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Transport of ^{85}Sr and ^{137}Cs under induced clay suspensions

Purush TerKonda

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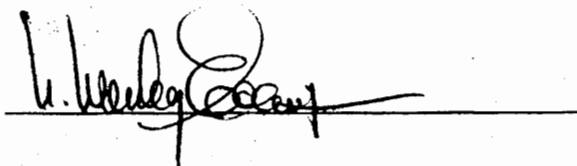
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TRANSPORT OF ^{85}Sr AND ^{137}Cs UNDER
INDUCED CLAY SUSPENSIONS

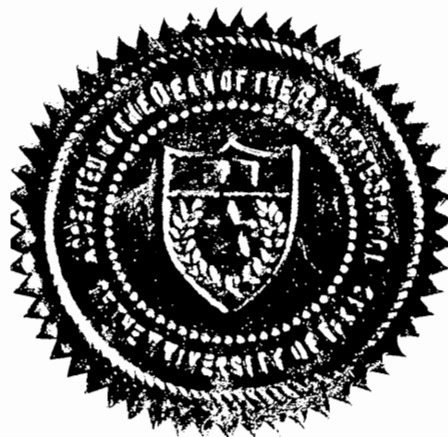
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TRANSPORT OF ^{85}Sr AND ^{137}Cs UNDER
INDUCED CLAY SUSPENSIONS

by

Krishnier Purushothaman, B.E., M.Sc.

DISSERTATION

Presented to the Faculty of the Graduate School of
The University of Texas in Partial Fulfillment
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TRANSPORT OF ^{85}Sr AND ^{137}Cs UNDER
INDUCED CLAY SUSPENSIONS

Publication No. _____

Krishnier Purushothaman, Ph.D.
The University of Texas, 1968

Supervising Professor: Earnest F. Gloyna

The purpose of this study was to evaluate the effect of induced clay suspensions on the transport of ^{85}Sr and ^{137}Cs in an aqueous environment.

An instrumented flume was used as a model river to simulate an environment typical of a slow-moving, unpolluted, turbid stream. Modified longitudinal dispersion relationships were used to describe the transport of radionuclides in turbid streams. Data were derived from instantaneous release of ^{85}Sr and ^{137}Cs with suspended Attapulgate and Kaolinite clays. Continuous release of ^{137}Cs with an Attapulgate clay suspension helped to establish uptake and transport characteristics for one environmental condition. Aquaria and laboratory studies preceded the model river experiments to define more specifically some of the physical and chemical characteristics.

Suspended clay in the model river system produced a significant reduction in the transport of ^{137}Cs . Conversely, the transport of ^{85}Sr was not affected appreciably since most of the ^{85}Sr remained in solution.

CONCLUSIONS

1. ^{85}Sr and ^{137}Cs when released instantaneously into a turbid stream is removed from solution. The transport can be approximated by a modified longitudinal dispersion equation:

$$C(x,t) = \frac{M}{A\sqrt{4\pi D_x t}} e^{-\frac{(x-ut)^2}{4D_x t}} e^{-Rt}$$

where $C(x,t)$ = radionuclide concentration with respect to time and distance, dpm/ml

M = total activity released, dpm

A = area of stream cross-section, ft^2

D_x = longitudinal dispersion coefficient, ft^2/min

t = time, min

u = average velocity of water, ft/min

x = distance downstream from point of release, ft

R = sorption coefficient, per hour.

2. The observed modal concentrations of ^{85}Sr and ^{137}Cs in a turbid stream typical of these studies, can be estimated from:

$$C_{OP} \text{ in dpm/ml} = \frac{3.53 \times 10^{-4} M}{A\sqrt{4\pi D_x (x/u)}} e^{-ax}$$

where a = transport coefficient, per ft .

3. The transport coefficient, " a " and the sorption coefficient, " R " are dependent on the amount of turbidity in the stream, settling rate of suspended solids, sorption, nature of radionuclide, and hydrodynamic characteristics. The values of " a " for ^{137}Cs were found to be four times greater as compared to those for ^{85}Sr in presence of Attapul-gite clay suspensions.

4. The relationship between the specific activity of ^{137}Cs on clay and in water is directly dependent on the clay concentration, radionuclide concentration, and velocity, and may be expressed:

$$S_y = d (S_w)^c$$

where S_y = specific activity of radionuclide on suspended clay, dpm/gm
 S_w = specific activity of radionuclide in water, dpm/ml
 d, c^w = constants depending on the nature of radionuclide and type of clay introduced.

5. As shown in aquaria studies, the bottom sediment concentration factor, C_s , and the plant (Vallisneria) concentration factor, C_p , for ^{85}Sr are functions of contact time. The relationships may be expressed:

$$C_s = 29 t^{0.475}$$

and $C_p = 235 t^{-0.128}$

where t = contact time, days.

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CHAPTER I

INTRODUCTION

The future development of nuclear power plants poses interesting problems in waste management and disposal, particularly in the area of water resource development. Nuclear power facilities, as manufacturing installation, may release waste products under certain conceivable conditions. These wastes or radionuclides enter a stream and are transported downstream during which time further dilution or possible concentration occurs.

During transport, physical and chemical reactions occur. The radionuclides may undergo changes in state, remain in solution, or interact with the various components of the aquatic system. Experience based on studies involving the Clinch, Mohawk, Savannah, and Columbia Rivers indicates that certain radionuclides released to these streams will become associated with suspended sediments. However, data derived from these systems seem to be limited and do not permit the development of a usable transport equation. Hence, this study was undertaken to investigate the effect of induced clay suspensions on the aquatic transport of radionuclides.

Objectives

The main purpose was to investigate the effect of an induced turbidity load on the transport of ^{85}Sr and ^{137}Cs in an aquatic environment. Specifically, it was desired to obtain a meaningful sorption-concentration relationship under steady and unsteady release conditions.

Scope

Laboratory experiments were designed to acquire basic information regarding the behavior of ^{85}Sr and ^{137}Cs in an aqueous system.

In particular, the laboratory study involved:

- development of sorption-concentration relationship with different concentrations of Attapulgite clay, different levels of radionuclide concentrations, and same pH; and

- determination of sorption differences between Lake Austin sediment suspensions and Attapulgite clay suspensions.

Experiments involving closed ecosystems included studies designed to establish the amount of uptake and release, and the distribution of ^{85}Sr in water, sediments and plants. The experiments involved the instantaneous release of ^{85}Sr into a non-flowing aquaria system containing an Attapulgite clay suspension.

Model river experiments were conducted, simulating a slow-moving, unpolluted, turbid stream. Attapulgite and Kaolinite clays were used to create the turbidity in the three model river experiments. In the first experiment, ^{85}Sr and ^{137}Cs was released instantaneously, and Attapulgite clay was released continuously. In the second experiment, Kaolinite clay and ^{137}Cs were substituted for the materials used in the first experiment. In the third experiment, ^{137}Cs and Attapulgite clay were released under steady-state conditions.

CHAPTER II

LITERATURE EVALUATION

This chapter deals with the evaluation of relevant literature concerned with the sorption of ^{85}Sr and ^{137}Cs by suspended and bottom sediments, the important factors affecting sorption of these radionuclides from the point of physical, chemical, and hydrodynamic characteristics, and the nature of sorption processes involved in the transport of these radionuclides.

The transport of radioactive materials by surface water is affected by the dispersal of dissolved and suspended matter in water and the sorption by natural materials in contact with water. The dissolved solids and suspended solids transported by the surface water may not move at the same rate. The particles which remain in suspension are probably transported at the velocity of the water, and, therefore, at the same rate as dissolved matter (1). The coarser materials which are not maintained constantly in suspension, will, however, tend to settle, and hence, travel at a slower rate. It is therefore necessary to know the extent of sorption by the suspended solids in the stream, in order to predict the rate of movement of radioactive substances.

Sorption of ^{85}Sr and ^{137}Cs by Suspended and Bottom Sediments

Several studies have been made in determining the removal of strontium and cesium by suspended and bed sediments and soils (13, 29, 31, 34, 40, 58, 59, 68, 88).

Carritt and Goodgal (8) studied the uptake of radioactive strontium in solution by suspensions of Roanoke River and Chesapeake Bay sediments. The initial concentration of activity in their experiments were several orders of magnitude in excess of the recommended MPC* values. Barker (1) investigated the sorption due to ion exchange of ^{137}Cs and ^{90}Sr by kaolinite, illite, halloysite, and two types of montmorillonite. Sediment concentrations and contact time were constant at 4000 mg/l and 17 hr. in these experiments. Comparison between the results of carrier-free and the carrier experiments indicated that both the concentration of radioactive substance and the concentration of sodium and calcium affect the amount of ^{90}Sr and ^{137}Cs sorbed.

The results from experiments conducted by Lackey, et al. (45) with five types of turbidity-causing materials - muck, silt, clay, fuller's earth, and silica sand - indicated that clay and fuller's earth removed $^{137}\text{CsCl}$ most readily by adsorption.

Goldsmith and Middlebrooks (28) studied the ^{90}Sr and ^{137}Cs removals by slurring with Yazoo and Zilpha clays that are abundantly available throughout the state of Mississippi. Both clays provided good removals of ^{137}Cs approximately 90 percent at 250 mg/l clay concentration. Between the two clays, Zilpha clay was more efficient in removing ^{90}Sr to the extent of 97 percent with clay concentration of 4000 mg/l compared to 57 percent removal by Yazoo clay under identical conditions.

The experiments conducted by Garder and Skulberg (26), both in

*Maximum permissible concentration

the laboratory and in a model recipient with postglacial marine clay from the River Nitelva area, Norway, indicated that appreciable amounts of ^{134}Cs and ^{137}Cs were sorbed to the sediments, and that the sorption of ^{89}Sr was insignificant. ^{134}Cs was used in the laboratory experiments and ^{137}Cs , in the model recipient. The removals of radionuclides from solutions was mainly due to the sorption by suspended clay minerals. Laboratory experiments were done with 16, 32, 64, 128, and 256 mg/l of H_2O_2 treated clay suspensions in distilled water and in River water with isotope concentrations between 0.5 $\mu\text{C}/\text{l}$ and 1 $\mu\text{C}/\text{l}$ (1110 and 2220 dpm/ml). The results are shown in Tables 2-1 and 2-2.

Factors Affecting Sorption

The factors affecting the nature and extent of sorption of radiostrontium and radiocesium in a stream are dependent upon a large number of variables. Some of them are the physical nature and type of sediment, composition of sediment, the hydrogen-ion concentration of the solution, and presence of other ions. The transport of sorbed radioactive components by fluvial sediments depends on hydrodynamic characteristics such as velocity and turbulence, sedimentation, and resuspension of the deposited sediments.

Most surface waters contain varying amounts of turbidity, usually in the form of fine clay or silt particles (87).

Clay Minerals and Their Structure: The component which enters into the sorption reaction in most of the cases is the clay fraction of the sediment. The sorption of radionuclides on to clay has been

Table 2-1. Sorption of ^{134}Cs and ^{89}Sr by Clay Suspended in
Distilled Water at 1-Day Contact Time (26)

Radionuclide	Clay Concentration (mg/l)	Activity Sorbed (%)	f_s/M (gm^{-1})	f_l/V ($10^{-3}/\text{ml}$)	$K_d = \frac{f_s/M}{f_l/V}$ (ml/g)
^{134}Cs	16	20.0	125	8.00	15,600
	32	31.0	97	6.90	13,600
	64	43.0	67	5.70	11,800
	128	58.2	45.5	4.18	10,800
	256	65.2	25.4	3.48	7,300
^{89}Sr	16	2.5	15.6	9.75	1,600
	32	4.6	14.4	9.54	1,500
	64	6.2	9.7	9.38	1,030
	128	9.5	7.4	9.05	820
	256	14.5	5.7	8.55	660

f_s = fraction of activity associated with clay

f_l = fraction of activity remaining in solution

M = weight of clay, gm

V = volume of solution, ml

Table 2-2. Sorption of ^{134}Cs and ^{89}Sr by Clay Suspended
in River Water at 1-Day Contact Time (26)

Radionuclide	Clay Concentration (mg/l)	Activity Sorbed (%)	f_s/M (gm^{-1})	f_l/V ($10^{-3}/\text{ml}$)	$K_d = \frac{f_s/M}{f_l/V}$ (ml/g)
^{134}Cs	16	12.5	78.3	8.75	8,900
	32	19.0	59.3	8.10	7,300
	64	26.0	40.6	7.40	5,500
	128	39.0	30.5	6.10	5,000
	256	50.0	19.5	5.00	3,900
^{89}Sr	16	1.5	9.38	9.85	950
	32	2.5	7.80	9.75	800
	64	5.0	7.80	9.50	800
	128	8.5	6.65	9.15	725
	256	14.5	5.70	8.55	660

f_s = fraction of activity associated with clay

f_l = fraction of activity remaining in solution

M = weight of clay, gm

V = volume of solution, ml

intensively studied for many years by soil scientists (29). Considerable work has been reported on the use of clay minerals for the removal of ^{89}Sr , ^{90}Sr , and ^{137}Cs from water (5, 6, 16, 33, 37, 47, 57, 84, 86, 87, 93, 91).

Grim (30) described clays as aggregates of extremely minute crystalline particles of one or more species of a small group of minerals known as the clay minerals. Baver (2) defined clays as disperse systems of the colloidal products of weathering in which secondary mineral particles of smaller than $2\ \mu$ predominate. The upper size limits of clays is established on the basis that particles smaller than $2\ \mu$ exhibited Brownian movement in aqueous suspension.

The three important types of clay minerals and their structural differences have been reported elsewhere (42, 63). Each type of clay has a preference for particular elements. The sorption studies at ORNL (89) using kaolinite, illite, montmorillonite, and vermiculite clays have shown the following order of preference for various radio-nuclides:

	Increasing Selectivity →			
Kaolinite	^{60}Co	<	^{137}Cs	< ^{85}Sr < $^{95}\text{Zr-Nb}$
Illite	^{85}Sr	<	$^{95}\text{Zr-Nb}$	< ^{60}Co < ^{137}Cs
Montmorillonite	$^{95}\text{Zr-Nb}$	<	^{60}Co	< ^{137}Cs < ^{85}Sr
Vermiculite	^{60}Co	<	^{85}Sr	< ^{137}Cs

Some of the data from ORNL studies (82) on the uptake of ^{137}Cs and ^{85}Sr by various clay minerals and Clinch River sediments in distilled

water have been reported by Reynolds and Gloyna (64). Illite and vermiculite had the greatest affinity for ^{137}Cs ; whereas montmorillonite had the strongest affinity for ^{85}Sr .

The sorptive capacities of clay minerals vary according to their structures and chemical composition (10, 28, 39). The results of Oi and Ohashi (60) indicated that at low ^{137}Cs concentrations (less than 10^{-6}M) a preferential structural adsorption occurs, producing an increase in ^{137}Cs removal. An explanation of this structural adsorption is described by Tamura and Jacobs (95), Schulz and his associates (80), and Jacobs (35). Tamura and Jacobs (94) showed that exchange and surface area are secondary to the c-axis spacing in predicting the cesium sorption behavior of clays.

Particle Size and Surface Area: The most important single sediment characteristic affecting the extent of sorption is probably the particle size (69). Particle size is important in determining both the extent and type of sorption. Sorption by sediments is roughly proportional to the surface area. The relationship of particle surface to particle size is known as specific surface - surface area per unit mass of soil. The variation of specific surface of spherical sediment particles with particle diameter is shown in Figure 2-1 (70).

From the studies made on river samples of suspended and bed materials, it is shown that the exchange capacities of the samples to be distributed among the sand, silt and clay components varied as follows (40):

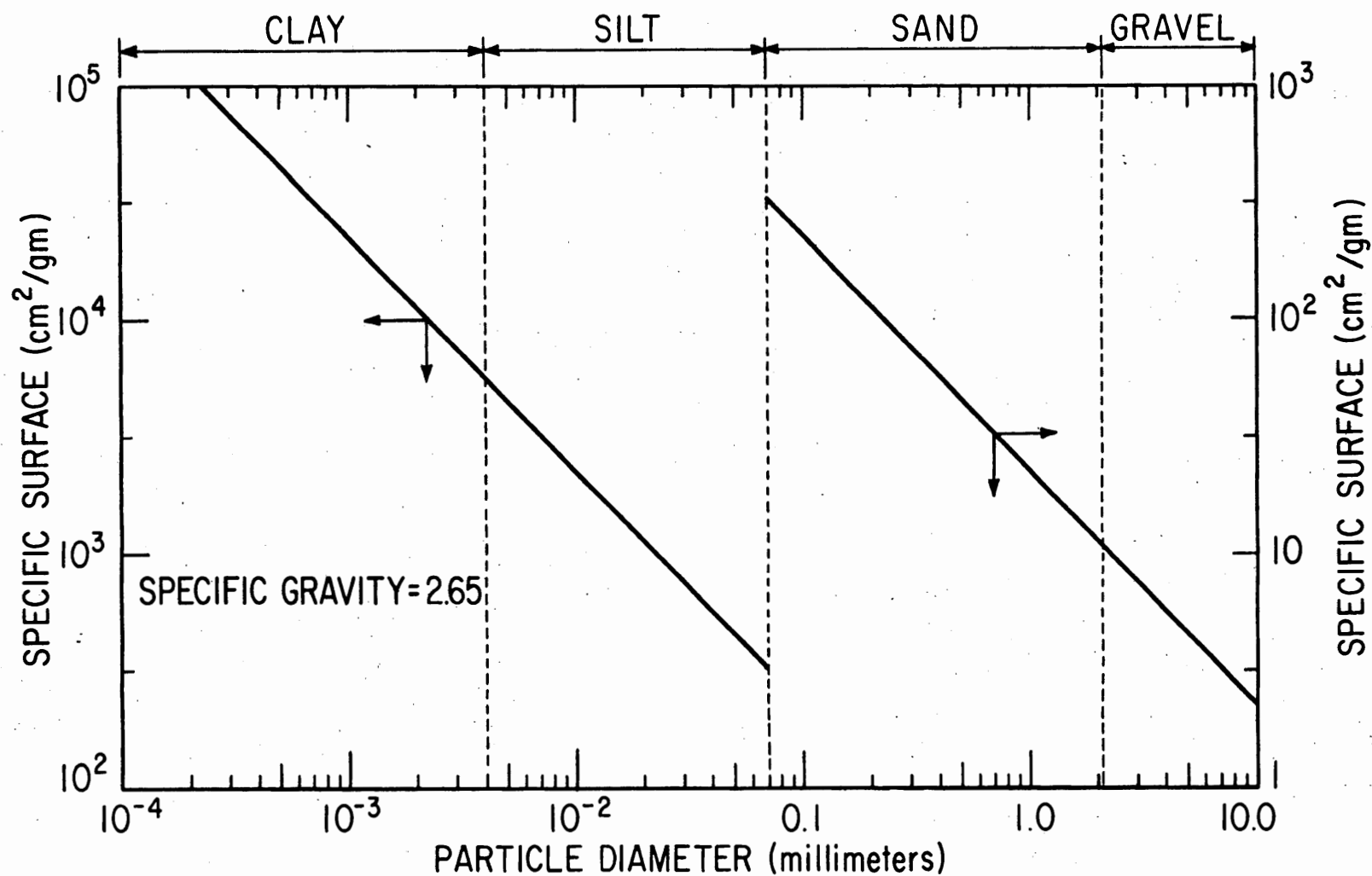


FIG.2-1. VARIATION OF SPECIFIC SURFACE OF SPHERICAL SEDIMENT PARTICLES WITH PARTICLE DIAMETER (70)

	<u>Bed Material</u>			<u>Suspended Material</u>		
	<u>Sand</u>	<u>Silt</u>	<u>Clay</u>	<u>Sand</u>	<u>Silt</u>	<u>Clay</u>
	(%)	(%)	(%)	(%)	(%)	(%)
Average	25	45	31	10	21	68
Maximum	92	85	62	36	45	90
Minimum	4	3	2	1	8	52

As the particle size increases, the chemical activity of soil tends to decrease (74). This fact may be instrumental in effecting adsorption as the principal mechanism by sands, and ion exchange as the dominant mechanism by clays. The exchange capacities of various fractions varied with the size as shown below (40):

Sand	0.5 - 18.9 meq/100 gm
Silt	9.6 - 35.4 meq/100 gm
Clays	20.4 - 77.6 meq/100 gm

The cation exchange capacities and size ranges of some clay minerals are given in Table 2-3.

Table 2-3. Cation Exchange Capacities and Size Ranges of Certain Clays (73)

Name of Clay Mineral	Cation Exchange Capacity (meq/100 gm)	Approximate Size Range (μ)
Kaolinite	3 - 15	0.5 - 2.0
Montmorillonite	80 - 125	0.01- 0.1
Illite	20 - 40	---
Attapulgite*	20 - 30 (27)	0.04- 2.0*

*From Technical Information No. 1004 of Minerals and Chemicals Philipp Corporation (Revised June 1964)

Colloidal State and Flocculation: Particles with an equivalent spherical diameter or Stokes diameter smaller than 2 microns are considered to fall in the colloidal size range (24, 98).

Due to the fact that clay particles take part in the transport of radionuclides, the colloidal state and flocculation of these particles are important. The dispersion and cohesion of colloidal sediment in river has been described by Krone (44).

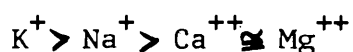
The role of flocculation is one of hindering transport by increasing the settling velocity of suspended particles (42). Since the colloidal clay particles remain in suspension for a longer time, they will be transported further downstream, possibly to a point where flocculation occurs. The factors favorable for the flocculation of colloidal particles are physical aspects such as concentration, collision and cohesion of suspended particles, and the chemical characteristics such as presence of electrolytes, pH, and nature and valence of ions.

pH: The effect of pH on strontium and cesium uptake has been reported in the literature (11, 12, 36, 58, 59, 63, 82, 92). Notably, Reynolds and Gloyna (63) reported that the removal of ^{89}Sr increased at higher pH values and that of ^{137}Cs was less sensitive to pH changes. McHenry (59) states that ^{90}Sr removal was optimum at a pH of approximately 10. Since the chemical behavior of strontium is similar to that of calcium, it may be expected that optimum removals of ^{90}Sr by coprecipitation would occur at high pH values (28). Cheng and Hamaguchi (12) report that the range for optimum removal of ^{137}Cs by most clays is from pH 3 to pH 8. Below pH 3, hydrogen ions compete

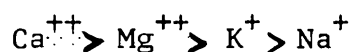
with ^{137}Cs ions, whereas at high pH, other excess cations may also offer competition to the ^{137}Cs ions. The results of Jacobs (37) and Lacy (48) showed that the increased hydrogen ion concentration decreases ^{137}Cs sorption.

Presence of Ions: Spitsyn and Gromov (83) found the presence of $\text{CO}_3^{=}$ and $\text{SO}_4^{=}$ anions in solution to be detrimental to the adsorption of ^{90}Sr by clay.

The monovalent cations are more inhibitive to ^{137}Cs uptake by the river sediment than the divalent cations (63). The order of competition is



In the case of ^{89}Sr uptake, the divalent ions are more inhibitive than the monovalents; the order of competition is



The dependency of cesium sorption upon the total concentration of cations in water has been cited in the literature (1, 37, 68, 69). Because of the similarity in chemical behavior of strontium and calcium, the predictions concerning the strontium sorption would have to also consider the sodium-calcium ratio in the water.

Hydrodynamic Characteristics: The dispersion, transport, and concentration of radioactive substances in streams are controlled by factors like velocity and turbulence, transport of suspended sediment, settling of the radioactive sediment, and under suitable conditions, resuspension of the settled deposits.

The dispersion of radionuclides in the flowing streams by both molecular and turbulent diffusion is a common occurrence, and by far, turbulence is the predominant mechanism causing dispersion (69). The degree and extent of sorption reaction depend on the concentration of contaminant, and in turn, concentration is dependent on dilution as accomplished through diffusion and turbulent mixing processes.

Sayre and Hubbell (77) found that the dispersion of bentonite clay was not significantly different from that of Rhodamine-B dye under the particular set of experimental conditions they described. The experiments were conducted in a canal, 1600 ft. long, 7 ft. bottom width, and $1\frac{1}{4}$ to 1 side slopes. The water discharge was 24 cfs, the water depth was 1.7 ft., and the water-surface slope was 0.00012.

Patterson and Gloyna (62) worked with one version of the dispersion equation,

$$C = \frac{M}{A\sqrt{4\pi D_x t}} e^{-\frac{(x-Ut)^2}{4D_x t}} \quad (2-1)$$

and an empirical formula for the longitudinal dispersion coefficient is as follows:

$$D_x = 0.8e^{0.034U\sqrt{A}} \quad (2-2)$$

where

- C = concentration of tracer
- M = mass of the tracer released
- A = cross-sectional area
- D_x = longitudinal dispersion coefficient
- t = time
- x = distance
- U = mean velocity in the longitudinal direction

The sediment particles in turbid streams are kept in suspension by turbulence. The velocity of flow alone cannot maintain the particles permanently suspended in a stream (21). Lateral variations in suspension concentrations usually occur if sand is the transported load, but they are minor for clays and silts.

The vertical distribution of suspended sediment concentrations at a stream cross-section may be calculated with Equation 2-3 (22).

$$\frac{C_y}{C_a} = \left[\frac{a(d-y)}{y(d-a)} \right]^{\frac{V_s}{0.40u^*}} \quad (2-3)$$

where C_y = suspended solids concentration at a distance y above bed
 C_a = measured suspended solids concentration at reference distance a above bed
 d = total water depth
 V_s = settling velocity of the particular grain size
 u^* = shear velocity of flow

Observations show that clay and silt will have uniform vertical distribution for all practical purposes (76). The concentration of sand in a vertical section of a stream will increase from top to bottom.

If the velocity of a stream decreases, part of the sediment load will be deposited on the bottom. In cases where deposition occurs radioactivity may concentrate to undesirable levels. During floods, the contaminated deposits may be resuspended. Sorption of radionuclides by suspended matter has been reported the greatest during periods when the discharges and suspended solids concentrations are high (20).

Sorption Processes

The processes by which most radionuclides associate with clay minerals have been described in the literature as adsorption, absorption, and ion exchange (9, 47, 71, 88, 96).

Physical Sorption: Sorption on sand surfaces is primarily physical, and the sorption capacity may be due to clay particles adhering to the sand surfaces. This has been demonstrated by a 90 percent reduction of sorptive capacity of clean sand after ultrasonic cleaning (40). Reynolds and Gloyna (65) reported a reduction of about 75 percent. The physical sorption occurring with colloidal clay particles is secondary to other types of sorption particularly ion exchange (75).

Ion Exchange: Ion exchange is a type of chemisorption, generally associated with the clays, and the fine-grained soils. The exchange reactions are described by selectivity coefficients or mass action constants which are developed under equilibrium conditions.

The sorption mechanism of ^{137}Cs and ^{85}Sr by clays and stream sediments has been explained as an ion exchange phenomenon (6, 37, 90, 92, 96).

Distribution Coefficient and Selectivity Coefficient: The distribution coefficient describes the relative affinity of an ion for the solid and liquid phases of a sorption system, while the selectivity coefficient describes the exchange reaction between two phases independent of ionic concentration. Equilibrium conditions are implied in

the development of both coefficients.

Clinch River studies (83, 88) on sediments and minerals have shown that sorption could be expressed by distribution coefficient, K_d , given as

$$K_d = \frac{f_s/M}{f_l/V} \quad (2-4)$$

where f_s = activity fraction associated with the sorbent
 M = weight of the sorbent, gm
 f_l = activity fraction remaining in the solution
 V = volume of solution, ml

The distribution coefficients and selectivity coefficients of ^{137}Cs and ^{85}Sr with various clay minerals, river, lake, and bay sediments under different environmental conditions have been reported elsewhere (14, 26, 53, 66, 82).

Trace Concentration: The amount of radioactivity released into streams are governed by the MPC* values, with a factor of safety of about 10 (70), and consequently, the radionuclides are present in waters at "trace" concentrations, far below the exchange capacities of sorbents in the system, and thus only a small fraction of the exchange sites are occupied (43). Hence, the concentration of ionic species in the solid and liquid phases remain practically unchanged during the sorption process due to the presence of a trace component.

Schubert and Conn (78) state that, in a true cation exchange process for a trace component, K_d is constant when M/V (Equation 2-4)

*Maximum permissible concentration

is varied, and decreases when the bulk electrolyte concentration is increased. It also decreases with the addition of a complexing agent to the solution. For an adsorption process, the K_d value varies directly with M/V which is in contrast to the exchange process.

The results of studies by Reynolds and Gloyna (66) showed that K_d isotherm for ^{137}Cs did not approach linearity for low sediment concentrations up to ~ 150 mg/l, although the ^{137}Cs used was carrier-free. The K_d isotherm for ^{89}Sr was not entirely linear for sediment concentrations varying from 50 to 2000 mg/l.

Transport of Radiostrontium and Radiocesium by Suspended Sediments

The radionuclides discharged from White Oak Dam included ^{90}Sr and ^{137}Cs which have been found in varying proportions in Clinch and Tennessee River waters and silts (17, 56). A study of radionuclide movement in White Oak Creek covered a period with widely ranging stream discharges and sediment loads (17). Large stream flows transported the greatest amounts of ^{137}Cs and ^{90}Sr . Largest fractions of ^{137}Cs were sorbed with greatest suspended loads, regardless of discharge. Almost no ^{137}Cs was sorbed on to the particles larger than $9\ \mu$. The sorption of ^{90}Sr was high only at times of high suspended loads and was mainly on particles smaller than $9\ \mu$. The field data demonstrated that ^{90}Sr was transported almost entirely in solution in White Oak Creek and the rivers beyond (55).

The U.S. Geological Survey and the Oak Ridge National Laboratories have done sorption studies using suspended bed sediments in an

attempt to evaluate the transport of radionuclides in streams (68,88). Lackey (46) indicated that differences between the transport of radionuclides in the Clinch-Tennessee Rivers and the Columbia River was probably due to differences in the suspended load. The turbid water of the Clinch and Tennessee were thought to transfer radioactivity to the bottom more rapidly than the clear Columbia River waters. Lackey et al. (45) described a highly turbid stream to be a decontaminating medium due to dispersal and sedimentation of clays with sorbed radionuclides.

In the Mohawk River studies (25), ^{137}Cs was believed to be quickly removed from solution by clay material and deposited wherever diminishing velocities occurred.

Reynolds and Gloyna (63), and Clanton and Gloyna (14) reported investigations of selected rivers, lakes, and bays in Texas to determine the role of sediments in the uptake of ^{89}Sr and ^{137}Cs from water. They found the radionuclides associated with river sediments decreased when the sediments were transported into the bays.

Klingeman and Kaufman (42) investigated the transport of radionuclides with suspended sediment in estuarine systems. Particular emphasis was given to fission products of recent fallout origin, ^{95}Zr - ^{95}Nb . The transport, deposition, and redistribution of recent river suspensions within portions of the San Francisco Bay system were followed with the help of large masses of labelled river suspended sediment by ^{95}Zr - ^{95}Nb .

Parker (61) proposed an analytical expression for radionuclide concentration including sediment and biota. He suggested the addition of e^{-Nt} to the Fickian dispersion expression and called N the "River-uptake coefficient".

Shih and Gloyna (80) proposed a transport function based on the sorption-desorption model for instantaneous release of ^{85}Sr without the addition of any pollutant. This promises to be a good starting point wherein the physical, chemical, and biological sorption mechanism as well as the release process are given a mathematical form. The transport function developed by Shih and Gloyna is given in Equation 2-5.

$$C(x,t) = \frac{Mx}{2AU\sqrt{\eta D_x}} e^{\left(\frac{Ux}{2D_x} - k_1 t\right)} N(x,t) \quad (2-5)$$

$$\text{where } N(x,t) = \int_0^t I_1 \left[2\sqrt{dk_1 k_s} (t-\tau)\tau \right] \sqrt{\frac{dk_1 k_s}{t-\tau}} \frac{1}{\tau} e^{-\left(\frac{x^2}{4D_x\tau} + B\tau\right)} d\tau$$

This equation may be expressed in a simpler form (Equation 2-6), provided the term $N(x,t)$ is assumed to be a constant instead of a variable with respect to time. Equation 2-6 becomes

$$C(x,t) = \left\{ \text{Constant} \right\} \frac{Mx}{2AU\sqrt{\eta D_x}} e^{\frac{Ux}{2D_x}} e^{-k_1 t} \quad (2-6)$$

another form of dispersion equation with the additional term $e^{-k_1 t}$ (where k_1 is known as mass transfer coefficient). Hence, in practical

engineering application, addition of various coefficients to the dispersion equation for different environmental factors in a stream may provide suitable approximations.

CHAPTER III

LABORATORY STUDIES

These studies were designed to obtain basic information regarding the behavior of ^{85}Sr and ^{137}Cs in trace concentrations with Attapulgitic clay and bottom sediment. Known concentrations of Attapulgitic clay and bottom sediment were mixed with known levels of radioactivity for a given period while maintaining the pH constant.

Attapulgitic clay was used because it is an active example of the inorganic suspended material which may be present in some southeastern and southwestern streams. The bottom sediments were from Lake Austin, and these represent typical bed-load material that can be transported in stream systems. The bed material used in these laboratory studies was also used in the model river.

The specific objective of these laboratory studies were four fold:

1. develop sorption-activity relationships with different concentrations of Attapulgitic clay and Lake Austin sediment at different levels of ^{137}Cs activity;
2. determine sorption-activity relationships with known concentrations of Attapulgitic clay and Lake Austin sediment at known levels of ^{85}Sr activity;
3. establish sorption differences between Lake Austin sediment and Attapulgitic clay suspensions; and
4. calculate K_d isotherms for ^{85}Sr and ^{137}Cs .

Preparation of Clay Suspension

Attapulgite clay was obtained from Georgia under the trade name of Attagel 150. The typical chemical analysis of the Attapulgite clay and its typical properties are given in Tables 3-1 and 3-2. The size of Attapulgite clay particles ranged from 0.04 to 2.0 microns as shown in Figure 3-1 with an average size of 0.12 micron (equivalent settling diameter).

Table 3-1. Typical Chemical Analysis of Attapulgite Clay*
(Volatile-Free Basis)

Constituent	Percent
Silicon (SiO_2)	68.0
Aluminium (Al_2O_3)	12.0
Magnesium (MgO)	10.5
Iron (Fe_2O_3)	5.0
Calcium (CaO)	1.7
Phosphorous (P_2O_5)	1.0
Potassium (K_2O)	1.0
Titanium (TiO_2)	0.7
Trace Elements	0.1

Note: 1) The major constituents shown in the analysis are combined as complex magnesium aluminium silicate and do not exist as free oxides

2) Heavy metals and Arsenic 10-100 ppm

*From Technical Information No. 1004 (Revised June 1964)
of Minerals & Chemicals Philipp Corporation

The stock suspension of Attapulgite was prepared in model river water, the analysis of which is shown in Table 3-3, and the

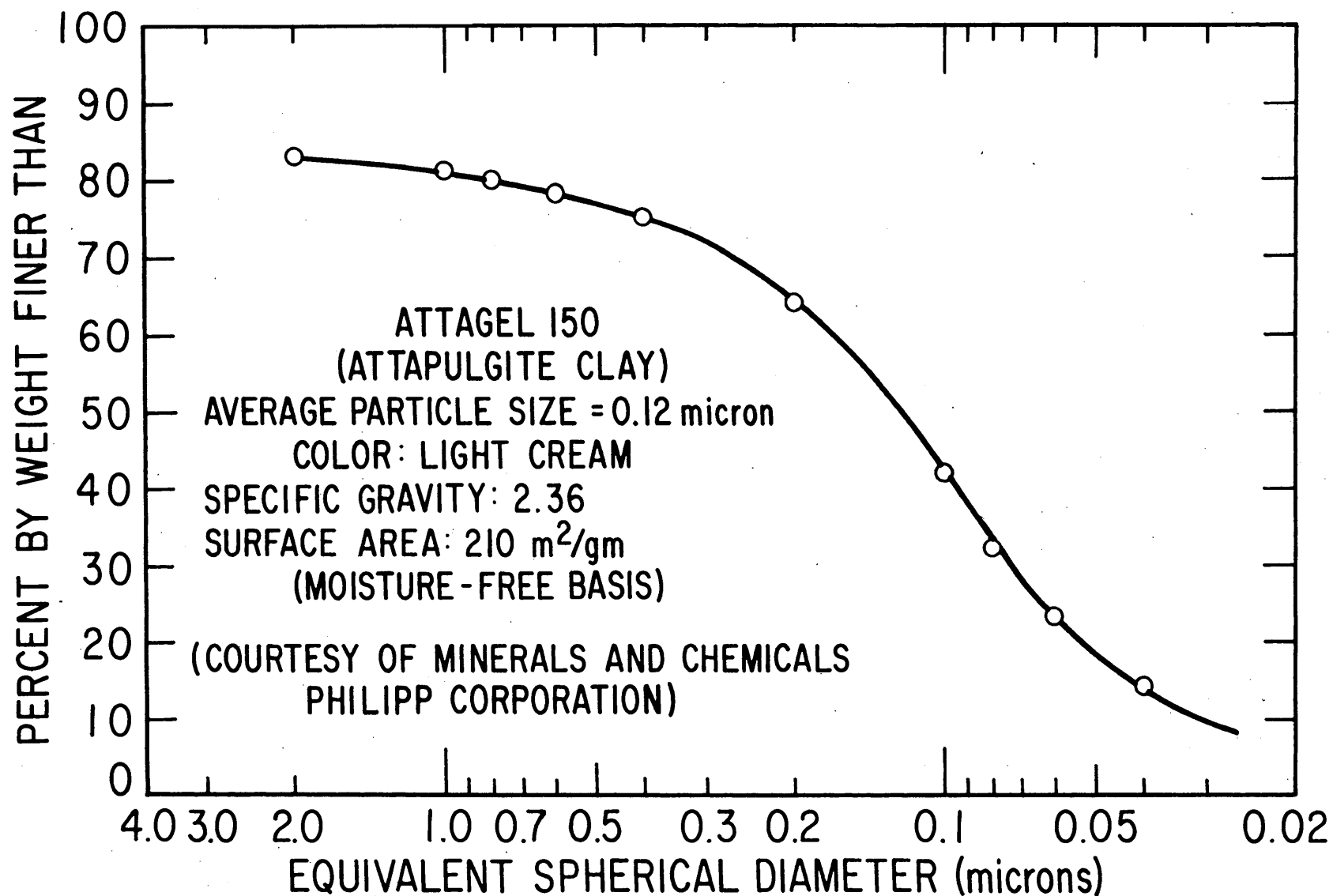


FIG.3-1. PARTICLE SIZE DISTRIBUTION CURVE-ATTAGEL 150

concentration determined gravimetrically (13.6 gm/liter). For each experiment, aliquots from this stock solution were placed in test bottles to give the desired clay concentrations.

Table 3-2. Typical Properties of Attagel 150*

Particle Shape	Needle-like ultimate particle shape
Average Particle Size	0.12 micron
pH	7.5-9.5
Color	Light cream
Specific Gravity	2.36
Surface Area	210 m ² /gm

*From Technical Information No. 1004 (Revised June 1964) of Minerals & Chemicals Philipp Corporation

Table 3-3. Analysis of Model River Water

Parameter	Average Concentration (mg/l)
Total Alkalinity as CaCO ₃	225
Hardness as CaCO ₃	261
pH	8.1
Specific Conductance	640*
Suspended Solids	10
Na ⁺	35
K ⁺	1
Ca ⁺⁺	44

*in μ mhos/cm

Preparation of Lake Austin Sediment

The Lake Austin sediment was dried under infrared heat lamps before it was ground to pass through a 200-mesh sieve. The non-clay minerals were quantitatively estimated by comparing with quartz and calcite standards, and the clay minerals were analyzed by x-ray diffraction. The approximate composition of the sediment is given in Table 3-4.

Table 3-4. Composition of Lake Austin Sediment

Type of Fraction	Percent
Non-clay Minerals (#325 mesh sieve):	
Quartz	36
Calcite	18
Dolomite	6
Clay Minerals (< 2 μ):	
Montmorillonite	20
Mica	10
Kaolinite	8
Organic Matter	2

For each experiment, the required amount of sediment was placed in test bottles to give the desired sediment concentrations.

Exchange Capacity

The cation exchange capacity of Attapulgite clay (Attagel 150) and Lake Austin sediment was determined by the tagged cesium method (3). The results of tests are as follows:

Material	Average Cation Exchange Capacity (meg/100 gm)
Attapulgate Clay	23
Lake Austin Sediment	35

Experimental Procedure

The ^{85}Sr and ^{137}Cs slurry tests were conducted under identical conditions in 4-ounce glass bottles. Each batch comprised of three sets of 4 bottles each. Each set consisted of a control bottle containing no sediment or clay, and three bottles with known sediment or clay concentrations. Details are given in Table 3-5. Model river water was used for all the experiments. Master solutions of ^{85}Sr and ^{137}Cs were prepared using distilled water with specific activity of 44,400 dpm/ml.

Aliquots of Attapulgate clay stock solution were added to give concentrations of 62.5, 125, and 250 mg/l; calculated amounts of the radionuclides were put into the 4-ounce bottles containing water; and then the total volume of solution was made up to 100 ml. In the case of Lake Austin sediments, preweighed dry sediment samples were added to the bottles in place of clay suspension.

The contents were mixed thoroughly after the bottles were capped and placed in a reciprocating bottle shaker for continuous agitation. At intervals of 1 hour, 1 day, 3 days, and 7 days, the bottles were removed from the shaker and the contents centrifuged

Table 3-5. Details of Experimental Set-up
for ^{85}Sr and ^{137}Cs

Batch #1 for ⁸⁵ Sr			Batch #2 for ¹³⁷ Cs	
Set	Concentration (*MPC Level)	Clay or Sediment Concentration (mg/l)	Concentration (**MPC Level)	Clay or Sediment Concentration (mg/l)
I	0.1	0	0.2	0
	0.1	62.5	0.2	62.5
	0.1	125	0.2	125
	0.1	250	0.2	250
II	1.0	0	2.0	0
	1.0	62.5	2.0	62.5
	1.0	125	2.0	125
	1.0	250	2.0	250
III	2.0	0	4.0	0
	2.0	62.5	4.0	62.5
	2.0	125	4.0	125
	2.0	250	4.0	250

*Maximum permissible concentration of ^{85}Sr in water for occupational exposure is 10^{-3} $\mu\text{c/ml}$ (2220 dpm/ml) according to NBS Handbook No.69

**Maximum permissible concentration of ^{137}Cs in water for occupational exposure is 2×10^{-4} $\mu\text{c/ml}$ (444 dpm/ml) according to NBS Handbook No. 69

for 15 minutes at $1060 \times G$. One milliliter of the supernatant liquid was pipetted out and placed in aluminium planchets, dried under heat lamps, and counted for gamma activity. The bottles, after sampling, were capped and returned to the shaker. The sorption of the specific radionuclide by clay or sediment was calculated by the difference between the control bottle activity and the activity of supernatant liquid after centrifugation. Losses of radioactivity to the glass bottle walls were negligible (63).

Since these studies were done to develop a better understanding of the sorption mechanism of ^{85}Sr and ^{137}Cs the pH was carefully monitored. The model river water throughout the experiment exhibited a pH of 8.1 ± 0.1 .

Radioactivity Measurement

Since both ^{85}Sr and ^{137}Cs are gamma emitters with 0.513 and 0.662 Mev energies respectively, the activity in the planchets were counted using a 4" diameter NaI (Thallium activated) crystal and a 256-channel gamma spectrometer. Statistical accuracy was obtained by providing sufficient time. The "drift" of the gamma spectrometer was minimized by calibrating the instrument with ^{137}Cs standard each day. The selected energy span was 1.0 Mev for the 256-channel group size. Accordingly the ^{137}Cs peak of 0.662 Mev energy was adjusted to channel 169.

The ^{85}Sr and ^{137}Cs data were corrected for background, counting efficiency, and decay to determine the fraction of radionuclide

concentration associated with the solid phase (clay or sediment). The procedures have been described elsewhere (80).

Results

The results of the sorption experiments of ^{85}Sr and ^{137}Cs are presented in this section. The effect of sorbent concentration (in this case clay or sediment) at a particular radionuclide concentration and the influence of this sorbent concentration on the uptake of ^{85}Sr and ^{137}Cs are given. The sorption-activity relationships and K_d isotherms for ^{85}Sr and ^{137}Cs uptake are discussed.

^{137}Cs Uptake: Increases in either Attapulgitic clay or Lake Austin sediment concentrations enhanced the sorption of ^{137}Cs at any one of the three activity levels, 0.2, 2.0, or 4.0 MPC. The results are presented in Tables 3-6 and 3-7 for Attapulgitic clay and Lake Austin sediment respectively. The uptake values by Lake Austin sediments were always higher as compared to Attapulgitic clay, Figures 3-2 and 3-3. This was due to the presence of clay minerals, montmorillonite, mica, and kaolinite, yielding a higher cation exchange capacity compared to Attapulgitic. The differences were small at higher concentrations of suspension.

The sorption of ^{137}Cs on Attapulgitic clay and Lake Austin sediment increased with contact time, Figures 3-4. After seven days of contact, the uptake by Attapulgitic was 86 percent and that by Lake Austin sediment was 91 percent at 250 mg/l concentrations. Hence, the contact time is important in evaluating ^{137}Cs transport.

Table 3-6. Cesium-137 Sorption by Various Clay Concentrations
at Different Activity Levels

Attapulgitte Clay Concentration = 62.5 mg/l

Activity Level	Variable	Contact Time			
		1 Hour	1 Day	3 Days	7 Days
0.2 MPC	Sorption (%)	28.7	30.8	35.8	40.0
	K_d (ml/gm)	6430	7100	8920	10,650
	*S.A. (dpm/gm) $\times 10^6$	0.37	0.39	0.46	0.51
2.0 MPC	Sorption (%)	45.0	45.3	50.0	50.0
	K_d (ml/gm)	13,100	13,250	16,000	16,000
	S.A. (dpm/gm) $\times 10^6$	5.95	6.00	6.6	6.6
4.0 MPC	Sorption (%)	53.8	55.7	58.8	59.0
	K_d (ml/gm)	18,600	20,100	22,800	23,000
	S.A. (dpm/gm) $\times 10^6$	12.90	13.40	14.10	14.20

Attapulgitte Clay Concentration = 125 mg/l

0.2 MPC	Sorption (%)	40.6	43.4	48.5	50.8
	K_d (ml/gm)	5460	6120	7540	8250
	S.A. (dpm/gm) $\times 10^6$	0.26	0.28	0.31	0.33
2.0 MPC	Sorption (%)	65.8	67.7	71.7	75.0
	K_d (ml/gm)	15,400	16,800	20,300	24,000
	S.A. (dpm/gm) $\times 10^6$	4.35	4.50	4.75	4.95
4.0 MPC	Sorption (%)	69.0	73.2	76.3	77.6
	K_d (ml/gm)	17,800	21,800	26,800	27,700
	S.A. (dpm/gm) $\times 10^6$	8.30	8.80	9.15	9.33

Table 3-6 (continued)

Attapulgitte Clay Concentration = 250 mg/l					
0.2 MPC	Sorption (%)	51.4	52.8	53.2	55.0
	K_d (ml/gm)	4220	4470	4650	4900
	S.A. (dpm/gm) $\times 10^6$	0.16	0.17	0.18	0.18
2.0 MPC	Sorption (%)	79.8	82.7	83.3	86.0
	K_d (ml/gm)	15,800	19,100	20,000	24,600
	S.A. (dpm/gm) $\times 10^6$	2.54	2.74	2.76	2.84
4.0 MPC	Sorption (%)	82.3	83.8	84.5	86.4
	K_d (ml/gm)	18,600	20,700	21,800	25,400
	S.A. (dpm/gm) $\times 10^6$	4.95	5.03	5.08	5.20

*Specific Activity

Note: Model river water, pH = 8.1, Temp. = 22°C

Table 3-7. Cesium-137 Sorption by Various Sediment
Concentrations at Different Activity Levels

Lake Austin Sediment Concentration = 62.5 mg/l

Activity Level	Variable	Contact Time			
		1 Hour	1 Day	3 Days	7 Days
0.2 MPC	Sorption (%)	54.0	60.0	62.0	75.0
	K_d (ml/gm)	18,300	24,000	26,100	48,000
	*S.A. (dpm/gm) $\times 10^6$	0.86	0.96	0.99	1.20
2.0 MPC	Sorption (%)	65.3	68.2	71.2	77.3
	K_d (ml/gm)	30,100	34,300	39,600	54,500
	S.A. (dpm/gm) $\times 10^6$	8.65	9.02	9.43	10.23
4.0 MPC	Sorption (%)	66.4	70.3	75.4	82.5
	K_d (ml/gm)	31,600	37,800	49,000	74,500
	S.A. (dpm/gm) $\times 10^6$	17.60	18.70	20.00	22.00

Lake Austin Sediment Concentration = 125 mg/l

0.2 MPC	Sorption (%)	62.0	74.0	76.0	78.0
	K_d (ml/gm)	13,060	22,800	25,300	28,400
	S.A. (dpm/gm) $\times 10^6$	0.50	0.59	0.61	0.63
2.0 MPC	Sorption (%)	74.7	76.8	77.4	83.1
	K_d (ml/gm)	23,600	26,500	27,400	39,300
	S.A. (dpm/gm) $\times 10^6$	4.95	5.08	5.12	5.50
4.0 MPC	Sorption (%)	78.2	80.5	81.5	88.7
	K_d (ml/gm)	28,700	33,000	35,200	62,800
	S.A. (dpm/gm) $\times 10^6$	10.40	10.70	10.83	11.79

Table 3-7 (continued)

Lake Austin Sediment Concentration = 250 mg/l					
0.2 MPC	Sorption (%)	68.0	76.0	78.2	80.0
	K_d (ml/gm)	8500	12,700	14,300	16,000
	S.A. (dpm/gm) $\times 10^6$	0.27	0.30	0.31	0.32
2.0 MPC	Sorption (%)	82.3	83.4	85.6	89.1
	K_d (ml/gm)	18,600	20,000	23,800	32,700
	S.A. (dpm/gm) $\times 10^6$	2.72	2.76	2.84	2.95
4.0 MPC	Sorption (%)	83.4	87.7	89.5	91.3
	K_d (ml/gm)	20,100	28,500	34,100	42,000
	S.A. (dpm/gm) $\times 10^6$	5.55	5.83	5.95	6.07

*Specific Activity

Note: Model river water, pH = 8.1, Temp. = 22°C

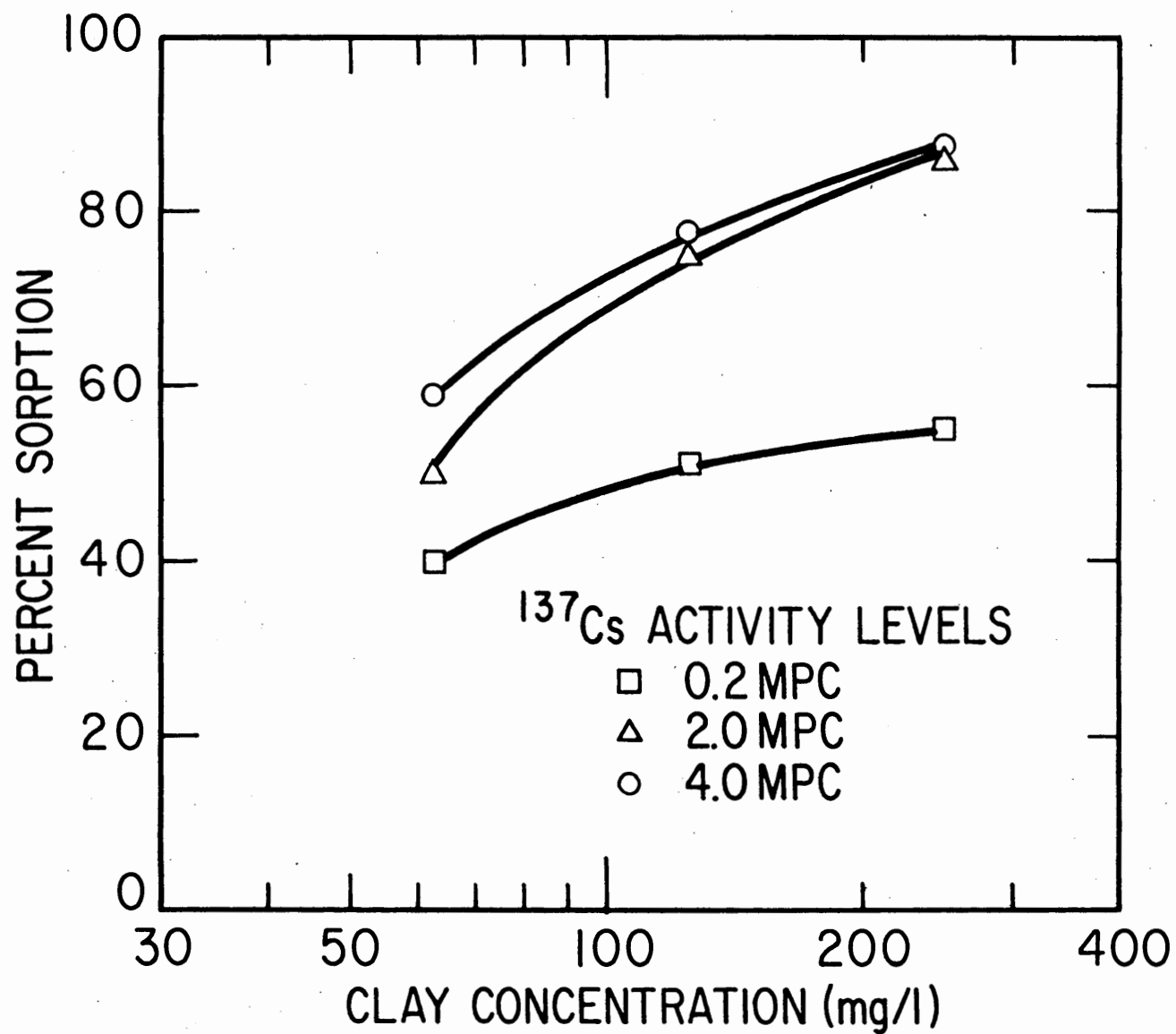


FIG.3-2. SORPTION OF ^{137}Cs BY ATTAPULGITE

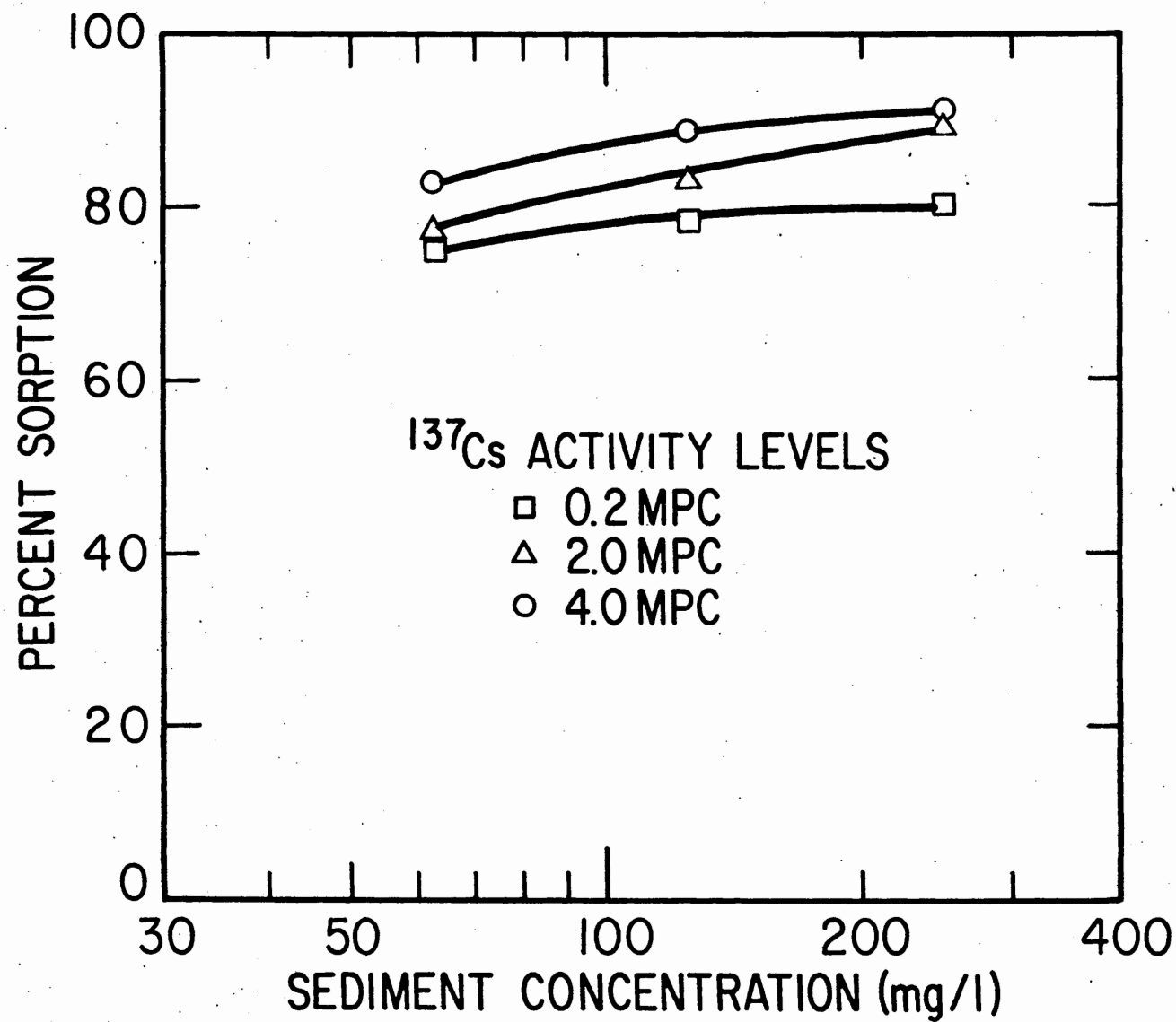
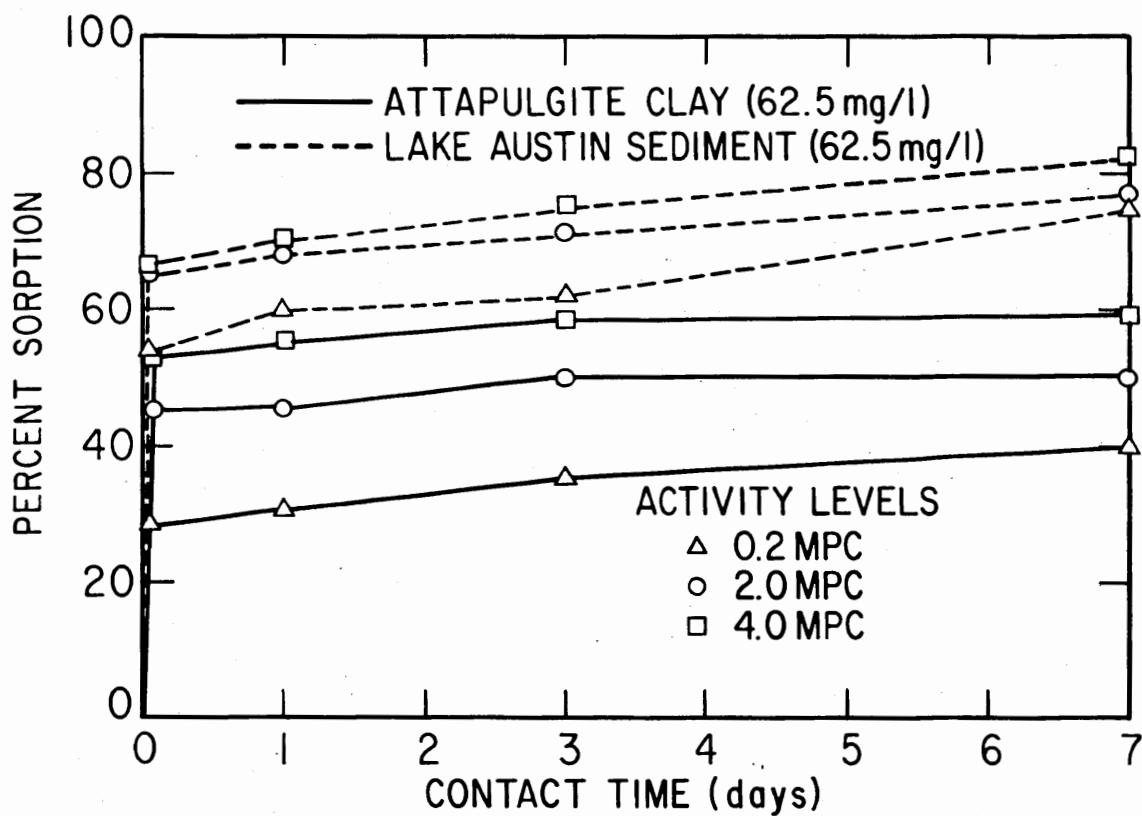
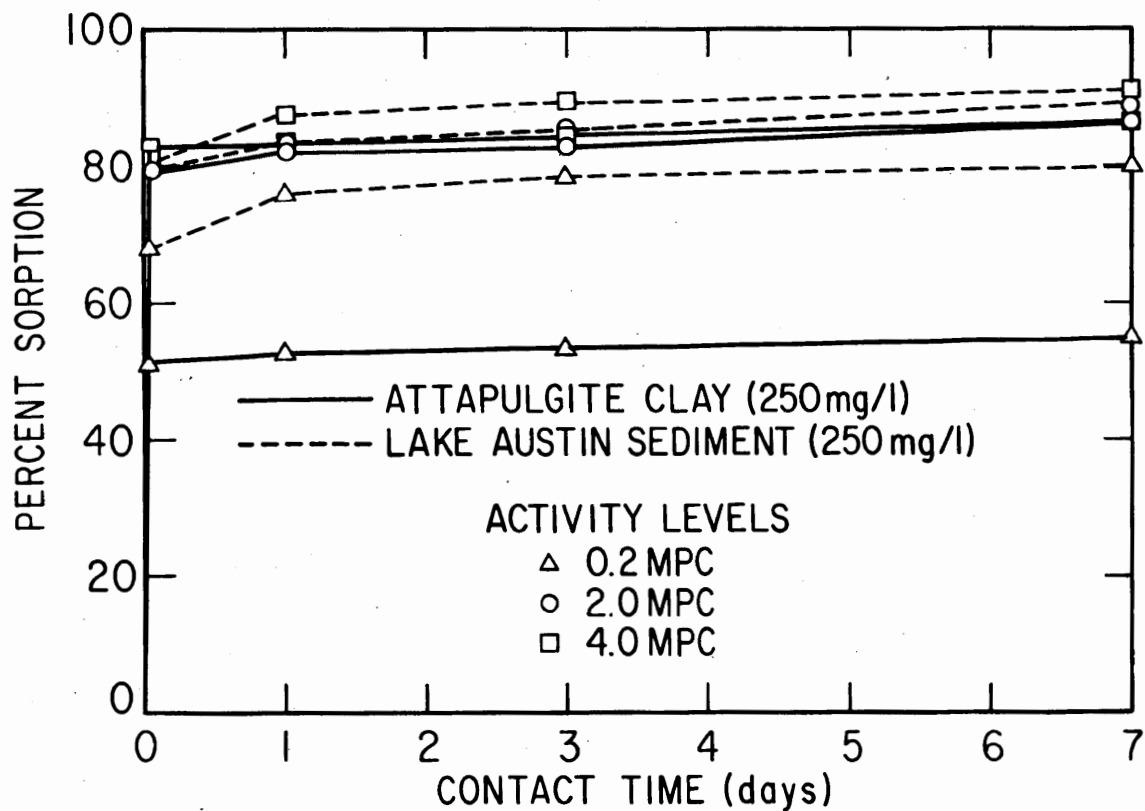


FIG.3-3. SORPTION OF ^{137}Cs BY LAKE AUSTIN SEDIMENT

FIG.3-4. UPTAKE OF ^{137}Cs

As seen from Figure 3-4, the higher the ^{137}Cs concentration, the greater the percent sorption by Attapulgitic and Lake Austin sediment. This uptake was found to be important for both short and long contact times.

^{85}Sr Uptake: An increase in suspended Attapulgitic clay and Lake Austin sediment did not appreciably effect the percent of uptake of ^{85}Sr irrespective of concentration. The results are shown in Figure 3-5 and Tables 3-8 and 3-9. It was found that the sorption of ^{85}Sr at 0.1 MPC levels by both types of suspension was negligible at all sediment concentrations. Though the uptake by Attapulgitic clay was slightly greater than that by Lake Austin sediments, the differences were very small, Figure 3-5. The reduced sorption by the sediment may be due to the presence of calcite and dolomite. The Ca^{++} and Mg^{++} present in the sediment were probably highly competitive with ^{85}Sr as reported by Reynolds and Gloyna (66).

The ^{85}Sr in terms of MPC activity levels did not affect the uptake by Attapulgitic clay and Lake Austin sediment suspensions at any concentration with contact time. The data are presented in Figure 3-6.

Sorption-Activity Relationships: The sorption-activity relationship for ^{137}Cs with both Attapulgitic clay and Lake Austin sediment was found to follow the typical Freundlich-type reaction (Figure 3-7). The reaction may be expressed empirically by the exponential relationship:

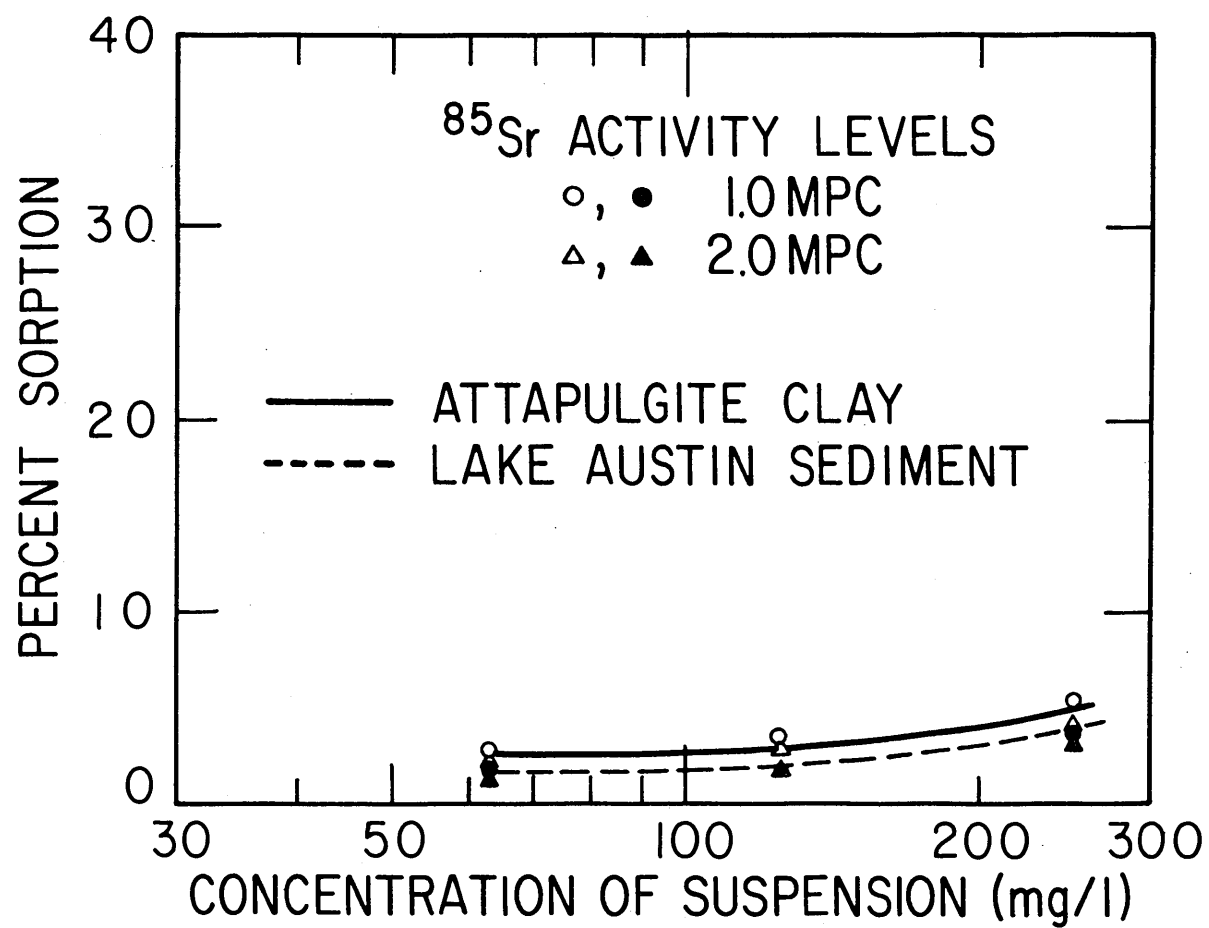


FIG. 3-5. SORPTION OF ^{85}Sr

Table 3-8. Strontium-85 Sorption by Various Clay Concentrations
at Different Activity Levels

Attapulgitte Clay Concentration = 62.5 mg/l

Activity Level	Variable	Contact Time			
		1 Hour	1 Day	3 Days	7 Days
1.0 MPC	Sorption (%)	2.0	2.2	2.2	2.3
	K_d (ml/gm)	326	360	360	376
	*S.A. (dpm/gm) $\times 10^6$	0.76	0.84	0.84	0.87
2.0 MPC	Sorption (%)	2.3	2.4	2.8	2.8
	K_d (ml/gm)	376	397	466	466
	S.A. (dpm/gm) $\times 10^6$	1.75	1.82	2.13	2.13

Attapulgitte Clay Concentration = 125 mg/l

1.0 MPC	Sorption (%)	2.8	2.9	3.1	3.3
	K_d (ml/gm)	230	239	256	273
	S.A. (dpm/gm) $\times 10^6$	0.53	0.55	0.59	0.63
2.0 MPC	Sorption (%)	3.0	3.2	3.3	3.5
	K_d (ml/gm)	247	264	273	290
	S.A. (dpm/gm) $\times 10^6$	1.14	1.22	1.26	1.33

Table 3-8 (continued)

Attapulgate Clay Concentration = 250 mg/l					
	Sorption (%)	3.7	3.9	4.1	4.1
1.0	K_d (ml/gm)	154	162	171	171
MPC	S.A. (dpm/gm) $\times 10^6$	0.35	0.37	0.39	0.39
	Sorption (%)	3.8	3.9	4.9	5.4
2.0	K_d (ml/gm)	158	162	206	228
MPC	S.A. (dpm/gm) $\times 10^6$	0.72	0.74	0.93	1.02

*Specific Activity

Note: Model river water, pH = 8.1, Temp. = 22°C

Table 3-9. Strontium-85 Sorption by Various Sediment
Concentrations at Different Activity Levels

Lake Austin Sediment Concentration = 62.5 mg/l

Activity Level	Variable	Contact Time			
		1 Hour	1 Day	3 Days	7 Days
1.0 MPC	Sorption (%)	1.0	1.1	1.2	1.2
	K_d (ml/gm)	161	178	194	194
	*S.A. (dpm/gm) $\times 10^6$	0.39	0.43	0.47	0.47
2.0 MPC	Sorption (%)	2.0	2.1	2.2	2.4
	K_d (ml/gm)	326	343	360	394
	S.A. (dpm/gm) $\times 10^6$	1.62	1.70	1.78	1.95

Lake Austin Sediment Concentration = 125 mg/l

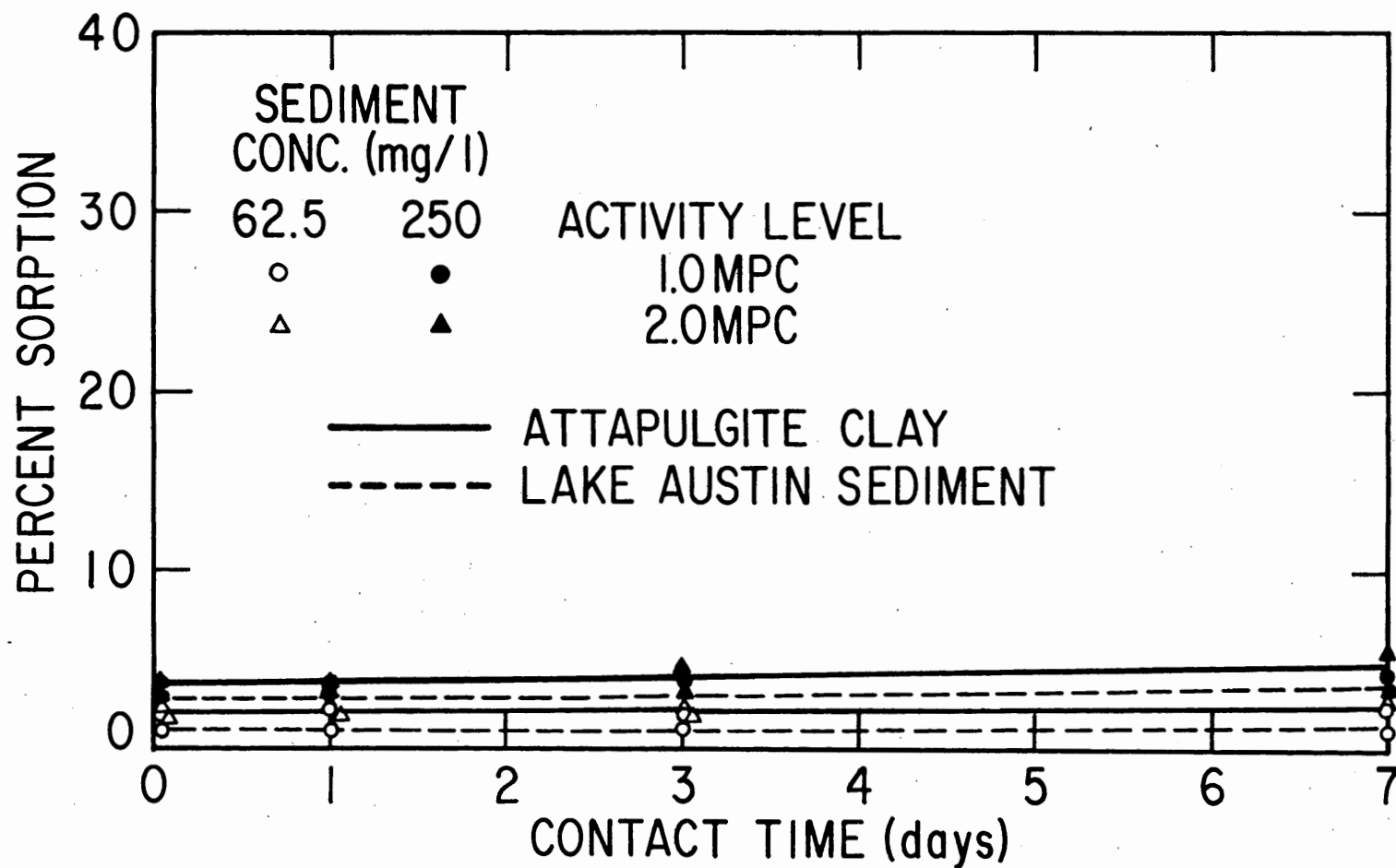
1.0 MPC	Sorption (%)	1.6	1.8	1.8	1.9
	K_d (ml/gm)	130	147	147	155
	S.A. (dpm/gm) $\times 10^6$	0.31	0.35	0.35	0.37
2.0 MPC	Sorption (%)	2.6	2.8	3.2	3.4
	K_d (ml/gm)	214	230	264	282
	S.A. (dpm/gm) $\times 10^6$	1.05	1.13	1.30	1.38

Table 3-9 (continued)

Lake Austin Sediment Concentration = 250 mg/l					
	Sorption (%)	3.0	3.2	3.4	3.5
1.0	K_d (ml/gm)	124	132	140	145
MPC	S.A. (dpm/gm) $\times 10^6$	0.29	0.31	0.33	0.34
	Sorption (%)	3.2	3.4	3.4	3.8
2.0	K_d (ml/gm)	132	141	141	158
MPC	S.A. (dpm/gm) $\times 10^6$	0.65	0.69	0.69	0.77

*Specific Activity

Note: Model river water, pH=8.1, Temp. = 22°C

FIG. 3-6. UPTAKE OF ^{85}Sr

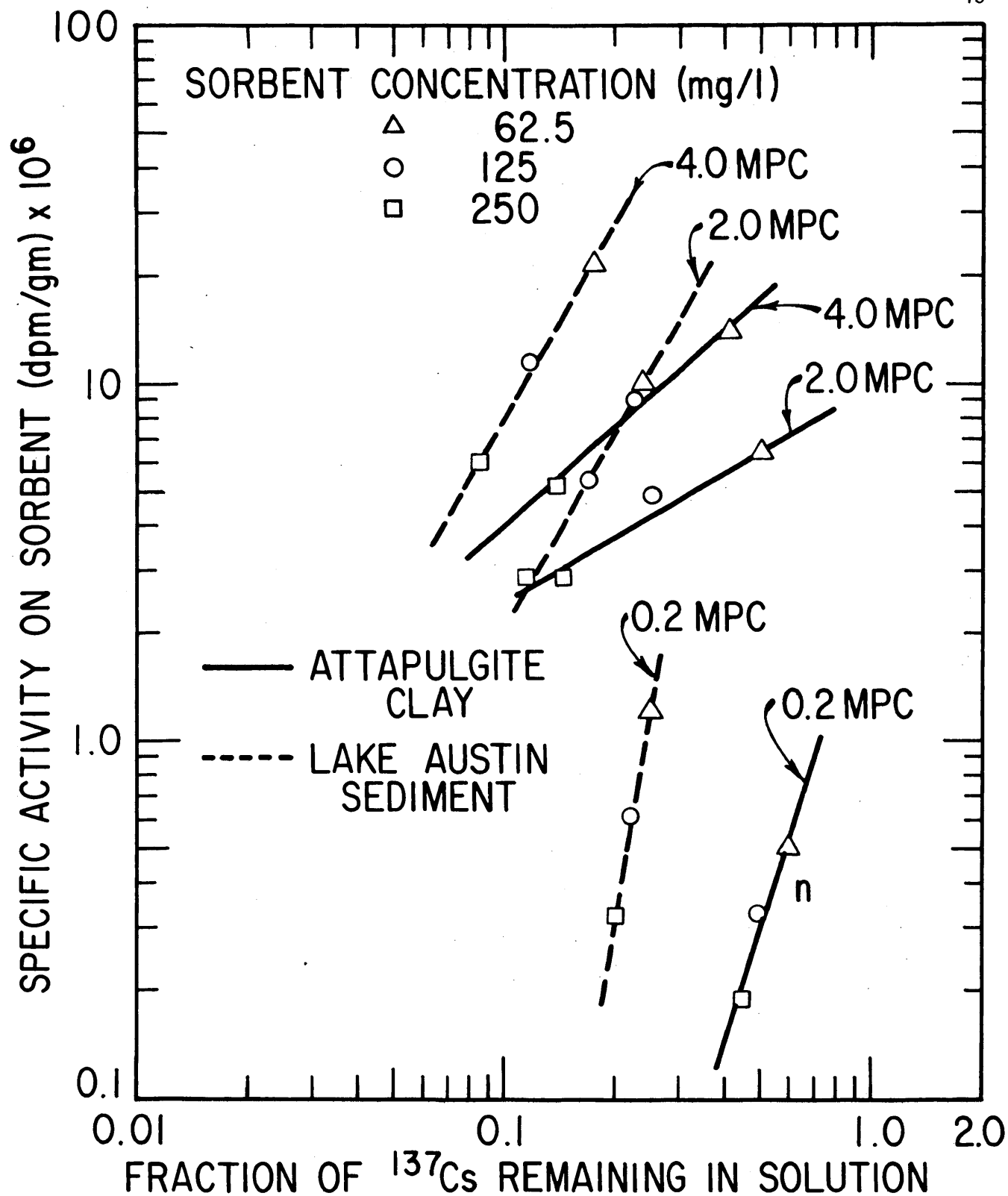


FIG. 3-7. SPECIFIC ACTIVITY ON SORBENT AT DIFFERENT ^{137}Cs ACTIVITY LEVELS

$$\frac{x}{M} = k C^n \quad (3-1)$$

where

- $\frac{x}{M}$ = specific activity on sorbent (sorbed activity per unit weight of sorbent), dpm/gm
- x = C_o minus C , which is the concentration of activity sorbed, or the initial concentration of sorbable activity, C_o , minus final concentration of sorbable activity, C , dpm/ml
- M = amount of sorbent added to the volume of solution, mg/l
- C = concentration of activity remaining, dpm/ml, or fraction of activity remaining, C/C_o
- k, n = constants depending on the sorbent and radionuclide investigated.

Straub and others (87) reported the same type of exponential relationship for ^{85}Sr , ^{91}Y , and ^{144}Ce in studies involving clay soils.

Figure 3-7 shows the fraction of ^{137}Cs remaining in solution versus specific activity on sorbent. Based on test of seven day duration, it is seen that the specific activity on sorbent was low at lower ^{137}Cs concentrations, and increased with higher values. The higher the sorbent concentration, the lower the specific activity on the sorbent. As indicated earlier, the greater sorption capacities of Lake Austin sediment are shown by the larger uptake (Tables 3-6 and 3-7).

The sorption-activity relationship for ^{85}Sr was found to follow the Freundlich-type reaction as reported by Straub and others (87). Based on test of seven days duration, there was no variation in plots indicating that the sorption of ^{85}Sr was not affected by concentration (Figure 3-8). Also, little difference was found in ^{85}Sr sorption between Lake Austin sediment and Attapulgate clay.

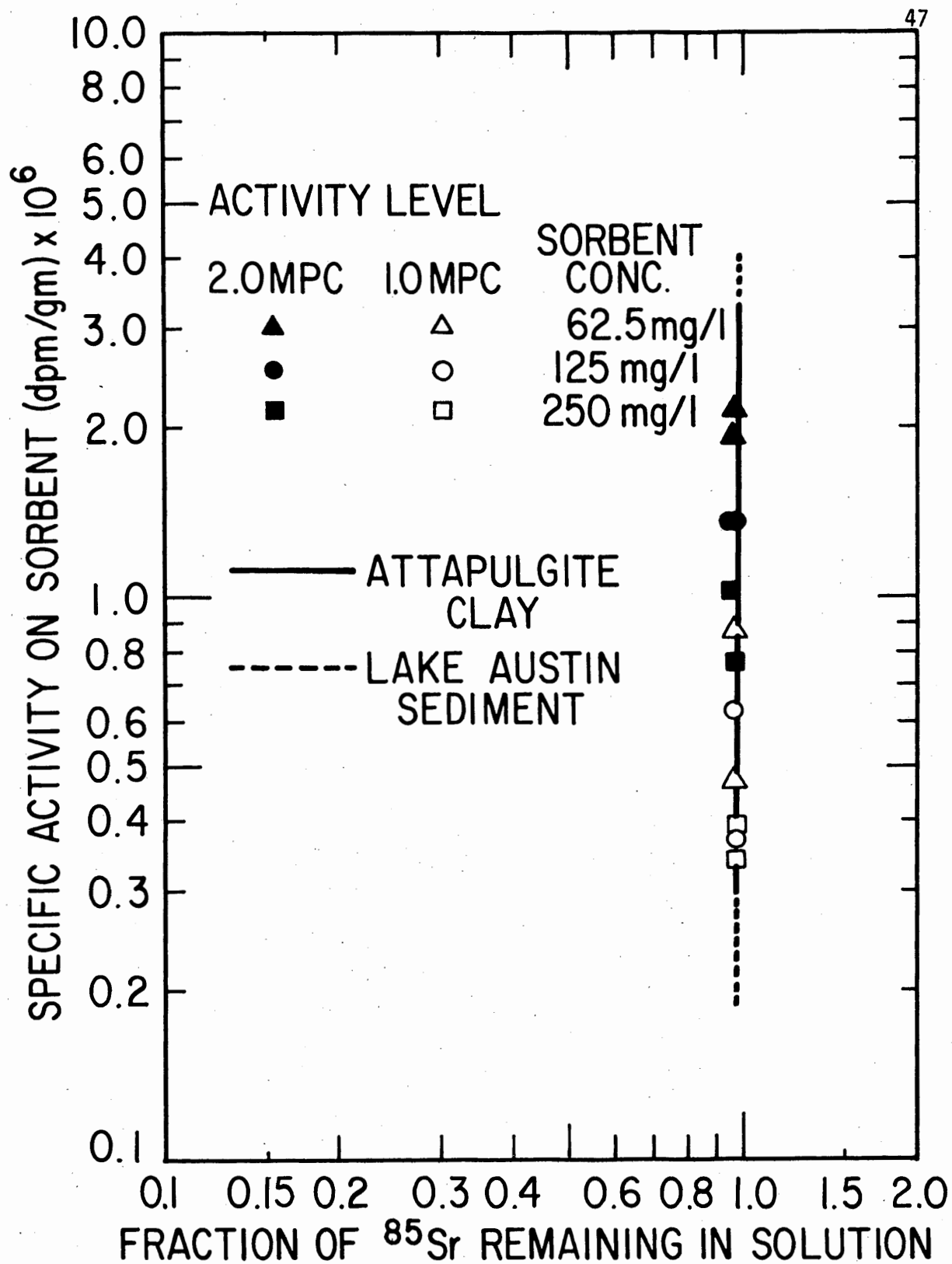


FIG. 3-8. SPECIFIC ACTIVITY ON CLAY AT DIFFERENT ^{85}Sr ACTIVITY LEVELS

K_d Isotherms For ⁸⁵Sr and ¹³⁷Cs: When ⁸⁵Sr and ¹³⁷Cs are present in trace concentrations, equilibria relationships may be represented by a distribution coefficient, K_d (42, 63). The data obtained with carrier-free ¹³⁷Cs are shown in Figures 3-9 and 3-10. Notably, the K_d isotherms of Attapulgitic clay and Lake Austin sediment were not linear. However, Reynolds and Gloyna (66) also reported non-linearity of K_d isotherm for ¹³⁷Cs at low sediment concentrations up to 150 mg/l (Figure 3-11). The data of Garder and Skulberg (26) showed a similar type of non-linear K_d isotherm for ¹³⁷Cs at sediment concentrations ranging from 16 to 256 mg/l (Table 2-1 and Figure 3-11).

Schubert and Conn (78) pointed out that K_d varies directly with the change in M/V (sediment to water concentration) for an adsorption process of a trace component. The K_d values for ¹³⁷Cs, in general, decreased with increased M/V values for sediment concentrations reported as seen in Figures 3-9 and 3-10, and Garder's data in Figure 3-11. As the ¹³⁷Cs concentrations were increased, the K_d values increased.

The ⁸⁵Sr data are given in Figure 3-12. The K_d isotherm was not linear, and was not affected by the type of sorbent and ⁸⁵Sr concentration. Higher sediment concentrations caused a reduction in the K_d values (Tables 3-8 and 3-9). No linearity was observed by Reynolds and Gloyna for the entire sediment concentration range, 50-2,000 mg/l. This non-linearity was explained to be due to an increase in Ca⁺⁺ and Mg⁺⁺ concentration, and thereby inhibiting strontium uptake (66). The results of Garder and Skulberg indicated a similar decrease in K_d values at higher sediment concentrations (Tables 2-1 and 2-2).

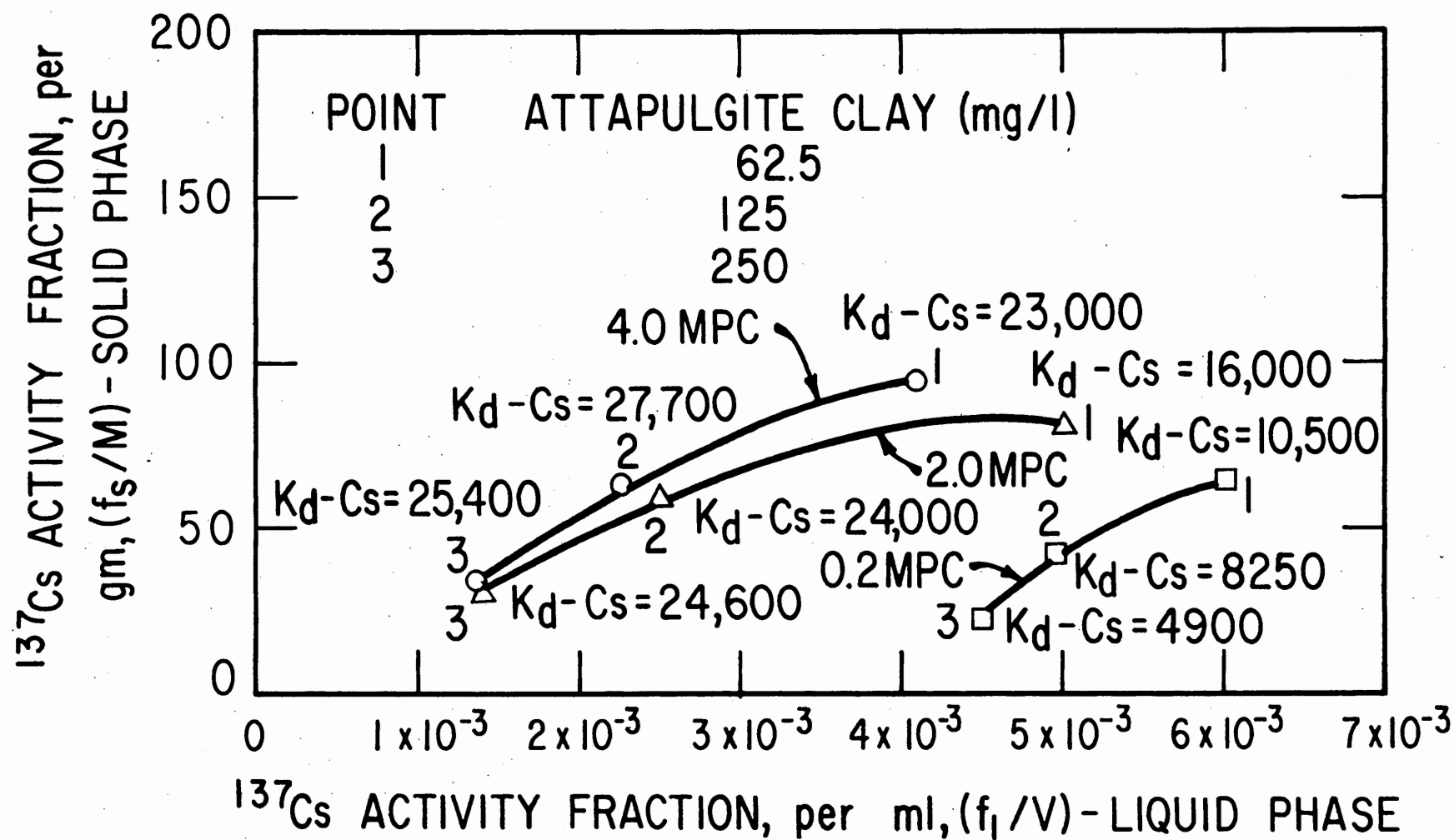


FIG. 3-9. K_d ISOTHERM FOR ^{137}Cs SORPTION

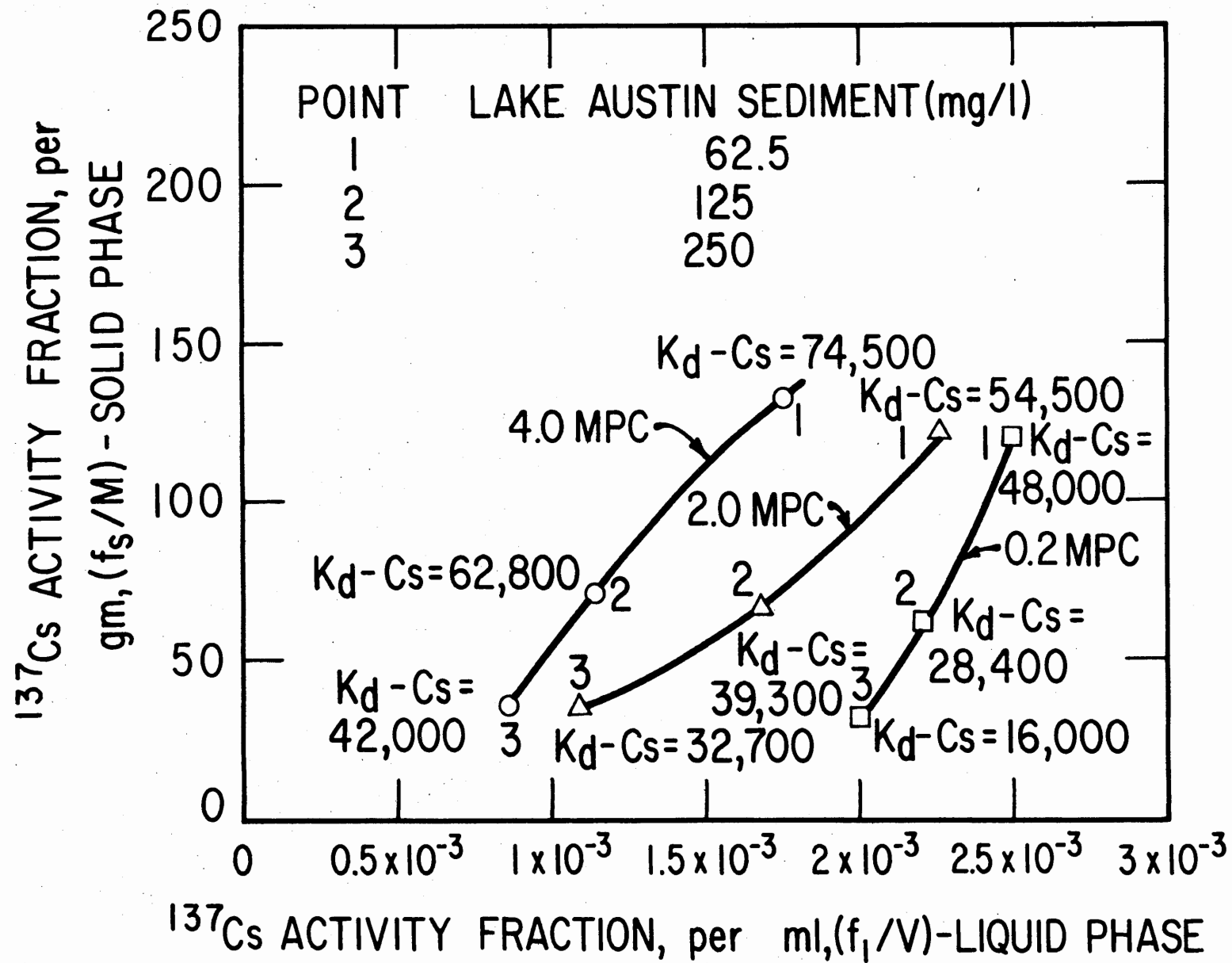
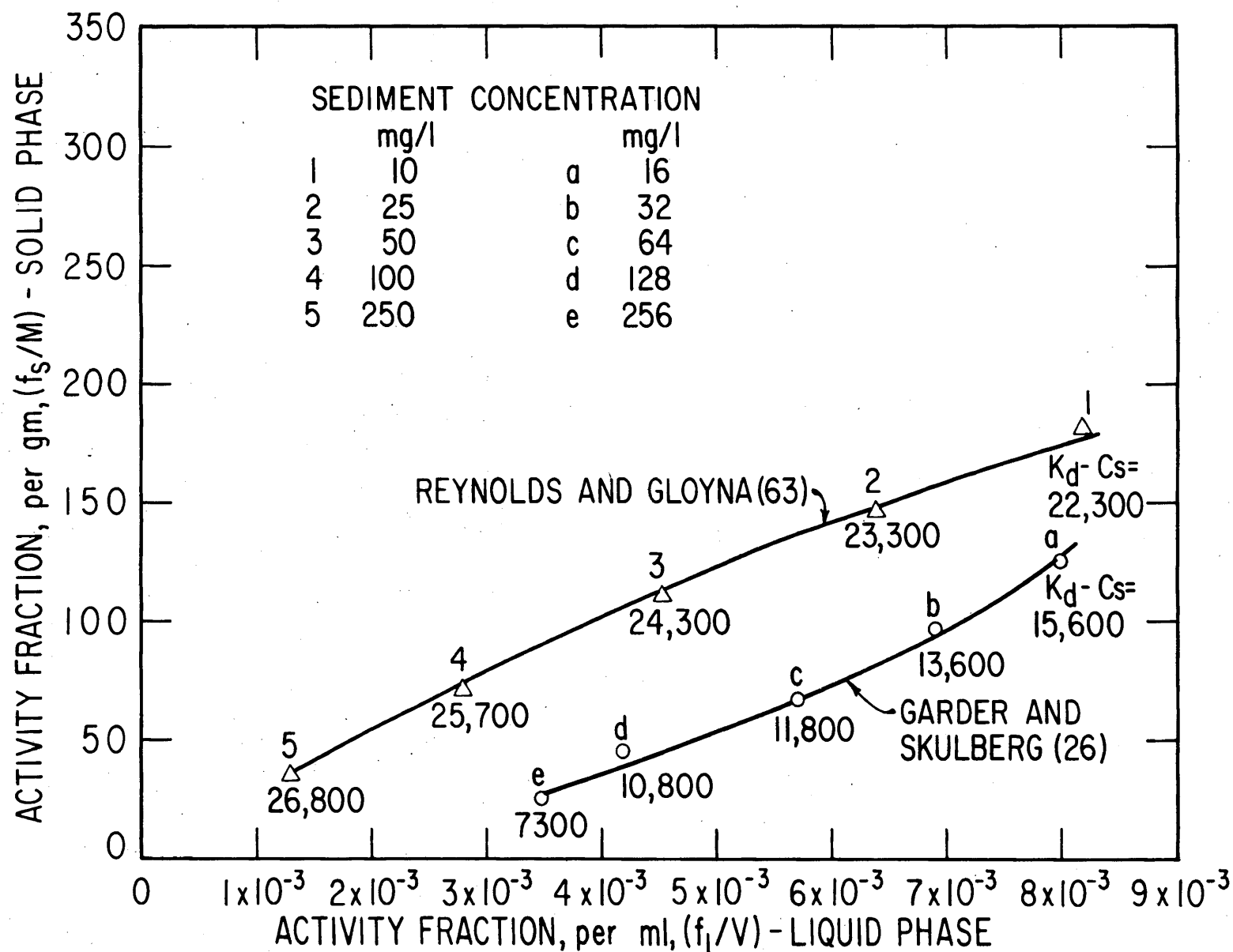


FIG. 3-10. K_d ISOTHERM FOR ^{137}Cs SORPTION

FIG. 3-II. K_d ISOTHERM FOR CESIUM UPTAKE

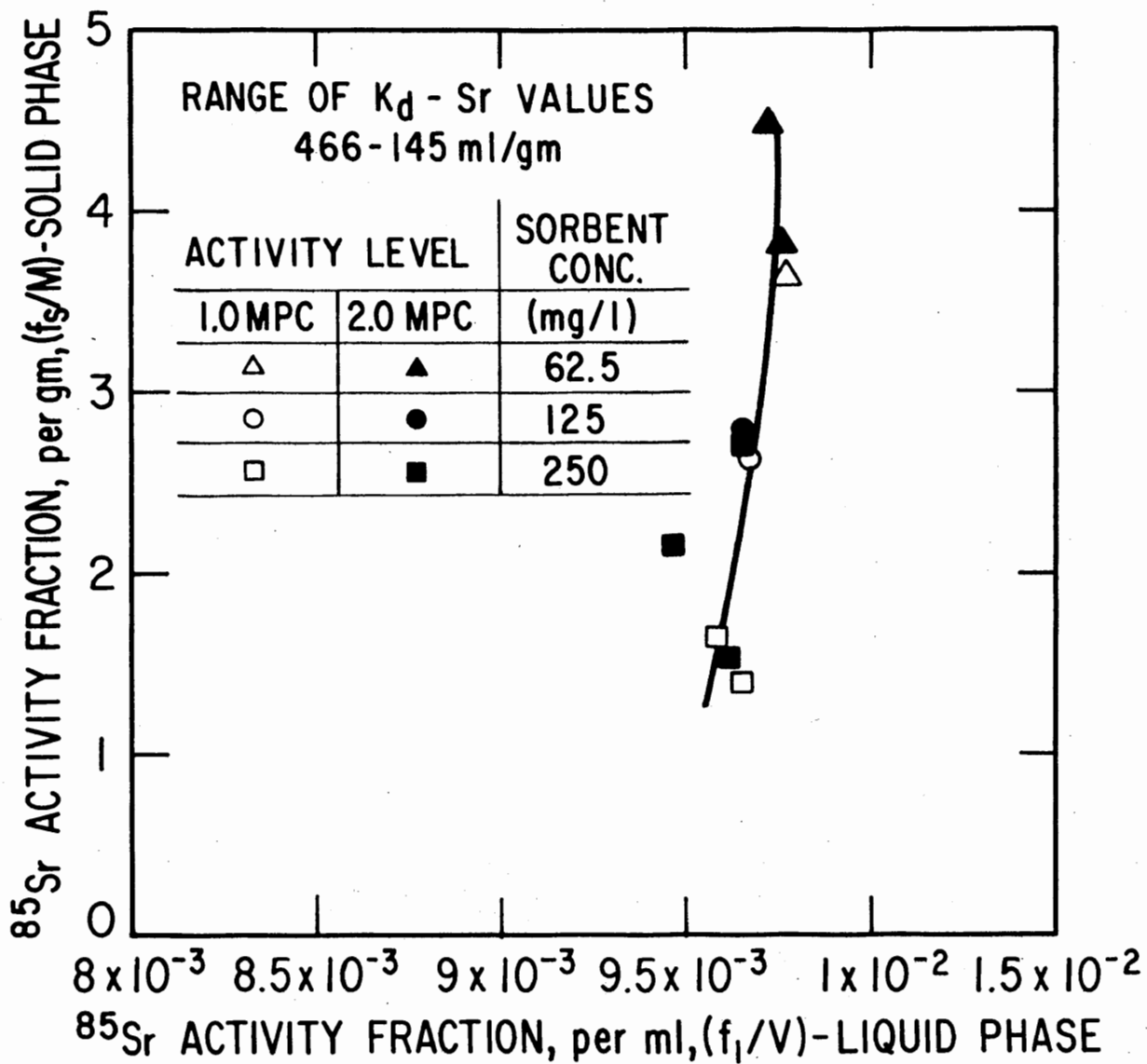


FIG. 3-12. K_d ISOTHERM FOR ^{85}Sr

CHAPTER IV

^{85}Sr IN A NON-FLOWING AQUARIA SYSTEM

This study was undertaken to examine the distribution of ^{85}Sr in ponded water, bottom sediments, and one selected rooted plant, Vallisneria. The Attapulgate clay concentration, temperature, and light intensity were all controlled.

Experimental Setup

Four aquaria were used. Each unit was 60 cm long, 26 cm wide, and 36 cm deep. To simulate natural mixing each unit was equipped with a stirrer immersed 7 cm below the surface. A layer of Lake Austin sediment was placed in each unit and Vallisneria was transplanted to the sediment by placing the plants in 4-ounce plastic containers. These containers were open at both ends and could be pushed into the sediments until only 1 cm was exposed. The depth of bottom sediments was about 8 cm, and a constant freeboard was maintained in each unit by adding distilled water periodically to compensate for evaporative losses. The temperature was maintained at $24^{\circ} \pm 0.5^{\circ}\text{C}$. Fluorescent lights provided 600 foot-candles of illumination 12 hours per day. The physical nature of aquaria and other details are presented in Table 4-1.

Table 4-1. Physical Details of Aquaria

Unit No.	Water Depth (cm)	Average Bottom Sediment Depth (cm)	Total Biomass (gm)	Initial ^{85}Sr Concentration (MPC)*	Initial Clay Suspension (mg/l)
1	21	8	6.5	0.48	0
2	22	8	3.0	0.47	50
3	22	8	0.9	0.72	0
4	22	7	2.0	0.71	50

*Maximum permissible concentration of ^{85}Sr in water for occupational exposure is 10^{-3} $\mu\text{c/ml}$ (NBS Handbook No. 69)

Sampling Techniques and Sample Preparation

The aquaria were allowed to stabilize for two weeks before ^{85}Sr was released instantaneously. After injection of ^{85}Sr , samples of water, bottom sediments, and Vallisneria were collected over a period of 32 days. Frequent sampling was undertaken during the first day.

A direct sampling procedure was adopted. Ten milliliters of water were withdrawn from each aquarium and transferred directly to aluminium planchets for drying under heat lamps and counting. Since Unit nos. 2 and 4 contained a known concentration of clay, an additional 75 ml of water were withdrawn from each unit and placed into 4-ounce plastic bottles. To determine the ^{85}Sr associated with the suspended fraction, this 75 ml aliquot was centrifuged at $1060 \times g$ for 10 minutes, and following separation 10 ml of the supernatant was placed into a planchet. Inorganic suspended solids were determined by

filtering the remaining volume through a 0.45 μ HA millipore filter, and igniting the filter in a muffle furnace at 600°C for 10 minutes.

The leaves of the Vallisneria were collected with clean steel tongs and placed in paper cups. These samples were dried under infra-red heat lamps, ground, placed into aluminium planchets, and weighed. The bottom sediment samples were obtained with a sediment sampler consisting of a lucite cylinder, an aluminium plunger and a steel sample holder (4, 80, 99). After collection, each sediment core was frozen. Then the top three inches of the frozen core was removed, dried at 110°C for about 24 hours, pulverized, transferred to an aluminium planchet, stabilized with lucite-in-acetone solution, and counted for gamma activity.

To evaluate the migration of ^{85}Sr , the frozen sediment cores were cut into five slices at depths of 1/4, 1/2, 1, 2, and 3 inches from the water-sediment interface. Each slice was dried, ground, and weighed.

^{85}Sr in Water

The concentration of ^{85}Sr in water decreased with increasing time after release, Figure 4-1. The same pattern of variation has been reported elsewhere (80). The ^{85}Sr ultimately became associated mostly with the bottom sediments. Very little ^{85}Sr was taken up by Vallisneria. The specific activity of ^{85}Sr in water, dissolved and suspended fraction, (aquaria nos. 2 and 4 containing clay suspensions) was higher than that in the control units (aquaria nos. 1 and 3).

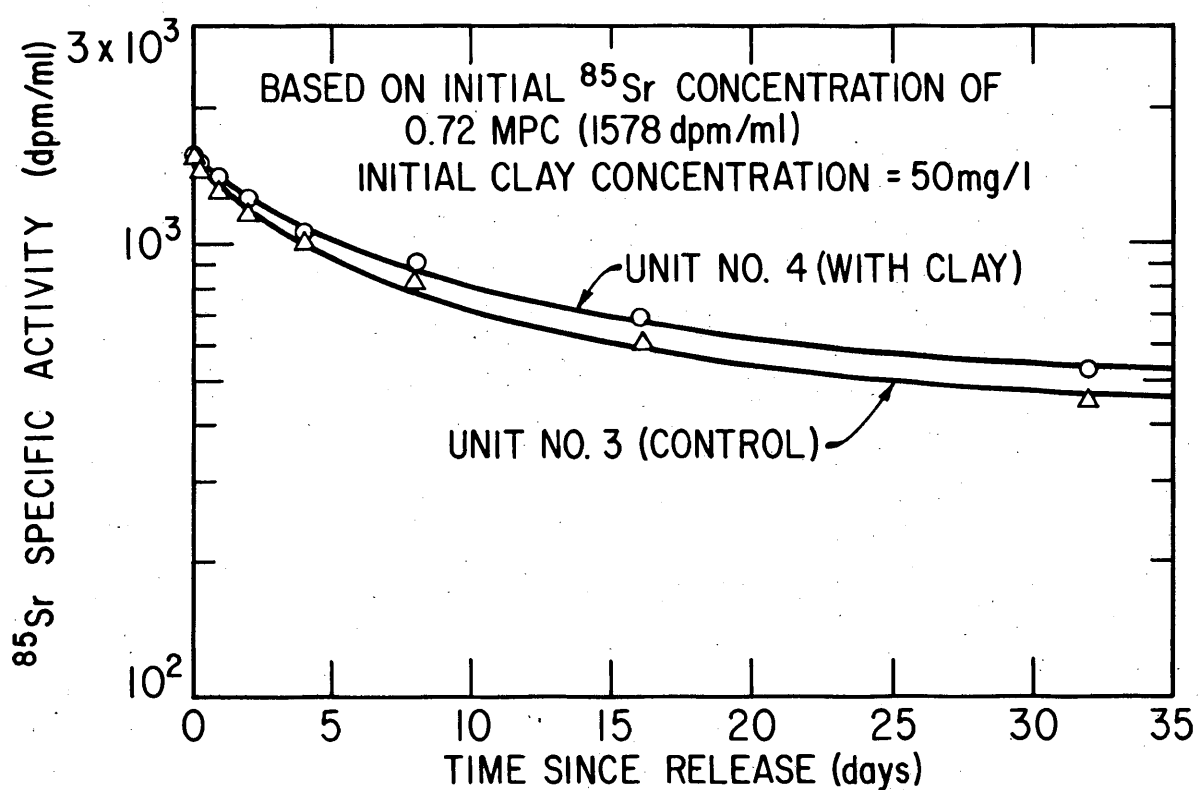
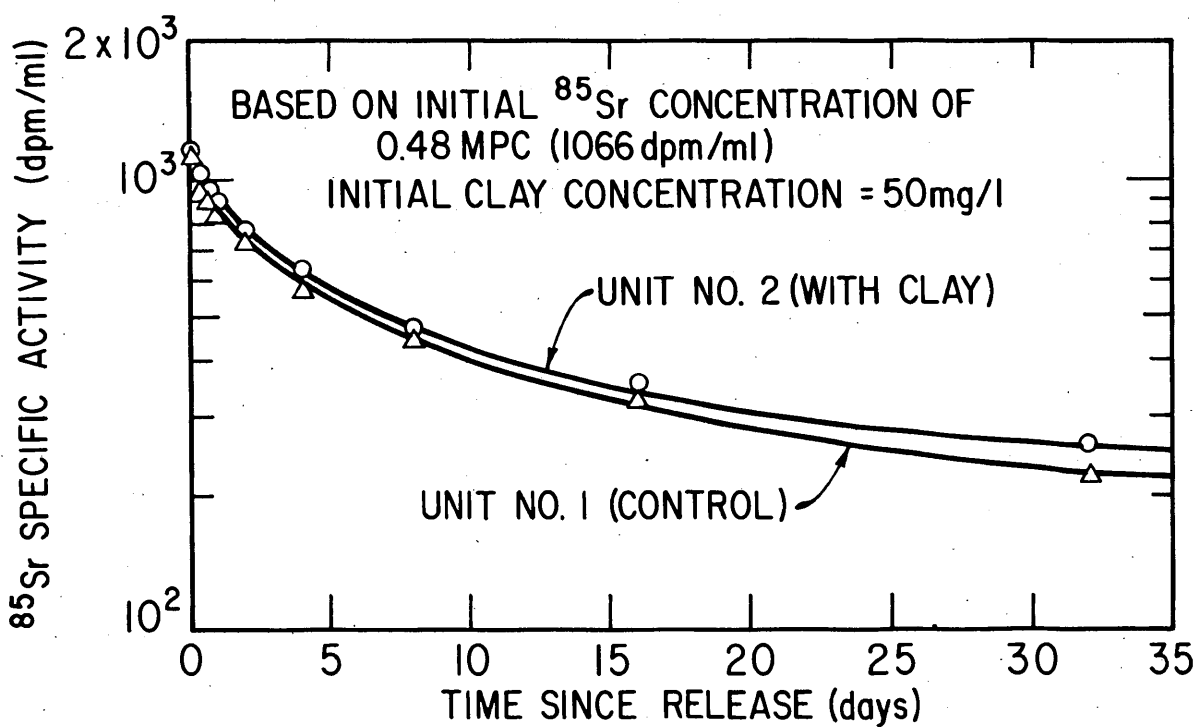


FIG. 4-1. ^{85}Sr IN WATER OF NON-FLOWING ECOSYSTEM

The clay concentration in the units 2 and 4 dropped gradually to zero after 2 days due to sedimentation.

The total uptake by bottom sediments and Vallisneria is shown in Table 4-2. As observed in the earlier study (80), the rate of decrease of ^{85}Sr in water approached quasi-equilibrium condition by the end of the experiment.

^{85}Sr in Bottom Sediments

The ^{85}Sr associated with bottom sediments increased with time as shown in Figure 4-2.

Table 4-2. Total Decrease of ^{85}Sr in Water Phase of Aquaria

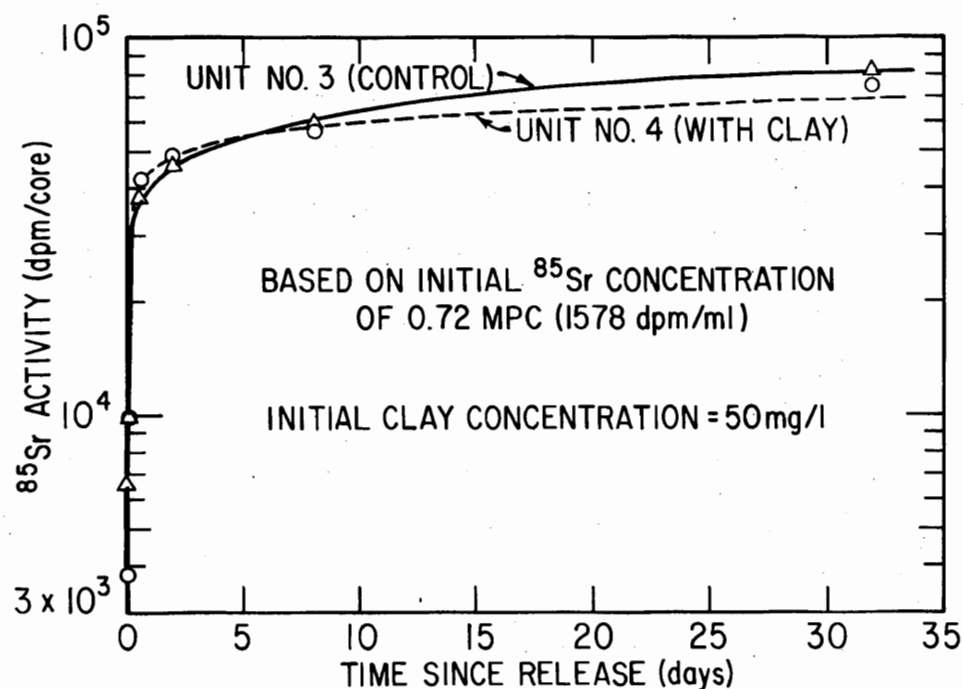
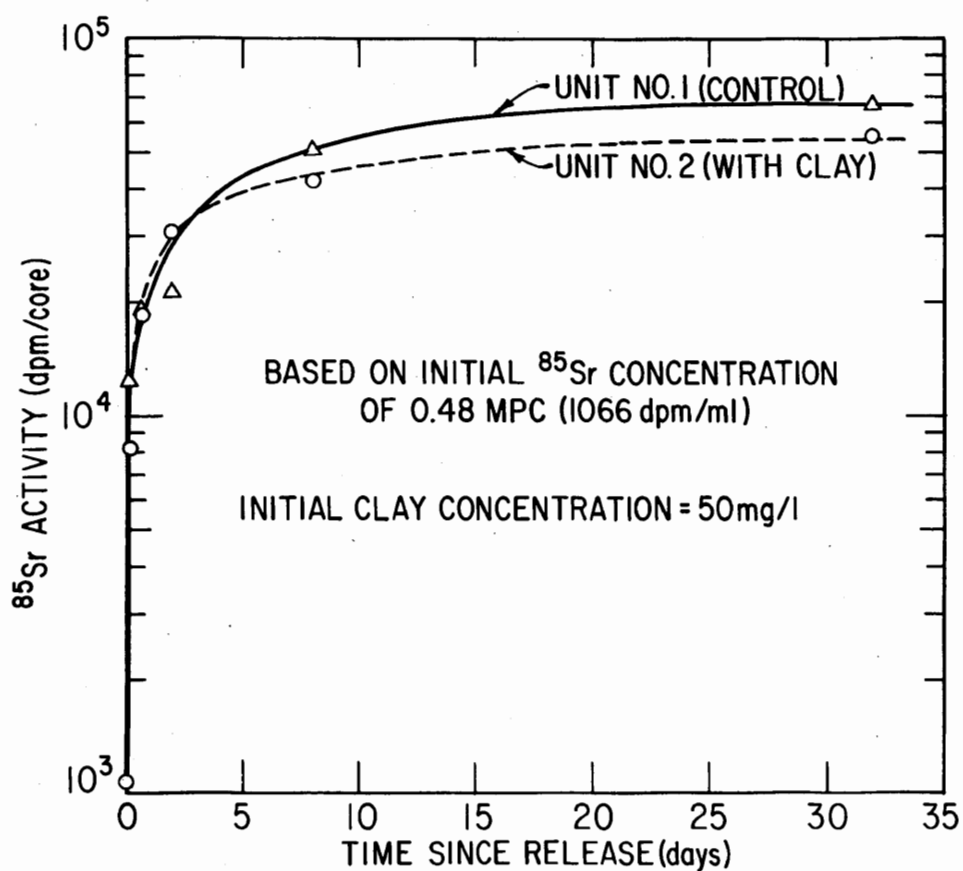
Unit No.	Total Decrease of ^{85}Sr (%)
1	77*
2	80*
3	70**
4	72**

* Based on initial ^{85}Sr concentration of 0.48 MPC

** Based on initial ^{85}Sr concentration of 0.72 MPC

^{85}Sr Migration into the Sediments

The migration of radionuclides into the sediments through interstitial water has been reported by others (4, 7, 67, 80). This migration was reported to be a first-order reaction, Shih and Gloyna (81). The derivation of Equation 4-1 is reported elsewhere (80).

FIG. 4-2. ^{85}Sr IN BOTTOM SEDIMENTS

$$N(d,t) = N_0 \exp \{-m(t)d\} \quad (4-1)$$

where $N(d,t)$ = total number of radionuclides per unit area at depth, d , and contact time, t
 N_0 = total number of radionuclides per unit area at sediment-water interface
 $m(t)$ = probability of retention of radionuclide in the sediment per unit depth at time t

To facilitate analysis, the depth, d , may be expressed in terms of sediment weight, w , as shown in Figure 4-3. The average dry weight of a sediment core of one inch depth having a cross-section of 0.6 sq. in. was found to be 11.0 gm. By rewriting Equation 4-1 it is possible to calculate the specific activity of the sediment (dpm/gm), Equation 4-2.

$$S(w,t) = S_0 \exp \{-m'(t)w\} \quad (4-2)$$

where $S(w,t)$ = specific activity of dried sediment at depth with cumulative weight, w , from the interface, dpm/gm
 S_0 = interface specific activity of sediment, dpm/gm
 $m'(t)$ = penetration constant per gm weight corresponding to 0.09 in. depth, depending on contact time, t , gm^{-1}

The migration of ^{85}Sr into the sediments is shown in Figure 4-4, and the migration or penetration constant, m' , for the units are given in Table 4-3. The m' values were found to be practically the same for the units with and without Attapulgitic clay suspensions. The ^{85}Sr concentration in the water did not appear to have affected the migration constants. Shih and Gloyna (80) reported no variation in the m' values for different ^{85}Sr concentration in water.

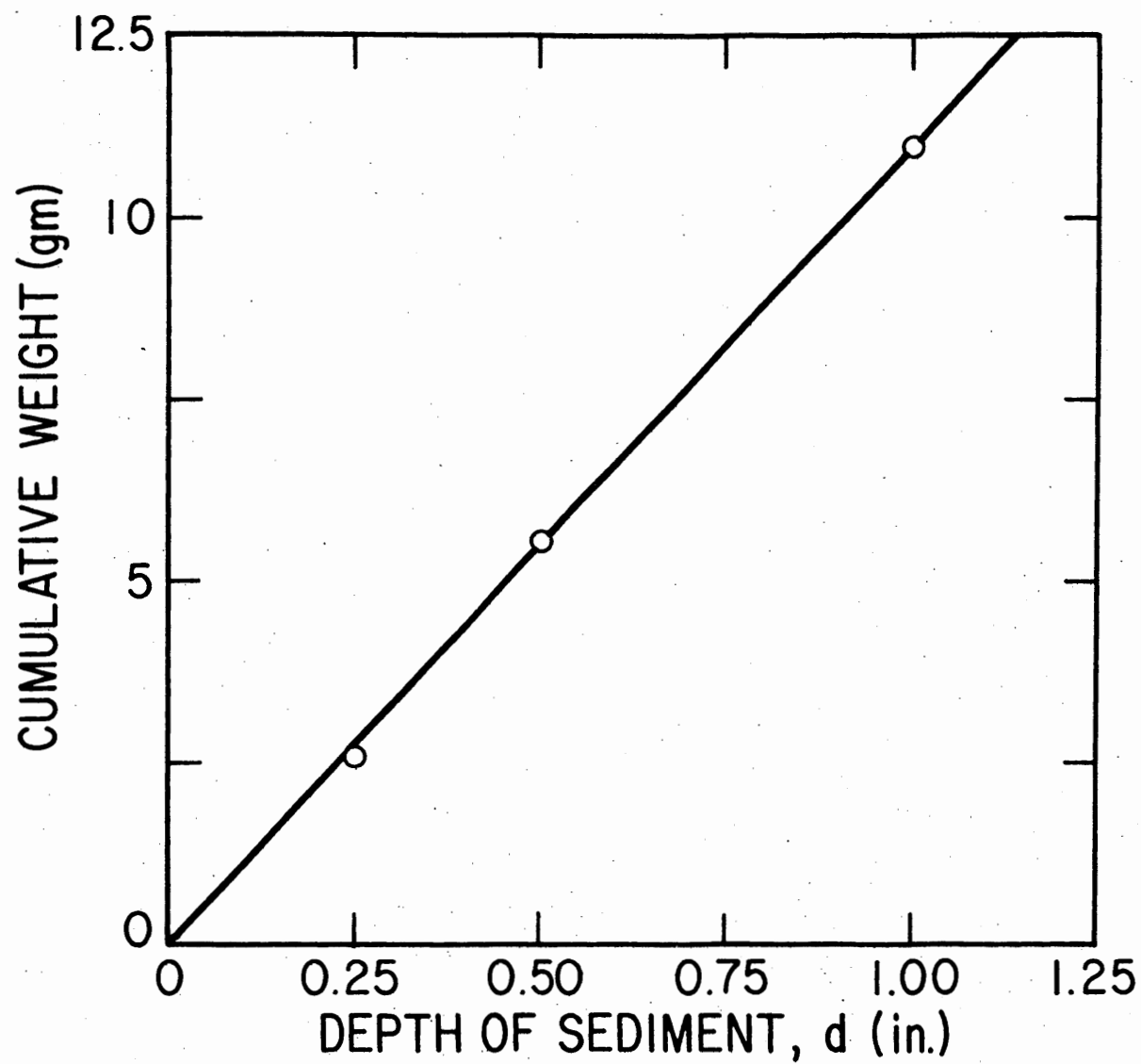


FIG. 4-3. RELATIONSHIP BETWEEN SEDIMENT DEPTH AND ITS WEIGHT

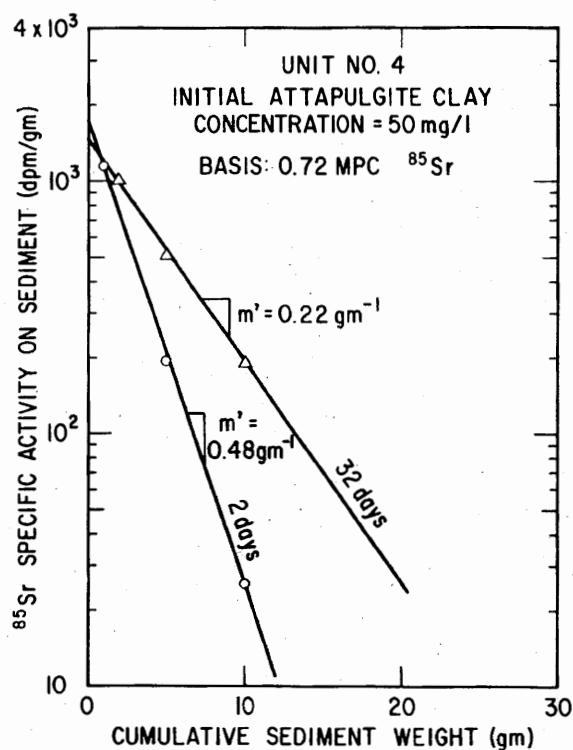
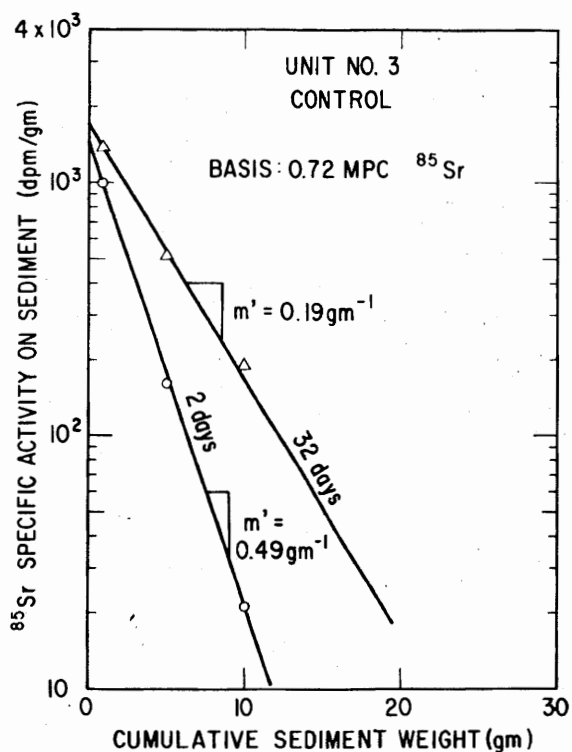
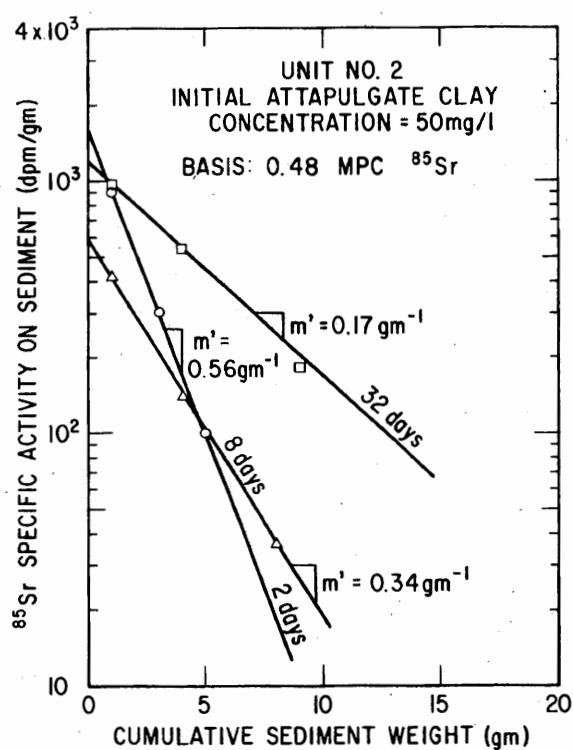
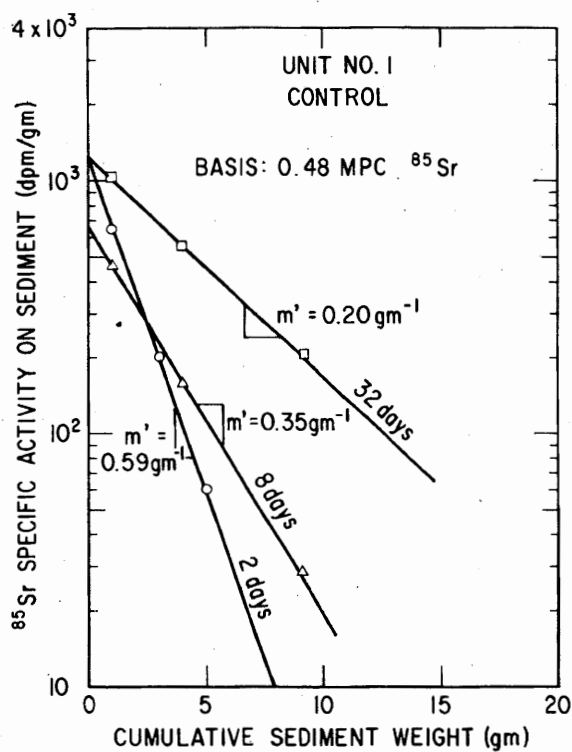


FIG. 4-4. MIGRATION OF ^{85}Sr INTO SEDIMENTS

^{85}Sr in Vallisneria

The uptake of ^{85}Sr by Vallisneria was found to be relatively fast increasing for a few hours after the release of ^{85}Sr . The plants released the activity after about eight hours, Figure 4-5. The same type of uptake and release pattern was observed in both the control

Table 4-3. ^{85}Sr Migration Constants (gm^{-1}) For
Bottom Sediments in Contained Systems

Unit No.	^{85}Sr Concentration (MPC)	Contact Time (days)			Remarks
		2	8	32	
1	0.48	0.59	0.35	0.20	control
2	0.47	0.56	0.34	0.17	with clay*
3	0.72	0.49	--	0.19	control
4	0.71	0.48	--	0.22	with clay*

*Initial clay concentration of 50 mg/l

units, and in the units containing clay suspensions; however, the ^{85}Sr concentration in plants was higher in the units containing the clay. After 8 days, there was no difference in the gamma activity of plants between the control and other units. It appears that the role of Vallisneria in the uptake of ^{85}Sr is not significant.

Distribution of ^{85}Sr

The uptake of ^{85}Sr by bottom sediments and Vallisneria was due to both physical and chemical sorption; however, the influence of plants was negligible. The ratio of ^{85}Sr in the sediment core to that

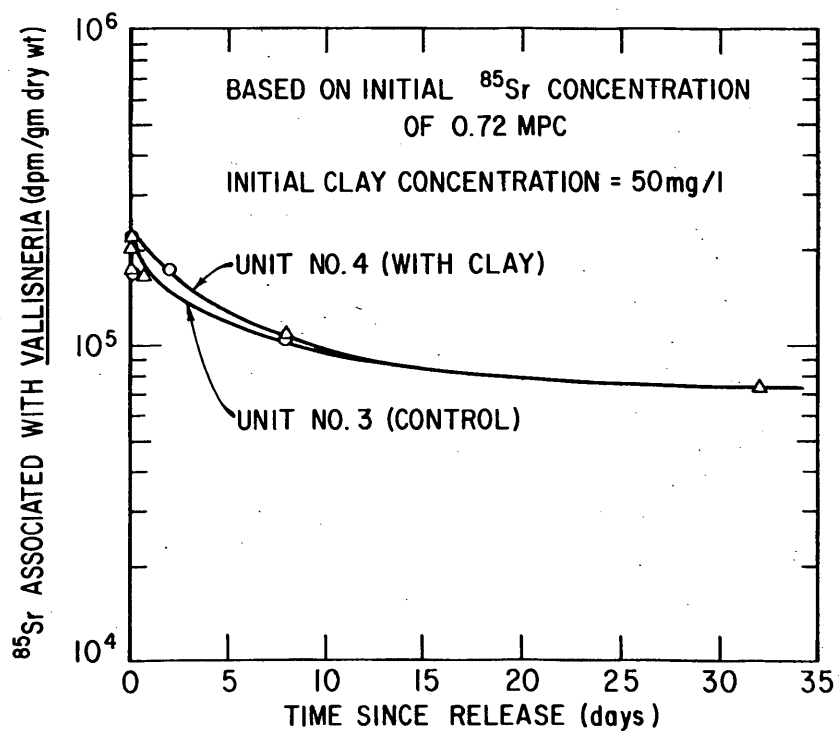
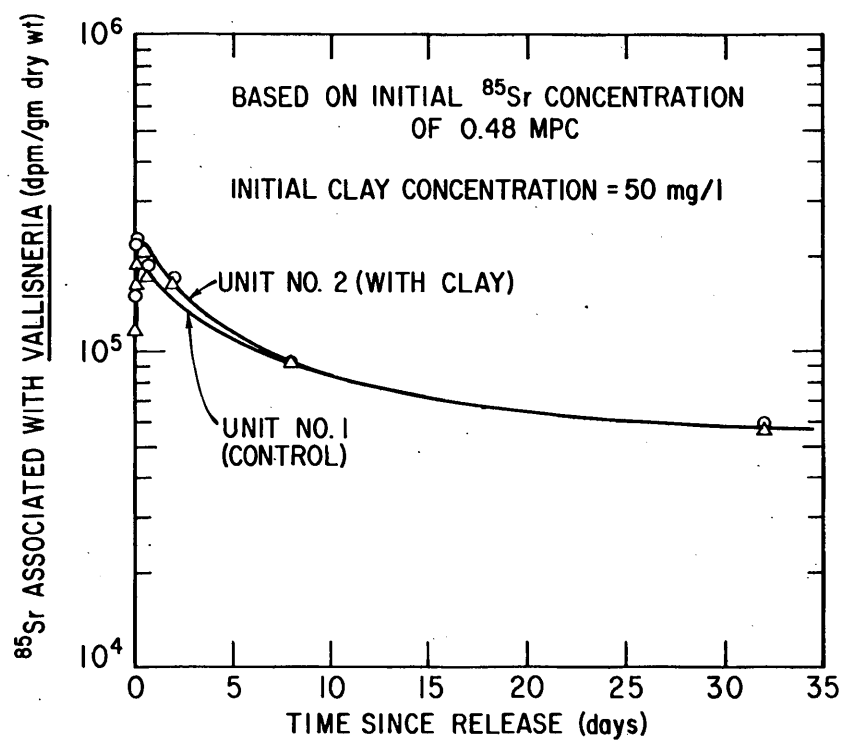


FIG. 4-5. VARIATION OF ^{85}Sr IN VALLISNERIA

in water is given as C_s and is the slope of the line in Figure 4-6.

Table 4-4 shows that the concentration factors for sediments and plants did not increase in the presence of clay suspensions.

Table 4-4. Concentration Factors For ^{85}Sr in Sediments and Plants

Type of Aquaria	Contact Time (days)	C_s of Bottom Sedi- ments, $\frac{\text{dpm per core}}{\text{dpm per ml}}$	C_p of <u>Vallisneria</u> , $\frac{\text{dpm per gm}}{\text{dpm per ml}}$
Control	2	38	200
	8	77	162
	30	156	155
Presence of Clay*	2	40	240
	8	79	192
	30	138	155

* Initial clay concentration of 50 mg/l

The functional relationships between the concentration factors, C_s and C_p , and contact time, t , may be expressed as:

$$C_s = 29 t^{0.475} \quad (4-3)$$

and
$$C_p = 235 t^{-0.128} \quad (4-4)$$

where t = contact time, days. Notably, the sediment concentration factor, C_s , increases while the value of C_p decreases with time. Such relationships were also reported previously (80).

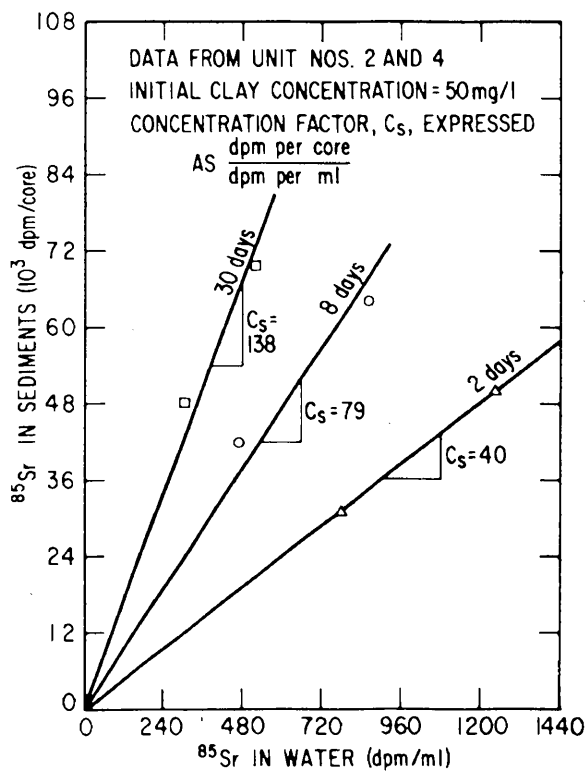
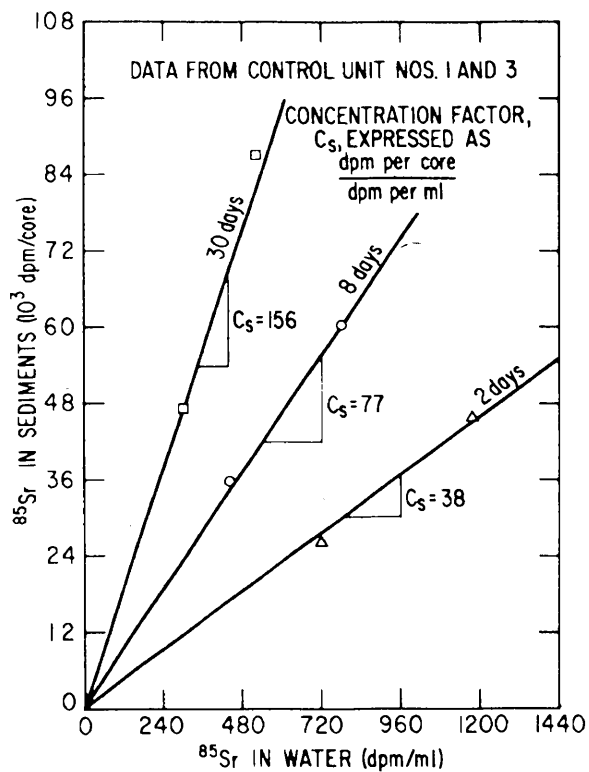


FIG. 4-6. ^{85}Sr UPTAKE BY PONDED BOTTOM SEDIMENTS

CHAPTER V

MODEL RIVER STUDIES

This chapter deals with the model river studies. Three major experiments are described: one steady-state release of ^{137}Cs and Attapulgate clay; and two instantaneous releases, (a) ^{85}Sr and ^{137}Cs with Attapulgate clay, and (b) ^{137}Cs with Kaolinite clay. The buildup and release of radionuclides on the sediments and a rooted plant under turbid stream conditions are included.

Description of Model River

The two unsteady release experiments were conducted in a model river consisting of a built-up rectangular, metal flume with dimensions, 200 ft. long, 2.5 ft. wide, and 2.0 ft. deep. The slope of the flume was adjustable from 0 to 0.006 ft./ft. by means of screw jacks, but for these experiments the slope was kept constant at 0.00021 ft./ft. The details of this flume have been reported in the previous studies (99). The water depth was regulated at 15 inches by an adjustable weir located at the outlet. A flow of 100 liters/min. was maintained during the two instant release tests. Under these conditions, the average velocity was 1.1 ft./min. Lake Austin sediment was placed in the flume as bottom sediments to an average depth of 6 inches. Macro rooted-aquatic plants, Myriophyllum, Potamogeton, and Vallisneria, were transplanted along the flume to simulate a dynamic ecosystem.

The continuous release study was conducted in a dual-channel system. Each channel was 200 ft. long, 1.25 ft. wide, and 2.0 ft. deep. These channels will be referred to as the east channel and west channel. The inflow of water to the flume was controlled through a system consisting of a constant head tank, gate valves, stilling basins, perforated baffles, two triangular V-notch weirs and manometers as shown in Figures 5-1 and 5-2. The rate of flow was calculated by:

$$Q = 0.181 M^{2.6} \quad (5-1)$$

and
$$Q = 0.440 H^{2.3} \quad (5-2)$$

where Q = flow rate, liters/min.
 M = manometer reading, cm
 H = head above the weir crest, cm.

A flow of 50 liters/min. was maintained in each channel with an average velocity of 1.7 ft./min. The corresponding water depth was 10 inches controlled by outlet weirs.

East and west channels contained approximately 6.5 inches of bottom sediments. The rooted plant, Vallisneria, was planted in each channel by placing the plants in open cylindrical containers. The open cylinders were pushed into the sediment so that only 2 cm were exposed.

Ground water, Table 3-3, was used in these tests, but it was stored in a one-half million gallon supply reservoir prior to use, providing an opportunity for the establishment of a typical plant and animal population.



FIG. 5-1. DUAL-CHANNEL MODEL RIVER

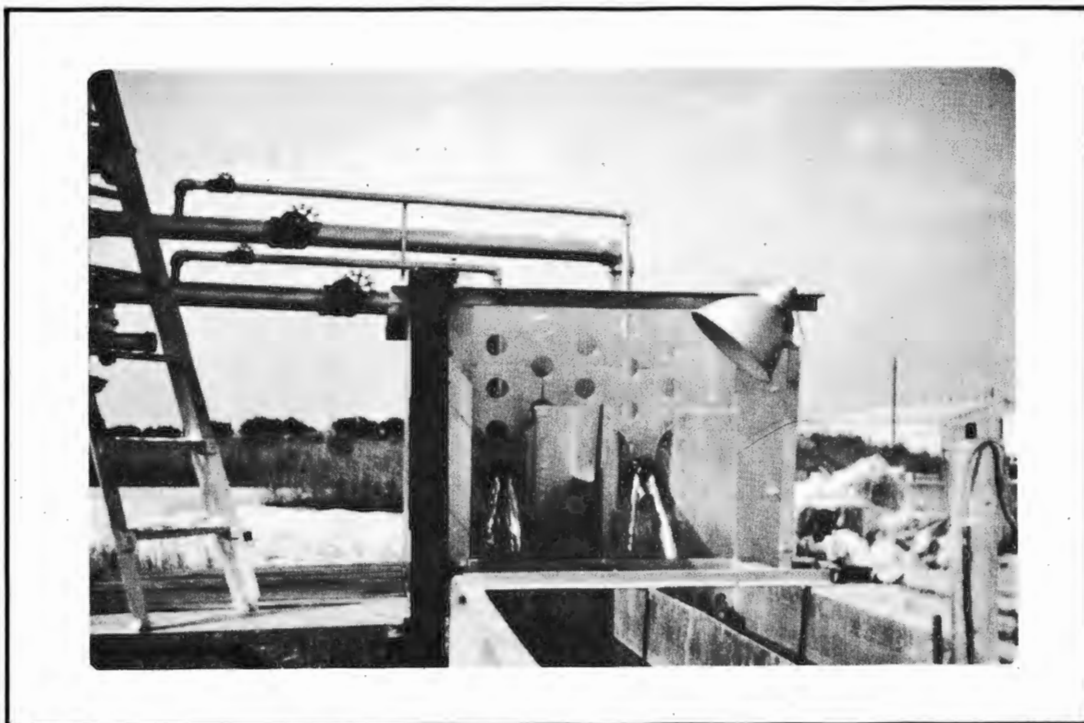


FIG. 5-2. INLET SYSTEM

Another reservoir of equal capacity served as a monitoring tank for the effluent.

Instantaneous Release Experiments

The instantaneous release studies involved two major experiments. Both of these were conducted in the uncompartmented flume. In the first experiment, 2 mc of $^{137}\text{CsCl}$ was injected rapidly in the model river at mid-water depth and 10 ft. from the inlet. Three hours prior to the release of radioactivity, a slurry of Georgia Kaolinite (Table 2-3) was injected at a rate of 8 pounds (dry wt.) per hour. This injection rate was continued for 12 hours. The influent suspension was mixed vigorously in a 40-gallon tank prior to injection.

Water and sediment samples were collected from sampling stations at 10, 40, 90, 140, and 185 ft. from release point. Plant samples were obtained from a 20 ft. long section, beginning at the 155 ft. marker.

For the second experiment 2 mc of $^{85}\text{SrCl}_2$ and 2 mc of $^{137}\text{CsCl}$ were released in a short burst. Due to the large number of samples produced in the previous experiment, this test was carried out using the second 100-ft. section of the model river. Attapulgate slurry was introduced continuously at the 100-ft. point to give a clay concentration of 100 mg/l at the release point. An electric stirrer was located in the flume at the release point. This experiment was conducted over a period of 14 days. A total of four sampling stations, one upstream and three downstream from the release point, were

established. Water, sediment, and Vallisneria samples were taken from selected stations during the experiment.

The sampling locations, type of samples collected and other details for both experiments are summarized in Table 5-1.

Sampling: Water samples were collected in 4-ounce polythene bottles with the aid of a vacuum system (80, 99). These samples were refrigerated, later 10 ml. aliquots were transferred to aluminium planchets and counted. The radioactivity associated with the suspended clay in water was determined by difference measurements, ie. counts of liquid before and after centrifugation at $1060 \times G$ for 10 minutes. The suspended clay concentration in water was determined by filtering a known volume of sample through millipore filters having pore openings of 0.45 microns.

Sediment samples were obtained and processed in the same manner as described in Chapter IV.

Plant samples were collected in 4-ounce glass bottles, and dried at 103°C . A one-half gram sample of the dried plant material was used for radionuclide assay.

Analysis: Usually four gamma emitters were released during each experiment. For this purpose a 512-channel Gamma spectrometer was used in conjunction with a 4-inch diameter thallium-activated NaI crystal and shielded chamber. The counting geometry was uniform, and an energy span of 2.0 Mev was selected for the 256-channel group.

Table 5-1. Instantaneous Release Experiments

Particulars	Experiment I	Experiment II
Duration of Experiment	19 days	14 days
Depth of Water	15 in.	15 in.
Flow Rate	100 liters/min.	100 liters/min.
Type of Clay Load	Kaolinite	Attapulgate
Radionuclide Added:		
⁸⁵ Sr	2.0 mc	2.0 mc.
¹³⁷ Cs	2.0 mc	2.0 mc
Sampling Stations:		
(ft. from release point)		
1	10 (W, S)	- 10 (W, S)
2	40 (W, S)	5 (W,S,P)
3	90 (W, S)	50 (W)
4	140 (W, S)	95 (W,S,P)
5	185 (W, S)	

W = water samples; S = sediment samples; P = plant samples

Note: (a) water samples in Expt. I collected at 2, 7, and 13 in. below water surface

(b) water samples in Expt. II collected at 5 and 10 in. below water surface

The output data were displayed on punched paper tapes, transferred to magnetic tapes, and translated into disintegrations per minute per sample content by the computer. Corrections for background, counting efficiency, contribution from other gamma emitters, self-absorption, and decay were incorporated in the computer programs (80). The counting efficiency and contribution coefficient were determined by counting known radionuclide standards. The counting efficiencies and contribution coefficients are given in Table 5-2.

Transport: The two instantaneous release tests were conducted in the undivided, single channel model river. ¹³⁷Cs was released with Kaolinite while ⁸⁵Sr and ¹³⁷Cs were released with Attapulgate.

Table 5-2. Coefficients For Radioactivity Analysis

Isotope	Influenced Isotopes				Counting Efficiency (%)
	^{85}Sr	^{137}Cs	^{58}Co	^{65}Zn	
^{85}Sr	1.0000	0.0	0.0	0.0	19.9
^{137}Cs	0.0755	1.0000	0.0	0.0	13.5
^{58}Co	0.4079	0.0839	1.0000	0.0	13.2
^{65}Zn	0.2456	0.1817	0.1954	1.000	4.7

The transport of ^{137}Cs was greatly reduced by the presence of the clay suspensions. However, this was not true for ^{85}Sr . The ^{137}Cs that was sorbed onto the suspended clays which settled to the bottom was mainly responsible for the high retention of ^{137}Cs in the model river. The transport of ^{137}Cs is indicated in Table 5-3. A slightly greater sorption of ^{137}Cs was noted in the Attapulgitite tests. The total percentage retained with Attapulgitite was 30%, and with Kaolinite it was 24%.

Water Phase: The time-concentration curves for ^{137}Cs show a distribution pattern which exhibits long die-away tails, Figures 5-3 and 5-4. Such dispersion patterns have been observed in natural streams (97).

The observed concentrations of the suspended Kaolinite and Attapulgitite in the flume are shown in Figure 5-5. The average influent and outlet Kaolinite concentration was 390 mg/l and 95 mg/l respectively. Thus, 76% of the added Kaolinite was either deposited on

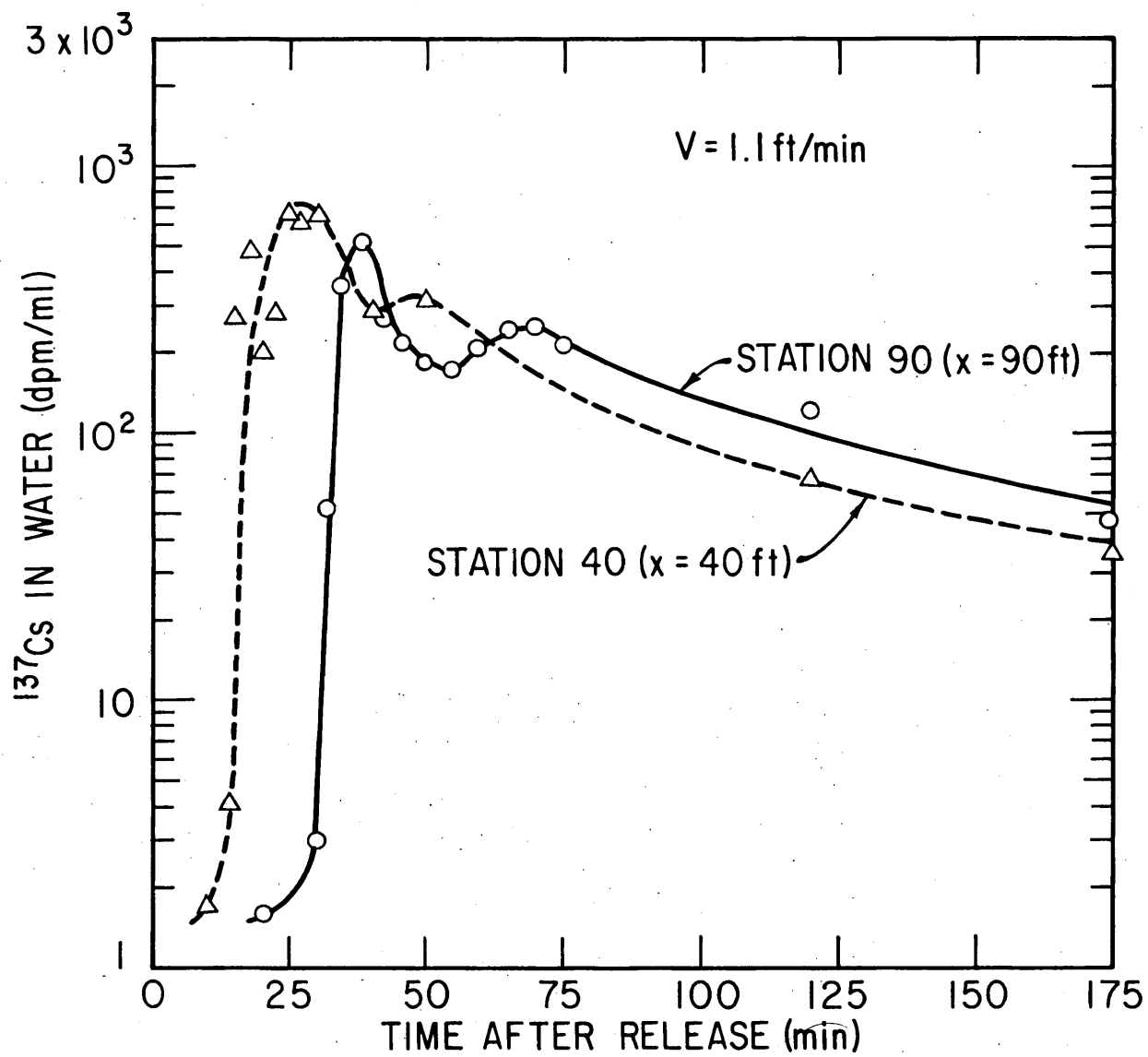


FIG.5-3. ^{137}Cs CONCENTRATION IN KAOLINITE SUSPENSIONS

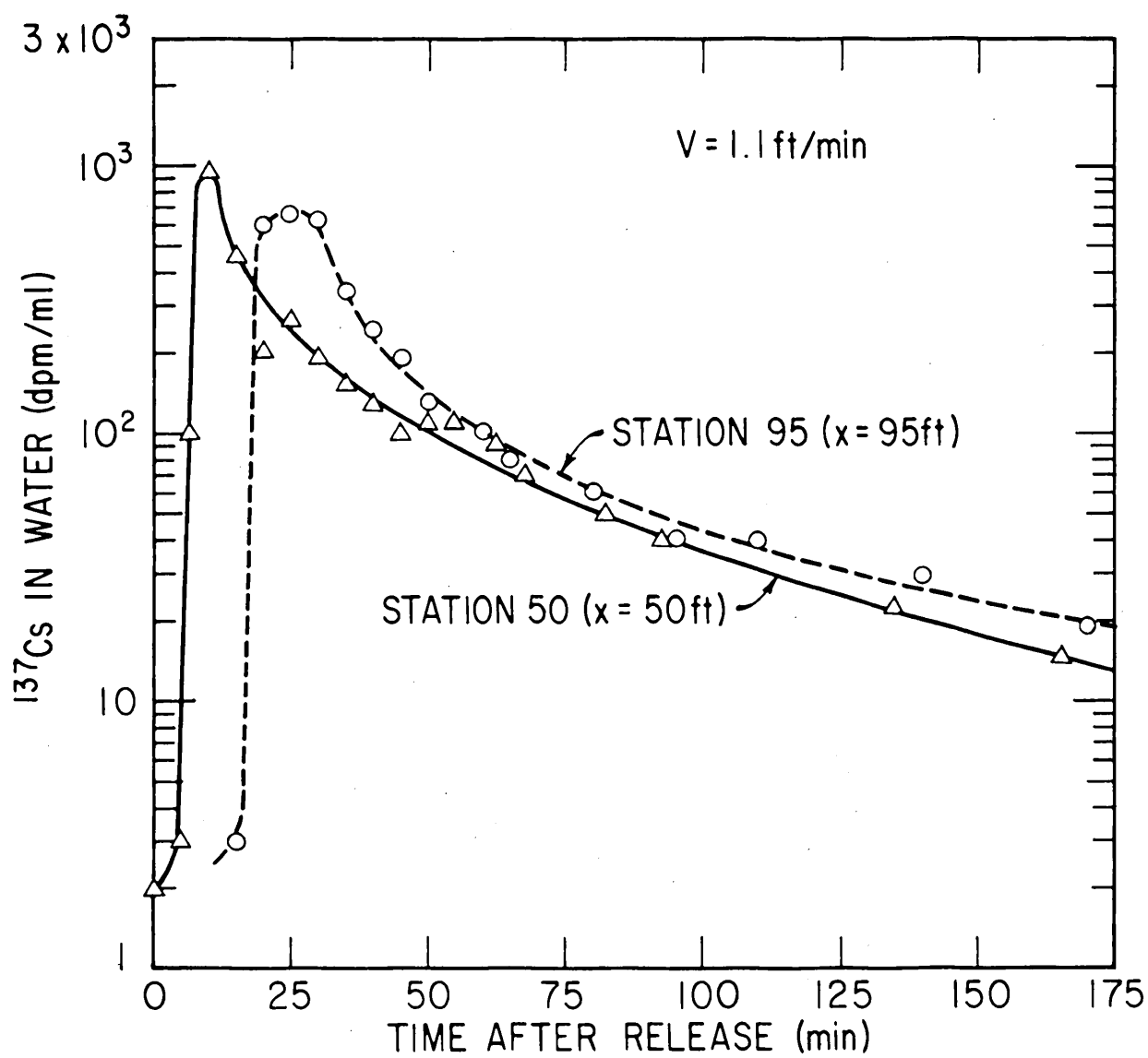


FIG.5-4. ^{137}Cs CONCENTRATION IN ATTAPULGITE SUSPENSIONS

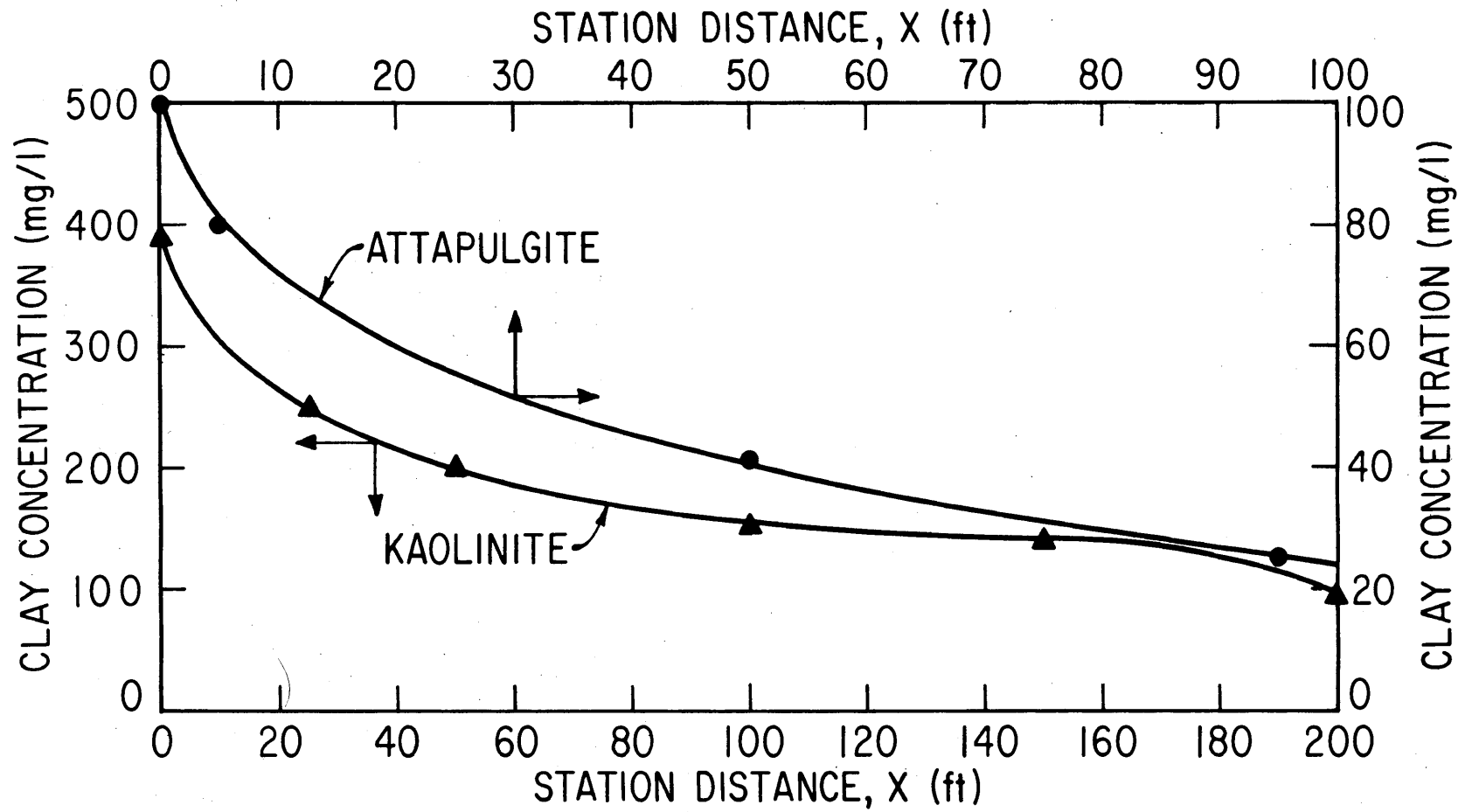


FIG. 5-5. CLAY CONCENTRATION ALONG THE MODEL RIVER

surfaces, or settled to the bottom. In the case of Attapulgite clay, the average influent concentration was 100 mg/l and at the outlet only 25 mg/l was detected.

Table 5-3. Transport Under Induced Suspended Clay Load#

Component	Percentage of Total Radionuclide Added		
	*Attapulgite Load		**Kaolinite Load
	^{85}Sr	^{137}Cs	^{137}Cs
Total Water	95	70	76
Bottom Sediments	2	20	16
Plants	3	2	2
Flume Walls	--	8	6

Instantaneous release of radionuclides

* Attapulgite clay load was released continuously and 75 percent of clay settled to the bottom

** Kaolinite clay was released for 12 hours and 75 percent of clay settled to the bottom

The amount of the ^{137}Cs carried in water is indicated in Tables 5-4 and 5-5. The percent ^{137}Cs passing each station was determined by measuring the area under the time-concentration curves. The fraction of initial amount passing each station decreased with distance. The uptake of ^{137}Cs by Attapulgite was higher than that by Kaolinite. It was found that 67% of ^{137}Cs in the water was associated with the suspended Attapulgite clay as compared to 25% for Kaolinite.

Table 5-4. Transport of ^{137}Cs in Model River Water
(Kaolinite Load)

Distance From Release Point (ft.)	Percentage of Initial ^{137}Cs Passing Each Station (%)
10	96
40	90
90	84
140	80
185	76

Table 5-5. Transport of ^{85}Sr and ^{137}Cs in Flume Water
(Attapulgitite Load)

Distance From Release Point (ft.)	Percentage of Initial Amount Passing Each Station (%)	
	^{85}Sr	^{137}Cs
5	99	95
50	96	80
95	95	70

The relationship between the specific activity of ^{137}Cs on suspended clay and in water is shown in Figure 5-6 and may be expressed by Equation 5-3:

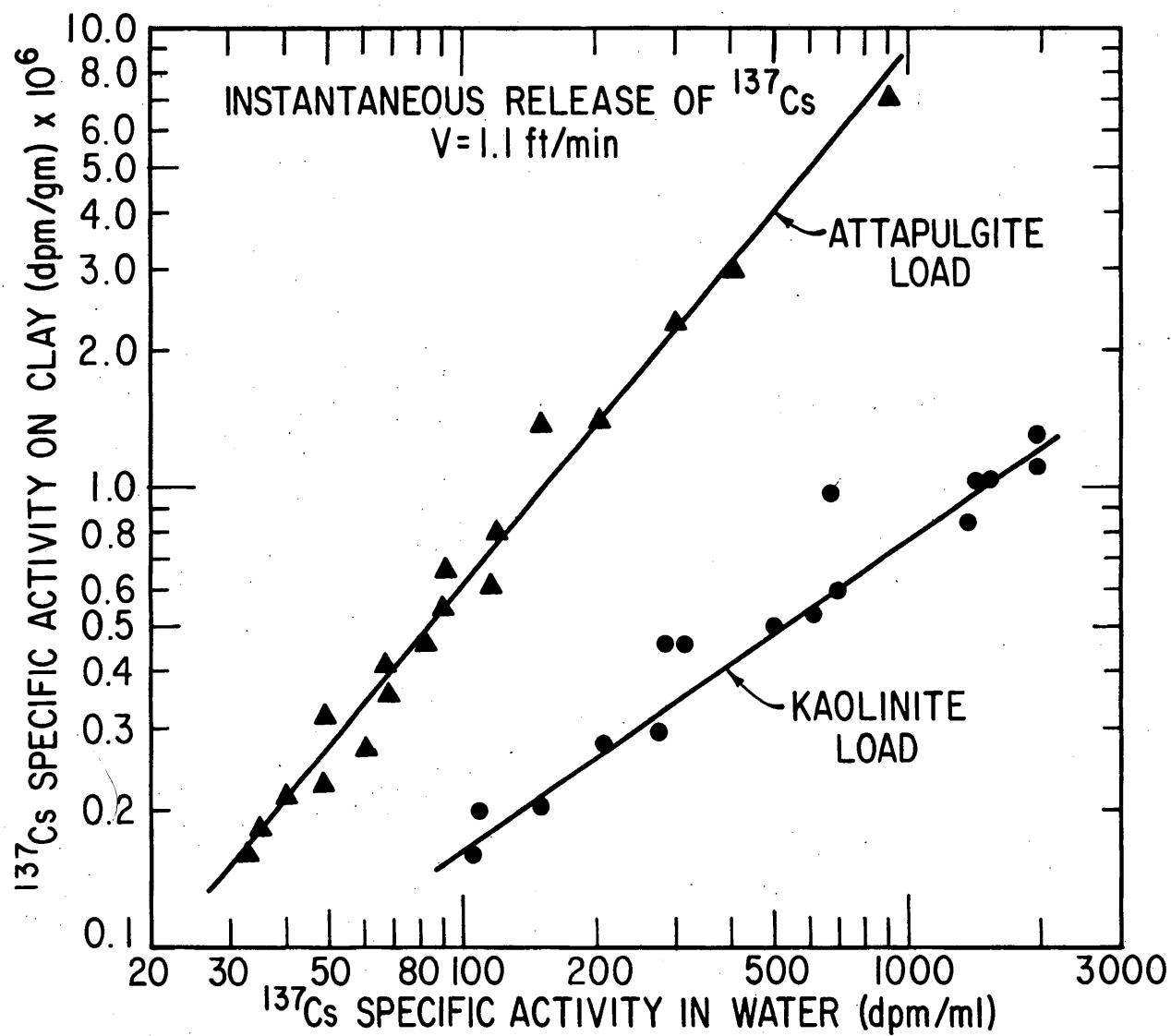
$$S_y = b (S_w)^c \quad (5-3)$$

where S_y = specific activity of radionuclide on suspended clay, dpm/gm
 S_w = specific activity of radionuclide in water, dpm/ml
 b, c = constants depending on the radionuclide and the type of clay suspension.

The decrease of ^{85}Sr in the water (dissolved and suspended fraction) was only 5% in the Attapulgate test, even though 75% of the clay settled in the model river. It was found that 6% of ^{85}Sr in the water was on suspended clay.

The relationship between the specific activity of ^{85}Sr on suspended clay and in water did not follow the type of reaction as described by Equation 5-3 (Figure 5-7). This may be due to different sorption mechanism involved other than physical sorption.

Information concerning the peak ^{85}Sr and ^{137}Cs concentrations in suspension under induced clay is given in Tables 5-6 and 5-7. The ^{137}Cs peak specific activities in suspensions under Kaolinite were smaller compared to ^{137}Cs peaks under Attapulgate along the model river, Figure 5-8.

FIG. 5-6. SORPTION-ACTIVITY RELATIONSHIP- ^{137}Cs

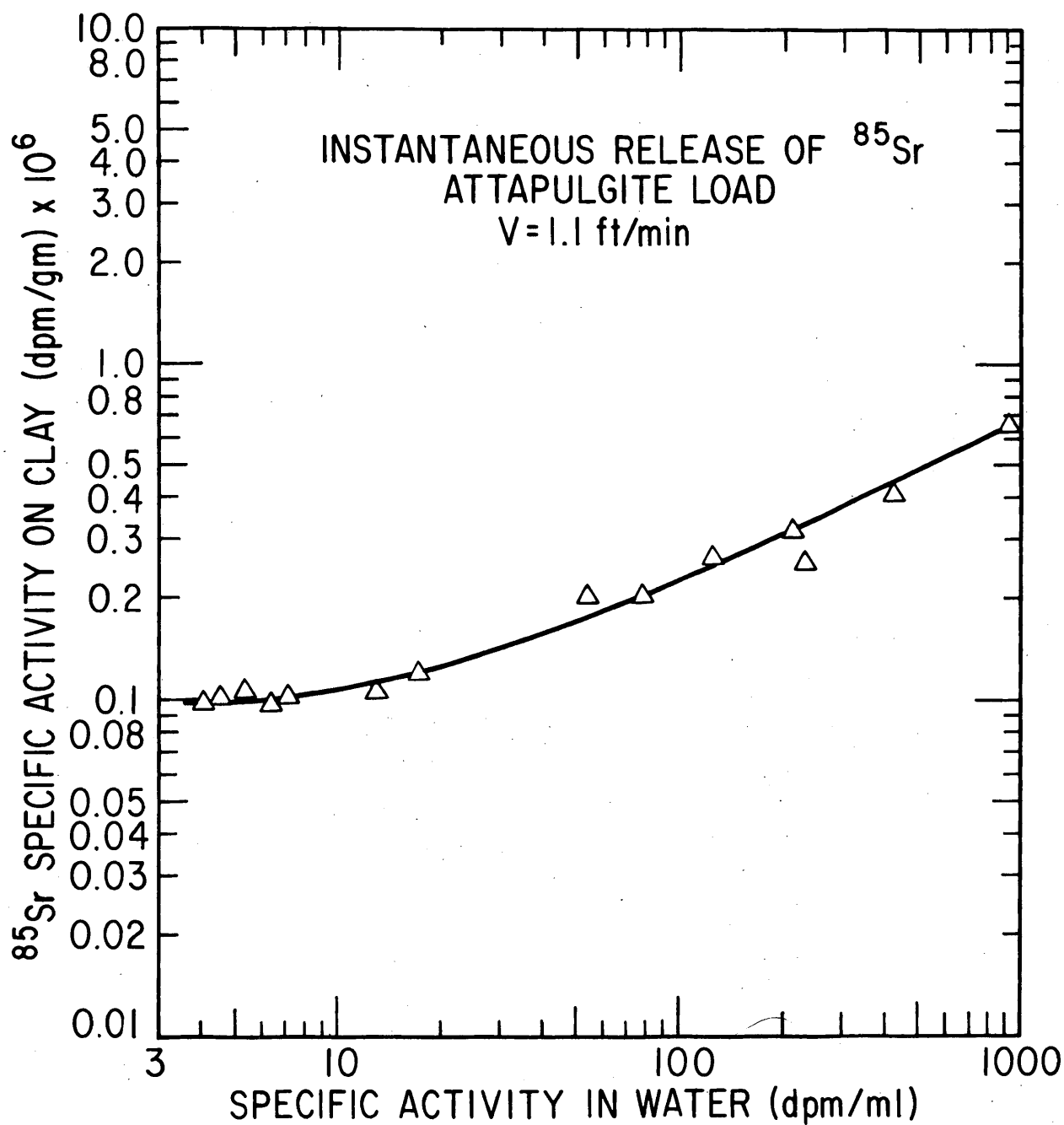


FIG.5-7. SORPTION - ACTIVITY
RELATIONSHIP - ^{85}Sr

Table 5-6. Radioactive Plume Characteristics
(Kaolinite)

Distance From Release Point (ft.)	Calculated Time To Peak (min.)	Observed Time To Peak (min.)	Observed Peak Concentration of ^{137}Cs (dpm/ml)
10	9.1	10.0	2006
40	36.4	26.0	870
90	81.8	90.0	438

Table 5-7. Radioactive Plume Characteristics
(Attapulgate)

Distance From Release Point (ft.)	Calculated Time To Peak (min.)	Observed Time To Peak (min.)	Observed Peak Concentration (dpm/ml)	
			^{85}Sr	^{137}Cs
5	4.6	3.5	2939	2896
50	45.5	40.0	885	762
95	86.4	75.0	634	453

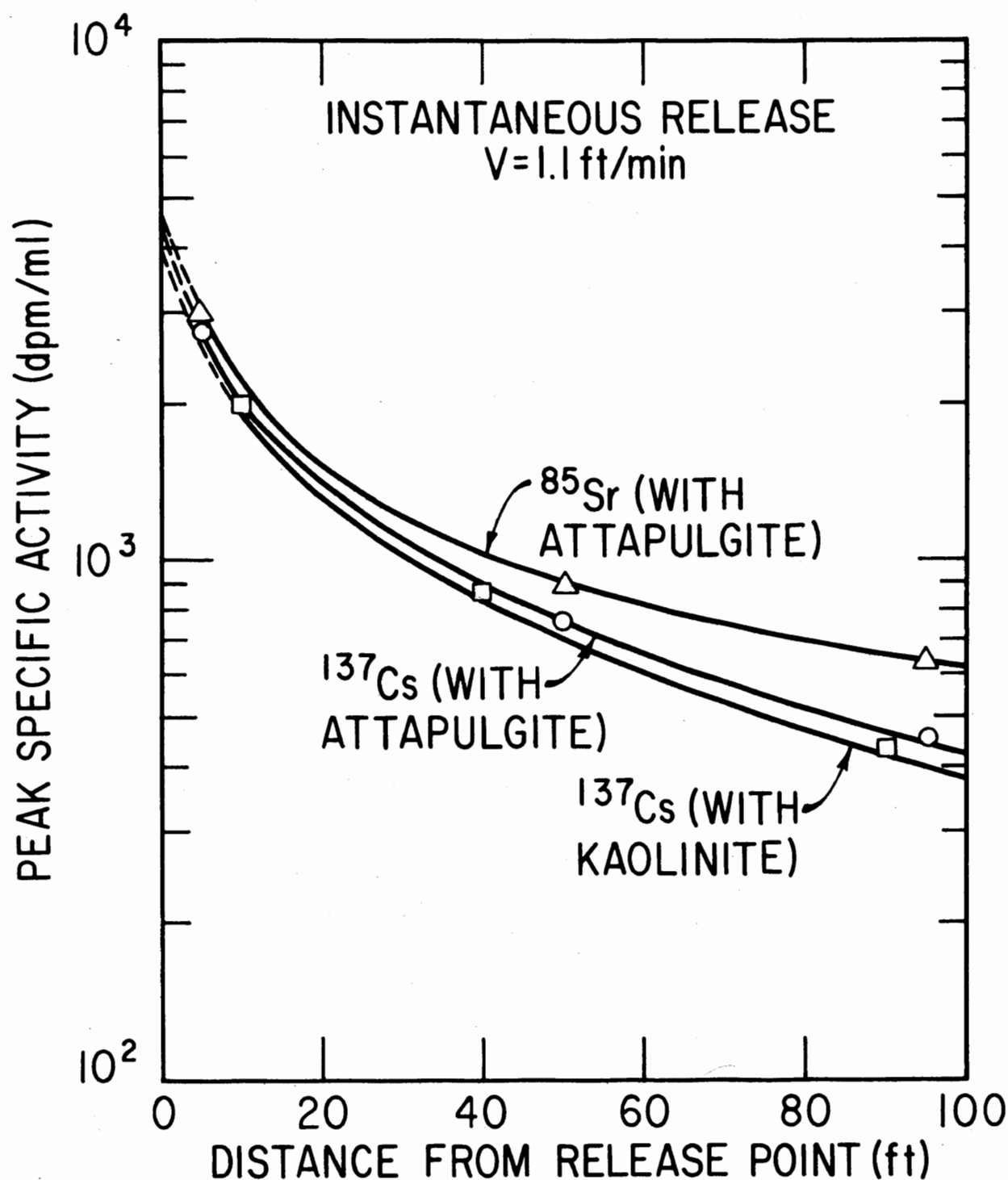


FIG.5-8. PEAK SPECIFIC ACTIVITY IN SUSPENSION VERSUS DISTANCE

Sediment Phase: Figure 5-9 shows that the variation of ^{85}Sr and ^{137}Cs associated with bottom sediments followed a pattern similar to that observed in the water phase. The migration function, Figure 5-10, of ^{85}Sr and ^{137}Cs in sediments was found to compare with aquaria results (Chapter IV). The penetration constants, m' , for ^{85}Sr were found to be 0.485 and 0.220 gm^{-1} , respectively, at 2 and 14 days contact time, indicating probable diffusion of ^{85}Sr through the interstitial water contained in the bottom sediments. In comparison the penetration constants for ^{137}Cs were calculated to be 0.80 and 0.60 gm^{-1} , respectively, for 2 and 14 days of contact.

Plant Phase: Quantitative data of the model river experiments indicated that the biomass sorbed a small fraction of the total amount of radionuclides released. The uptake of ^{85}Sr and ^{137}Cs by the plant mass was estimated to be 3% and 2% under Attapulgitite loading, and of ^{137}Cs was 2% under Kaolinite load as given in Table 5-3. The retention of ^{85}Sr and ^{137}Cs by Vallisneria was insignificant. Hence, plants as collection reservoirs for ^{85}Sr and ^{137}Cs play a minor role as compared to bottom sediment deposition, particularly when the streams are turbid.

The uptake of ^{85}Sr and ^{137}Cs by Vallisneria was immediate but the residence time was short. Most of the radioactive material was released to the water after the passage of the plume. The initial sorption of ^{137}Cs was less than the ^{85}Sr , Figure 5-11. However, this lack of uptake may be attributed to the fact that ^{137}Cs was sorbed onto the clay rapidly and therefore was not immediately available to the plants.

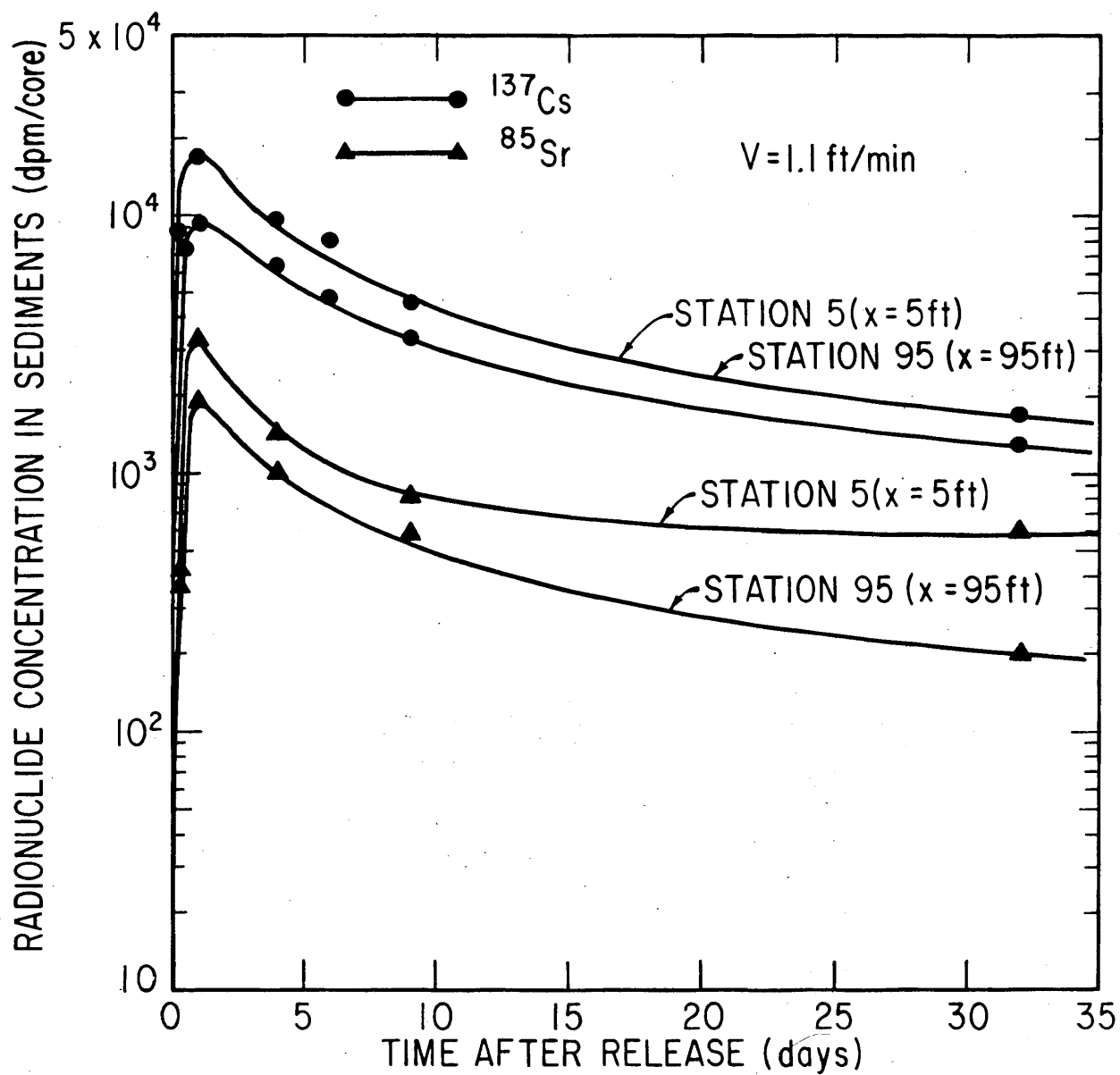


FIG. 5-9. VARIATION OF ACTIVITY ASSOCIATED WITH SEDIMENTS UNDER INDUCED SUSPENDED CLAY LOAD

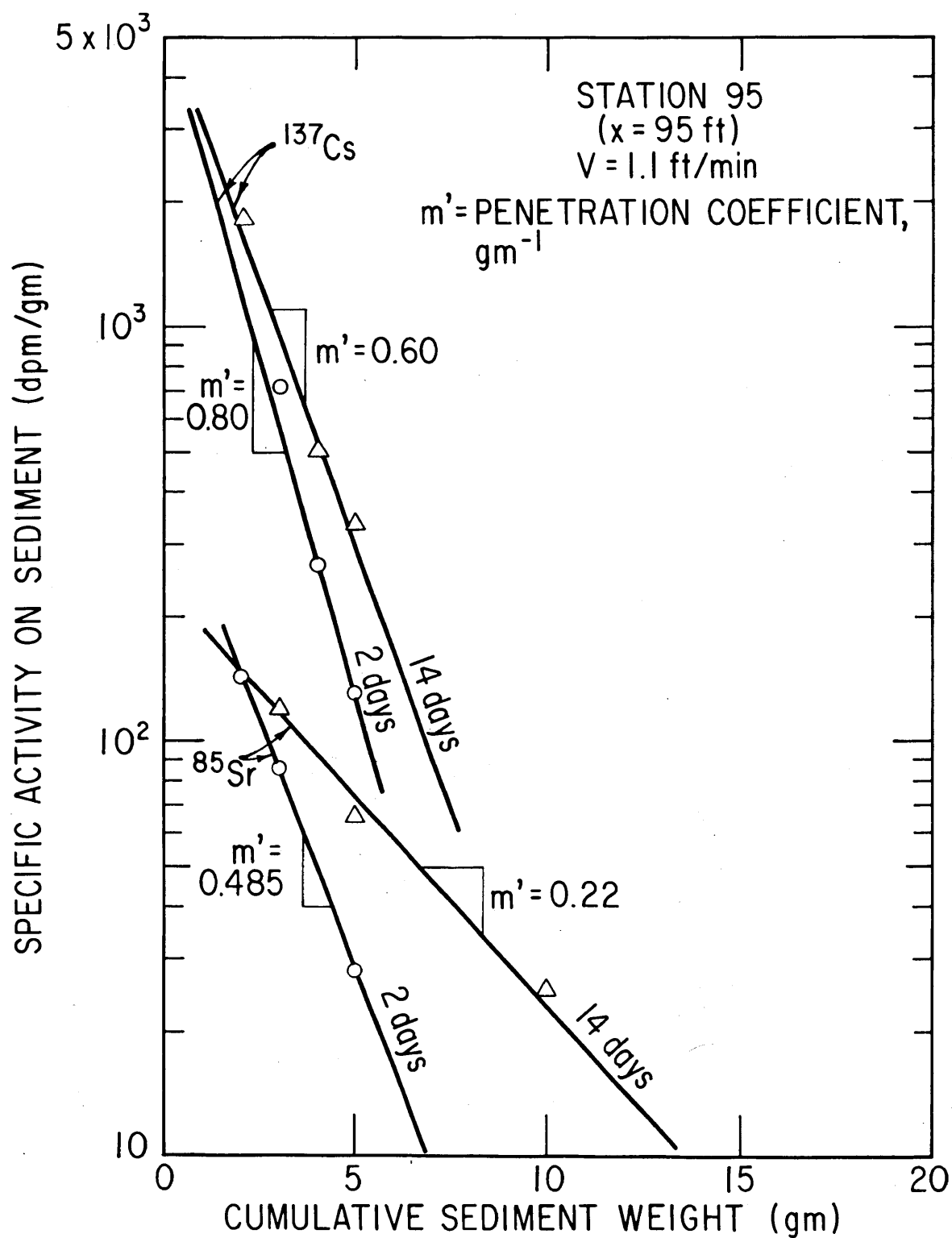


FIG.5-10. PENETRATION COEFFICIENTS UNDER INDUCED CLAY SUSPENSIONS

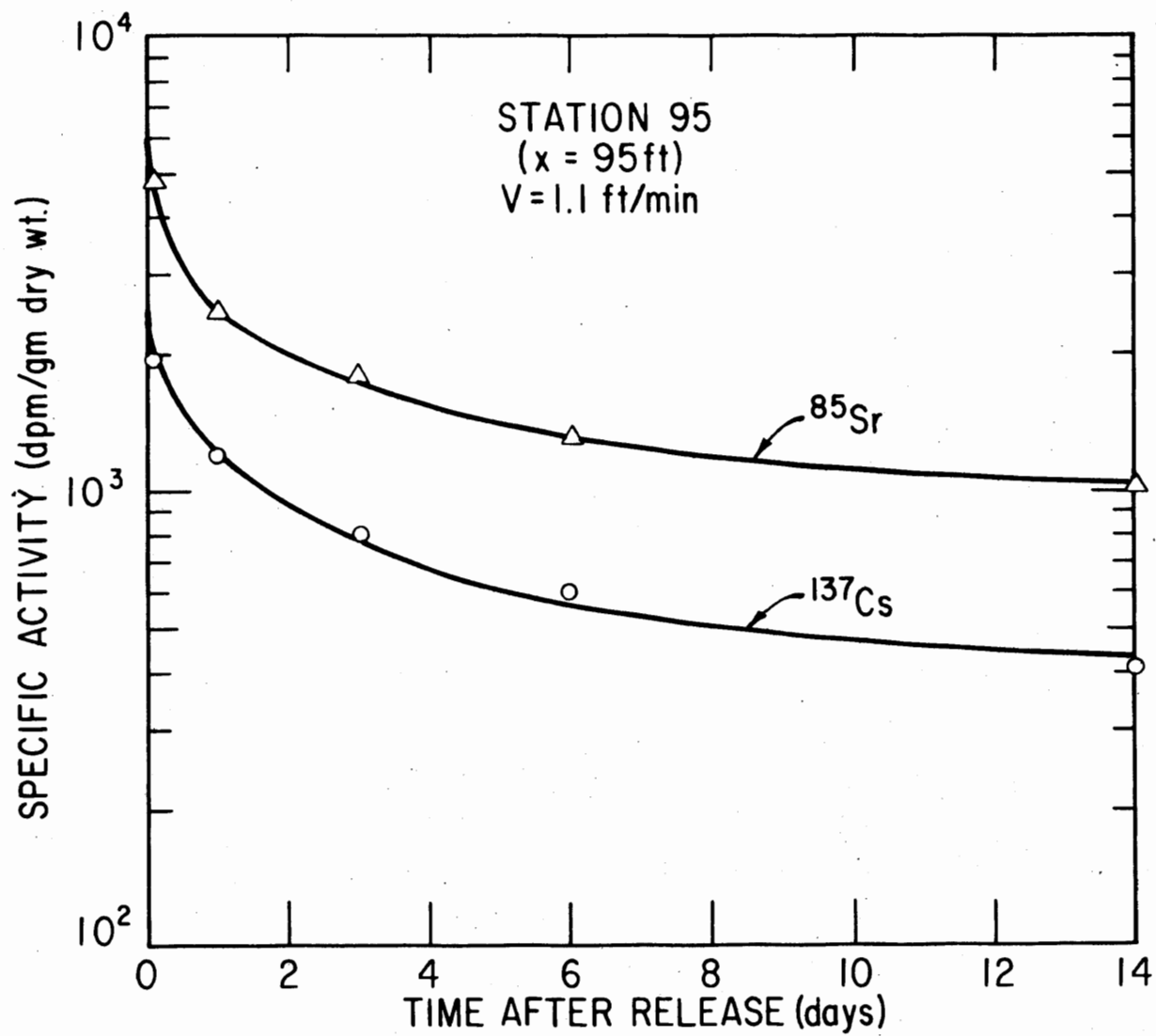


FIG. 5-II. RADIONUCLIDE UPTAKE BY VALLISNERIA

Continuous Release Experiment

The steady-state experiment was conducted utilizing the second 100-ft. section of the dual-channel flume system. The east channel was kept as a control, and an Attapulgite clay suspension was added to the west channel at the 100-ft. point. Electrical stirrers were located in both channels one foot upstream of the release point. The clay was introduced to the west channel for five hours, starting one hour before ^{137}Cs release and stopping at the termination of the four-hour ^{137}Cs release test. The average feed rate of the clay suspension was 3170 mg/min. The clay concentration as determined by gravimetric analysis was 13.6 gm/l. The east and west channels received 0.5 MPC (222 dpm/ml) of ^{137}Cs for a period of four hours totaling 1.2 mc for each channel. The sampling stations, type of samples obtained, and other details of this experiment are summarized in Table 5-8.

Sampling: The details of sample collection and processing are presented elsewhere (7, 80).

Analysis: The radionuclide content of samples was determined by gamma spectroscopy. Pertinent information regarding the counting efficiencies and contribution coefficients for ^{51}Cr , ^{85}Sr , and ^{137}Cs are given in Table 5-9, and the self-absorption factors for ^{137}Cs are indicated in Figure 5-12.

Transport: A discussion of the general distribution and transport of ^{137}Cs in the model river under induced suspended Attapulgite clay will be given in this section. ^{137}Cs was released in both channels

Table 5-8. Details of Steady-State Experiment

Particulars	East Channel	West Channel
Dates of Experiment	11-18-66 to 12-4-66	11-18-66 to 12-4-66
Duration	16 days	16 days
Depth of Water	10 in.	10 in.
Flow Rate	50 liters/min.	50 liters/min.
Type of Clay Load	no clay	Attapulgate
Radionuclide Added: ¹³⁷ Cs	1.2 mc	1.2 mc
Sampling Stations: (ft. from release point)		
1		10 (W,S)
2		30 (W,S)
3		70 (W,S)

W = water samples; S = sediment samples

Note: Water samples were collected at 8 inches below water surface

Table 5-9. Coefficients For Radionuclide Assay

Isotope	Influenced Isotopes			Counting Efficiency (%)
	⁵¹ Cr	⁸⁵ Sr	¹³⁷ Cs	
⁵¹ Cr	1.0	0	0	3.0
⁸⁵ Sr	0.1376	1.0	0	17.8
¹³⁷ Cs	0.1680	0.0676	1.0	11.6

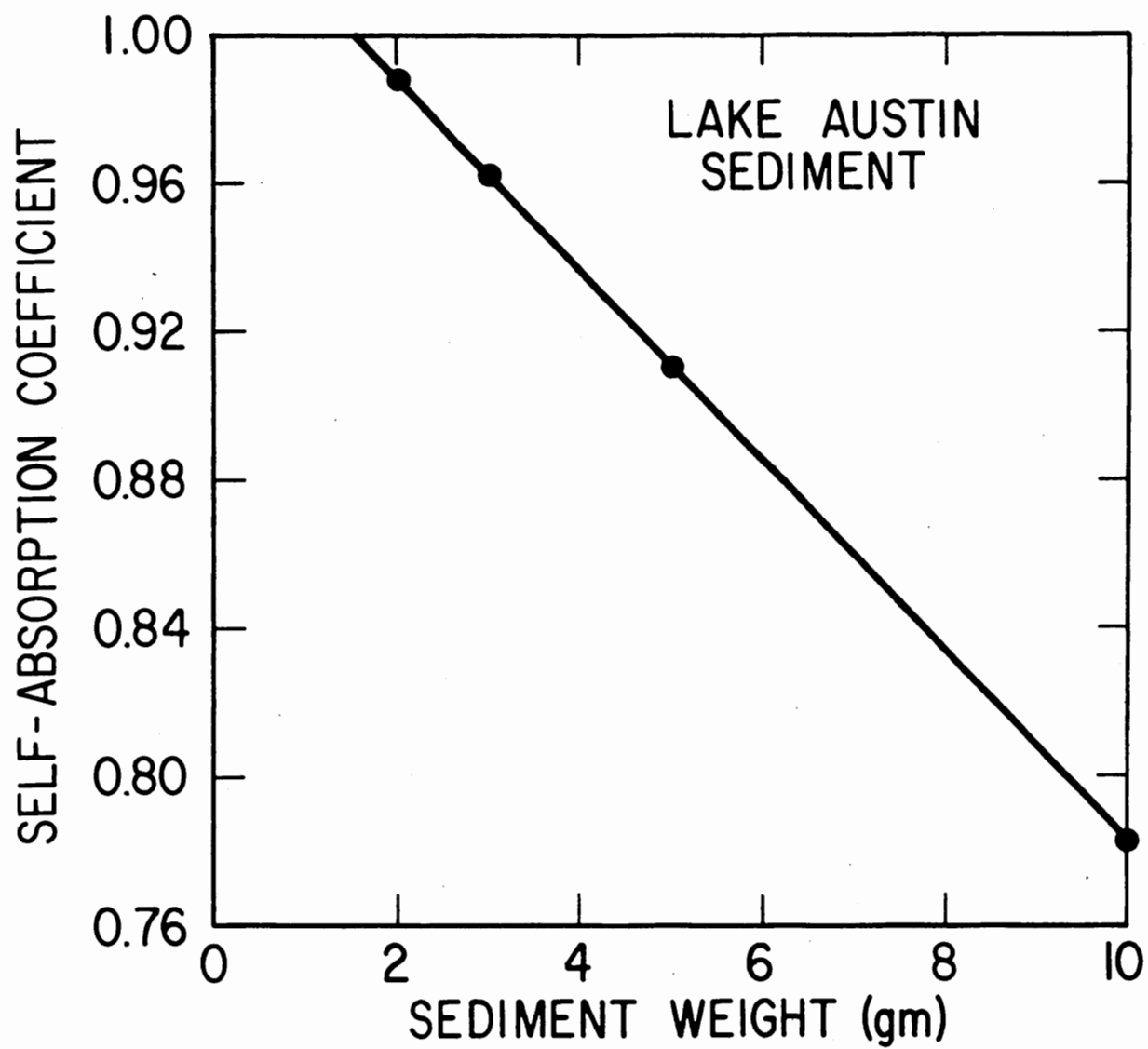


FIG. 5-12. SELF-ABSORPTION FACTORS
FOR ^{137}Cs

for four hours which was equivalent to an average of four detention periods, using an average flow of 74 ml/min. Attapulgite clay suspension was added at an average rate of 233 ml/min. to the west channel keeping the east channel as control.

The major postulate involved in this experiment was that the suspended clay would remove a large percentage of ^{137}Cs from suspension. As a result of this, the ^{137}Cs concentration in the clay-water mixture would be higher due to the suspended clay, and consequently, the transport of ^{137}Cs would be increased. However, if a portion of the clay settles, more retention of the added ^{137}Cs would be expected. The results of the test demonstrated such higher retention of ^{137}Cs in the turbid channel as shown in Table 5-10.

^{137}Cs in Water: The ^{137}Cs concentration in suspension during steady-state release conditions reached a fairly uniform level after one detention period (1 hour) at the farthest downstream station, with even shorter times required for stations nearer the release point. The observed steady-state concentrations are presented in Table 5-11. The ^{137}Cs concentration in suspension at steady state in both channels and the time-concentration of ^{137}Cs are shown in Figures 5-13 and 5-14. In the turbid channel (west), the radionuclide concentration in suspension decreased with distance from the release point due to removal by sorption on the clay and subsequent sedimentation. On the clear water side (east), the ^{137}Cs concentration in suspension decreased slightly along the model river. The average concentration at steady state in the east channel was 195 dpm/ml; the average west channel concentration was 222 dpm/ml.

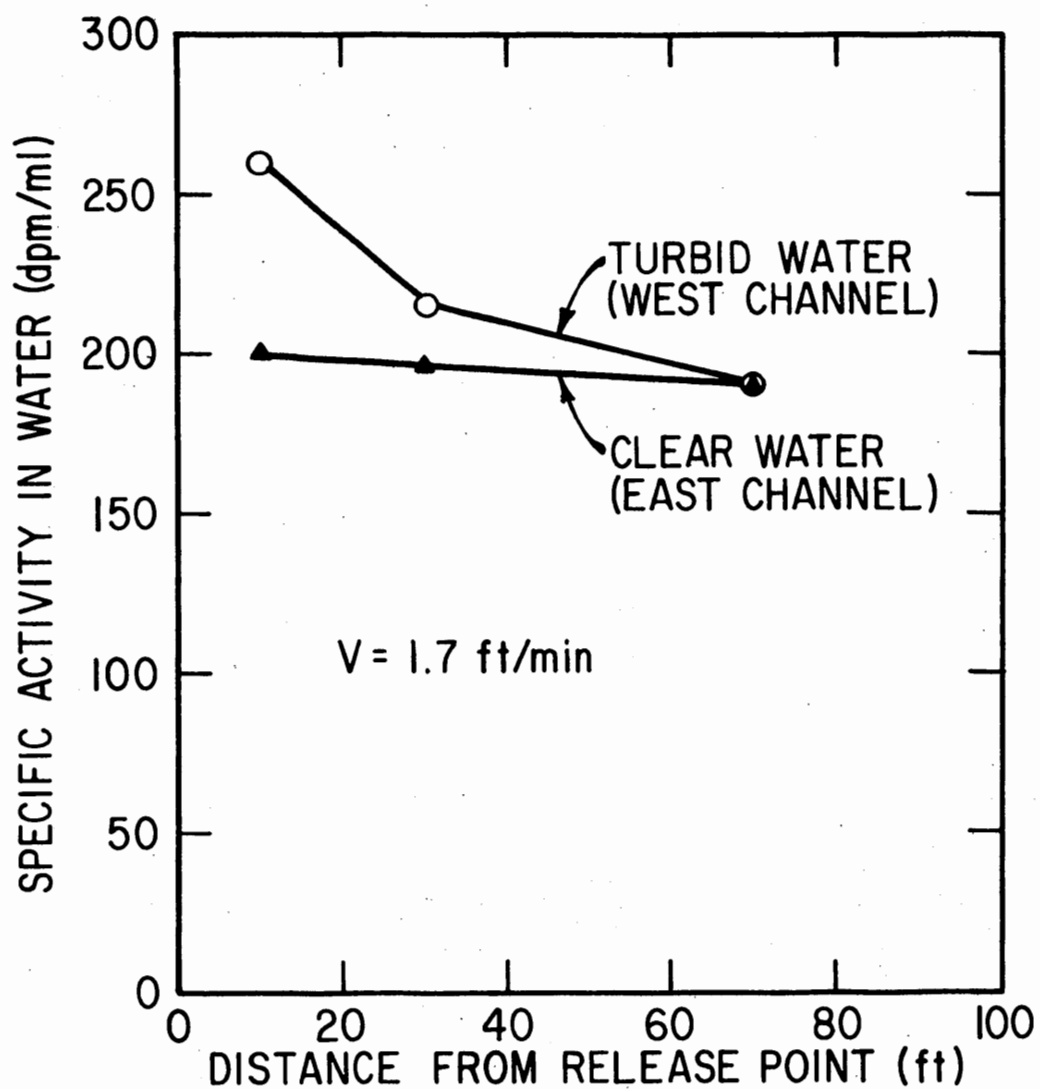


FIG. 5-13. ^{137}Cs CONCENTRATION AT STEADY STATE

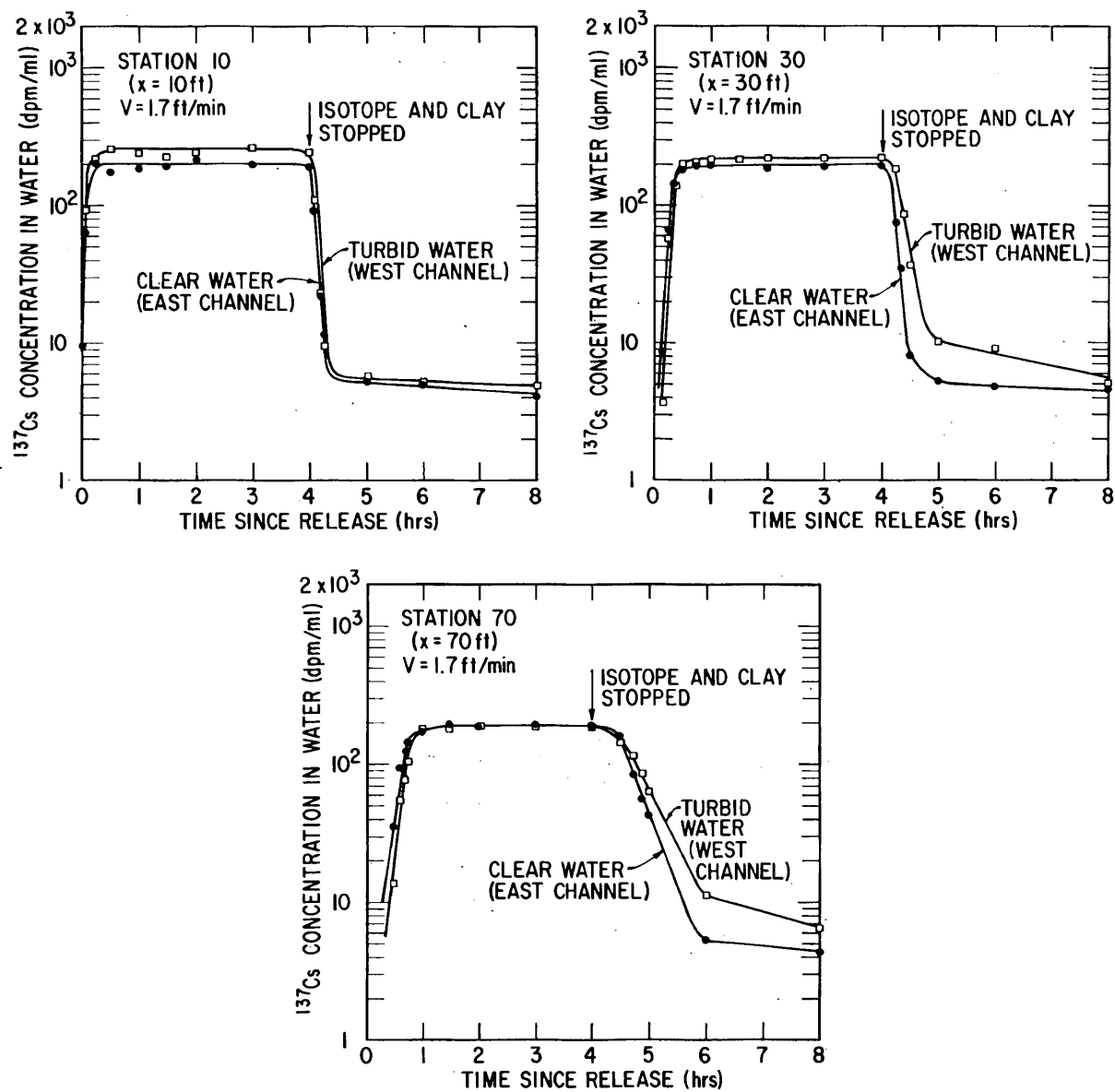
FIG.5-14. TIME-CONCENTRATION OF ^{137}Cs

Table 5-10. Transport Under Steady-State Release Conditions*

Phase	Percent of Total ^{137}Cs Added	
	East Channel- Control	West Channel- With Suspended Clay
Water:		
Discharged at End of Release	75	67
In the Model River	12	17
Bottom Sediments	8	15
Flume Walls	5	1

* Both isotope and Attapulgite clay released continuously

The steady-state ^{137}Cs concentrations in the turbid water were always higher than those on the control side at 10-ft. and 30-ft. stations; however, at 70-ft. stations, the concentration in both channels was about the same. These results indicate that the ^{137}Cs was sorbed onto the clay in the first portion of the west channel; then the clay began to settle and remove ^{137}Cs from the suspension. The slight decrease of the ^{137}Cs concentration in the suspension of the control channel indicates that there was uptake by the bottom sediments.

The concentration of Attapulgite clay in the west channel is shown in Figure 5-15. The average influent clay concentration of 63 mg/l was reduced to 39 mg/l at the outlet. Of the clay which was added to the west side, 87% passed 10-ft. station, 73% passed 30-ft. station, and 62% passed 70-ft. station. This means that 38% of the total clay

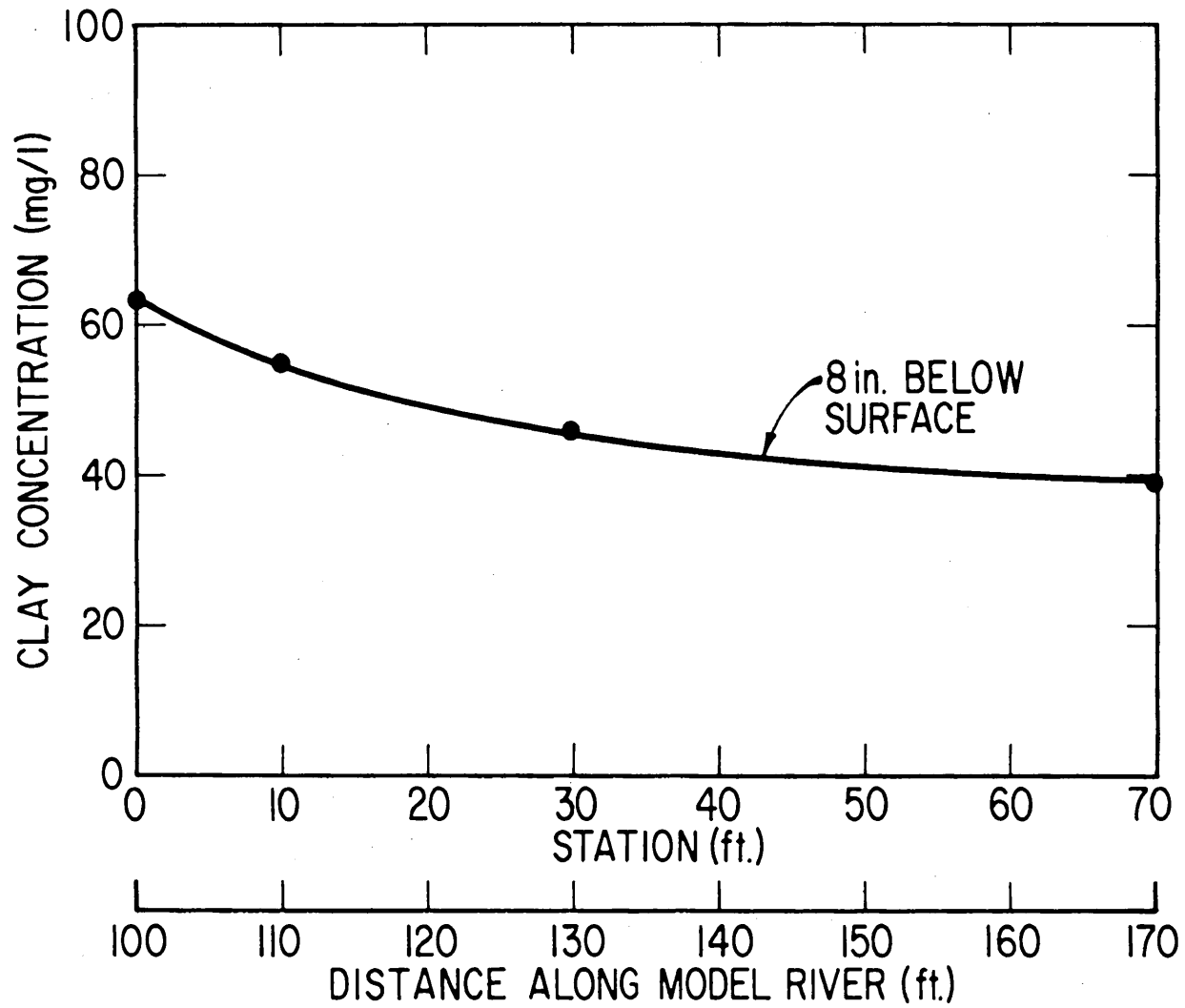


FIG. 5-15. ATTAPULGITE CLAY CONCENTRATION
ALONG MODEL RIVER

Table 5-11. ^{137}Cs in Suspension Under Steady-State Conditions

Distance From Release Point (ft.)	East Channel (Control)		West Channel (With Suspended Clay)		Ratio: <u>West Eql.</u> <u>East Eql.</u>
	^{137}Cs Concen- tration** (dpm/ml)	Fraction* ^{137}Cs	^{137}Cs Concen- tration** (dpm/ml)	Fraction* ^{137}Cs	
10	200	1.000	260	1.000	1.30
30	195	0.975	215	0.825	1.10
70	190	0.950	190	0.730	1.00

* Fraction of concentration at 10 ft.

** At steady-state

settled and was responsible in reducing the transport of ^{137}Cs in the west channel.

The percentage of ^{137}Cs in suspension associated with the suspended clay was determined by the filtration procedure. Approximately 8% of the ^{137}Cs in suspension was associated with suspended matter on the control side. In the turbid channel, about 73% of the ^{137}Cs in suspension was sorbed onto the suspended clay.

Obviously, within certain limits, the more ^{137}Cs in the suspension the more will be adsorbed on the clay. Within the limits of this experiment, the amount of ^{137}Cs on the clay and in the clay-water mixture was controlled by a linear relationship. This plot is shown in Figure 5-16.

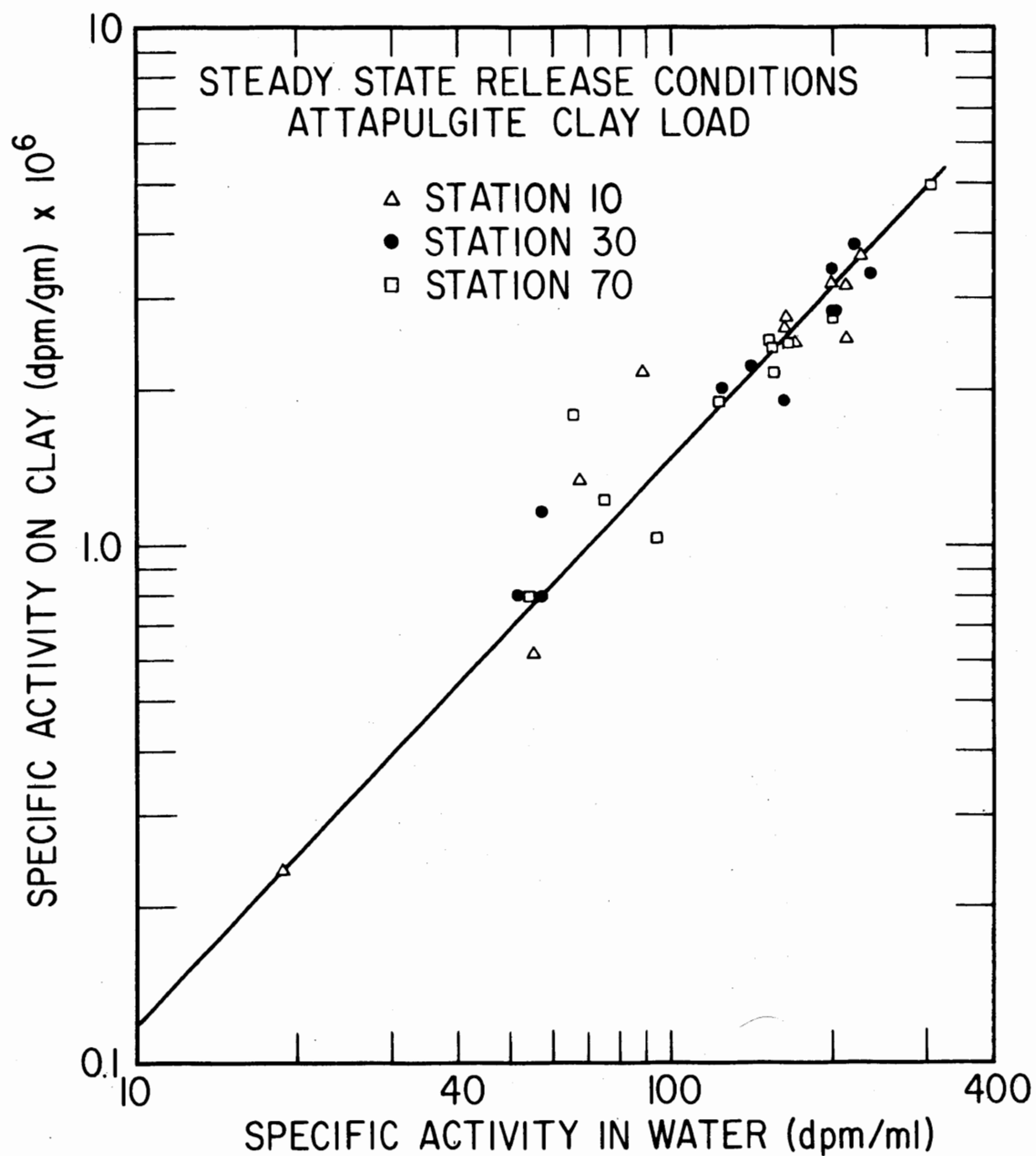


FIG. 5-16. SORPTION - CONCENTRATION
RELATIONSHIP - ^{137}Cs

^{137}Cs in Bottom Sediments: The effect of the induced suspended clay on the concentration of ^{137}Cs is indicated in Table 5-12 and Figure 5-17. The bottom sediments in the turbid channel contained nearly twice as much ^{137}Cs as did the sediments in the control channel.

The amount of ^{137}Cs associated with the bottom sediments increased on both sides, after stopping the release of ^{137}Cs . The deposited clay with sorbed activity was responsible for this. It is assumed that as more suspended clay and more ^{137}Cs were added, the bottom sediment activity on the west side would have continued to increase.

The penetration of ^{137}Cs into the bottom sediment was examined to determine the extent of migration. The results are presented in Figure 5-18 for 10-ft. and 70-ft. stations. The higher values of the penetration constants, m' , in the turbid channel indicate that more of

Table 5-12. Amount of ^{137}Cs in Sediment Phase

Time (days)	Percentage of ^{137}Cs Added	
	East Channel-Control	West Channel-Turbid
0*	8	15
2	9	17
4	12	18
14	9	16
16	7	15

*Values same as the 4-hour values at the end of radionuclide release period

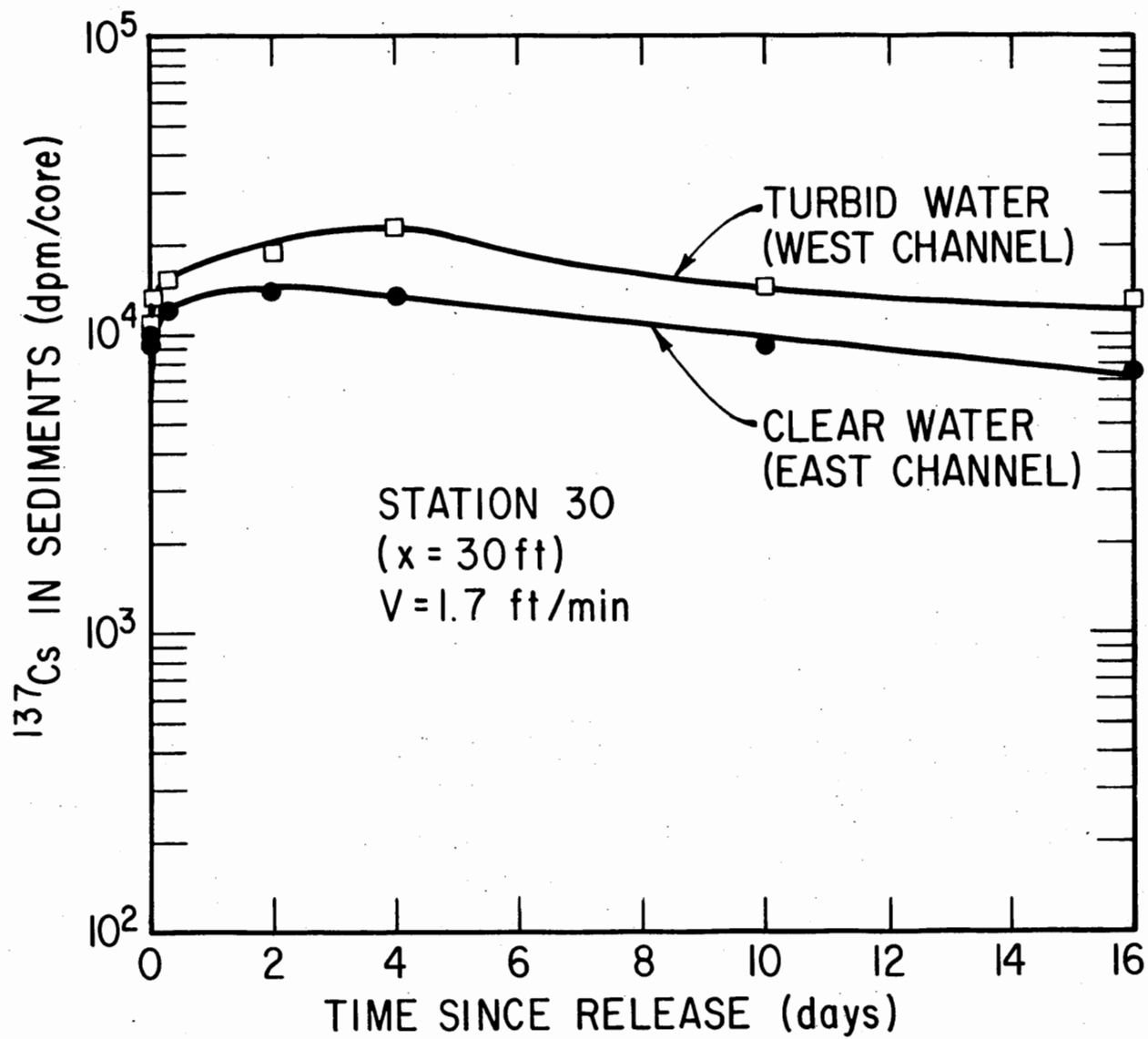


FIG. 5-17. ^{137}Cs ASSOCIATED WITH SEDIMENTS

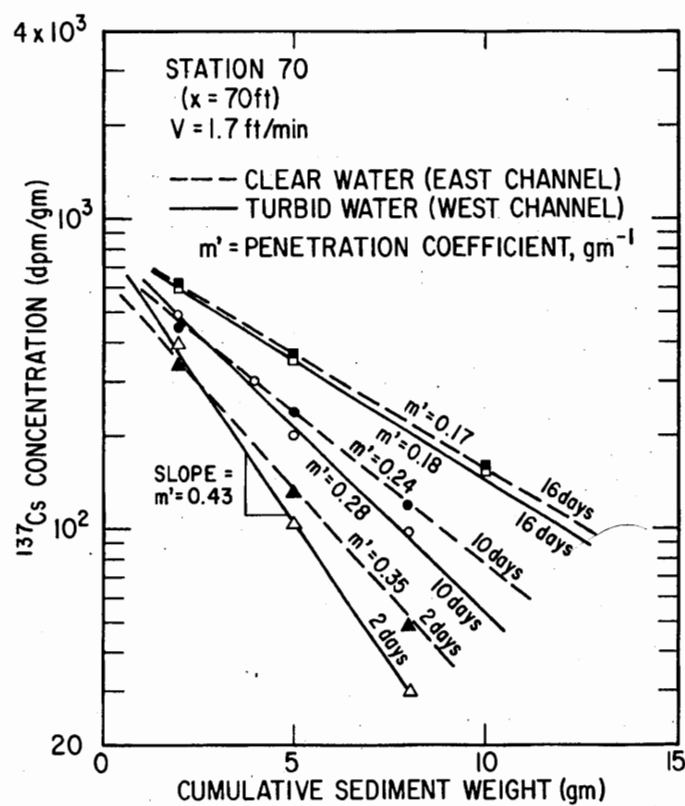
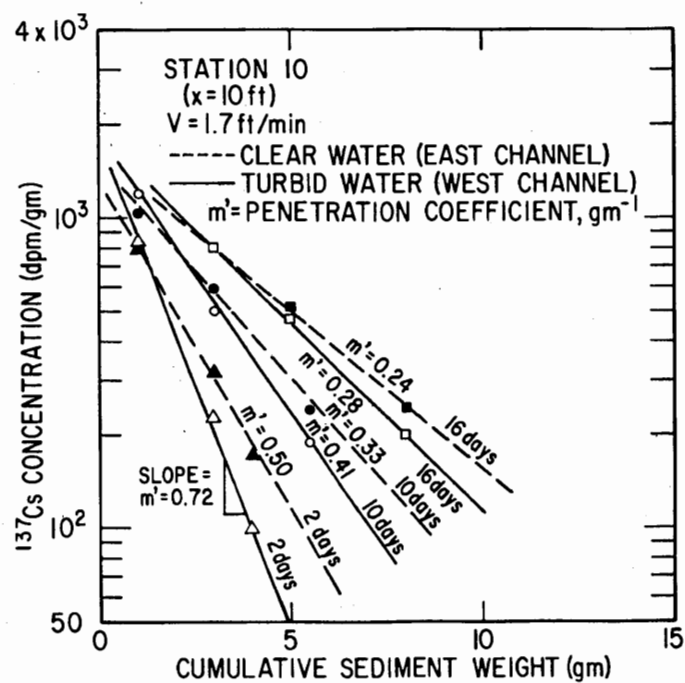


FIG. 5-18. ^{137}Cs PENETRATION INTO BOTTOM SEDIMENTS

^{137}Cs was retained in the sediments due to the layer of deposited clay. As the contact period increased, the m' values decreased indicating a release of ^{137}Cs from the bottom sediments. The clay concentrations were lower at the 70-ft. station and hence the layer of deposited clay was also thin giving lower m' values. These results support the fact that deposited clay will tend to increase the amount of ^{137}Cs retained by the bottom sediments.

CHAPTER VI

DISCUSSION

Modified longitudinal dispersion equations can be used to describe the transport of ^{85}Sr and ^{137}Cs in turbid streams. The transport equations are presented in terms of observed peak radionuclide concentrations with respect to time and position in the flume.

Also, an empirical equation can be written which will describe the distribution of suspended clay along a model river during steady-state release conditions.

Modified Dispersion Equation

The development of a modified dispersion equation describing the transport of radionuclides in a turbid stream is relatively complex. Yet, the basis for describing the transport of any soluble, non-reactive tracer material, instantaneously released, may be expressed by a two-dimensional, longitudinal dispersion relationship, Equation 6-1 (62):

$$C_T(x,t) = \frac{M}{A\sqrt{4D_x t}} e^{-\frac{(x-ut)^2}{4D_x t}} \quad (6-1)$$

where

$C_T(x,t)$	= calculated concentration of tracer in water (mass per unit volume)
M	= total mass of tracer released
A	= area of stream cross-section, ft^2
D_x	= longitudinal dispersion coefficient, ft^2/min .
t	= time, min.
u	= average velocity of water, ft/min .
x	= distance downstream from point of release, ft.

The peak tracer concentration can be estimated by Equation 6-1 for a modal time, t_m , which is equal to

$$t_m = x/u \quad (6-2)$$

Hence, the calculated peak tracer concentration, C_{TP} , may be expressed by Equation 6-3.

$$C_{TP} = \frac{M}{A \sqrt{4\pi D_x t_m}} \quad (6-3)$$

However, due to the introduction of the suspended clay into the model river, Equation 6-3 must be modified to account for the discrepancies found between the calculated and observed peak concentrations of ^{85}Sr and ^{137}Cs . Such discrepancies develop as a result of radionuclide sorption on clay and subsequent settling of this clay, and radionuclide sorption by both bottom sediments and plants.

Disparities for ^{85}Sr and ^{137}Cs in the model river with Attapul-gite and Kaolinite clay suspensions follow a very well defined pattern, Figure 6-1. The ratio of the observed to calculated peak radionuclide concentrations at each station are shown in Tables 6-1 and 6-2. The slope of the plot, Figure 6-2, is called the transport coefficient, "a". The peak radionuclide concentration that might occur at any distance downstream from the point of release in a turbid stream can be estimated from Equation 6-4.

$$C_{OP} = C_{TP} e^{-ax} \quad (6-4)$$

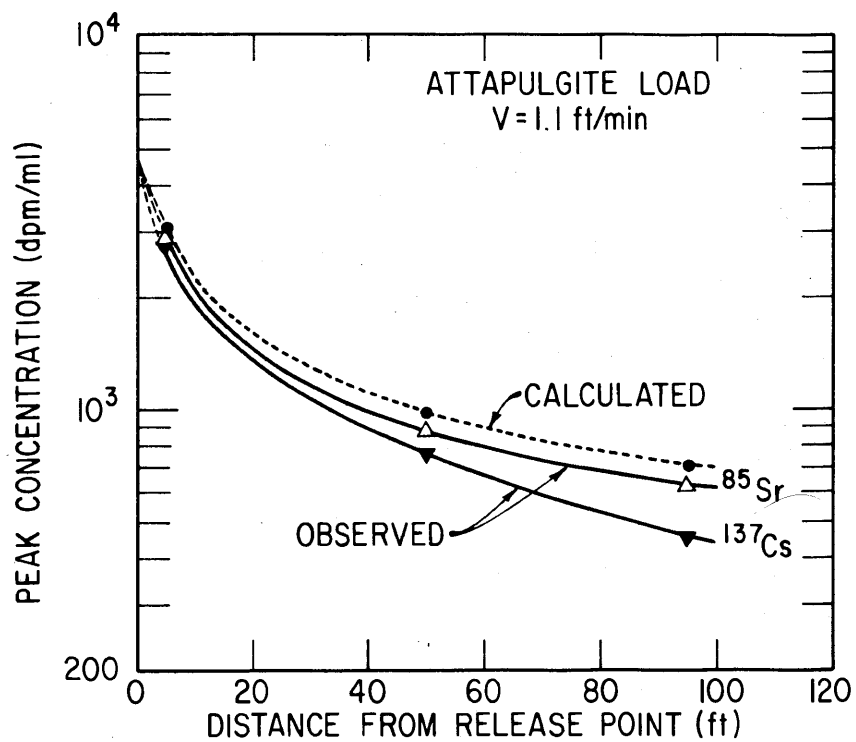
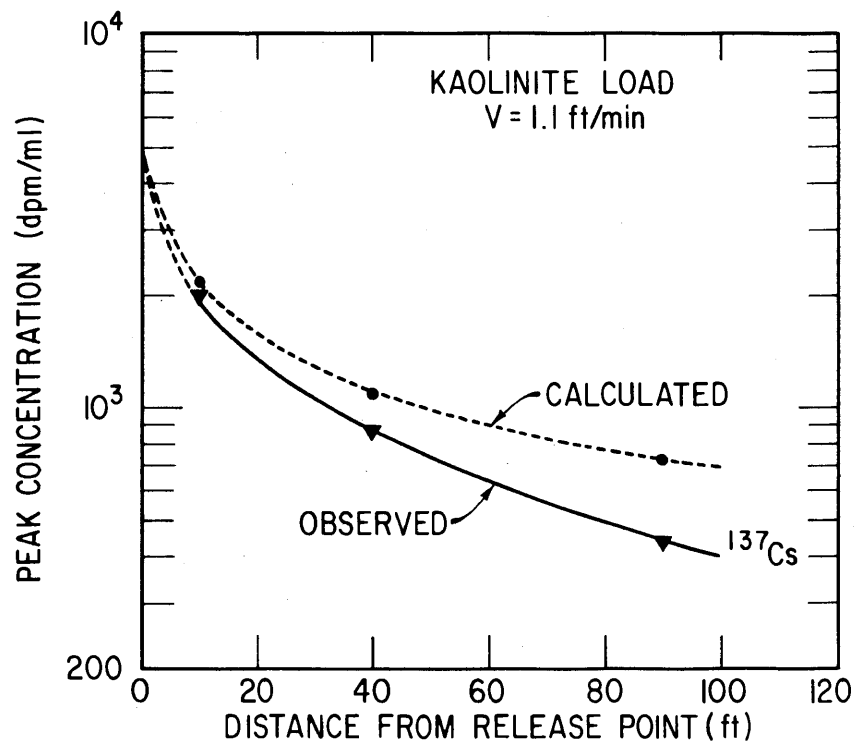


FIG. 6-1. DISPARITY BETWEEN CALCULATED AND OBSERVED VALUES

Table 6-1. Calculated and Observed Peak Concentrations
in the Model River (Kaolinite)

Station, x (ft.)	Modal Time, t_m (min.)		Peak ^{137}Cs Concentration (dpm/ml)		Ratio: $\frac{\text{Observed Peak, } C_{OP}}{\text{Calc. Peak, } C_{TP}}$
	Observed	Calculated	*Observed	Calculated	
10	10.0	9.1	2006	2180	0.92
40	26.0	36.4	870	1090	0.80
90	90.0	81.8	438	730	0.60

*Average values

Table 6-2. Calculated and Observed Peak Concentrations
(Attapulgitite)

Station, x (ft.)	Modal Time, t_m (min.)		Peak Concentration (dpm/ml)			Ratio: $\frac{\text{Obs. Peak, } C_{OP}}{\text{Calc. Peak, } C_{TP}}$	
	Obs.	Calc.	Observed*		Calc.	^{85}Sr	^{137}Cs
			^{85}Sr	^{137}Cs			
5	3.5	4.6	2939	2896	3080	0.95	0.94
50	40.0	45.5	885	762	975	0.91	0.78
95	75.0	86.4	634	453	708	0.89	0.64

*Average values

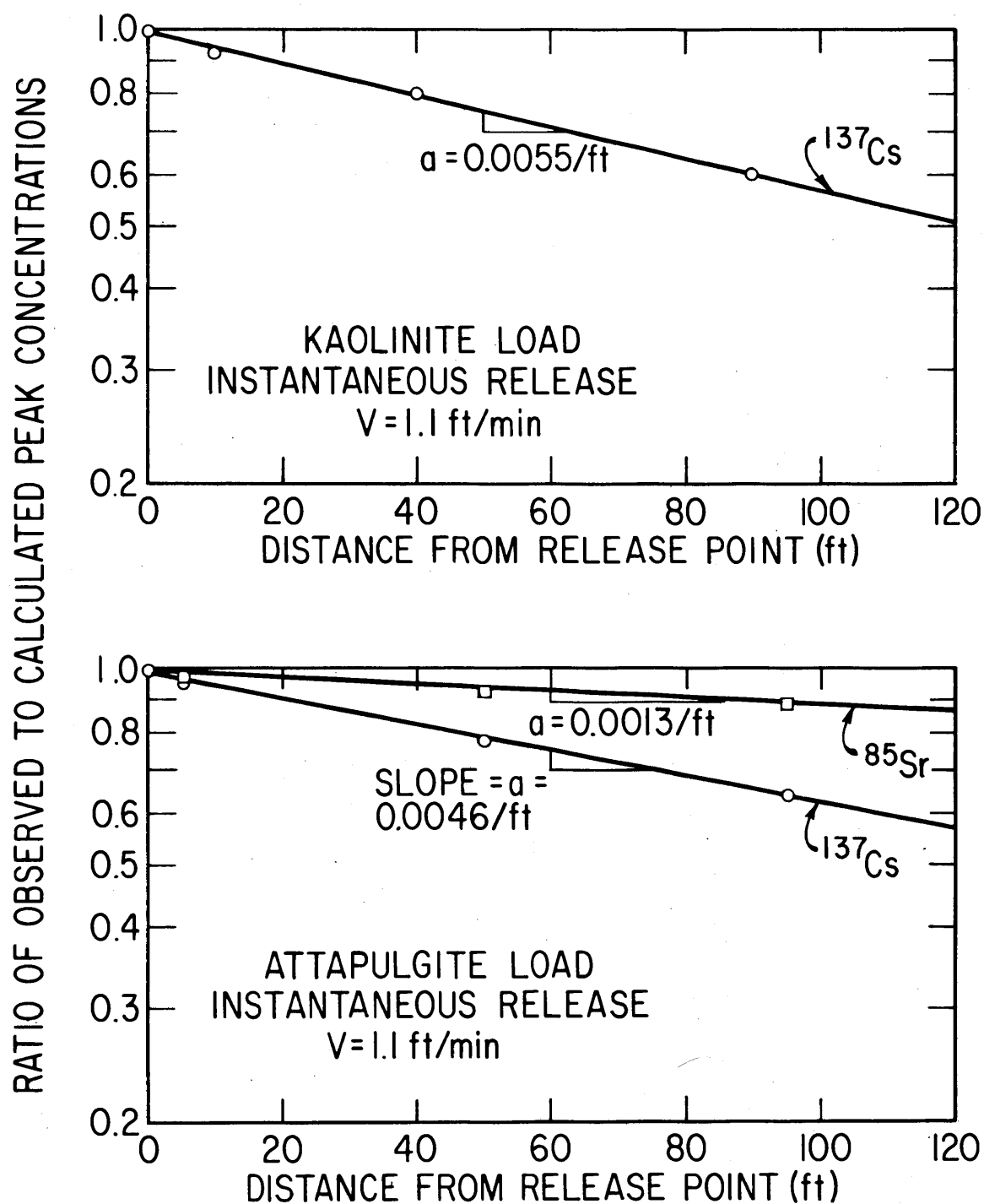


FIG. 6-2. RELATIONSHIP BETWEEN DISTANCE DOWNSTREAM AND OBSERVED PEAK CONCENTRATIONS

where C_{OP} = observed peak radionuclide concentration in water, dpm/ml
 C_{TP} = calculated peak radionuclide concentration in water, dpm/ml
 a = transport coefficient, per ft.
 x = distance downstream from release point, ft.

Equation 6-4 can be rewritten in terms of Equation 6-3:

$$C_{OP} = \frac{M}{A\sqrt{4\pi D_x t_m}} e^{-ax} \quad (6-5)$$

or

$$C_{OP} \text{ in dpm/ml} = \frac{3.53 \times 10^{-4} M}{A\sqrt{4\pi D_x t_m}} e^{-ax} \quad (6-6)$$

The factor, 3.53×10^{-4} , converts ft^3 into cm³.

Values of "a" as established for ^{85}Sr and ^{137}Cs under these particular experimental conditions are summarized in Table 6-3.

Whereas Equation 6-6 provides estimates for modal concentration, it is also possible to calculate the reduction in radionuclide concentration passing a station in a turbid stream through the use of a modified longitudinal dispersion equation. The fractions of initial ^{85}Sr and ^{137}Cs passing established sampling stations are indicated in Tables 6-4 and 6-5. Figure 6-3 shows the correlation between the fraction of initial amount passing a station and mean flow-through time. The slope of the plot is called the sorption coefficient, R. The value of R is dependent on the amount of turbidity in the stream, nature of radionuclide, and hydrodynamic conditions of the stream. Values of the sorption coefficients as established for ^{85}Sr and ^{137}Cs

Table 6-3. Transport Coefficients, "a"

Radionuclide	Induced Suspended Clay Load	
	Attapulgate (ft. ⁻¹)	Kaolinite (ft. ⁻¹)
⁸⁵ Sr	0.0013	----
¹³⁷ Cs	0.0046	0.0055

in the model river are summarized in Table 6-6. A generalized equation for the relationship shown in Figure 6-3 is:

$$\frac{C(x,t)}{C_T(x,t)} = e^{-Rt} \quad (6-7)$$

where $C(x,t)$ = radionuclide concentration at any instant, t , and
at a particular station, x
 $C_T(x,t)$ = radionuclide concentration from dilution only with
respect to time and position
 R = sorption coefficient, per hour
 t = time, hour.

It is possible to write Equation 6-8 to describe the effects of both dilution and sorption:

$$C(x,t) = \frac{M}{A\sqrt{4\eta D_x t}} e^{\frac{-(x-ut)^2}{4D_x t}} e^{-Rt} \quad (6-8)$$

For conditions other than those described in these experiments it is necessary to reevaluate the coefficients. The major physical

Table 6-4. Transport Characteristics of ^{85}Sr and ^{137}Cs
(Attapulgate)

Station, x (ft.)	Mean Flow-Through Time (min.)	Percent of Initial Concentration Passing*	
		^{85}Sr	^{137}Cs
5	4.6	99	95
50	45.5	96	80
95	86.4	95	70

*Corrected for radioactive decay

Table 6-5. Transport of ^{137}Cs (Kaolinite)

Station, x (ft.)	Mean Flow-Through Time (min.)	Percent of Initial Concentration Passing*
10	9.1	96
40	36.4	90
90	81.8	84

*Corrected for radioactive decay

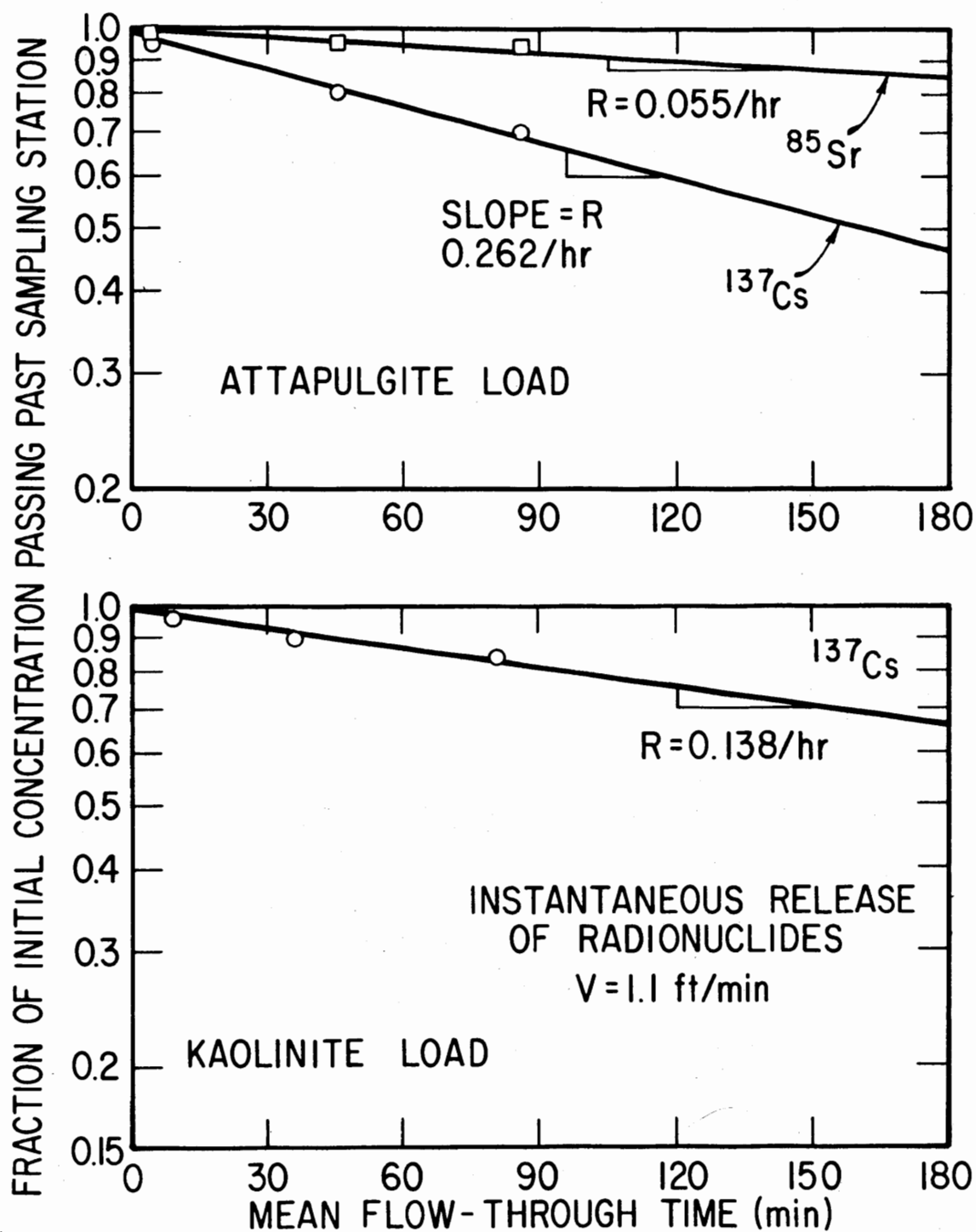


FIG. 6-3. RELATIONSHIP OF MEAN FLOW-THROUGH TIME TO FRACTION OF INITIAL CONCENTRATION PASSING

Table 6-6. Sorption Coefficient, R, Per Hour

Isotope	Induced Suspended Clay Load	
	Attapulgate	Kaolinite
^{85}Sr	0.055	---
^{137}Cs	0.262	0.138

and hydrodynamic variables of a turbid stream must be established and the constants "a" and R determined.

Longitudinal Distribution of Clay Load

Calculation of the radioactivity budget for the stream would be incomplete without considering the case of a tributary carrying suspended sediment. Information regarding the distribution of tributary sediment, and the sorptive capacity of the suspended sediment is required.

The nature of concentration and load variation of the suspended sediment in the stream is important because it is indicative of the capacity or lack of capacity of the stream to transport a given amount of radioactive waste. A distribution of the Attapulgate clay suspension was obtained in the model river during steady-state release tests. The variation of total amount of clay released as it moved downstream is shown in Figure 6-4. The data used for the plot are given in Table 6-7.

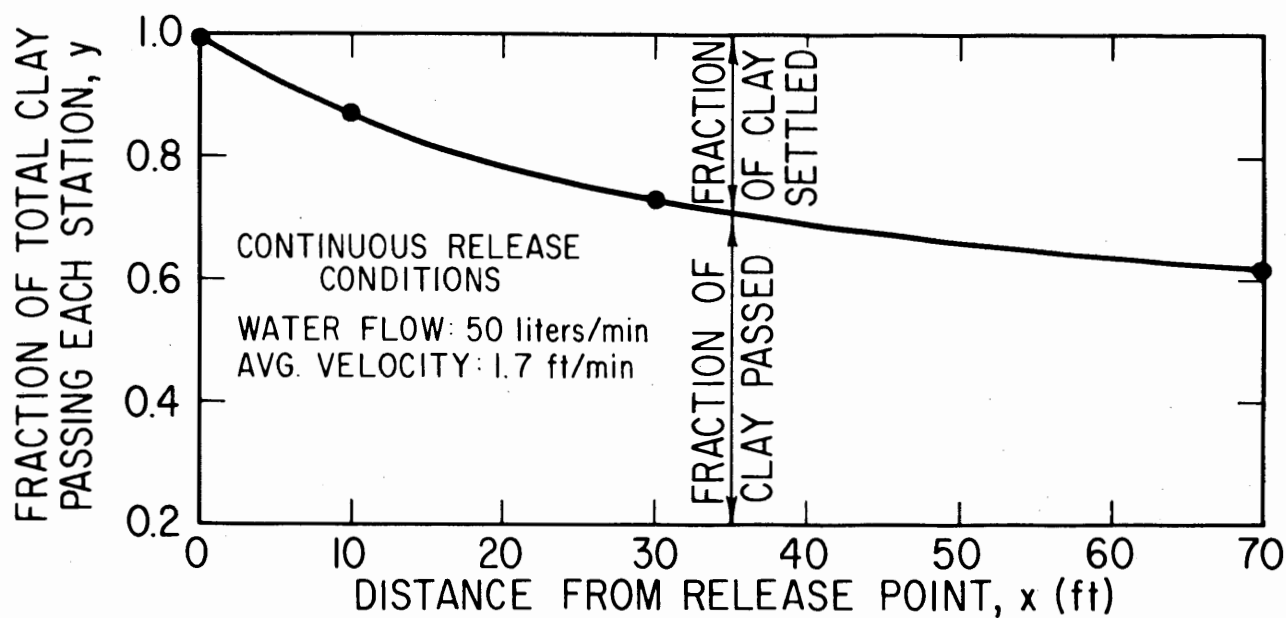


FIG. 6-4. VARIATION OF TOTAL AMOUNT OF CLAY RELEASED WITH DISTANCE

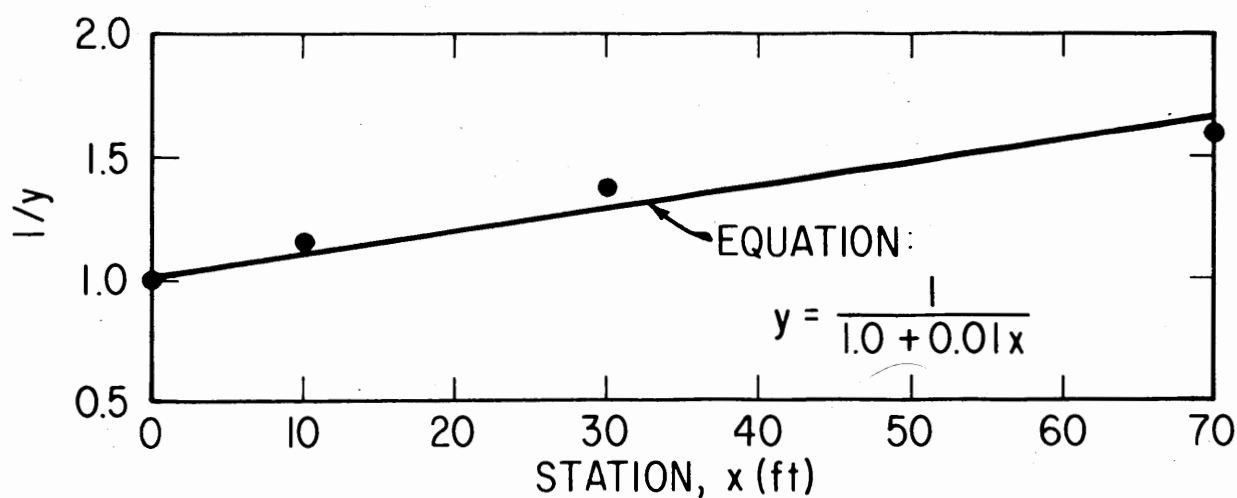


FIG. 6-5. RELATIONSHIP BETWEEN FRACTION OF CLAY PASSED AND DISTANCE

The plot indicated that the settling characteristics exhibited by the clay were typical of flocculent particles. A linear relationship (Figure 6-5) between the fraction of total clay passing, y , and station, x , was obtained through replotting of the data in Figure 6-4. For this special case the empirical relationship shown in Equation 6-9 is applicable. For any other stream, it will be

$$y = \frac{1}{1.0 + 0.01x} \quad (6-9)$$

where y = fraction of total clay passing a station, x ft. from the release point

necessary to know the type and settling characteristics of the suspended sediment, velocity of the stream, and nature of ions present in the stream water.

Table 6-7. Details of Clay Distribution in the Continuous Release Experiment - West Channel*

Station, x (ft.)	0	10	30	70
Average Clay Concentration (mg/l)	63	55	46	39
Percent Passing Each Station (%)	100	87	73	62
Total Amount of Clay Passing Each Station (gm)	760	663	555	472
Gm. of Clay Settled Between 2 Stations	97	108	83	

*During the 4-hour period of ^{137}Cs release

Suggestion for Future Research

Future research can be directed toward determining the extent of sorption by organic and inorganic fractions of suspended and bottom sediments. Also the effect of recycling of the effluent on the radio-nuclide uptake by bottom sediments can be investigated.

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