable of processing four liters of water per minute and the battery, fully charged, provided approximately eight hours of sample collection.

The ion exchange column consisted of two resin compartments in series as described by Mann and Casso (1981). The two compartments are analyzed separately, and the collection efficiency is given by (Mann and Casso, 1981)

$$\varepsilon = 1 - \frac{C_2}{C_1} \qquad (1)$$

where

ε = collection efficiency of each resin compartment,

C_1 = activity in the top compartment, and

C_2 = activity in the bottom compartment.

In laboratory tests, the collection efficiency for Cs-137 was greater than 97% at a flow rate of 5 L/min. The field samples

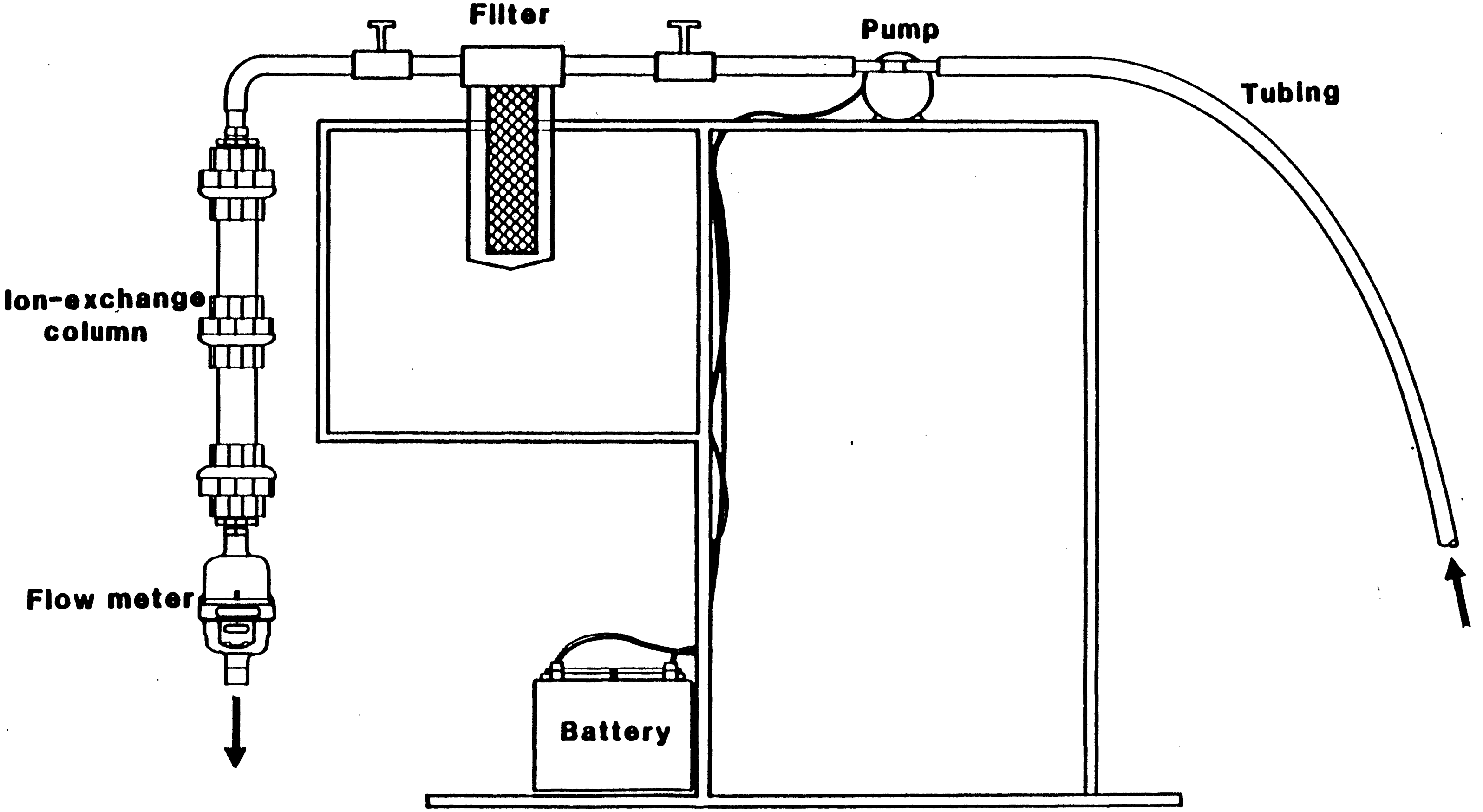


Figure 3. Field sampling system for separating dissolved and particulate phase Co-60 and Cs-137

were collected at a flow rate of 4 L/min and no detectable activity was found in any of the bottom compartments.

Samples were processed as follows. Cartridge filters were transferred from the field in plastic bags and air dried for four hours upon return to the laboratory. Each filter was placed in a 1000 mL pyrex beaker and ashed in a muffle furnace for 12 hours at 450°C. After ashing, the dried sediment was transferred to a preweighed aluminum dish for sediment mass determination and for gamma spectrometry. Resin columns were transported from the field intact. For analysis, the resin from each compartment was transferred to a 500 mL wide mouth jar which was filled to 250 mL with distilled water.

The methodology for separating dissolved and particulate phase radionuclides requires that an operational definition of these terms be established. Dissolved phase radionuclides are those species which become concentrated on the ion exchange resin. Thus, radionuclides which are in solution, but in non-ionic form, are not collected and do not contribute to the dissolved phase concentration. While it is likely that cesium is in ionic form, the form of Co-58 and Co-60 is not known. Particulate radionuclides are operationally defined as those species which are present with the dried sediment after the ashing process. Radionuclides associated with particles that are too small to be captured in the filter do not contribute to the particulate phase concentration. In addition, any radionuclides associated with organic matter or other volatile form are lost in the ashing process. While the latter consideration is probably not important for cesium and cobalt, the significance of the former is not known. The effect would be to underestimate the particulate phase concentration.

Tritium

The acquisition of a liquid scintillation counter (Model LS 3801, Beckmann Instruments, Irvine, CA) by the Department of Environmental Systems Engineering in the fall of 1985 permitted the scope of the research to be expanded to include tritium measurements. The objectives were to determine the relative magnitude of H-3 concentrations throughout the upper sub-basin and to determine the temporal and spatial distribution in Hartwell Lake. For a period of two months samples were collected on an almost daily basis from four locations in Hartwell Lake and on a weekly basis in Lakes Keowee and Jocassee. Additional samples were obtained from Russell Lake, Clarks Hill Lake, and tributaries of the Seneca river arm of Hartwell Lake. The spatial distribution in Hartwell Lake was determined by collecting over 50 samples throughout the entire lake.

All samples were collected in 20 mL plastic bottles, where they were stored until analysis. Six milliliter aliquots were mixed with six milliliters of liquid scintillation cocktail (Ready-Solv MP, Beckmann Instruments, Irvine, CA). The samples were counted for four hours.

Laboratory Studies

The experimental procedures were adapted from those of Duursma and Bosch (1970), Schell et al. (1981b), and Hairr (1974).

Bottom sediments needed for laboratory measurements were obtained from two locations on Hartwell Lake using a benthos sediment corer with 5 cm diameter polycarbonate tubes. That section of each core at a depth of 10-15 cm beneath the sediment surface was chosen for use. Stock suspensions of the sediments were prepared by wet sieving to retain the fraction of particles having diameters less than 63 µm. The stock suspensions were stored in glass jars at 4°C and the solids concentration for each suspension was determined using standard procedures (APHA, 1980).

Fifty liters of Hartwell Lake water were collected at field sampling location 7. The water was pumped from a depth of 6 meters, filtered in the field using a 293 mm Millipore filtering device containing a 0.8 µm filter, and retained in a 60 liter acid washed plastic container. The stock lake water was stored at 4°C and the pH determined three days after collection. Adsorption experiments were performed by spiking 200 mL lake water samples with Co-60 and Cs-137, adjusting pH to its initial value, and adding sediment. The suspension was stirred constantly and duplicate 3 mL aliquots were taken at designated time intervals. The aliquots were mixed with 17 mL of distilled water for gamma spectrometry.

Sample Analysis

Gamma Spectrometry

The gamma spectrometry system consisted of a coaxial high purity germanium detector (17.6% NPR, Princeton Gamma-Tech, Princeton, NJ) housed in a 10 cm lead shield, a linear spectroscopy grade amplifier (Model 2021, Canberra Industries, Meridan, CT), and either a 1024 channel pulse height analyzer (Series 30, Canberra Industries, Meridan, CT) or a 4096 channel pulse height analyzer (Model 7450, EG&G Ortec, Oak Ridge, TN). All gamma spectra were corrected for background, and net counts in full energy peaks determined. Counting efficiencies (Table II) were determined with NBS traceable or EPA reference standards as discussed below.

The counting efficiency for the resin samples used in water column sampling was determined using liquid Cs-137 and Co-60 reference standards (Quality Assurance Division, Environmental Protection Agency, Las Vegas, NV). The geometry was approximated by placing 125 ml of resin in a clean 500 mL glass jar containing 250 mL of distilled water and spiking with known amounts of Cs-137 and Co-60. Field samples were counted for 100,000 seconds due to the low activities present.

Counting efficiency for dried suspended sediment in aluminum counting dishes was determined using two different methods. Initially, a two gram sample of a reference river sediment (SRM 4350B, Columbia River Sediment, National Bureau of Standards, Washington, DC) was used. Because of the low Cs-137 activity in this standard, the counting uncertainty was almost 40% and the efficiency was checked using a 47 mm filter standard (No. 11001A-37, Analytics, Inc., Atlanta, GA). The efficiency using the latter was approximately 30% higher than the former and the uncertainty was almost an order of magnitude lower. Because of the lower uncertainty, the efficiency of the filter standard was

Table II. Gamma detection efficiencies for various counting geometries

Sample Geometry	Counting Efficiency of Radionuclide (%)			
	Co-58 (810 keV)	Co-60 (1172 keV)	Cs-134 (605 keV)	Cs-137 (662 keV)
500 ml solid in 1 L Marinelli	0.69 ± 0.04	0.49 ± 0.03	0.90 ± 0.05	0.83 ± 0.05
500 ml resin jars	NM	0.26 + 0.02	NM	0.76 ± 0.06
47 mm filter on counting dish	2.4 + 0.01	1.00 + 0.01	NM	3.00 ± 0.01
2g river sediment on counting dish	NM	NM	NM	2.30 ± 0.90
20ml liquid in 125 ml Nalgene bottle	NM	0.70 + 0.03	NM	2.00 ± 0.07

NM = not measured.

used for analyses. Counting time for the suspended sediment samples was 100,000 seconds.

Counting efficiency for the bottom sediment samples was determined using a 500 mL solid standard contained in a 1 L Marinelli beaker (No. 15689-37 and No. 11003-37 , Analytics, Inc., Atlanta, GA). Bottom sediments were typically counted for approximately 10,000 seconds. All samples collected in 1982 were analyzed initially using a 1024 channel analyzer. In the summer of 1985, the Department of Environmental Systems Engineering purchased a 4096 channel analyzer. Due to limited resolution of the 1024 channel analyzer, Cs-134 concentrations in the initial analyses were based on its 570 keV peak, for which the minimum detectable concentration was approximately 350 pCi/kg. The 4096 channel analyzer permitted analysis of Cs-134 using its 605 keV peak, for which the minimum detectable concentration was less than 100 pCi/kg. To verify the Cs-134 concentrations and to check reproducibility of the Co-60 and Cs-137 analyses, a number of samples were reanalyzed with the 4096 channel analyzer.

Counting efficiency for the 20 mL volumes used in the laboratory experiments was determined using EPA references standards. Counting times were typically 1000-5000 seconds.

Liquid Scintillation

Tritium calibrations were made using EPA reference standards (Quality Assurance division, Las Vegas, NV). Background was determined with fossilized water supplied by EPA and the background count rate was approximately 25 counts per minute. Samples were counted for four hours.

IV. RESULTS AND DISCUSSION

Radionuclide Distributions in the Study Area

Sediment Dredge Data

To facilitate the presentation and discussion of bottom sediment data, the results are grouped according to location of the samples. These groups are as follows (refer to Figure 2):

1. Keowee River from the Oconee Nuclear Station (SC 183) to Lawrence Bridge (SC 27).
2. Keowee River from Lawrence Bridge (SC 27) to the junction with Twelve Mile Creek.
3. Twelve Mile Creek from SC 133 to the junction with the Keowee River.
4. Junction of the Keowee River and Twelve Mile Creek north of US 123.
5. Seneca River from US 123 to SC 37.
6. Seneca River from SC 37 to junction with the Tugaloo River.
7. Tugaloo River from GA 184 to junction with the Seneca River.
8. Main body of Hartwell Lake from the junction of the Seneca and Tugaloo Rivers to the dam.
9. Miscellaneous locations outside of Hartwell Lake.

Presented in Table III are concentrations of Co-58, Co-60, Cs-134, Cs-137 and, for comparison purposes, naturally occurring K-40. No entry indicates that the sample was not analyzed for the given radionuclide. The entry <MDC indicates that the spectrum was examined visually and no peaks were observed for the particular radionuclide. As discussed in the Methodology section, some

Table III. Radionuclide concentrations in bottom sediments

General Location	Sample No.	Date	CONCENTRATION (pCi/kg)					Specific Location or Buoy No.	Notes
			Co-58	Co-60	Cs-134	Cs-137	K-40		
Keowee River: SC 183 to SC 27	6	6/15/82	<MDC	<MDC	<MDC	366	19880		sand
	7	6/15/82	<MDC	<MDC	257	687	19770		sand
	8	6/15/82	<MDC	<MDC	<MDC	366	16880		sand
	75	6/15/82		346	1332	3176	34380		
Keowee River: SC 27 to Twelve Mile Creek	1	6/15/82	95	600	608	4593	13630	Issaqueena Cove	
	2	6/15/82	48	194	321	1309	8160	Issaqueena Cove	
	67	11/24/82		155	705	1004	13740	Issaqueena Cove	
	68	11/24/82		808	512	2811	20550	Issaqueena Cove	
	69	11/24/82		703	1307	5338	23540	Issaqueena Cove	
	70	11/24/82		1320	1472	6870	21800	Issaqueena Cove	
	71	11/24/82		385	1017	3280	24010	Issaqueena Cove	
	72	11/24/82		983	1293	3559	21580	Issaqueena Cove	
	73	11/24/83		1460	1214	5756	20170	Issaqueena Cove	
	78	6/15/82			415	1477		Issaqueena Cove	
	79	6/15/82			1074	6971		Issaqueena Cove	
	3	6/15/82	128	428	1960	3990	22330	Near Bayshore	
	74	6/15/82			1597	4351		Near Bayshore	
	76	6/5/82			1470	3832		Near Bayshore	
	84	10/15/85		743	455	4930		Near Bayshore	
Twelve Mile Creek	28	9/4/82		410	435	3240	5580		
	29	9/4/82	45	483	717	3052	5380		
	30	9/4/82	40	<MDC	87	481	11120		
	31	9/4/82	60	307	284	1944	8580	12MC4	
	32	9/4/82		883		3335		12MC4A	
Junction of Keowee River and Twelve Mile Creek	4	6/15/82	125	1635	1354	6643	13180	K1	
	5	6/15/82	90		1061	4720	10310	K1	
	77	6/15/82			1137	4723		K1	
	9	6/29/82	70	720	628	3611	7000	K12M	

Table III (Continued)

General Location	Sample No.	Date	Co-58	Co-60	Cs-134	Cs-137	K-40	Specific Location or Buoy No.	Notes
	10	6/29/82	84	1116	805	4099	8450	K12M	
	11	6/29/82	69	907	733	3879	7520	K12M	
	33	9/4/82		185	495	1374	18220	K12M	
	82	10/15/85		711	<MDC	3190		K12M	
	83	10/15/85		704	<MDC	3280		K12M	
Seneca River US 123 to SC 37	12	6/29/82	86	1539	1028	5949	1000	S99	
	13	6/29/82	93	1386	860	5443	9970	S98	
	14	6/29/82	73	928	963	4870	6090	S92	
	15	6/29/82	63	1230	1052	5745	6880	S91	
	34	9/4/82		1538	1063	6043	9730	RR Trestle	
	35	9/4/82		393	<MDC	1972	10960	RR Trestle	
	36	9/4/82		692	619	4512	8570	S97	
	37	9/4/82	30	188		2003		S97	
Seneca River SC 37 to junction with Tugaloo River	17	6/29/82	55	528	515	2948	7020	S79	
	16	6/29/82	50	409	648	3404	6880	S77	
	44	9/26/82		<MDC		1046	4380	S77	
	45	9/26/82		428	515	4028	5370	S74	
	46	9/26/82		<MDC		919	1580	S73	
	47	9/26/82		<MDC	<MDC	1007	7970	S72A	
	59	10/28/82		362	463	4096	5070	S66	
	58	10/28/82		229		3591	6530	S55	
	57	10/28/82		88	<MDC	2150	8960	S45	
	56	10/28/82		<MDC		391	15090	S34/35 traverse (shoreline)	
	55	10/28/82		<MDC	<MDC	1918	6860	S34/35 traverse (forest)	
	54	10/28/82		<MDC		270	12880	S34/35 traverse (shoreline)	
	53	10/28/82		69	637	3340	17530	S34/35 traverse (channel)	
	52	10/28/82		103	332	4406	12180	S34/35 traverse (channel)	
	51	10/28/82		108	<MDC	2969	7690	S34/35 traverse (channel)	
	50	10/28/82		103	393	2774	8620	S34/35 traverse (channel)	
	49	10/28/82		88	384	2014	8620	S34/35 traverse (channel)	
	48	10/28/82		<MDC		1558	14890	S34/35 traverse (channel)	

Table III (Continued)

General Location	Sample No.	Date	Co-58	Co-60	Cs-134	Cs-137	K-40	Specific Location or Buoy No.	Notes
	18	7/7/82	<MDC	<MDC	<MDC	396		S18	sand
	19	7/7/82	28	<MDC	379	3434	8310	S8	
	20	7/7/82	33	<MDC	483	2272	8690	S2	
	21	7/7/82	<MDC	<MDC	<MDC	157	5700	S1	
	27	7/26/82		<MDC	<MDC	1729	7900	S1/S2	
Tugaloo River: GA 184 to Seneca River	22	7/7/82	<MDC	<MDC	<MDC	1612		T1	
	60	7/26/82	<MDC	<MDC	<MDC	1750			
	65	10/30/82	<MDC	<MDC	<MDC	868	19260	T43/T44	
	66	10/30/82	<MDC	<MDC	<MDC	942	8210	T26/T27	
	61	10/30/82	<MDC	<MDC	<MDC	978		T74	
	62	10/30/82	<MDC	<MDC	<MDC	984		T69	
	63	10/30/82	<MDC	<MDC	<MDC	413	8110	T59	
	64	10/30/82	<MDC	<MDC	<MDC	420	10450	T53/T54	
	40	7/17/82	<MDC	<MDC	<MDC	750			
	41	7/17/82	<MDC	<MDC	<MDC	1150			
	80	7/17/82	<MDC	<MDC	<MDC	351			
Main body of Lake	24	7/26/82	<MDC	<MDC	<MDC	1954	8430	21/22	
	25	7/26/82	<MDC	<MDC	<MDC	3369	8000	15	
	26	7/26/82	<MDC	<MDC	<MDC	2040	7240	9	
Locations outside Hartwell Lake	38	7/26/82	<MDC	<MDC	<MDC	1242	11860	Lake Issaqueena	
	39	7/26/82	<MDC	<MDC	<MDC	960		Lake Issaqueena	
	42	7/17/82	<MDC	<MDC	<MDC	1207		Lake Keowee	
	43	7/71/82	<MDC	<MDC	<MDC	1191		Lake Keowee	
	81		<MDC	<MDC	<MDC	288		Howard Creek	

of the sediment samples, approximately 25%, were analyzed initially in 1982 using a 1024 channel analyzer and were reanalyzed in 1986 with a 4096 channel analyzer primarily as a check on Cs-134 concentrations, although Co-60 and Cs-137 were also analyzed. Agreement between the initial determinations of Cs-137 and Co-60 concentrations and the reanalyses was excellent, generally within 10%. Most of the Cs-134 analyses were within 10-20%. Where the two differed by more than 25%, the reanalysis is reported. Observations are given below for each of the anthropogenic radionuclides.

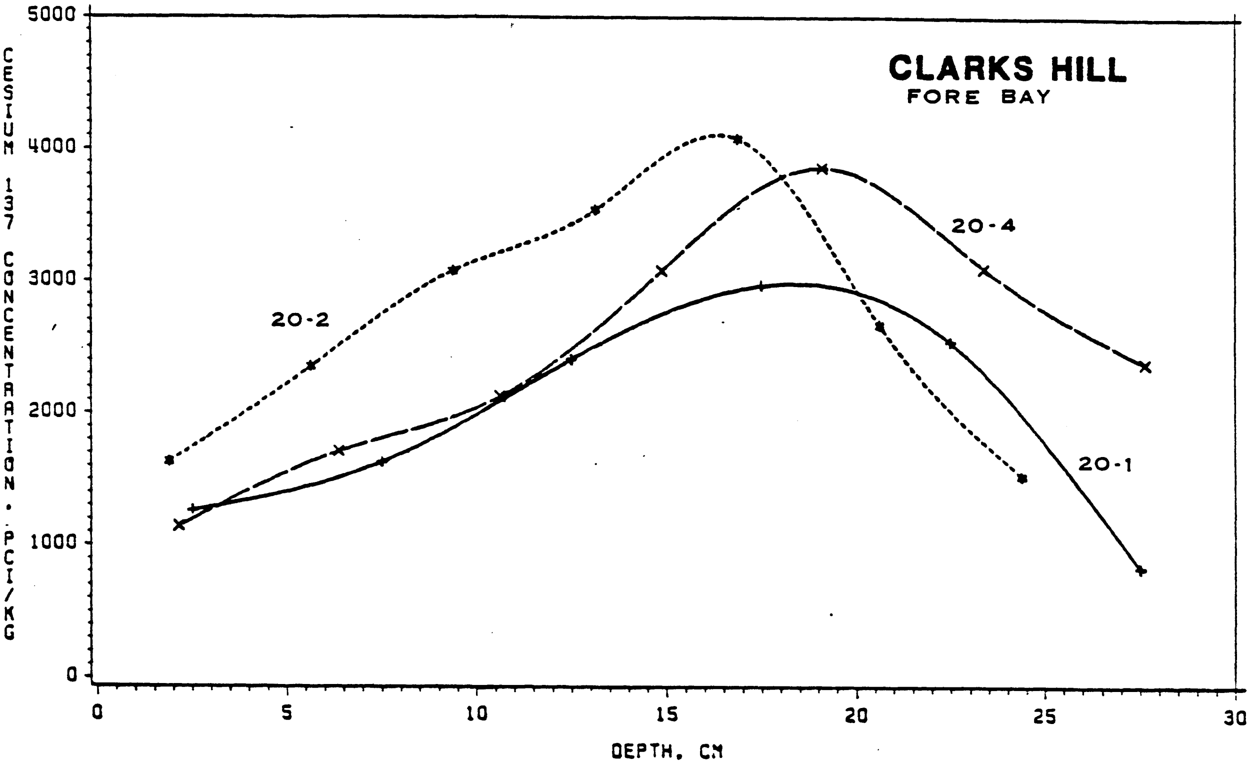
<u>Cs-137</u>: The Cs-137 concentrations in Hartwell Lake bottom sediments ranged from a low of approximately 400 pCi/kg to a high of over 6000 pCi/kg. With one exception, the concentration was low, (<1000 pCi/kg) in the section of the Keowee River between the Oconee Nuclear Station and SC 27. The flow rate in this section of the river exhibits large fluctuations due to periodic operation of the hydroelectric facility on Lake Keowee. Much of the sediment deposited during periods of low flow would be subject to scouring and resuspension during periods of high flow. The three sediment samples having low concentrations were quite coarse and sandy, while the one with high concentration (>3000 pCi/kg) was clayey. Between SC 27 and the Twelve Mile Creek junction, concentrations were typically in the 3000 - 6000 pCi/kg range. In this section the river widens considerably, allowing sedimentation of smaller particles, which have a greater surface area to mass ratio and therefore usually show greater activities on a mass basis

compared to larger particles. Particle size characteristics of samples analyzed from this area show 10 to 50 per cent of the mass below 2 µm characteristic diameter (Polansky, 1984), indicating significant amounts of the clays typical of the local geology comprise the sediments. Data for Twelve Mile Creek above its junction with the Keowee River were variable, depending on clay content and the distance upstream. In the Seneca River between US 123 and SC 37, the concentration was consistently high, ranging from 2000 to 6000 pCi/kg and the sediments were largely clay. From SC 37 to the Tugaloo junction, the concentration in clay sediments began to decrease somewhat, ranging between 1000 and 4000 pCi/kg. A traverse across the river at navigational buoys S35 and S36 showed high concentration/high clay content in the center of the channel and low concentration/low clay content near the shoreline. In the Tugaloo, the concentration ranged from less than 500 to almost 2000 pCi/kg. In this section, even those samples with high clay content had low concentrations. In the main body of the lake, the concentrations were in the 2000 - 3000 pCi/kg range. These samples were all from the midchannel region and had a high clay content. Samples from Lake Issaqueena and Lake Keowee had high clay contents and concentrations on the order of 1000 pCi/kg. The Howard Creek sample was quite sandy and had a low concentration.

Co-58, Co-60, and Cs-134: The results for Co-58, Co-60, and Cs-134 generally follow the same trends as the Cs-137 data. As with Cs-137, the highest concentrations were found in the sections between SC 27 and SC 37 and were in the samples with high clay content. In this section the Cs-134 and Co-60 concentrations ranged from 500 to 1500 pCi/kg and the Co-58 concentrations ranged from 50 to 100 pCi/kg. The concentration decreased significantly in the section between SC 37 and the Tugaloo junction, with Co-60 and Cs-134 decreasing to the 100 to 500 pCi/kg range and Co-58 being below the detection limit. None of the radionuclides were detected in the Tugaloo arm of the Lake, the main body of the lake, or locations outside the lake.

Sediment Core Data

Sediment core data for Hartwell and Clarks Hill are given in Figure 4. It can be seen that there was a difference in the concentration profiles from the two lakes. In Clarks Hill, the concentration reached a maximum at a depth between 15 and 20 cm, while the Hartwell core showed a plateau at a depth of approximately 15 cm followed by a sharp increase. The maximum in the Clarks Hill concentration profile was undoubtedly due to the surge in weapons testing that occurred shortly before the ban on atmospheric tests in 1963. The peak at a depth of 15 to 20 cm is consistent with an estimated deposition rate on the order of one cm/yr. For the Hartwell core, the rise in concentration from 42 cm to the plateau at 15 cm was probably due to fallout, and the subsequent increase was probably due to releases from the Oconee Nuclear Station, which has dominated inputs to the lake since the mid 1970's. It is interesting that the concentration in the upper portions of the Clarks Hill core were on the order of 1000 pCi/kg



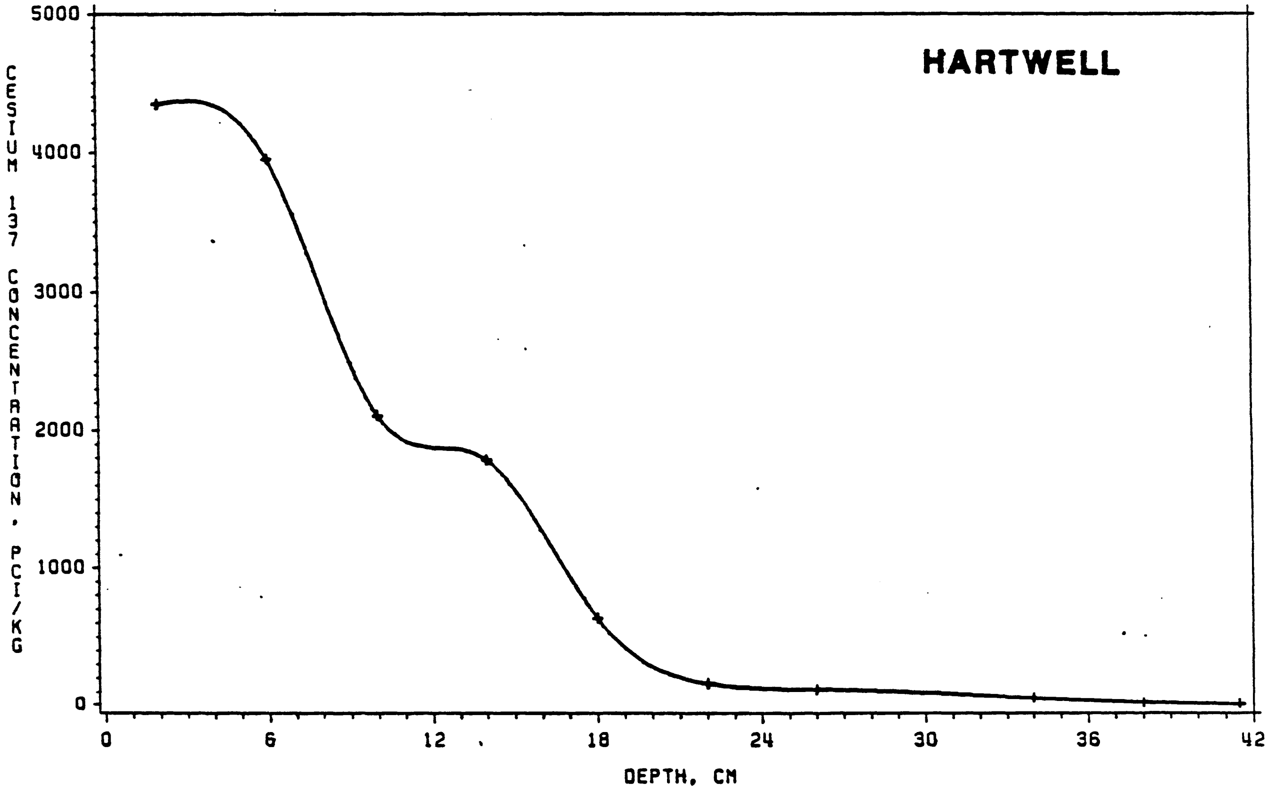


Figure 4. Concentration of Cs-137 as a function of depth for sediment cores from Hartwell and Clarks Hill Lakes

as found in Lake Issaqueena, Lake Keowee, and the Tugaloo River arm of Hartwell Lake.

Water Column Data

Because of low concentrations, the radionuclides were difficult to detect in the dissolved and particulate phases of the water column. Whereas some success was achieved in determining Cs-137 concentrations in both phases, it was only possible to detect Co-58 and Co-60 in a few of the particulate samples.

Cs-137: The Cs-137 data obtained from each of the field samples are given in Table IV. Cesium-137 in the dissolved phase was detected in all samples except at the Tugaloo River location. Adsorbed Cs-137 concentrations were below the detection limits at the first three locations due to the small mass of suspended sediment collected on filter. At subsequent locations larger volumes of water were processed to obtain a sufficient amount of sediment for radioassay. Consequently, adsorbed Cs-137 concentrations were successfully measured in samples 4 through 11 and 13.

Cesium-137 was below the MDC in both the dissolved and adsorbed phases in the Tugaloo River (sample 12). The two known sources of Cs-137 to Hartwell Lake are fallout due to weapons testing and liquid effluents released from the Oconee Nuclear Station. Ritchie et al. (1972) reports fallout to be evenly distributed over regional watersheds. Therefore, the concentrations of Cs-137 due to fallout in the Tugaloo River and in the remainder of Hartwell Lake should be nearly equal. The absence of detectable Cs-137 concentrations in the Tugaloo River thus suggests that

Table IV. Water column concentrations of Cs-137, Co-60 and Co-58

Sample No.	Location (Buoy NO.)	Cs-137		Co-60	Co-58	
		Date	Dissolved (pCi/L)	Particulate (pCi/kg)	Particulate (pCi/kg)	Particulate (pCi/kg)
1	Hartwell: US 123 (K12MC)	5/17/84	0.29 ± 0.03	<MDC[b]	<MDC[d]	<MDC[e]
2	Hartwell: SC 183	5/17/84	0.20 ± 0.03	<MDC[b]	<MDC[d]	<MDC[e]
3	Hartwell: Near dam (1/2)	6/2/84	0.05 ± 0.01	<MDC[b]	<MDC[d]	<MDC[e]
4	Hartwell: US 123 (K12MC)	6/2/84	0.05 ± 0.02	3400 ± 2200	<MDC[d]	<MDC[e]
5	Hartwell: US 123 (K12MC)	6/14/84	0.04 ± 0.01	17400 ± 2100	<MDC[d]	<MDC[e]
6	Hartwell: Issaqueena Cove	6/14/84	0.03 ± 0.01	4000 ± 1600	<MDC[d]	12,600 ± 1300
7	Hartwell: Seneca R.(S89/91)	6/28/84	0.05 ± 0.02	5000 ± 1000	<MDC[d]	2800 ± 300
8	Hartwell: Seneca R.(S89/91)	6/28/84	0.04 ± 0.02	6100 ± 1200	<MDC[d]	4300 ± 400
9	Hartwell: US 123 (K12MC)	10/12/84	0.05 ± 0.01	5400 ± 300	1980 ± 200	1200 ± 100
10	Hartwell: Seneca R.(S96/97)	10/13/84	0.05 ± 0.01	4400 ± 400	1300 ± 100	600 ± 50
11	Hartwell: Seneca R.(S96/97)	10/13/84	0.04 ± 0.01	4200 ± 300	1200 ± 100	800 ± 50
12	Hartwell: Tugaloo (T39)	10/13/84	<MDC[a]	<MDC[c]	<MDC[d]	<MDC[e]
13	Hartwell: US 123 (K12MC)	10/85	0.13 ± 0.02	4200 ± 1700	<MDC[d]	NM
14	Hartwell: US 123 (K12MC)	10/85	0.12 ± 0.02	NM	<MDC[d]	NM
15	Hartwell: Bayshore	10/85	0.09 ± 0.02	NM	<MDC[d]	NM
16	Russell: Landing north of SC 72	5/85	0.009 ± 0.002	1600 + 700	<MDC[d]	NM
17	Clarks Hill: Landing north of Bobby Brown	5/85	0.009 ± 0.004	320 + 270	<MDC[d]	NM

[a] 0.02 pCi/L
[b] 20,000 pCi/kg
[c] 1800 pCi/kg
[d] 2300 pCi.kg for a 1 g sample
[e] 1000 pCi/kg for a 1 g sample

Cs-137 due to fallout is below the minimum detectable concentration of 0.02 pCi/L in the remainder of Hartwell Lake. The Hartwell dissolved concentrations ranged from 0.03 to 0.29 pCi/L, with a majority of measurements in the 0.04 to 0.13 pCi/L range. The concentrations in Russell and Clarks Hill were both less than 0.01 pCi/L.

The average Cs-137 particulate phase concentration was 4,600 ± 1,000 pCi/kg in samples 3 through 11 (excluding 5) and 13. This concentration is in the range of the bottom sediment data for the corresponding section of the lake. A slightly higher concentration on suspended sediments compared to bottom sediments may result from a smaller particle size distribution of sediments in the water column. Compared to the bottom sediments, the smaller suspended particles have a greater surface area to mass ratio and therefore a greater capacity per unit mass to adsorb Cs-137.

The 17,400 pCi/kg concentration measured on suspended sediment in sample 5 is more than a factor of three greater than the average Cs-137 concentration of 4,600 pCi/kg. One explanation for this unusually high concentration is the possible presence of "hot" particles in the sample. Although Cs-137 concentrations of this magnitude were not found in the bottom sediments, "hot" particles in those samples, if present, would be less significant as they were diluted in the 400 to 500 grams that was usually counted. However, in the water column study, a sediment mass on the order of one gram was counted. Therefore, activities associated with "hot" particles present in the sample could conceivably have a significant effect on the total Cs-137 activity measured on the suspended sediment.

<u>Co-60 and Co-58</u>: Dissolved phase concentrations of Co-58 and Co-60 were below detection limits in all of the samples. Particulate phase Co-58 was detected in samples 6-11 and particulate phase Co-60 was detected in samples 9-11. The average concentrations were 1,900 pCi/kg and 1,500 pCi/kg, respectively. Average bottom sediment concentrations for this general area of the lake were considerably lower, approximately 70 pCi/kg for Co-58 and 800 pCi/kg for Co-60. The lower concentrations in bottom sediment are to be expected since they represent several years of deposition during which time significant decay of Co-58 (t_H = 71.3 d) and Co-60 (t_H = 5.2 y) would occur. The 12,600 ± 1,300 pCi/kg of Co-58 measured in sample 6, similar to the high Cs-137 measurement in sample 5, is approximately three times greater than any other Co-58 adsorbed value. Here again, one possible explanation is the presence of "hot" particles.

<u>H-3</u>: Two sets of H-3 data are presented. The first set (Figure 5) consists of a series of measurements obtained over a two month period in Hartwell Lake (at SC 93, US 123 and SC 27 and in Clemson University tap water) and in Lakes Keowee and Jocassee. In Lakes Keowee and Jocassee the concentrations, which ranged from 100 to 400 pCi/L, were consistent with the levels observed due to fallout in the northern hemisphere (Kathren, 1983). Concentrations for the SC 93, US 123 and Clemson University tap water samples were significantly higher than fallout levels, ranging

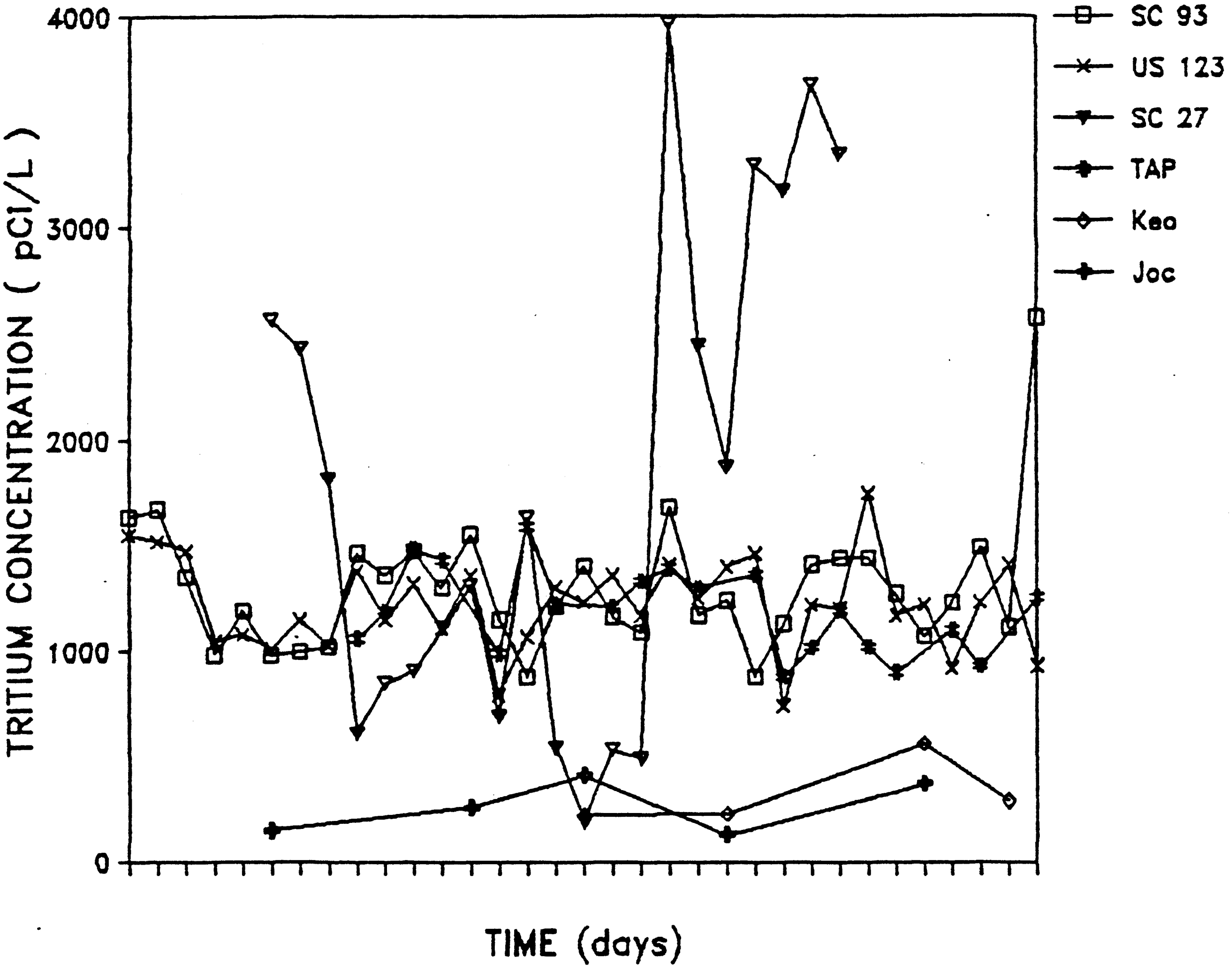


Figure 5. Concentration of H-3 at various locations during the time period 4/17/86-6/13/86

between 1000 and 1500 pCi/L. The day to day variations were modest and, with the exception of the final SC 93 sample, the three samples generally tracked together. These data are in sharp contrast to those for the SC 27 location, where the concentration ranged from 500 to 4000 pCi/L and there were large daily fluctuations. These fluctuations were probably due to the periodic releases of liquid effluent from the Oconee Nuclear Station, resulting in the transport of a pulse of radioactivity down the river. This conclusion is supported by data on 4/24/86 in which the concentration at SC 183 was found to be 9600 pCi/L and at SC 27 it was 640 pCi/L. The absence of large fluctuations at US 123 suggest significant mixing and consequent damping of the pulse at the junction of the Keowee River and Twelve Mile Creek.

Results of measurements taken throughout Hartwell Lake are presented in Table V. In the Tugaloo arm of the lake and in the main body of the lake, concentrations were generally below the detection limit. In the Seneca River arm of the lake, the concentrations were higher, ranging between 1000 and 1500 pCi/L except in the uppermost section of Coneross Creek (COC10/11), where the concentration was less than 1000 pCi/L. The concentration results in the Keowee River/Twelve Mile Creek section between SC 133 and SC 27 are interesting. The concentration increased from 1300 pCi/L at SC 133 to 2500 pCi/L at the junction of Twelve Mile Creek and the Keowee River, and then decreased to 1400 pCi/L at SC 27.

Table V. Tritium concentrations in Hartwell and Russell

Hartwell Lake 7/10/86		Russell Lake 7/23/86	
Location	Concentration	Location	Concentration
Main Lake 1/2	140 + 160	R3/4	600 + 150
Main Lake 6/7	210	R15/20	520
Main Lake 10/11	250 + 200	R25/30	440
Main Lake 17/18	210	R47/48	980
Main Lake 23/24	170	R54/55	910
Tugaloo Ts/T2	630	R83/86	920
Tugaloo T3/T4	630	SC 184	910
Sadler's Creek SAC2	320		
Sadler's Creek SAC5	690		
Camp Creek CAC1	850		
Deep Creek DC3A/4	1110		
Deep Creek DC23M/26M	1180		
Seneca River S20/21	1200		
Coneross Creek CO2/3	1190		
Coneross Creek CO6/7	1100		
Coneross Creek CO10/11	620		
Twelve Mile Creek SK12MC	2560		
Twelve Mile Creek 12MC4	2010		
Twelve Mile Creek SC 133	1350		
Keowee River K1	1870		
Keowee River K2	1960		
Keowee River K3	2220		
Keowee River K3A	1490		
Keowee River K3B	1470		
Keowee River at SC 27	1380		

Observations Based on Field Data

The most obvious observation based on the results presented above is that reactor releases have a significant impact on the distribution of anthropogenic radionuclides in Hartwell Lake. This influence of Oconee releases is seen in the presence of Co-58, Co-60 and Cs-134, the only significant source of which is the reactor, in the Seneca River arm of the lake. It is also seen in the elevated concentrations of H-3 and Cs-137 in this arm of the lake relative to concentrations in Lakes Keowee and Issaqueena. Second, the results suggest that particulate phase Cs-137 in Russell Lake, Clarks Hill Lake and the upper stretches of the Tugaloo River arm of Hartwell Lake is due solely to weapons fallout and is not affected by the reactor releases. This is based on the similarity of bottom sediment concentrations among samples from these areas. Specifically, concentrations in the near-surface sections of the Clarks Hill cores and in Tugaloo, Keowee and Issaqueena dredge samples suggest the weapons contribution of Cs-137 to be approximately 1000 pCi/L. Using the value of distribution coefficient measured in Hartwell implies the weapons contribution in dissolved Cs-137 to be approximately 0.01 pCi/L. This is consistent with the value of 0.009 pCi/L measured in Russell and Clarks Hill and with values reported for the Savannah River near North Augusta (SRL, 1980). However, measurements in Issaqueena or Keowee to confirm this number are not available.

Third, the weapons contribution to H-3 concentration is on the order of 100 to 500 pCi/L. This is based on the data from

Lakes Keowee and Jocassee and is consistent with an estimate by Kathren (1983).

Fourth, adsorption plays a significant role in the transport of Cs-137, as evidenced by the suspended and bottom sediment concentration data. The use of field and laboratory data to quantify adsorption is discussed below.

Finally, based on the tritium data a picture of the transport of radionuclides from Oconee is developing. Above US 123 there appear to be large temporal and spatial variations due to advection and dispersion of the periodic effluent pulses. However, below US 123 the variations are modest and might well be approximated using compartmental models. A simple compartmental model is evaluated in the final part of this section.

Adsorption of Cs-137 and Co-60 by Water Column Particulate Matter

Quantification and Significance

Radionuclides entering a lake can be subject to a variety of transport and transformation processes, including dissolution and biological uptake. One of the potentially important interactions involves solid surfaces. Particles in the water column and sediments of lakes can adsorb Cs-137, Co-60 and other radionuclides and chemicals. While adsorbed, the fate of the adsorbed materials is tied to the fate of the particles. Under certain conditions, a significant fraction of the total loading of a radionuclide can be present in the adsorbed phase, hence attempts to predict the fate and distribution of adsorbable materials must include consideration of both the rate and extent of adsorption and desorption. Parameterization and quantification of sorption processes is requisite for inclusion in fate and distribution models as well as development of a general understanding of when sorption processes are likely to be important. Finally, predictive radionuclide transport models which have been verified with field adsorption data are needed. Although there is much evidence that suspended particles can effectively remove dissolved radionuclides from solution (Haushild et al., 1973; Haushild et al., 1975; Onishi et al., 1981), there are few models which include adsorption, and field data on radionuclide adsorption are limited (NRC, 1977).

Sorption processes may be significant to the fate and effects of radionuclides for many reasons. For example, dissolved phase radionuclides are generally more available to biota than particulate phase radionuclides. Bioaccumulation of radionuclides is also possible. Particle scavenging of radionuclides and subsequent settling can be an effective removal mechanism of radionuclides from the water column. However, once in the sediments, sediment "focussing" processes can result in uneven distribution of particles and creation of "hot-spots" of activity of radionuclides. Also, resuspension or removal of sediments containing radionuclides may result in their redistribution and possible release of radionuclides back to the water column.

In this investigation, sorption of Cs-137 and Co-60 in Hartwell Lake was considered. Three main areas were addressed, as follows:

1. review of the conceptual framework of parameterization and quantification of sorption processes, mainly the distribution coefficient (K_d),
2. general consideration of factors that promote the significance of adsorption, and
3. development of field and laboratory techniques for measuring distribution coefficients (K_d's) of Cs-137 and Co-60, and collection and comparison of field and lab data on K_d's for Cs-137 and Co-60 for Hartwell Lake.

The discussion below is presented to correspond to these major areas of this part of investigation.

Parameterization and Quantification of Sorption

Adsorption occurs due to adsorbate-adsorbent attractions, which can be chemical (chemical adsorption) or physical (physical adsorption) in nature (Stumm and Morgan, 1981). Elucidation of the mechanism active in a particular case can be difficult, but is very important to fundamental understanding and modeling of factors that affect the process. The extent of adsorption is dependent upon several variables, including the nature of the adsorbent and the adsorbate, available surface area of the adsorbent, concentration and types of solutes in solution, time, and temperature (Onishi et al., 1981, Sayre et al., 1963). Sayre et al. (1963) reported that particle size distribution, which is directly related to surface area for a given mass, is the most important sediment characteristic affecting adsorption of ionic radionuclides on clays. The concentration of other ions in the solution, including hydrogen ions (pH), is also an important characteristic since competing ions may affect adsorption of the target ion.

Traditionally, researchers studying the interactions of ionic trace constituents, such as most radionuclides, have used ion exchange concepts, which involve the displacement of ions sorbed at the surface of a solid by other ions of like sign, to interpret results (Onishi et al., 1981). Ion exchange is considered to be the dominant adsorption process due to the dominance of an adsorption mechanism related to charged surfaces and the presence of other ions, such as Na^+, to act as dominant counter ions.

The relative bonding energies of monovalent cations in ion-exchange reactions are, according to the Hofmeister series (Sayre et al., 1963);

$$Cs^+ > Rb^+ > K^+ > Na^+ > Li^+ \qquad (2)$$

Since Cs-137 is one of the most abundant long-lived fission products, and potassium and sodium are among the most abundant exchangeable cations on clay mineral surfaces, the relative positions of the ions in the Hofmeister series is of practical significance in waste solutions containing fission products (Sayre et al., 1963). The Hofmeister series predicts Cs-137, released to surface waters in or transformed to the ionic form, could exchange with sodium or potassium present on clay mineral sediments even when the Cs-137 is at a lower concentration.

It can be shown (Onishi et al., 1981; Vandeven, 1985) that when a) the concentration of Cs^+, or other target ion, is much lower than the concentration of Na^+, or the major solution ion(s); b) the ratio of the activity coefficients of Cs^+ in the solution and solid phases is constant, and c) the system is at equilibrium, the extent of adsorption can be quantified by a simple distribution coefficient (K_d), as follows:

$$K_d = \frac{(Cs^+)_{adsorbed}}{(Cs^+)_{solution}} \qquad (3)$$

If the amount adsorbed, $(Cs^+)_{adsorbed}$, is in units of mg/g, and the solution concentration is in units of mg/mL, the resultant units of K_d are mL/g. For radionuclides, the following expression is useful:

$$K_d(\text{mL/g}) = \frac{\text{Radionuclide concentration solid (pCi/g)}}{\text{Radionuclide concentration dissolved (pCi/mL)}} \qquad (4)$$

Historically, the quantification of radionuclide adsorption has been expressed in terms of the distribution coefficient, or K_d, which can be derived theoretically for an ideal ion-exchange reaction. However, due to the chemical complexity of most aquatic systems, radioactive ions such as Cs^+-137 must compete for exchange sites with other cations in the water such as potassium, calcium, and magnesium. In addition, radionuclides may be present in the particulate phase due to processes other than ion-exchange, such as specific sorption by metal hydroxide coatings or adsorption by organic matter. Also, attainment of system equilibrium can be difficult to ascertain in natural aquatic systems, and even in laboratory systems, as will be discussed below. Therefore, use of a distribution coefficient to quantify radionuclide adsorption is often done on an operational basis and is therefore site specific, making a field determination useful if at all possible, and making careful interpretation of field and lab data important. Combinations of these factors result in reported K_d's for Cs-137 ranging from 50 to 80,000 mL/g and for Co-60 ranging from 1000 to 71,000 mL/g (Onishi et al., (1981).

Circumstances Promoting Adsorption

It follows from the above discussion that adsorption is likely to be important when the affinity of the target ion is great for the adsorbent and when the concentration of adsorbent is high. Although there are many factors and interactions to consider (beyond the scope of this discussion - see references such as Stumm and Morgan, 1981, and Onishi et al., 1981) the major considerations in an initial assessment of the potential importance of adsorption to the fate of a radionuclide are the tendency of the radionuclide to be adsorbed (parameter: K_d) and the amount of available adsorbent (parameter: suspended solids concentration).

Consequently, accurate determination of both the K_d and the adsorbent concentration can be important. The relationships between K_d, adsorbent concentration, and fraction of adsorbate adsorbed can be conveniently summarized in a plot (Figure 6). Note the variation in fraction adsorbed that result over the range of published K_d values for Cs^+ of 50 to 80,000 (Onishi et al., 1981), hence the need for improved measurements. The effects of increases in K_d and adsorbent concentration are also evident, as well as the indication that if K_d is low (e.g. $<10^3$ mL/g) it is unlikely a majority of the radionuclide will be adsorbed, and if

K_d is high (e.g. $>10^5$), suspended sediment concentrations of 10 mg/L or more will result in most of the radionuclide being adsorbed. Figure 6 and the combined effects of increases in K_d and adsorbent concentrations on fraction adsorbed also infer that under some conditions the fraction adsorbed will not vary significantly with changes in K_d or adsorbent concentration. Figure 7 is more explicit in this regard showing fraction adsorbed vs the product of K_d times the suspended sediment concentrations. When this product is greater than approximately 10, fraction adsorbed is relatively less sensitive to K_d and adsorbent concentration. (See Vandeven, 1985, for derivation of these plots.)

The majority of available radionuclide distribution coefficients have been determined from laboratory studies. Data from field investigations are very limited. Ideally, radionuclide distribution coefficients should be determined under natural field conditions, but this is often difficult due to low radionuclide concentrations or the unavailability of suitable field equipment (Schell et al., 1981a).

Field and Laboratory Results of K_d for Cs-137 and Co-60 in Hartwell Lake

The procedures for obtaining field measurements of dissolved phase and particulate phase radionuclide concentrations have already been discussed in the methodology section. Development of these techniques was significant in that they made it possible to obtain field derived K_d data, which was not available for Hartwell Lake (or many other sites), and allowed comparison of field and

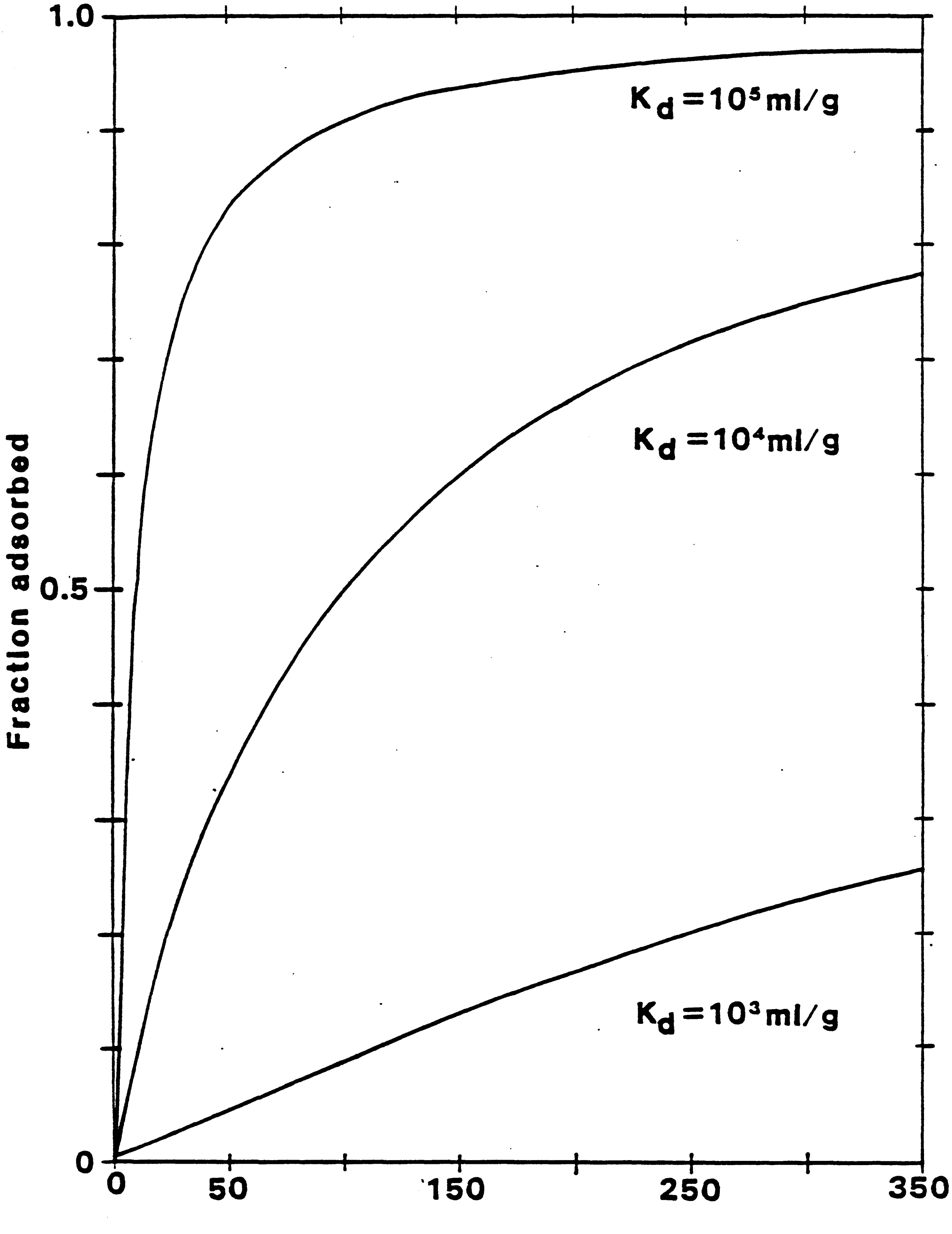


Figure 6. Theoretical fraction adsorbed vs suspended sediment concentration

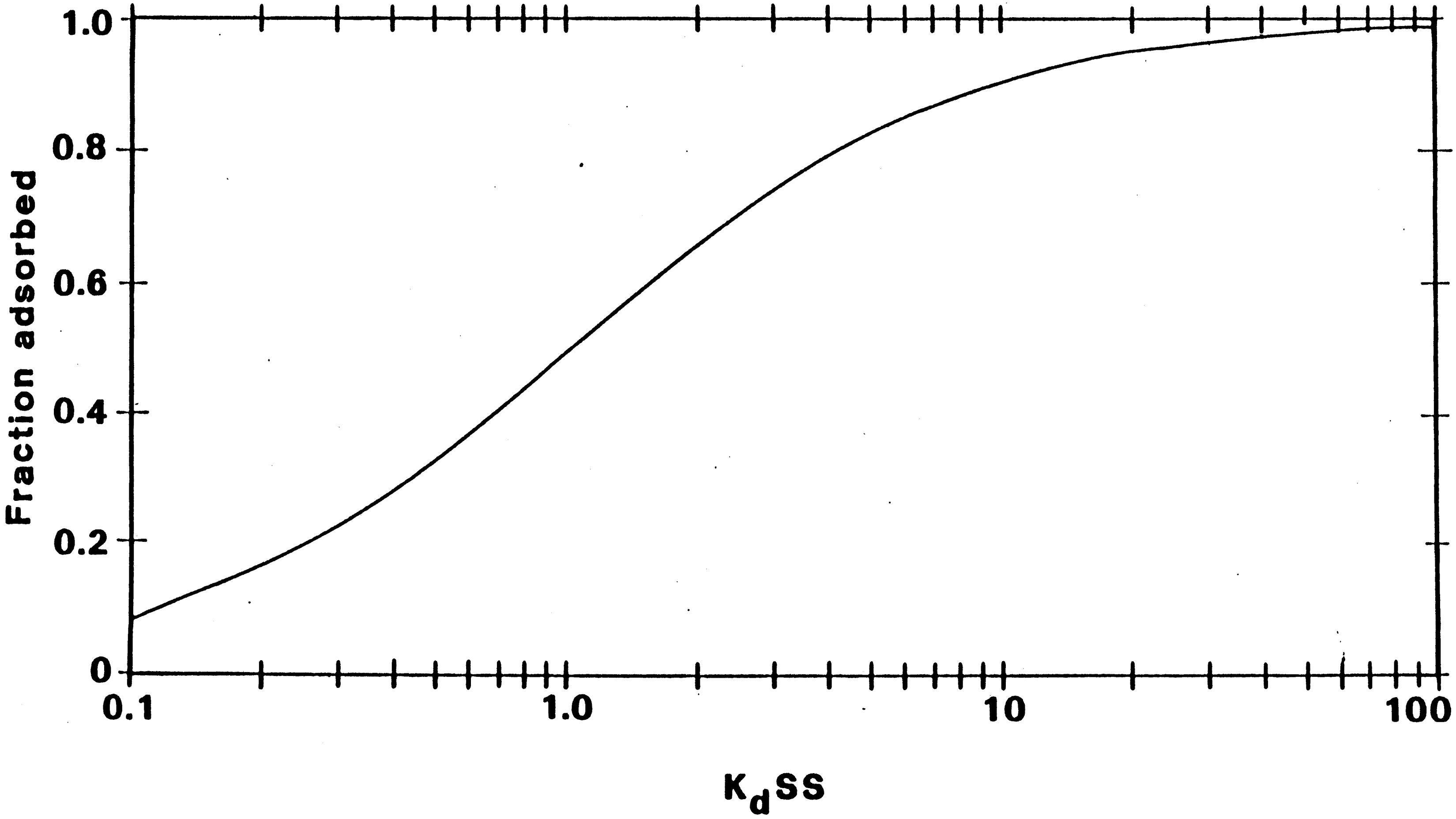


Figure 7. Theoretical fraction adsorbed vs K_dSS

laboratory K_d data for evaluation of laboratory techniques. Results obtained elucidated several potential problems and indicate more investigations of this type are needed to adequately understand and quantify radionuclide adsorption. Variability in previously published data and the disparity between published laboratory and field K_d values, with field results generally higher than lab results, as shown in Table VI, also provide impetus to further investigation.

Laboratory determinations of K_d were made using modifications of the techniques of Duursma and Bosch (1970) and Schell et al. (1981b). Details of the experimental procedures are available in Vandeven (1985). Three experiments were conducted to investigate the effect of contact time and suspended sediment concentration on the adsorption of Cs-137 and Co-60 by suspensions of Hartwell Lake sediments. The choice of conditions to be examined in each experiment was based on results obtained from all previous experiments and from data collected in the field.

In Experiment 1, Hartwell Lake water spiked with Cs-137 and Co-60 was contacted with lake sediment at suspended sediment concentrations of 10 mg/l, 50 mg/l, 200 mg/l, and 1000 mg/l. Cesium-137 and Co-60 concentrations were 800 pCi/ml and 180 pCi/ml, respectively. Eight duplicate sample aliquots were removed during a total contact time of 380 hours.

Experiment 2 was conducted to examine the effect on Cs-137 adsorption over a wider range of suspended sediment concentrations. Lake sediment was used at concentrations of 10.4 mg/l, 1,040 mg/l, and 10,400 mg/l. A larger Cs-137 concentration, 1600 pCi/ml was

Table VI. Cesium and cobalt fresh water distribution coefficients

Isotope	K_d(ml/g)	Reference	Comments
FIELD STUDIES			
Cs-137	2×10^5	Jinks & Wrenn, 1976 Linsalata et al., 1984	[Cl] = 0.025 mg/L
Cs-137	2×10^3	Beasley & Jennings, 1984	[SS] = 60 mg/L (0.3 μm Millipore)
Cs-137	3.5×10^4	Shure & Gottschalk, 1975	[SS] = 5 mg/L
LABORATORY STUDIES			
Cs-134	3900 - 8900	Garder & Skulberg, 1964	K_d increases with decreasing concentration of sediment, SS = 16-256 mg/L
Cs-137	800 - 1000	Jenne & Wahlberg, 1968	Sediment particles less than 250 μm in diameter
Co-60	1400 - 2400		
Cs-137	500 - 1700	Schell et al., 1981a	K_d increases with decreasing concentration of sediment at T = 4°C and diameter < 64 μm
Co-57	3200	Clayton et al., 1981	T = 4°C, particle diameter < 64 μm

[SS] = Suspended sediment concentration
[Cl] = Chloride concentration

used in order to detect accurately the dissolved fraction of the 10,400 mg/l sample. Six duplicate sample aliquots were taken during a total contact time of 196 hours.

Experiment 3 was performed to examine the adsorption of Cs-137 at contact times of less than one hour. Two suspended sediment concentrations were used, 50 mg/l and 200 mg/l. Six sample aliquots were taken, four during the first hour, and a total contact time of 120 hours was obtained.

One significant problem was encountered in the laboratory determinations. The separation technique originally chosen was vacuum filtration using a Gelman membrane (0.45 um, GN-6) filter and a system which consisted of a borosilicate glass funnel clamped to a coarse-frit glass filter support. Upon filtering and washing sample aliquots, analysis of the two phases produced a combined radionuclide concentration significantly less than the initial concentration of the sample. The cause of the problem was traced to the filtering apparatus by placing the coarse-frit glass filter support on the detector. Although the amount could not be quantified, it was obvious from the activity in the filter support that the filter support was adsorbing a significant amount of both Cs-137 and Co-60, which resulted in an apparent decrease in the dissolved phase concentration. The problem was solved by using a polycarbonate filtering system with a polypropylene filter support which did not adsorb Cs-137 or Co-60.

Once a suitable separation technique was developed, the laboratory procedure was applied with a minimum of difficulty. Sample aliquots taken from the control samples during each experiment revealed that neither Cs-137 nor Co-60 was adsorbing to the container walls during shaking or to the filter or filter support during separation. Activity balances for each Cs-137 sample aliquot showed the sum of adsorbed and dissolved activities to be within 10 percent of the initial activity.

The fraction of Cs-137 adsorbed as a function of contact time during Experiment 2 is shown in Figure 8. Similar results were obtained in Experiment 1. It is seen that Cs-137 adsorption is characterized by an initial period of rapid adsorption that is followed by continued adsorption at a slower rate. For example, in Experiment 2 at a suspended sediment concentration of 10,400 mg/l, over 95 percent of the total activity was removed to the sediment within the first hour. Significantly, for lower suspended sediment concentrations the transition between the rapid and slower rates was more gradual. At 10.4 mg/l, approximately 5 percent was adsorbed in the first hour and 15 percent by 200 hours. Results for Experiment 3 were similar and also showed that essentially all of the amount adsorbed in 1 hour actually was adsorbed in less than 15 minutes.

Distribution coefficients as a function of time are presented in Figure 9 for Experiment 2. As seen from these and the previous figure, adsorption, and thus the distribution coefficient, was increasing throughout the experiments. The purpose of the laboratory studies was to obtain adsorption data in a well characterized, controlled environment which would aid in the interpretation of the field results. Therefore, these tests were designed to duplicate, as closely as possible, the conditions found in the

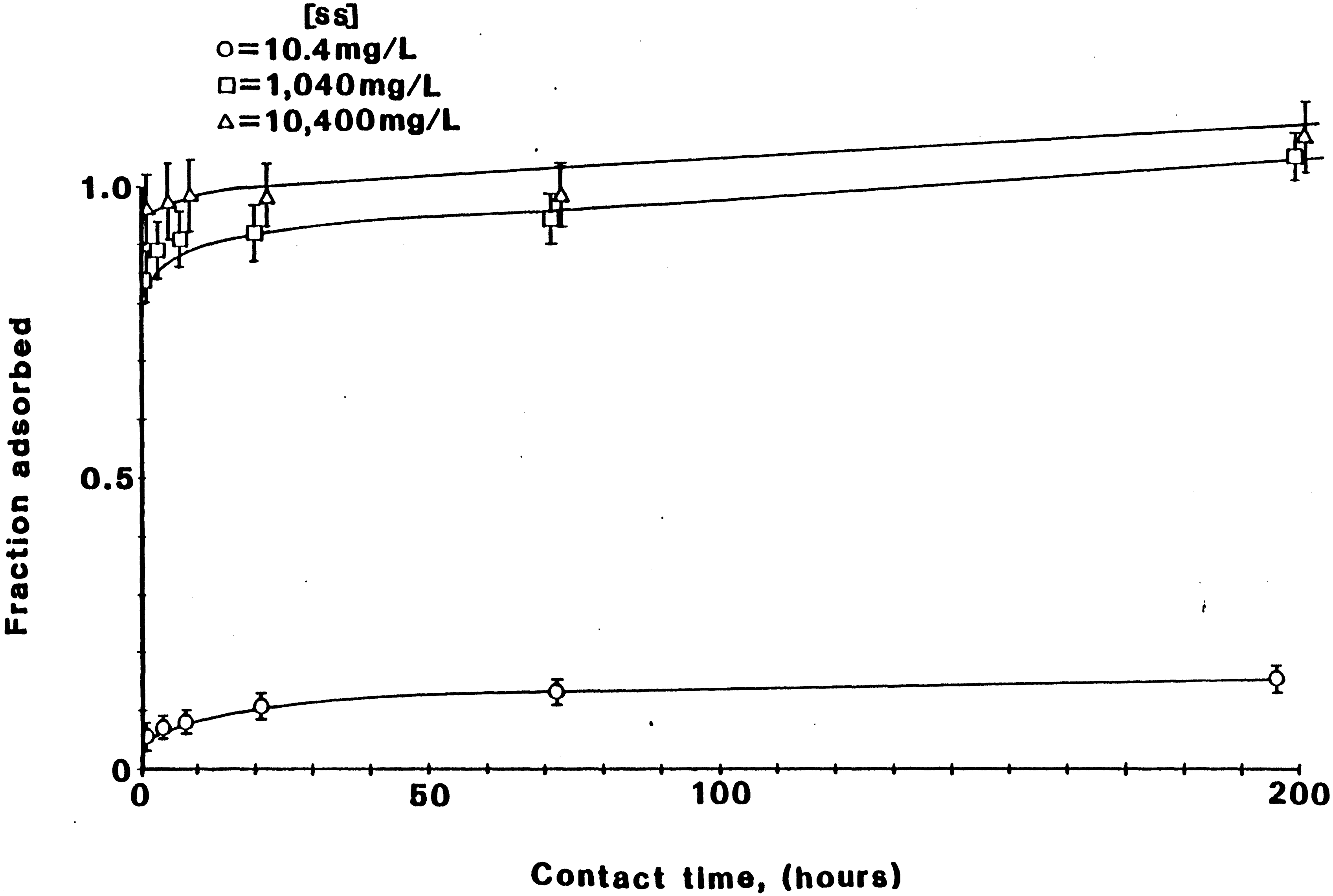


Figure 8. Fraction of Cs-137 adsorbed vs contact time in Experiment 2

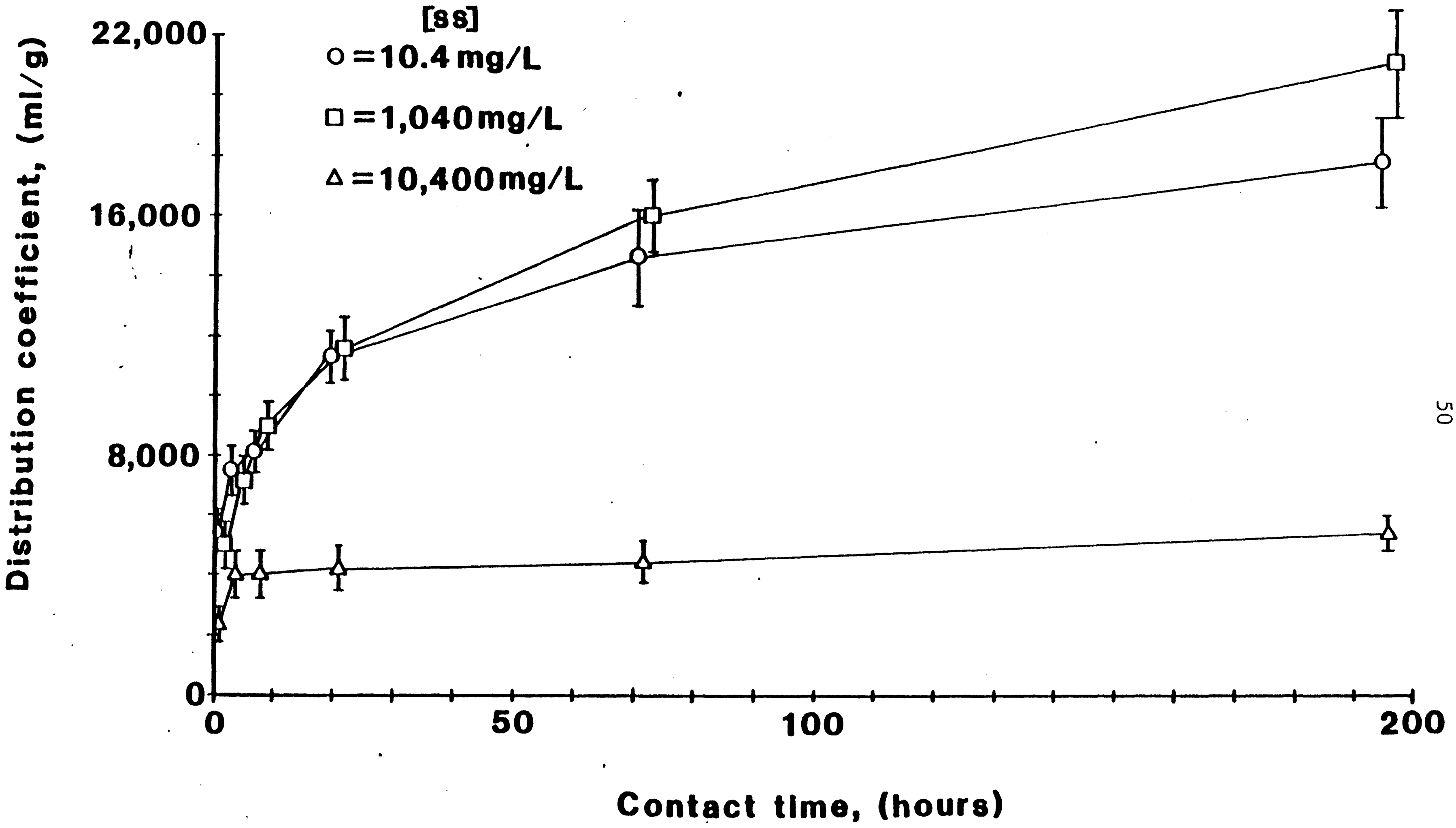


Figure 9. Cs-137 distribution coefficient vs contact time in Experiment 2

field. However, the contact time between radioactive species and suspended sediment in Hartwell Lake is unknown and, therefore, cannot be reproduced in the laboratory tests.

In order to relate laboratory data to results obtained in the field, laboratory results should be reported only after equilibrium is reached between radionuclides in the dissolved and particulate phases. A strict thermodynamic definition of a reaction at equilibrium is when its forward rate of reaction is equal to the rate of the reverse reaction (Stumm and Morgan, 1981). In these experiments, equilibrium would therefore be achieved when the rate of adsorption equaled the rate of desorption. Apparently, equilibrium in the strict sense was not reached during the course of these experiments. However, in adsorption experiments, system equilibrium is usually more subjectively defined. For example, the equilibrium distribution coefficient may be reported when adsorption appears close to its maximum value, usually estimated from a visual inspection of fraction adsorbed versus time curve. This subjective convention warrants a closer scrutiny of the results obtained and how these results are interpreted.

Referring back to Figure 7, it is clear that the effect of errors in fraction adsorbed related to errors in K_d on suspended sediment concentration are not linear over the entire range of the product $K_d[SS]$. Conversely, errors in K_d at a given suspended sediment concentration are not linearly related to errors in experimental determination of fraction adsorbed over the entire range of fraction adsorbed data. For example, consider K_d values from Experiment 2 for the 10.4 mg/L and 1040 mg/L suspended sediment concentrations at 20 hours, when they were both about 11,500 mL/g, and at 196 hours, when they were 18,000 mL/g and 21,000 mL/g, respectively. From Figure 7 it can be determined that a distribution coefficient increase from 11,500 mL/g to 18,000 mL/g at a suspended sediment concentration of 10.4 mg/L represents an increase in the fraction adsorbed from 0.107 to 0.158, a 48.0 percent increase. Correspondingly, a distribution coefficient increase from 11,500 mL/g to 21,000 mL/g at a suspended sediment concentration of 1,040 mg/L represents an increase in the fraction adsorbed from 0.923 to 0.956, or an increase of only 3.4 percent. When $K_d[SS]$ is greater than 10, i.e. when it attains a value >10 in an experiment, changes in fraction adsorbed at a given suspended sediment concentration are related to smaller changes in K_d than when $K_d[SS]$ is <10.

Although most researchers have assumed that equilibrium is reached between dissolved and adsorbed cesium within hours, the continuation of slower adsorption, and its significance, has been recognized. Jenne and Wahlberg (1968) reported that cesium initially displaces the readily exchangeable portion of potassium on clay particles, followed by a slow release of interlayer potassium or cesium. Results similar to those reported here were found by Evans et al. (1983). Their results indicated that an initial rapid adsorption of radiocesium occurs to the surface of clay particles, followed by the slow movement of radiocesium into the clay interlayer sites. They also suggest that, although this second process is slow relative to the first, it is rapid in comparison to the half life of Cs-137 and should be considered in transport studies. There is also field evidence indicating a fast and slow stage of cesium adsorption. Hakanson and Whicker (1975) added 1 kg of stable Cs-133 to a 5 hectare lake in Colorado and studied the transport of cesium over a 393 day period. They reported the loss of cesium occurred in two phases; the rapid phase removed 60 percent of the cesium from the water with a loss half-time of 12 hours, while the slow phase had a loss half-time of 130 days. Further development of kinetic modeling of radionuclide adsorption is introduced in Vandeven (1985).

The Co-60 experiments also suggested Co-60 was rapidly removed from solution to the sediment. At sediment concentrations of 50 mg/L, 200 mg/L, and 1,000 mg/L, Co-60 in the dissolved phase was below the MDC of 12 pCi/mL after 3.25 hours. Even at the lowest sediment concentration tested (10 mg/L), dissolved Co-60 was below the MDC after 28 hours. The distribution coefficient implied by these data is in excess of 1 x 10^6 mL/g. With a distribution coefficient of this magnitude, the K_dSS product would be greater than 10 even at a suspended sediment concentration of 10 mg/L. Therefore, cobalt partitioning between the dissolved and particulate phases will be sufficiently approximated, at suspended sediment concentrations ranging from 10 to 1,000 mg/L, using a distribution coefficient of 1 x 10^6 mL/g.

The importance of recognizing the existence of relatively slow kinetics for a fraction of radionuclides that are adsorbed relates to inferences about the mechanisms of adsorption, which shall not be addressed further here, and potential artifacts in laboratory data. For example, many results reported in the literature are not sufficiently substantiated by confirmation of attainment of equilibrium. Such artifacts can be a contributing factor to observations that K_d's seem to be dependent on suspended solids concentrations, contrary to thermodynamic predictions. However, this explanation probably does not fully account for all observations reported, for example, by Schell et al. (1981b), Aston and Duursma (1973), and O'Connor and Connolly (1980) (Vandeven, 1985). For the purposes of this investigation equilibration time is important in considering comparisons of field and laboratory K_d data, as discussed below.

An estimate of the field measured Cs-137 distribution coefficient was calculated for samples 4 through 11. Due to the high activity measured in sample 5, the distribution coefficient calculated for this sample was not considered to be representative and was not included in the calculation for the following discussion. The average distribution coefficient calculated from sample 4 and samples 6 through 8 was 115,000 mL/g, with an average counting uncertainty of 41,200 mL/g. The distribution coefficients calculated from these samples were not considered to be significantly different considering the large counting uncertainty of at least 40 percent. Another sampling trip was scheduled in an attempt to lower this uncertainty. This was achieved by processing a larger volume of water through the filter and thereby collecting a greater amount of sediment. The results of this effort are shown as samples 9 through 11. The average distribution coefficient of these samples was 89,400 mL/g with an average counting uncertainty of 23,000 mL/g.

The Co-60 distribution coefficient could not be quantified because dissolved concentrations were not detected in any of the samples. An explanation may be based on the results of the laboratory data, where the Co-60 distribution coefficient was found to be greater than 1 x 10^6 mL/g. It would therefore be expected that most of the Co-60 released would become associated with the solid phase. In addition, a relatively small amount of Co-60 has been released compared to Cs-137. Also, a significant fraction of cobalt may be released from reactors in particulate form, although this information is not available for cobalt released from the Oconee nuclear station. These two facts may explain why dissolved Co-60 and Co-58 were below detection limits in each sample.

Table VII lists Cs-137 distribution coefficients measured in the field and laboratory. The field distribution coefficients were calculated from dissolved and particulate phase concentrations given in Table IV. Results from locations 9, 10 and 11 are presented due to their lower uncertainty relative to the other field measurements, as discussed above. Laboratory measurements represent values for the longest contact time. The average field measurement of the Cs-137 distribution coefficient was 89,400 with an average counting uncertainty of 23,000 mL/g. Laboratory measurements ranged from a low of 5,400 ± 373 mL/g, obtained for a suspended sediment concentration of 10,400 mg/L and a contact time of 196 hours, to 27,530 ± 2,000 obtained for a suspended sediment concentration of 50 mg/L and a contact time of 380.5 hours.

A generally higher value for field distribution coefficient measurements, as compared to laboratory measurements, is evident

Table VII. Comparison of field and laboratory derived Cs-137 distribution coefficient data

Distribution Coefficient (mL/g)	Contact Time (hours)	Particle Size (µm)	Suspended Sediment Concentration (mg/L)
FIELD MEASUREMENTS			
85,000 ± 17,000	unknown	unknown	4.0
87,400 ± 23,900	unknown	unknown	4.0
95,900 ± 21,000	unknown	unknown	4.0
LABORATORY MEASUREMENTS			
24,750 ± 1,800	380.5	<63	10
27,530 ± 2,000	380.5	<63	50
22,580 ± 1,650	380.5	<63	200
15,420 ± 1,100	380.5	<63	1000
17,890 ± 1,250	196.0	<63	10.4
21,123 ± 1,800	196.0	<63	1,040
5,405 ± 373	196.0	<63	10,400
23,900 ± 1,670	120.0	<63	50
23,920 ± 1,675	120.0	<63	200
16,980 ± 420	24.0	<2	65
19,575 ± 490	168.0	<2	65

Note: Data for contact time and particle size are unavailable for field samples.

in the data in Table VII and is also found in the literature, as was shown in Table VI. Particle size differences between lab and field samples could not explain the differences in K_d (Vandeven, 1985). Equilibrium (contact) time was, however, found to be important.

The laboratory results indicated the Cs-137 distribution coefficient increased with contact time and was still increasing when the final sample aliquots were taken. This result has particular significance in comparison with the field measurements where contact times could not be determined. Although there is no method for determining how long a particular suspended sediment sample has been in the system, it is likely that field contact times were, on the average, much greater than the longest contact time obtained in the laboratory (16 days). Therefore, a possible explanation for the higher Cs-137 distribution coefficients measured in the field is the difference in contact times (also see Jenne and Walberg, 1968). Although it is not possible to obtain contact times for the field samples (equilibration times between dissolved radionuclides and particles), and they would vary greatly, it appears the field values were more representative of equilibrium K_d values than the laboratory values. In the laboratory experiments, none of the tests with suspended sediment concentrations below 1000 mg/L attained a value of the product $K_d[SS]$ greater than the practical limit of ten discussed previously.

Therefore, considering the field measurements did not vary appreciably, the average value of 89,400 ± 23,000 mL/g is a good approximation of the Cs-137 distribution coefficient in equilibrium between the dissolved and adsorbed phase. As discussed above, the ability to detect only adsorbed Co-60 and Co-58 in the field may be explained through the laboratory results. The Co-60 distribution coefficient in the laboratory experiments was found to be greater than 1 x 10^6 mL/g at a contact time of 28 hours. Therefore, even at a suspended sediment concentration of 10 mg/L, over 90 percent of the cobalt would be adsorbed within approximately one day. The small amounts of Co-60 released relative to Cs-137, the short half-life of Co-58, the large cobalt distribution coefficient, and the possibility that cobalt was released in particulate form all contribute to the inability to detect dissolved cobalt.

Although field and laboratory measurements of the Cs-137 distribution coefficient differed by approximately a factor of three, it is evident that adsorption of Cs-137 by suspended sediment is a significant process in Hartwell Lake even at low suspended sediment concentrations. The average Cs-137 distribution coefficient of 89,400 ± 23,000 mL/g measured in the field results in approximately 25 percent of the total Cs-137 being transported in the adsorbed phase at a suspended sediment concentration of 5 mg/L. Laboratory results showed that the Cs-137 distribution coefficient is constant over the range of suspended sediment concentrations found in Hartwell Lake (10 to 1,000 mg/L). Although during the present investigation suspended sediment concentrations in Hartwell Lake were not greater than 5 mg/L, concentrations greater than 5 mg/L are not unusual during and after storm events. At these higher suspended sediment concentrations the fraction of Cs-137 adsorbed would increase significantly.

Both the field and laboratory measurements suggest that radiocobalt (Co-60 and Co-58) is transported primarily in the particulate phase. Laboratory Co-60 distribution coefficients were calculated to be greater than 1×10^6 mL/g. A distribution coefficient of this magnitude results in over 90 percent of the activity being adsorbed at suspended sediment concentrations of 10 mg/L or greater.

In addition to the magnitude of the distribution coefficient, Co-60 adsorption was found to be extremely rapid. The adsorption of cobalt can be considered to occur faster than other lake processes such as sedimentation. These laboratory results are substantiated by the Co-60 and Co-58 field measurements. Neither Co-60 nor Co-58 was detected in the dissolved phase of any of the field samples, while adsorbed Co-60 and Co-58 were detected at several locations. However, caution must be exercised when interpreting field measurements of the distribution of cobalt between the dissolved and adsorbed phases. This is a consequence of the origin of this radionuclide. A large fraction of activation products such as Co-60 and Co-58 can be released in particulate form (Blanchard, 1980). Thus it is possible that the absence of dissolved Co-58 and Co-60 is due to their being released in particulate form.

Modeling

One of the objectives of this project was to utilize the field data to evaluate a simple compartmental model of radionuclide transport in the study area. In this section, such a model is developed and comparisons are given between its predictions and the field measurements of H-3 and Cs-137 concentrations. In the compartmental approach, a system is approximated as a series of discrete, interconnected compartments in which inputs are instantaneously and uniformly dispersed. The time rate of change of radionuclide concentration in a compartment is given by a first order differential equation containing rates of input, output, decay, and reaction. A conceptualization of the model is given in Figure 10 for a single compartment in which radionuclides in the dissolved phase are subject to adsorption by suspended particles which settle to the bottom. The rate of change of total activity in the water column, including both dissolved and particular phases, is

$$\frac{dC_T}{dt} = \begin{matrix}\text{Release Rate}\\ \text{from Oconee}\end{matrix} - \begin{matrix}\text{Hydraulic}\\ \text{Washout}\end{matrix} - \begin{matrix}\text{Radioactive}\\ \text{Decay}\end{matrix} - \begin{matrix}\text{Settling of}\\ \text{Particulate Phase}\end{matrix} \tag{4}$$

or mathematically

$$\frac{dC_T}{dt} = \frac{S}{V} - \frac{QC_T}{V} - \lambda C_T - \frac{v_s C_P}{Z} \; . \tag{5}$$

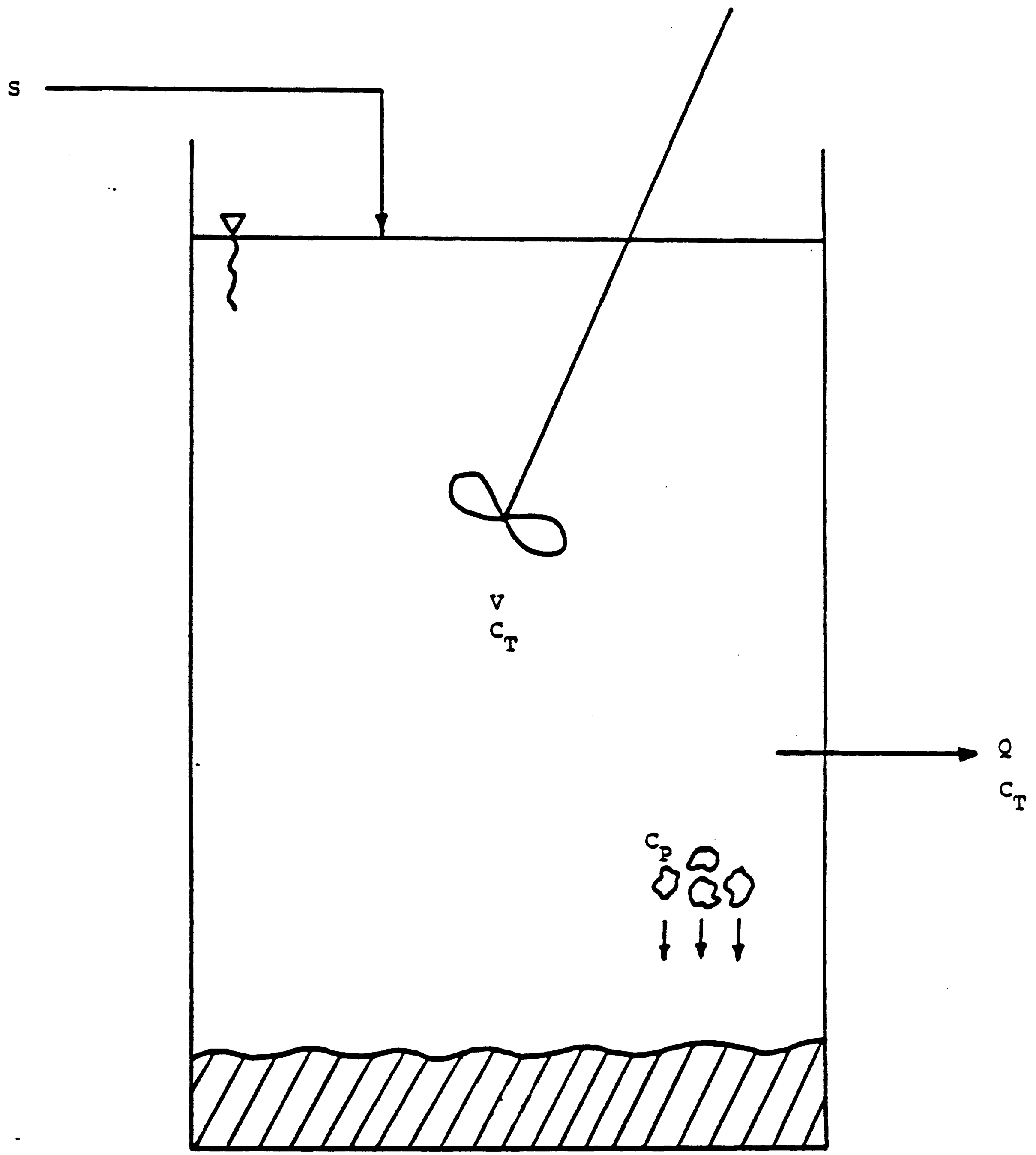


Figure 10. Single compartment model with adsorption, sedimentation, and flow

where

C_T = total water column concentration (pCi/L),

C_P = particulate phase concentration (pCi/kg),

C_D = dissolved phase concentration (pCi/L),

S = source rate (Ci/yr),

V = volume of compartment (L),

Q = flow rate (L/yr),

λ = radioactive decay constant (1/yr),

v_s = average settling velocity (m/yr), and

Z = water column mean depth (m).

Equation 5 can be expressed solely in terms of C_T by defining P as the fraction of activity in the particulate phase. It is given by

$$P = \frac{(K_d)(SS)}{(K_d)(SS)+1} , \tag{6}$$

where

K_d = distribution coefficient (mL/g), and

SS = suspended sediment concentration (mg/L).

Therefore

$$C_D = (1-P)\ C_T \quad . \tag{7}$$

Substituting Equation 7 into Equation 5 yields

$$\frac{dC_T}{dt} = \frac{S}{V} - [\frac{Q}{V} + \lambda + \frac{v_s P}{Z}]\ C_T \ . \tag{8}$$

Equations 7 and 8 may be combined to obtain a differential equation for dissolved phase concentration,

$$\frac{dC_D}{dt} = \frac{(1-P)S}{V} - \left[\frac{Q}{V} + \lambda + \frac{V_s P}{Z}\right] C_D \ . \tag{9}$$

While it is possible to derive a differential equation for particulate phase concentration in a manner analogous to that used in obtaining Eqn. (9), it is simpler to use the distribution coefficient, i.e.

$$C_P = K_d C_D \ . \tag{10}$$

The concentration response to slowly varying, continuous sources may be approximated by letting S remain constant, setting $dC_D/dt = 0$, and calculating the steady-state concentration. The resultant dissolved and particulate phase concentrations are, respectively,

$$C_D = S\left[\frac{1-P}{Q+\lambda V+Av_s P}\right] \tag{11}$$

and

$$C_P = S\left[\frac{P}{Q+\lambda V+Av_s P}\right] \ , \tag{12}$$

where the lake area, A, has been substituted for V/Z.

In applying these equations to calculate H-3 and Cs-137 concentrations in Hartwell Lake, it is found that the second and third terms of the denominator are small compared to Q and can be neglected. This reduces the number of parameters in the model from eight to four. The four parameters are flow rate, source rate, suspended sediment concentration, and distribution coefficient. The average flow rate over the Hartwell dam is $4x10^9$ m^3/y (U.S. Army Corps of Engineers, 1960). The mean flow rate in the Seneca River arm of the lake is probably on the order of one-half of this value, but an exact figure is not known. Source rates for H-3 and Cs-137 are given in Table I. Suspended sediment concentrations measured at five times during this investigation were all approximately 5 mg/L. While this value is probably representative for much of the time, it could be significantly higher than this following extended periods of rain. The distribution coefficient for H-3 was taken as zero; for Cs-137 it was taken to be approximately 90,000 mL/g (Table VII).

Presented as the solid line in Figure 11 is the predicted H-3 concentration as a function of flow rate for the average 1984-1985 Oconee release rate of 1260 Ci/y. The dashed lines represent the band of measurements for estimated reactor related tritium in the upper part of the Seneca River. The data are consistent with a flow rate between $1x10^9$ and $2x10^9$ m^3/y; which is reasonable for the Seneca River arm of the lake.

Results for the Cs-137 model are given in Figures 12a and 12b for dissolved and particulate phases, respectively. Radionuclide concentration is plotted as a function of suspended sediment concentration for source rates of 0.05 Ci/y and 0.2 Ci/y, which bound the 1983 and 1984 Oconee release rates of 0.15 Ci/y and 0.07 Ci/y. The flow rate for these calculations is $1.5x10^9$ m^3/y, based on the H-3 results.

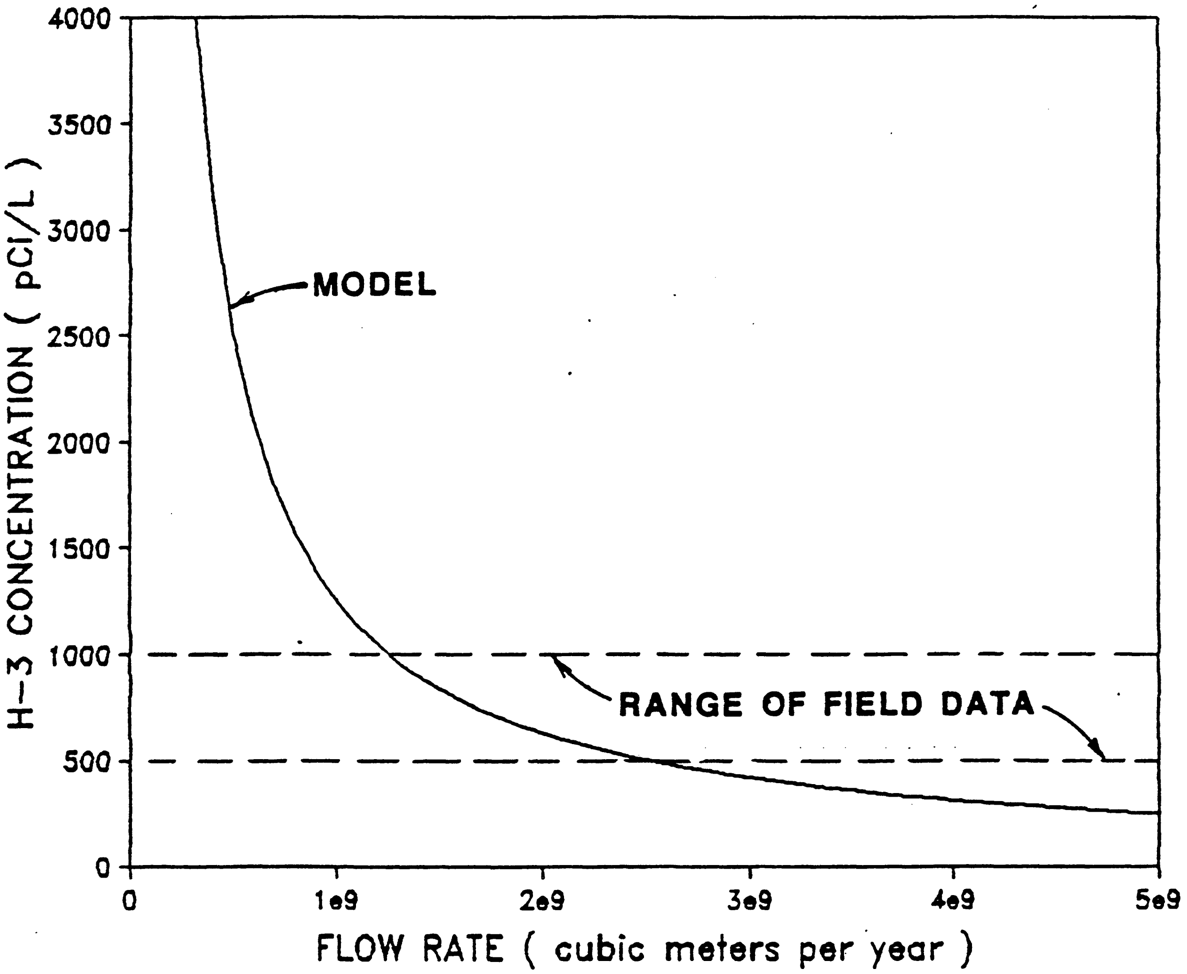
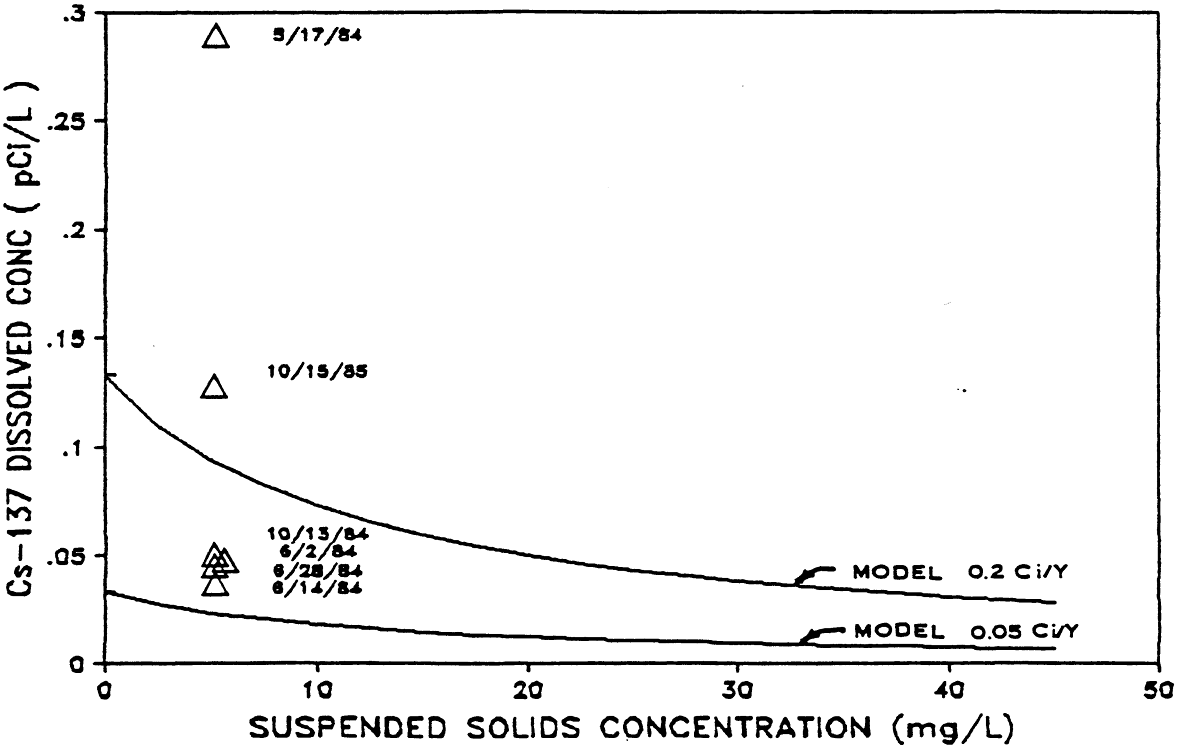


Figure 11. Comparison of H-3 field data with predictions of the compartmental model

Field data presented are for samples collected at the junction of the Keowee River and Twelve Mile Creek and for the upper Seneca River. The concentrations plotted in the figure are the values given in Table IV less the estimated contribution due to fallout. For the dissolved phase, this correction is 0.009 pCi/L based on the Russell and Clarks Hill data. For the particulate phase, the correction is 1000 pCi/kg based on the Tugaloo, Keowee, Issaqueena and Clarks Hill measurements. These data are plotted at a suspended sediment concentration of 5 mg/L. The field data are generally consistent with the prediction of the model. A majority of the measured dissolved phase concentrations and all but one of the measured particulate phase concentrations (the 17,400 pCi/kg outlyer discussed previously) lie within the source rate band. It must be recognized, however, that the location of the band depends on the value of flow rate. Calculations were also performed for flow rates of $1x10^9$ and $2x10^9$ m^3/y. The majority of dissolved phase concentrations remained within the band for both flow rates. The particulate phase concentrations remained in the band for $1x10^9$ m^3/y, but were centered about the upper band for $2x10^9$ m^3/y.

The comparisons presented in Figures 11 and 12, while not sufficient to validate the model, are certainly supportive of the compartmental approach. The good agreement between field data and these single compartment predictions for the upper portion of the Seneca River provides a basis for future application of a multicompartment, time dependent model to the remainder of Hartwell Lake.

(a)



(b)

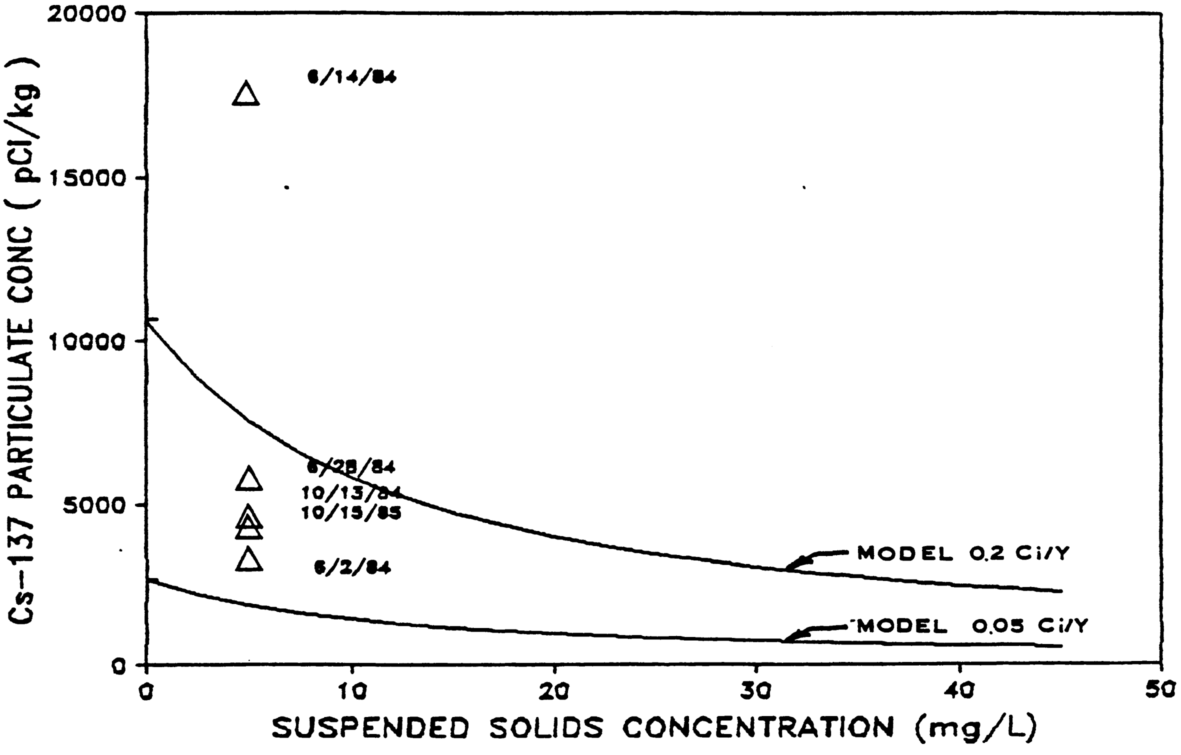


Figure 12. Comparison of (a) dissolved and (b) particulate phase Cs-137 field data with predictions of the compartmental model

V. SUMMARY

Hydrogen-3, Co-58, Co-60, Cs-134 and Cs-137 concentrations were measured in the upper sub-basin of the Savannah River system. The results indicate that the influence of release from the Oconee Nuclear Station are confined primarily to the Seneca River arm of Hartwell Lake. There was no evidence of the migration of Co-58, Co-60 or Cs-134 to the Tugaloo River arm of Hartwell Lake, the main body of the lake below the Seneca River/Tugaloo River junction, Russell Lake, or Clarks Hill Lake. While there appeared to be slightly elevated H-3 concentrations in Russell, the concentrations in the main body of Hartwell were on the order of those expected from weapons testing.

Adsorption was found to be a significant process affecting the transport of cesium radioisotopes. Laboratory measurements of the distribution coefficient for Cs-137 were significantly lower than those measured in the field, emphasizing the importance of field measurements in attempting to quantify the effects of adsorption in transport models.

Large temporal and spatial variations of H-3 concentrations were observed in the Keowee River arm of Hartwell Lake, but variations below the Keowee/Twelve Mile junction were modest. These results suggested that the concentration of reactor related radionuclides below this junction could possibly be estimated using the compartmental modeling approach.

A single compartment model was applied to the upper part of the Seneca River. Predictions of H-3, dissolved phase Cs-137, and particulate phase Cs-137 were compared to field measurements.

With a few exceptions, there was good agreement between the field data and the predictions of the model. These results suggest that the compartmental modeling approach might prove useful in estimating the fate and distribution of radionuclides in Hartwell Lake and throughout the upper sub-basin.

REFERENCES

1. APHA (American Public Health Association). 1980. Standard Methods for the Chemical Analysis of Water and Wastewater, 15th edition, Washington, D.C.

2. Aston, S.R. and E.K. Duursma. 1973. Concentration effects on Cs-137, Zn-65, Co-60 and Ru-106 sorption by marine sediments, with geochemical implications. Netherland Journal of Sea Research 6: 225-240.

3. Beasley, T.M. and C.D. Jennings. 1984. Inventories of Pu-239, Pu-240, Am-241, Cs-137, and Co-60 in Columbia River sediments from Hanford to the Columbia River estuary. Environmental Science and Technology 18: 207-212.

4. Clayton, J.R. Jr., T.H. Sibley and W.R. Schell. 1981. Distribution Coefficients for Radionuclides in Aquatic Environments: Effects of Dissolved Organic Compounds on the Distribution Coefficients of Co-57, Ru-106, Cs-137 and Am-241 (NUREG/Cr-1853, Vol. 1). NRC, Washington, D.C.

5. 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. Netherlands Journal of Sea Research 4: 395-469.

6. Eisenbud, M. 1973. Environmental Radioactivity. Second edition. Academic Press, New York, New York.

7. Evans, D.W., J.J. Alberts and R.A. Clark III. 1983. Reversible ion-exchange fixation of Cs-137 leading to mobilization from reservoir sediments. Geochimica et Cosmochimica Acta 47: 1041-1049.

8. Fabrikant, J.I. 1984. Nuclear energy, public health and public policy. Health Physics 46: 739-744.

9. Garder, K. and O. Skulberg. 1964. Sorption phenomena of radionuclides to clay particles in river water. International Journal of Air and Water Pollution 8: 229-241.

10. Hairr, L.M. 1974. An investigation of factors influencing radiocesium cycling in estuarine sediments of the Hudson River. Ph.D. Dissertation. School of Engineering and Science, New York University, New York, New York.

11. Hakonson, T.E. and F.W. Whicker. 1975. Cesium kinetics in a montane lake ecosystem. Health Physics 28: 699-706.

12. Haushild, W.L., G.R. Dempster Jr. and H.H. Stevens Jr. 1975. Distribution of radionuclides in the Columbia River streambed, Hanford Reservation to Longview, Washington. U.S. Geological Survey Professional Paper 433-0, Washington, D.C.

13. Haushild, W.L., H.H. Stevens Jr., J.L. Nelson and G.R. Dempster Jr. 1973. Radionuclides in transport in the Columbia River from Pasco to Vancouver, Washington. U.S. Geological Survey Professional Paper 433-N, Washington, D.C.

14. Jacobs, D.G. 1983. Man-made sources released to the environment. In Environmental Radioactivity, pp.27-37, National Council on Radiation Protection and Measurements, Bethesda, Maryland.

15. Jenne, E.A. and J.S. Wahlberg. 1968. Role of certain stream-sediment components in radioion sorption. U.S. Geological Survey Professional Paper 433-F, Washington, D.C.

16. Jinks, S.M. and M.E. Wrenn. 1976. Radiocesium transport in the Hudson River estuary. In Environmental Toxicity of Aquatic Radionuclides, pp.207-227, IAEA, Vienna, Austria.

17. Kathren, R.L. 1984. Radioactivity in the Environment. Harwood Academic Publishers, New York.

18. Krieger, H.L. and G.W. Frishkorn. 1971. Evaluation of ion-exchange surveilance sampler for analyzing radioactive liquid effluents. Health Physics 21: 591-595.

19. Linsalata, P., N. Cohen and M.E. Wrenn. 1984. Sources, behavior, and transport of Cs-137 in sediments and water of the Hudson River estuary. (Draft) New York University Institute of Environmental Medicine, New York, New York.

20. Luetzelschwaba, J.W. 1976. A field samling unit that uses an ion-exchange resin to concentrate radioactive ions in river water. Health Physics 31: 387-389.

21. Mann, D.R. and S.A. Casso. 1981. Report Number De-ACO-81EVI0694. Woods Hole Oceanographic Institute.

22. NRC. 1977. Estimating Aquatic Dispersion of Effluents from Accidental and Routine Reactor Releases (Regulatory Guide 1.113). U.S. NRC, Washington, D.C.

23. O'Connor, D.J. and J.P. Connolly. 1980. The effect of concentration of adsorbing solids on the partition coefficient. Water Reserach 14: 1517-1523.

24. Olsen, C.R., H.J. Simpson and R.M. Trier. 1981. Plutonium, radiocesium, and radiocobalt in sediments of the Hudson River estuary. Earth and Planetary Science Letters 55: 377-392.

25. Onishi, Y., R.J. Serne, E.M. Arnold, C.E. Cowan and F.L. Thompson. 1981. Critical Review: Radionuclide Transport, Sediment Transport, and Water Quality Mathematical Modeling and Radionuclide Adsorption/Desorption Mechanisms (NUREG/CR-1322), NRC, Washington, D.C.

26. Ritchie, J.C., J.R. McHenry, A.C. Gill and P.H. Hawks. 1972. Fallout Cs-137 in reservoir sediment. Health Physics 22: 98-99.

27. Russell, R.S. and R.S. Bruce. 1969. Environmental contamination with fallout from nuclear weapons. In Environmental Contamination by Radioactive Materials, pp. 15-26, IAEA, Vienna, Austria.

28. Savannah River Laboratory. 1984. Savannah River Plant Environmental Report for 1984 (DPSPU 85-30-1), Savannah River Laboratory, Aiken, South Carolina.

29. Sayre, W.W., H.P. Guy and A.R. Chamberlain. 1963. Uptake and transport of radionuclides by stream sediments. U.S Geological Survey Professional Paper 433-A, Washington, D.C.

30. Schell, W.R., A.L. Sanchez and T.H. Sibley. 1981a. Distribution Coefficients for Radionuclides in Aquatic Environments - Adsorption and Desorption Studies of Cs-137 (NUREG/CR-1852, Vol. 2), U.S. NRC, Washington, D.C.

31. Schell, W.R., T.H. Sibley, A.E. Nevissi, A.L. Sanchez, J.R. Clayton Jr. and E.A. Wurtz. 1981b. Distribution Coefficients for Radionuclides in Aquatic Environments - Methodology (NUREG/CR-1852, Vol. 1), U.S. NRC, Washington, D.C.

32. Shure, D.J. and M.R. Gottschalk. 1975. Cs-137 dynamics within a reactor effluent stream in South Carolina. In Radioecology and Energy Resources, D.E. Cusing Jr., Ed., pp.234-241, Dowden, Hutchinson and Ross, Stroudsburg, Pennsylvania.

33. Spigarelli, S.A. 1976. Cesium-137 activities in fish residing in thermal discharges to Lake Michigan. Health Physics 30: 411-413.

34. Stumm, W. and J.J. Morgan. 1981. Aquatic Chemistry. Second edition. John Wiley and Sons, New York New York.

35. U.S. Army Corps of Engineers, Savannah District. 1960. Hartwell project construction information. Savannah, Georgia.

36. Vandeven, J.A. 1985. The role of adsorption in radionuclide transport in Hartwell Lake. M.S. Thesis. Department of Environmental Systems Engineering, Clemson University, Clemson, South Carolina.