

RADIOACTIVITY TRANSPORT IN WATER--
TRANSPORT OF Sr⁸⁵ and Cs¹³⁷ IN AN AQUATIC ENVIRONMENT

Technical Report--10

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ABSTRACT

The transport of two radionuclides, Sr⁸⁵ and Cs¹³⁷, was observed in a model river containing lake sediments, typical rooted plants, and suspended plankton. Measurements of uptake and release of these radionuclides in aquatic systems following a single release were conducted at various locations and times during 120 days. In support of these radionuclide transport data, measurements were also made of pH, oxidation-reduction potential, dissolved oxygen, temperature, total and filtered light intensity, carbon dioxide, biomass geochemical analyses, and flow.

The results describe the relative uptake and transport by the components of the flume system. Graphical techniques were developed to describe the distribution of radionuclides in the liquid phase and analytical expressions were obtained to relate the measured detention to the amounts of plants, sediments, and suspended solids.

CONCLUSIONS

1. In a sluggish stream system, the greatest fraction of Sr^{85} and Cs^{137} will remain in the water phase; however, a significant amount of Sr^{85} and Cs^{137} will be temporarily removed by aquatic plants and bottom sediments. The radionuclides sorbed by aquatic plants will be released back into solution within a few days following the introduction of the radionuclides into the stream, whereas the radionuclides sorbed by bottom sediments will be released back into solution after a few months following the initial release.

2. The net transport of Sr^{85} and Cs^{137} across a given section of stream as used in this experiment can be formulated as follows:

$$F_x = F_0 e^{-kx},$$

where:

F_x = net transport,

F_0 = cumulative fraction of radionuclide discharged through the release point,

k = slope of line (semilog plot), and

x = distance downstream from the point of release.

3. The depth of penetration of Sr^{85} and Cs^{137} into the bottom sediments can also be defined in terms of a first order reaction by

$$C = C_0 e^{-pd},$$

where:

C = concentration of radionuclides at depth d ,

C_0 = concentration of radionuclides at interface,

d = depth of penetration, and

p = constant defined by the slope of the line.

4. The selectivity coefficients of Lake Austin sediments in the flowing system were found to be about 3.47×10^{-3} for K_{Ca+Mg}^{Sr} and about 3.35 gm/ml for K_{Ca+Mg}^{Cs} .

5. The range of fluctuations of Sr^{85} in various stream components was found to be larger than that of Cs^{137} . It appears that Sr^{85} is more readily released and resorbed by various stream components than Cs^{137} .

6. The relative uptake of Sr^{85} by aquatic plants was as follows:

Spirogyra sp. > Potamogeton sp. > Zannichellia sp. >

Myriophyllum sp. > Chara sp. > Cladophora sp.

The uptake of Cs^{137} exhibited the following order:

Cladophora sp. > Potamogeton sp. > Chara sp. > Spirogyra sp. >

Myriophyllum sp. > Zannichellia sp.

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CHAPTER I

INTRODUCTION

This study is concerned with the transport of Sr^{85} and Cs^{137} in a flume system. Similar studies using the radionuclides of Co^{58} and Zn^{65} were made simultaneously with the strontium and cesium investigations and are reported separately. (22,41) Reports related to this project are also available. (22,41,44)

Purpose

The objective of this research was to investigate the distribution and transport of radioactive strontium and cesium in a model river system. In addition, the purpose was to determine the influence of various environmental factors on the uptake and release of these two radionuclides.

Scope

The scope of this study included: (1) comparisons of the concentrations of Sr^{85} and Cs^{137} that were associated with suspended solids, sediments, aquatic plants, and the water phase; (2) measurement of the distribution of Sr^{85} and Cs^{137} in various phases of the aquatic system; (3) measurement of the penetration of radioactive Sr^{85} and Cs^{137} into the sediments; and (4) determination of the relationship between Sr^{85} and Cs^{137} and the aquatic system.

CHAPTER II
LITERATURE SURVEY

The disposal of radioactive wastes generally consists of the removal of the radionuclides by isolating them from the environment or diluting the waste to an acceptable level.⁽¹¹⁾ The dilution method can only be used for a few short-lived radioactive isotopes that are not significantly incorporated into the food chain. Usually, the removal of radionuclides from liquid wastes has been achieved by unit processes such as ion exchange, precipitation, sedimentation, and evaporation.⁽⁵²⁾

In streams radionuclides are accumulated in the biota, sediments, and suspended debris. Thus, there is the possibility that a waste having radionuclide concentrations below the permissible concentration for drinking water may be concentrated to significant levels by various components of a stream.⁽³⁷⁾

Sorption on Clays and Sediments

Sorption of radiostrontium and radiocesium from solutions by minerals may occur as a result of both adsorption and absorption. Adsorption is the uptake of a solute at the surface of the sorbent, whereas absorption is the penetration of a solute into the sorbent.⁽³³⁾ Adsorption and absorption are also classified as non-specific electrostatic adsorption and specific adsorption.⁽⁹⁾

Non-specific electrostatic adsorption occurs mainly in the region of the ionic double layer by attractive forces of an electrostatic nature. Non-specific adsorption is also termed "physical adsorption" since the adsorption is affected only by the surface charge density of the lattice, the valence of

the ion, and the concentration of electrolyte. Specific adsorption is a result of a strong chemical or geometrical interaction between the adsorbed ion and the surface of an adsorbing lattice. Specific adsorption is termed "chemical adsorption" since it is generally attributed to the presence of excess salts and valence forces.⁽⁵⁾

Physical sorption is believed to be the predominant method of sorption of isotopes by the coarser grained soils such as silts, sands, and gravels.⁽⁷⁵⁾ Ion exchange is a special type of chemical sorption in which ions in the solution replace ions held by the sorbent.⁽⁵⁾ Ion exchange is the predominant method of sorption of isotopes by the finer grained soils such as clays.⁽⁷⁵⁾

There are several isotherms used to express the sorption by a solid. The Freundlich isotherm, which is an empirical equation, is⁽⁵⁾

$$\frac{x}{m} = kC^n \quad (2-1)$$

where x , m , and C are the weight of sorbate, the weight of sorbent, and concentration of solute in the solvent at equilibrium, respectively. The constants, k and n , are determined by experimentation. The Freundlich formula has been found to apply to both physical and chemical sorption in dilute solution;⁽⁷⁵⁾ however, a long time may be required to reach an equilibrium condition.

Burt's isotherm takes into account time, t . In Eq. 2-2 the terms

$$\frac{x}{m} = kt^n \quad (2-2)$$

x , m , k , and n are the same as those used in Eq. 2-1.

For prolonged durations, Burt's formula may be modified into

$$\text{Log } \frac{\sigma}{\sigma-s} = kt^n, \quad (2-3)$$

where σ represents the saturation value of $\frac{x}{m}$ and s represents the value of σ at time t .

Another similar sorption equation is the Langmuir isotherm⁽⁵⁾ which is based on the amount of saturation occurring in a monomolecular layer of sorbed molecules on the sorbent surface, Eq. 2-4.

$$\frac{x}{m} = \frac{akC}{1+kC} \quad (2-4)$$

The term a is the weight of sorbed solute at saturation of the molecular layer. The Langmuir equation can apply to both physical and chemical sorption where the concentration of the sorbate at the liquid-solid interface is less than that required to cover the surface with a monomolecular layer.

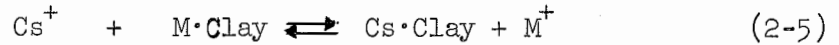
Many other isotherms have been obtained by numerous investigators. It has been found that the extent of sorption is a function of the specific nature of solid sorbent, the solute, the surface area available, the solvent, and the duration involved.⁽⁹⁾

Ion Exchange

Ion exchange is a type of chemical sorption particularly characteristic of clays, fine grained silts, and some special exchangers. This ability is caused by existence of replaceable cations and anions held in exchange sites. The replaceable ions have a charge opposite to the surplus charge within the exchanger structure, and these ions are taken into the exchange sites to compensate for the unbalanced electrical charge within the lattice of the exchanger. The exchangeable ions are also termed "counter ions."⁽¹⁹⁾ The reactions of ion exchange occur when the counter ions are displaced by other ions with the same charge.

Ion exchange reactions follow the law of mass-action⁽¹⁹⁾ and are stoichiometric and reversible. For example, the reaction between cesium and

a clay mineral containing a monovalent exchangeable cation may be expressed:



The equilibrium constant will be:

$$K_{\text{M}}^{\text{Cs}} = \frac{q_{\text{Cs}}}{C_{\text{Cs}}} \frac{C_{\text{M}}}{q_{\text{M}}}, \quad (2-6)$$

where q and C represent the respective cation concentration in the solid and liquid phases. Also, K_{M}^{Cs} is termed the selectivity constant or mass-action constant.

The distribution coefficient, K_{d} is defined as the ratio of the concentration of ionic species in solid and liquid phases; i.e., $K_{\text{d}} = \frac{q}{c}$.

The equilibrium constant therefore can be written

$$K_{\text{M}}^{\text{Cs}} = \frac{K_{\text{d}}^{\text{Cs}}}{K_{\text{d}}^{\text{M}}}. \quad (2-7)$$

In order to express the amount of radionuclides removed from solution by clay minerals or other exchangers, the distribution coefficient may also be written⁽⁴⁵⁾

$$K_{\text{d}} = \frac{f_{\text{s}}/M_{\text{s}}}{f_{\text{e}}/V}, \quad (2-8)$$

where f_{s} is the fraction of radionuclide activity sorbed to the solid phase and f_{e} is the activity remaining in the solution. M_{s} is the mass of exchanger and V is the solution volume.

The rate of ion exchange is dependent upon the diffusion of counter ions into and out of the structure of exchanger. Thus, the kinetics of ion exchange reactions differ from those of other chemical reactions.⁽¹⁹⁾

The kinetics of an ion exchange reaction depend upon the size of exchanger, extent of mixing, ionic concentration of the solution, temperature, and the ionic radius of the replaced ion. (1)

Cation Exchange by Clay Minerals: Since the capacity of an ion exchange material is a function of the number of fixed ionic sites that can enter into the exchange reactions, the exchange capacities of clay minerals depend upon the structures and chemical compositions of the minerals. (7) The most common crystalline clays are kaolinites, illites, montmorillonites, and vermiculites. The structures are composed of unit sheets consisting of octahedral and tetrahedral units arranged in layers. These clay minerals are frequently called three-layer clays. Usually, the octahedral unit consists of six oxygen or hydroxyl groups in an octahedron configuration with an atom of Mg, Al, or other metals in the center. The tetrahedral unit consists of four oxygens in tetrahedral arrangement usually with a silicon atom in the center. The unbalanced charges of the clays result from substitutions within the lattices. The usual substitutions are Al^{3+} for Si^{4+} or Mg^{2+} for Al^{3+} . (16)

The montmorillonites and vermiculites are expanding clays because the sheet-to-sheet spacings or C-spacings vary with the cations and water molecules present in the interlayer positions. The kaolinites and illites are non-expanding clays because the sheet-to-sheet C-spacings are fixed.

The expanding clays generally have the greatest exchange capacities. The cation exchange capacities of kaolinites and illites are only about 20 percent of those for the montmorillonites and vermiculites, because the ionic exchange sites of non-expanding clays are broken bonds around the

edges of the lattices and the replacement of hydrogens from the hydroxyl groups at the edges of the lattices.

The cation exchange capacities that have been found for crystalline clays are shown in Table 2-1.⁽¹⁶⁾ The magnitude of the ion exchange capacity is expressed in milliequivalents (meq) of exchangeable ions per 100 gm of exchanger at pH 7.

Table 2-1

Cation Exchange Capacities of Common Clays in Water

Clay	Exchange Capacity (meq/100 gm)
Kaolinite	3 - 15
Illite	10 - 40
Montmorillonite	80 - 150
Vermiculite	100 - 150

Anion Exchange by Clay Minerals: Most clay minerals, except some kaolinites and illites, have a smaller anion exchange capacity (AEC) than cation exchange capacity (CEC). The AEC results mainly from the substitution and absorption of the hydroxyl groups to the lattice edges. Generally, the AEC's of kaolinites and illites are of the same magnitude as the CEC's while the AEC's of montmorillonites and vermiculites are insignificant.

Factors Affecting Ion Exchange: The major factors affecting cation exchange are the selectivities of the exchange and exchanged cation, concentration of competing cations, pH values, time of reaction, particle

size of the exchanger, temperature, the nature of clay or counter ion, and the ionic or colloidal states of any contaminants.

In general the replaceability of a cation is dependent upon its position in the Hofmeister or lyotropic series;⁽⁴³⁾ i.e., exchangers tend to have greater selectivity for ions with higher valence, ions with smaller equivalent volume, polar ions that react strongly with fixed ionic groups in the mineral lattice, and ions participating least in complex formation.⁽⁹⁾

The magnitude of cation exchange decreases with decreasing pH because of the competition caused by the H^+ ion in the acidic range.⁽¹⁶⁾ The preference for particular ions by the clay minerals varies for different types of clays. For example, the selectivity order for various radionuclides by montmorillonites has been found to be $Sr^{85} > Cs^{137} > Co^{60} > Zn-Nb^{95}$, but the selectivity order for kaolinites has been found to be $Zn-Nb^{95} > Sr^{85} > Cs^{137} > Co^{60}$.⁽¹⁹⁾ Decreasing particle size increases the cation exchange capacity of non-expanding clays, such as the kaolinites and illites; however, the size of the mineral particles does not appreciably affect the CEC of the expanding clays. As temperature increases, the CEC of clay minerals decreases gradually, but within the temperature variation encountered in a stream, the effect of temperature is negligible.⁽¹⁶⁾ Organic matter sorbed by the soil increases the CEC because organic materials have an unusually high exchange capacity.

Biological Uptake of Radionuclides

Biological uptake of radionuclides by organisms may be achieved by sorption to the surface, engulfment, or metabolic processes. Elimination of radioactive materials in organisms may occur by surface exchange, excretion

through natural physiological channels, or through cell lysis after death. (26)

Biological uptake may lower the concentration of radioactive materials sorbed on sediments through metabolic competition. (18)

Biological uptake of radionuclides is affected by factors such as the trophic level of the food consumer, method of uptake, specificity of the organism for the isotope, pH value, temperature, light intensity, age and growth rate of the organisms involved, retention and elimination rate, rate of the reproductive processes, and the competition by other elements present.

The pH variations affect biological uptake indirectly because there is an optimum pH range for microbial and biological activity. Since temperature affects the rate of metabolism of microorganisms, plants, and cold-blooded animals, the seasonal differences in temperature will cause changes in the rate of radionuclide uptake. (26) Light intensity will affect the rate of radionuclide uptake since light intensity affects photosynthesis. (25)

The radioactive materials removed by the primary producers (the organisms in the first trophic level) are mainly in an ionized state, although a certain amount of particulate materials may be sorbed to surfaces. (25)

The degree of specificity for certain radionuclides by organisms varies from species to species. The concentrating ability of organisms is usually expressed by the concentration factor, K_c , which is

$$K_c = \frac{\text{concentration of activity in the organism.}}{\text{concentration of activity in the water}}$$

Bacteria have been found to possess the greatest concentrating ability among organisms. (26)

In an aquatic system, there are two main types of photosynthetic plants, the rooted or large floating plants and phytoplankton.⁽¹²⁾ The phytoplankton tend to concentrate the induced activity products to a greater extent than the fission products because the transition elements formed by fission have the ability to form complexes with the microplankton.⁽²⁸⁾ Furthermore, the phytoplankton are able to concentrate elements such as Co, Zn, and Fe to the extent necessary to meet the metabolic requirements of the higher aquatic organisms.⁽¹²⁾

Dispersion of Radioactive Nuclides in a River System

Because the percent uptake in a sorption reaction is dependent upon concentration,⁽²⁶⁾ the dilution of radioactive wastes brought about by diffusion and turbulent mixing processes affects the degree of uptake.⁽³⁸⁾ Also, the diffusion mechanisms are important in establishing radionuclide concentration in the sediments.⁽²⁵⁾

In turbulent streams, molecular diffusion, as represented by Fick's law, is insignificant in comparison with turbulent diffusion. However, turbulent diffusion is anisotropic and a simple coefficient cannot be used.

Gloyna and Patterson⁽³⁸⁾ found in flume studies that if $y = 0$ and $v = 0$, the solution of two-dimensional dispersion becomes

$$C = \frac{M}{4 \pi Z t \sqrt{D_x D_y}} \exp \left[-(x-Ut)^2 / 4 D_x t \right], \quad (2-9)$$

where;

M = mass of diffusant,

A = cross sectional area of flow,

Z = depth of flow,

$$D_x = \sigma_x^2 / 2T,$$

$$\sigma_T^2 = \frac{\sum_{i=1}^n C_i (t_i - T)^2}{\sum_{i=1}^n C_i},$$

U = mean velocity component along X axis,

C_i = concentration for each time increment,

T_i = time increment,

y = distance from source of flume in transverse direction, and

V = velocity component along the direction of y.

They also found that the long-term dilution of a mass of radionuclides instantaneously introduced into a stream can be represented by

$$C = M \cdot \exp [-(x-Ut)^2 / 4D_x t] / (A \sqrt{4 \pi D_x t}). \quad (2-10)$$

Thus, by flume studies and field measurements, the empirical formula for the longitudinal coefficient has been formulated by

$$D_x = 0.8 \exp (0.034 U \sqrt{A}). \quad (2-11)$$

Survey of Previous Uptake Studies

Previous studies have been made on the fates of some radionuclides in systems containing ion-exchange materials and in ecological systems. Most of these systems have not been of the continuous flow and controlled-environment type.

Uptake by sediments: Ion exchangers have been used as a means of disposal for radioactive wastes. ^(20,21) The most common exchangers used were clay minerals because of their abundance, durability, and low cost. The reported distributions of sorption capacity of bed materials from nine

rivers are shown in Table 2-2. (21)

Table 2-2
Sorptions Capacity of Bottom Sediments (%)

Range	<u>Bed Materials (%)</u>			<u>Suspended Materials (%)</u>		
	sand	silt	clay	sand	silt	clay
Average	25	45	31	10	21	68
Maximum	92	85	62	36	45	90
Minimum	4	3	2	1	8	52

The cation exchange capacities, CEC, of sand fractions were 0.5 to 18.9 meq/100 gm. Similarly, the CEC's of clays were 20.4 to 77.6 meq/100 gm. (24) The CEC of untreated sand was high when clay particles were encrusted on the surface of the sand.

The distribution coefficient, K_d , for strontium and cesium for various soils and minerals varied from 35 to 2,470 ml/gm and from 1290 to 100,000 ml/gm, respectively.

In the Clinch River study, (46) the CEC's of kaolinites and montmorillonites at the end of seven days of contact were essentially the same as after one hour. For illites, the CEC's were three and nine times greater after a seven-day contact period than after a one-day contact period. The Cs¹³⁷ sorption was relatively independent of pH, but Sr⁸⁵ sorption increased with increasing pH. (44,45)

Uptake of Sr⁹⁰ is affected by the concentration of sodium ions and total salt content; however, the presence of calcium will cause the greatest reduction in uptake. (44) Furthermore, the presence of stable isotopes of

the same element or other stable ions of the same group in the periodic table may influence the uptake of the radionuclides by sediments. (34,46)

Reynolds and Gloyna studied the transport of Sr^{89} and Cs^{137} in both freshwater and marine systems. (40) They found that the most important factors affecting the uptake of Sr^{89} and Cs^{137} by river and lake sediments were the CEC of sediments, the concentration of competing cations in water, and the mass-action coefficients for the system. The mass-action coefficients were relatively constant as long as the potassium concentration in the solution did not vary appreciably. The mass-action coefficients were relatively uniform for freshwater systems; however, this was not the case for marine waters. The average mass-action coefficients were:

$$\frac{K^{\text{Sr}}}{\text{Ca} + \text{Mg}} \approx 1.55$$

and

$$\frac{K^{\text{Cs}}}{\text{Ca} + \text{Mg}} \approx 5.7 \times 10^6 \text{ ml/gm.}$$

Because of the increase of competing cation concentration, some of the radionuclides in the river-borne sediments were released when the sediments were deposited in salt waters. The order of release was:

$$\text{Cs}^{137} > \text{Sr}^{85} > \text{Ru}^{103} > \text{Fe}^{59} > \text{Cr}^{51}.$$

The monovalent cations were more inhibitive to Cs^{137} uptake than the divalent cations, but the reverse was true for Sr^{89} uptake. The orders of the inhibition were as follows:

$$\text{K}^+ > \text{Na}^+ > \text{Ca}^{++} \approx \text{Mg}^{++} \quad \text{for } \text{Cs}^{137}$$

and

$$\text{Ca}^{++} > \text{Mg}^{++} > \text{K}^+ > \text{Na} \quad \text{for } \text{Sr}^{85}.$$

Reynolds and Gloyna show that the uptake of Sr^{89} and Cs^{137} is proportional to the concentration of sediment.

Jacobs⁽²³⁾ has found that Cs^{137} in a waste solution composed primarily of sodium and calcium salts is ordinarily sorbed by most clay minerals.

Tamura⁽⁴⁸⁾ et al. have studied methods of entrapping Cs^{137} in the interlayer positions of vermiculites. They concluded that the optimum Cs^{137} removal was obtained with an influent potassium concentration of 0.0005 M which gave a Cs^{137} K_d value of 366 ml/gm and a final CEC of 46.8 meq/100 gm. Also, they studied the CEC of heat-treated Gibbsite for Cs^{137} and Sr^{85} removal and found that the heat-treated Gibbsite had more affinity for Sr^{85} than Cs^{137} .

Biological Uptake: In the past few years, the biological uptake of radionuclides has been studied by many investigators.^(3,6,14) Some investigations involved the determination of the degree of decontamination of radioactive wastes achieved by biological systems while other studies investigated the amount of radioactive materials ingested by man through various food chains.

The biological uptake of Cs^{137} by various organisms has been reported.⁽²⁶⁾ Table 2-3 shows the concentration factors, K_c , of some plants and plankton. Algae have been reported to demonstrate the highest concentration factors. The uptake of aquatic plants was about 500 times that of rooted plants. The concentration of Cs^{137} in plants found in White Oak Lake at Oak Ridge⁽⁴⁸⁾ was considerably less than that found in

Table 2-3
 Concentration Factors of Cs¹³⁷ by Organisms

Organism	Concentration Factors, K _c	
	Experimental	Field
Green algae	400 - 4,000	1,200
Submerged plants	400 - 1,000	
Floating plants	250 - 600	70 - 240
Emergent plants	500 - 600	
Plankton	1,000 - 25,000	

the sediments. Some uptake data are shown in Table 2-4.

Table 2-4
 Uptake of Radionuclides by Various Plants

Plant Genus	Concentration (μμc/gm of oven dry wt.)		
	Cs ¹³⁷	Co ⁶⁰	Sr ⁹⁰
Festuca	1.60	1.42	0.38
Festuca (stem)	1.36	1.08	0.34
Polygonium	2.85	6.20	1.33
Tupatorium	1.50	1.99	0.38
Rumex	1.83	3.42	2.05

The concentrations of Cs¹³⁷ in plants such as Eloda, Myriophyllum, and Ceratophyllum are lower than concentrations found in some marine organisms,

Table 2-5.⁽³⁾

Table 2-5
Concentration Factors in Some Marine Organisms

Element	Concentration Factors (wet wt.-vol. basis)	
	Cs	Sr
Limacina	(0.2) \pm	--
Centropages	0.1 - 1	--
Calanus	0.1 - 1	0.8*
Ommestrepes	(0.1) \pm	(0.3)*
Saggita	--	70
Euphausia	--	(0.3)*

Values in parenthesis are for analyses made on closely related genera.

* Derived from Ca analysis and Sr/Ca ratio given by Chow and Thompson (1956).

\pm Based on analysis from K, assuming Cs/K ratio in the organism to be the same as that in water.

Burkholder⁽⁶⁾ has studied some plants from both fresh and marine waters that had been contaminated by fallout. Since all the submerged plants exhibited considerable radioactivity, it is believed that the radionuclides were removed from the water and not through the root system. Additional data are provided in Tables 2-6 and 2-7.

Table 2-6
Total β -Radiation in the Ash of Flowering Aquatic
Plants, Algae and Plankton

Organisms	No. of Samples	Beta Activity	
		Range (dpm)	Average (dpm)
Phanerogams (freshwater)			
Brasenia	1	310	310
Ceratophyllum	2	90 - 123	106
Elodea	2	120 - 399	259
Lemna	12	107 - 400	201
Myriophyllum	5	47 - 207	127
Nuphar	15	73 - 317	158
Nymphaea	7	137 - 506	238
Peltandra	14	43 - 600	235
Pontederia	8	106 - 1170	457
Potamogeton	3	80 - 340	170
Typha	2	56 - 247	151
Utricularia	3	242 - 1134	735
Wolffia	11	107 - 373	172
Phanerogams (marine)			
Zostera	10	23 - 164	78
Macro-algae (marine)			
Green	19	8 - 130	57
Red	12	60 - 332	167
Brown	12	93 - 240	156
Plankton and Micro-algae			
Marine	37	25 - 2193	466
Freshwater	21	20 - 750	215

Table 2-7
Total β and γ -Radioactivity in Representative
Aquatic Vegetation

Plants	Source	Beta and Gamma Activity	
		Total β (dpm)	Total γ (dpm)
Utricularia	Edgewood Pond, Saranac, N.Y.	625	1,230
Nymphae	Turtle Pond, Saranac, N.Y.	173	466
Vaucheria	Westwood Pond, Westwood, N.J.	323	874
Gracilaria	Long Island Sound, Conn.	139	143

Disintegrations per minute per gram of dried plants are based on a counting efficiency of β -radiation emitted from Potassium-40 and of γ -radiation from Cesium-137.

CHAPTER III

SAMPLE COLLECTION AND PROCEDURE

This chapter describes the model river system, sample collection methods, and analytical procedures.

Flume System

The model river system consisted of a metal flume; two large circular concrete tanks; and a pumping system. The rectangular flume was 200 ft long x 2.5 ft wide x 2 ft deep. The slope of the flume was adjustable through the range of zero to 0.006 ft/ft by using screw jacks. The details of the flume are shown in Figs. 3-1 and 3-2. The two reservoir tanks were 12 ft deep and each had a capacity of 500,000 gallons. One was used as a supply reservoir and the other as an effluent monitoring tank.

The supply pump was driven by a 10 hp, 860 rpm electric motor and was primed by a "wobble" pump. The capacity of the supply pump was about 1.71 cfs at a 35 ft head. The flow through the influent pipe was regulated by a gate valve and the depth of water in the flume was controlled by an adjustable weir at the outlet.

Aquatic Environment: The water in the reservoir was a mixture of well water from Balcones Research Center and treated city water. The supply water was hard since both sources had been in contact with the surrounding limestone formation. An analysis of the water is shown in Table 3-1.

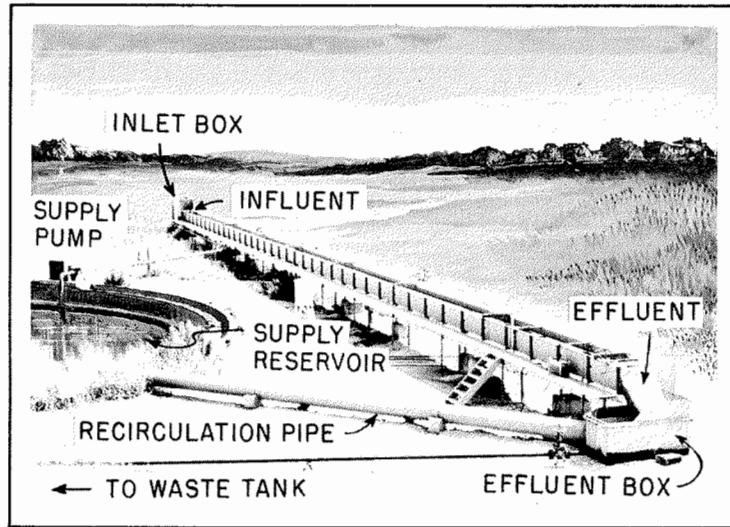
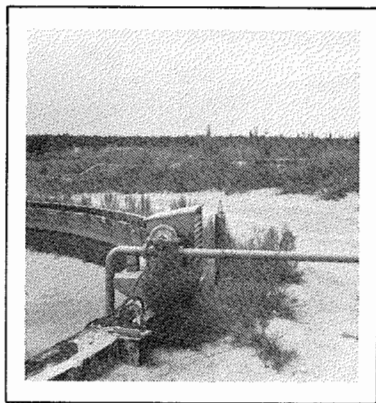
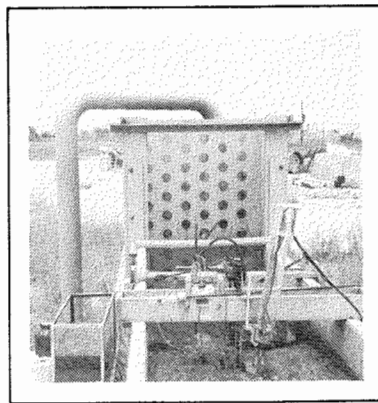


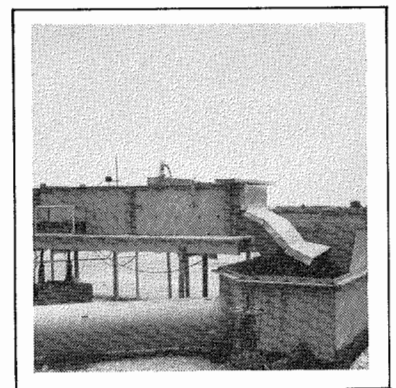
FIG. 3-1. RESEARCH FLUME



Supply Pump



Inlet



Outlet

FIG. 3-2 SUPPLY PUMP, INLET & OUTLET DEVICES ON RESEARCH FLUME.

Table 3-1

Typical Chemical Analysis

Item	Concentration (mg/l) (April 16)			Concentration (mg/l) (April 23)			Concentration (mg/l) (April 24)			Concentration (mg/l) (April 27)					
	Tank	Inlet	Mid	Eff	Inlet	Eff	Inlet	Eff	Inlet	Eff	Inlet	Eff	Input	Eff	
Suspended matter	--	--	--	--	94	60	40	72	124	20	72	70	74	70	
Turbidity as SiO ₂	5.6	9.2	7.8	9.2	7.8	8.6	5.2	5.6	6.4	5.9	5.2	10.1	7.8	7.8	
P (CaCO ₃)	20	20	25	25	15	15	20	10	15	15	22	17	20	17	
Alk. MO (CaCO ₃)	115	115	115	115	117	117	118	120	117	118	117	119	118	118	
Total Solids	1416	1341	1352	1512	1335	1500	1300	1430	1162	1180	1216	1425	1290	1340	
Non Volatile ASL	675	649	607	625	530	554	490	570	492	510	500	505	440	550	
Volatile matter	786	692	745	887	806	950	810	860	670	670	720	920	850	790	
Hardness (CaCO ₃)	850	825	825	840	825	845	845	850	815	840	840	840	845	850	
Free NH ₃ (N)	0.13	0.14	0.14	0.10	0.04	0.05	0.09	0.08	0.04	0.05	0.04	0.05	0.06	0.06	
Org. Nitrogen	0.5	0.5	0.4	0.4	1.1	1.3	1.9	2.0	1.1	0.3	1.0	2.4	2.2	2.1	
Na	70	69	70	70	69	70	--	--	70	69	69	69	69	69	
K	8.9	9.7	9.7	8.9	10.5	9.7	--	--	9.7	10.5	9.7	9.7	9.7	9.7	
Ca	84	86	84	87	92	92	92	93	92	93	92	93	92	92	
PO ₄	--	--	--	--	0.56	0.65	2.75	2.25	0.75	0.94	1.19	2.25	2.20	2.75	
Cl	607	607	612	612	604	612	605	607	612	607	617	608	615	612	
SO ₄															
Co															
					N e g l i g i b l e				t r a c e s						
					0.0025	0.0017	0.0017	0.0017	0.0017	0.0095	0.0025	0.0083	0.0033	0.0025	0.0042

Table 3-2

Results of Botanical Surveys

I. Macro-plants found growing in the research flume (April & May, 1964).

- **¹ Potamogeton sp. (3 types)
 ** Myriophyllum sp.
 * Echinodorus sp.
 ** Zannichellia sp. (2 types)
 ** Chara (a macro-alga)

II. List of algae collected from the flume. (Observation dates were: March 23; April 1; April 24; May 6; May 18.)

- (A) CYANOPHYTA (blue-green algae)
 (1) Spirulina sp. 3/23; 4/1; 4/24
 (2) Oscillatoria sp. 3/23; 5/6
 * (3) Merismopedia sp. 3/23; 4/1; 4/24; 5/6; 5/18
 (4) Gomphosphaeria sp. 5/6; 5/18
 (5) Lyngbya sp. 4/24
- (B) CHLOROPHYTA (green algae)
 order CHLOROCOCCALES
 (1) Cryptomonas sp. 3/23; 5/6
 * (2) Scenedesmus sp. (4) - 3/23; 4/1; 4/24; 5/6; 5/18
 * (3) Ankistrodesmus sp. 3/23; 4/1; 4/24; 5/6; 5/18
 (4) Selenastrum sp. 4/1
 (5) Dictyosphaerium sp. 4/1; 5/6; 5/18
 (6) Echinosphaerella sp. 4/1
 (7) Franceia sp. 4/1
 (8) Polyedropsis sp. 4/1
 order ULOTRICHALES
 * (1) Stigeoclonium sp. 3/23; 4/24; 5/6; 5/18
 order VOLVOCALES
 (1) Chlamydomonas sp. 3/23; 4/1
 (2) Pandorina sp. 3/23
 order CLADOPHORALES
 * (1) Cladophora sp. 3/23; 4/24; 5/6; 5/18
 (2) Coleochaete sp. 3/23; 5/6; 5/18
 (3) Aphanochaete sp. 3/23

¹ Number of asterisks indicates relative abundance (the more asterisks, the greater the frequency).

- order OEDOGONIALES
- * (1) Oedogonium sp. 3/23; 4/24; 5/6; 5/18
- order ZYGNEMATALES
- * (1) Spirogyra sp. 3/23; 5/6
- CHAROPHYTA
- ** (1) Chara contoria 3/23
- CHRYSOPHYTA
- * Class BACILLARIOPHYCEAE (diatoms)
 - (1) Gomphonema sp. 3/23; 4/1; 4/24; 5/6
 - (2) Cocconeis sp. 3/23; 4/1; 4/24; 5/6
 - (3) Synedra sp. 3/23; 4/1; 4/24; 5/6
 - (4) Navicula sp. 4/1
 - (5) Rhopalodia sp. 4/1
- III. List of organisms collected in water storage reservoir.
- * (1) Euglena (animal)
 - * (2) Scenedesmus sp. 4/1; 4/24; 5/6; 5/18
 - (3) Chlamydomonas sp. 4/1
 - * (4) Ankistrodesmus sp. 4/1; 5/6; 5/18
 - (5) Lyngbya sp. 4/24
 - (6) Navicula sp. 4/1; 4/24
 - (7) Spirulina sp. 4/1
 - (8) Cladophora sp. 5/6
 - (9) Stigeoclonium sp. 5/6
 - * (10) Daphnia sp. (animal) 4/1
- IV. Details of observation dates.
- (1) 3/23 = general collection from the flume - water and filamentous samples.
 - (2) 4/1 = 15 bucketsful of water from each of 10 stations located on flume and storage reservoir poured through plankton net. The samples were later cultured and the results observed on 5/6/64.
 - (3) 4/24 = slides 1 week old taken from stations on the flume and in the storage reservoir.
 - (4) 5/6 = observation of plankton samples cultured in BBM media taken on 4/1/64.
 - (5) 5/18 = observation of slides from stations located on the flume and in storage reservoir. These slides were over 3 weeks in growth age.
-

Table 3-3

Animals Taken From the Flume Ecosystem

Protozoa
Ciliophora
Ciliata
Holotrichida
Paramecidae
<u>Physalophrya</u> sp.
Coelenterata
<u>Hydra hymanae</u>
Annelida
Oligochaeta
Naididae
<u>Chaetogaster</u> sp.
Arthropoda
Crustacea
Cladocera
<u>Moinodaphnia macleayii</u> King
Ostracoda
Copepoda
Calanoida
Insects
Odonata
Coenagrionidae
Ephemeroptera
Oligoneuriellidae
Diptera
Chironomidae
<u>Chironomous</u> sp.
Mollusca
Gastropoda
<u>Lymnaea columella</u> Say

The sediment in the flume was taken from Lake Austin and was placed in the flume at depths of about 10 to 15 cm. X-ray diffraction showed the Austin Lake sediment to be composed of the following minerals:

<u>Mineral</u>	<u>Approximate Content (%)</u>
Calcite	50
Quartz	10
Dolomite	8
Clay mineral	30
Organic material	2

The clay fractions contained illites, vermiculites, and other degraded clays. The cation exchange capacity, as determined by the tagged cesium method, was about 29 meq/100 gm of sediment.

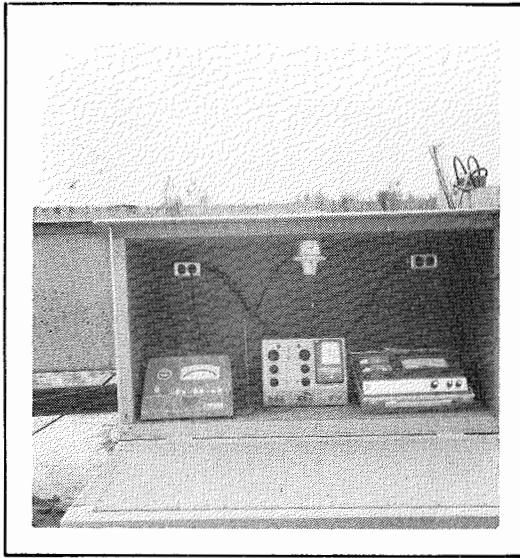
The aquatic plants in the flume were transplanted from several rivers, and the ecosystem had reached a quasi-equilibrium condition before the experiment was started. The major plants and animals in the flume at the time of the release of radionuclides are shown in Tables 3-2 and 3-3. During the first several weeks of the experiment, the dominant species were Myriophyllum, Chara, and Potamogeton, but at the end of the experiment Cladophora, Spirogyra, and Myriophyllum predominated. Of the plants mentioned above, only Cladophora and Spirogyra were floating phytoplankton.

Equipment Used: Equipment used during the experiments is listed in Table 3-4 and shown in Fig. 3-3.

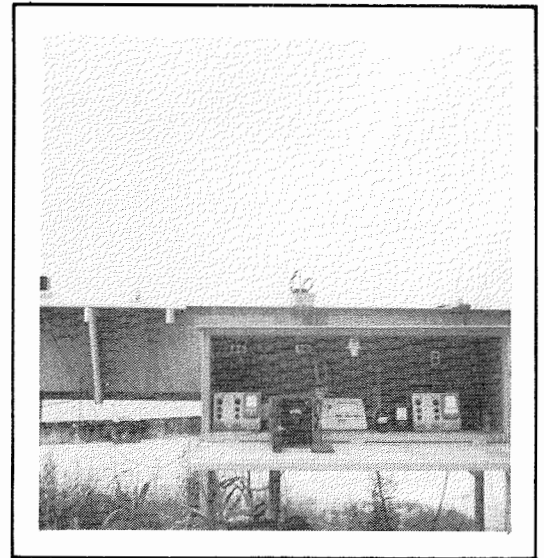
Hydraulic Characteristics: The diffusion and dispersion mechanism of flow was determined by use of Rhodamine B and a Turner Fluorometer. The calibration curve is shown in Fig. 3-4. For this test, the flow was 0.847 cu ft per min (24 liters per min). Based on an average cross sectional area of

Table 3-4
Instrumentation of Model River

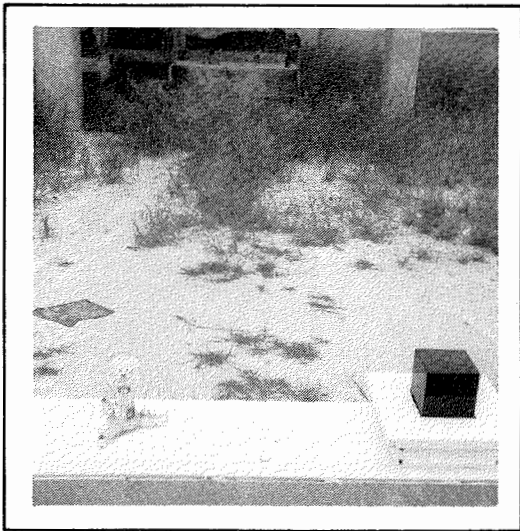
Equipment and Manufacturer	Location (Distance from Inlet)	No. Used	Measurement
Electronic Thermometer (Research Associates Model F7-2)	5' and 195'	2	Temperature
Galvanic Oxygen Cell	5' and 195'	2	Dissolved Oxygen
Expanded Scale pH Meter (Beckman, Model 76)	5' and 195'	2	pH
Zeromatic pH Meter (Beckman, Model 9600)	100'	1	ORP
Pyroheliometer (Eppley, 180)	100' (One with filter and one without)	2	Light Intensity
Dual Channel Record (YS1, Model 81)	5', 100', 195'	3	Continuous record for pH, D.O. and Light Intensity
Conductivity Cell (U-type with Self- contained Line Operator Portable Conductivity Bridge	100'	1	Conductivity
Fluorometer (Turner Model III)	195'	1	Dye
Scintillation Spectrometer (Nuclear Chicago)	195'	1	Evaluation of Radioactivity
Rectilinear Recorder	195'	1	Continuous Record of the Radioactivity in Solution
512-Channel Gamma Spectrometer (RCL)	Lab	1	Radioactivity - gamma (Gross)



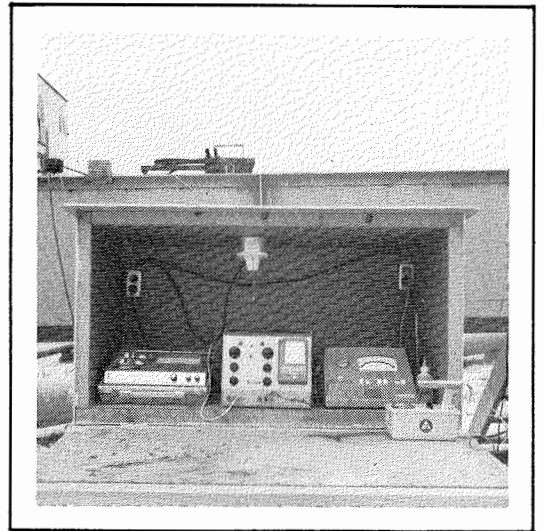
Inlet: pH Meter, Recorder,
Galvanic Cell Oxygen Analyzer



Midlength: Pyr heliometer,
Conductivity Cell, Zeromatic
pH Meter, 2 Electronic
Thermometers and Recorder



Midlength :2 Pyr heliometers



Outlet: Galvanic Cell Oxygen
Analyzer, Recorder, pH Meter

FIG. 3-3. INSTRUMENTATION.

2.5 sq ft (1 ft depth), the calculated mean velocity was 0.0057 ft per sec (0.342 ft per min). The slope of the flume was 0.00021 ft/ft.

One hundred mg of Rhodamine B was dissolved in one liter of water and released instantaneously at the mid-depth near the inlet. Samples were taken at the top, mid-depth, one-third of the depth from the bottom, and at the bottom at stations which were located 5, 50, 100, 150, and 195 ft from the release point. The fluorometer was located near the outlet and a continuous flow sampling apparatus provided a means of recording the concentrations directly.

Using the distribution of the dye along the flume as shown in Figs. 3-5 through 3-8, the concentration-time relationship can be described. By comparing the time-concentration curves of the 5-ft station and 50-ft station, it can be seen that thorough mixing was observed at the 50-ft station. This was caused by the presence of the rooted plants and phytoplankton.

The mean flow-through times and dispersion coefficients calculated by the dye release study are shown in Table 3-5. The dispersion coefficient varied from 0.022-0.059 ft²/sec, which agreed fairly well with prior investigations. (38)

Sampling Procedures

Water samples of 100 ml were taken at three different depths in the channel: namely, 2, 6, and 10 inches from the surface. The sampling apparatus is shown in Fig. 3-9. Water samples were refrigerated until duplicated aliquots of 10 ml could be transferred to aluminum planchets for processing and radioactivity analysis. The concentrations of Sr⁸⁵ and Cs¹³⁷ associated with the suspended materials were determined by the difference in the counts of the

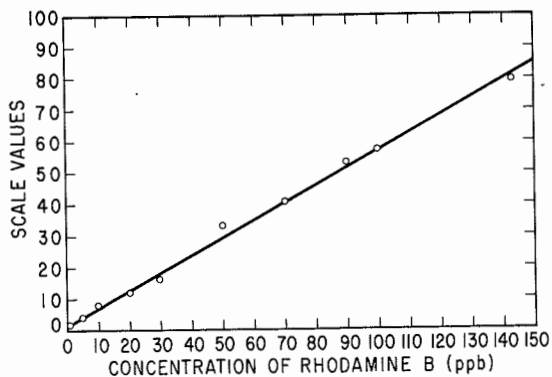


FIG. 3-4 LINEARITY OF RHODAMINE B

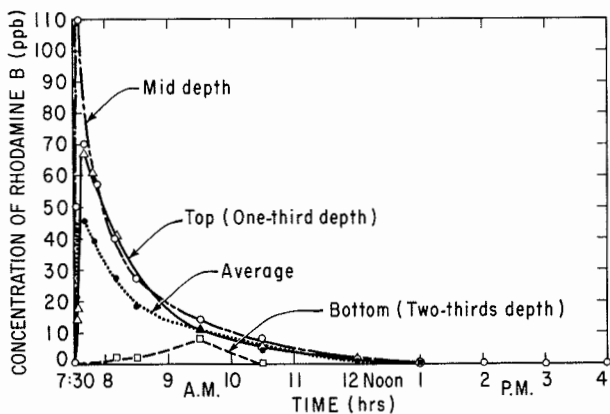


FIG. 3-5. VERTICAL DISTRIBUTION OF RHODAMINE B AT 5 FT. STATION

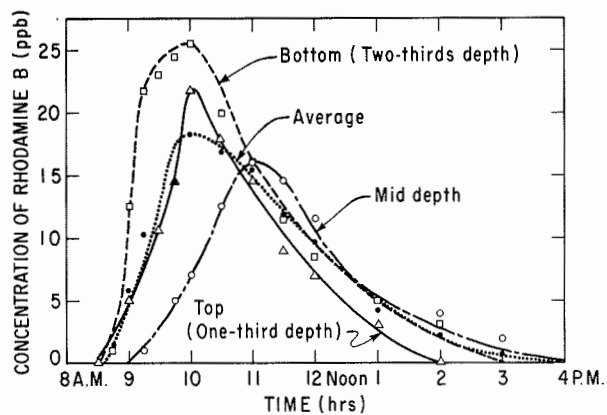


FIG. 3-6. VERTICAL DISTRIBUTION OF RHODAMINE B AT 50 FT. STATION

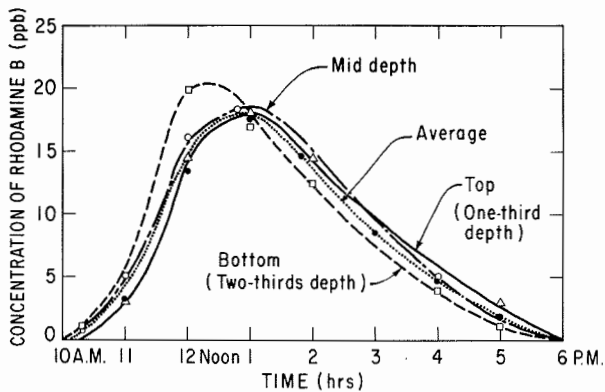


FIG. 3-7. VERTICAL DISTRIBUTION OF RHODAMINE B AT 100 FT. STATION

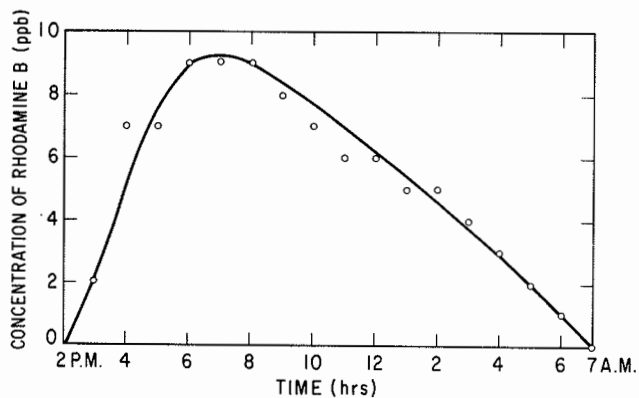


FIG. 3-8. CONCENTRATION OF RHODAMINE B AT EFFLUENT (200 FT. STATION)

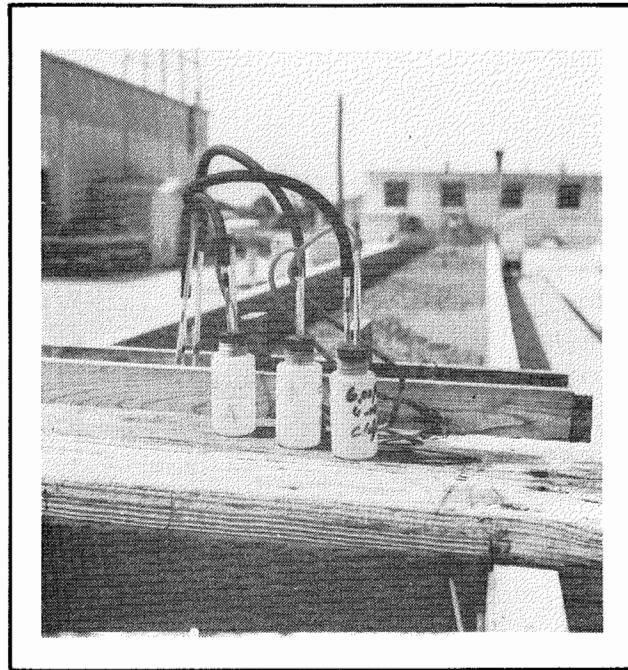
liquid phases before and after centrifugation (3000 G for 10 minutes).

Table 3-5
Mixing Characteristics

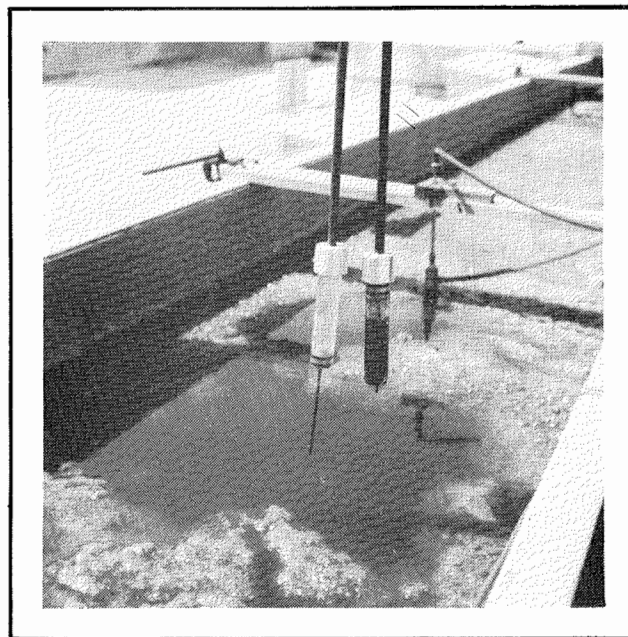
Station (Distance from inlet in ft)	T (hr)	σ_T^2 (hr ²)	σ_x^2 (ft ²)	D_x (ft ² /sec)
5	1.16	1.036	436.23	0.053
50	3.50	1.654	696.45	0.028
100	5.50	2.106	886.77	0.022
200	13.67	13.640	5743.39	0.059

- T = Mean flow through time (hr)
 D_x = Dispersion coefficient along X-axis (ft²/sec)
 σ_T^2 = Variance in time units (hr²)
 σ_x^2 = Longitudinal variance in length units (ft²)

The sediment sampler consisted of a lucite tube, an aluminum piston, and steel sample holder as shown in Fig. 3-9. The sediment samples were collected by forcing the lucite tube into the sediment. The piston in the tube helped to keep the core intact as the sampler was removed from the flume. After collection, both the sample and the tube were frozen. Then the top one inch of the frozen sample was pushed out and cut off. This section of sample was ashed at 600°C for 15 minutes, cooled, and from this material almost 3 gm, representing an equivalent depth of 0.7 cm, were transferred to an aluminum planchet for counting.



Water Samplers



Sediment Samplers

FIG. 3-9. SAMPLERS.

The plant samples were collected individually and washed with water; approximately one half of the sample was washed a second time in a weak solution of citric acid. In processing, the plant samples were ashed at 600°C, and a 100-mg portion was placed in a planchet for counting.

Counting Procedures

Since all the samples contained Sr⁸⁵, Cs¹³⁷, Zn⁶⁵, Co⁵⁸, and an impurity of Co⁶⁰, the multi-channel gamma spectrometer incorporating a four-inch diameter thallium-activated, NaI crystal in a shielded cabinet was used. The size of the cavity in the shield was 24 x 24 x 27 inches. The cabinet walls, top, and floor consisted of four inches of lead that were interlined with 0.75 inch magnesite, 0.063 inch copper, and 0.125 inch aluminum plates. All data were punched on paper tape and typed on regular printout paper.

In order to minimize the "drift" of the gamma spectrometer, a calibration test with a Co⁶⁰ standard was made during each counting period. The selected energy span was 2 Mev for the 256-channel group size. Co⁶⁰ peaks of 1.172 and 1.333 Mev were adjusted to channels 150 and 170. The values of the contribution of each isotope are reported in Chapter IV.

The counting efficiencies for Sr⁸⁵ and Cs¹³⁷ were 15.4 and 17.5 percent. Calibration was accomplished by counting Sr⁸⁵ and Cs¹³⁷ standards with known concentrations of the radionuclides. All the data were corrected for counting efficiency, background, absorption, decay, and contribution from other radionuclides. The correction coefficients for self-absorption were obtained by counting the same known concentration of Sr⁸⁵ and Cs¹³⁷ associated with different weights of sediments. Correction coefficients for the plant samples were 1.0 for Sr⁸⁵ and 0.99 for Cs¹³⁷. For sediment samples the

correction coefficients were 0.97 for Sr^{85} and 0.99 for Cs^{137} . The geometry remained the same for all samples.

Release Studies

In order to effect a release of radionuclides from the plants, 105 ft of the flume from near the influent was covered with boards on July 29, 1964. Water samples and sediment samples were taken at 50, 100, 150, and 195 ft from the inlet twice a day for the first week after the flume was covered, and plant samples were taken at a point 175 ft from the inlet once each day. After the first week, the sampling frequency decreased to once every two days. On August 17, 1964, the flow of the flume was stopped in an attempt to produce an anaerobic environment and thereby provide some information relative to release and changing environments.

In general, the collection and analyses program remain similar to the previously discussed procedures. Since Myriophyllum and Cladophora were predominant, only these two species were studied in detail.

CHAPTER IV
DATA ANALYSIS

The results of the water, sediments, and plant samples are described in detail in this chapter. The environmental factors are reported as they related to this study. However, no correlations were attempted since this effort is part of another study.

Description of the Gamma Spectrum

The gamma spectrum of each sample consisted of the photopeaks of Sr^{85} , Cs^{137} , Co^{58} , (Co^{60} impurity), Zn^{65} and Co^{60} . The radionuclides were released in the model river on April 24, 1964.

A typical gamma spectrum is shown in Fig. 4-1. The energy of the gamma ray for Sr^{85} is 0.51 Mev, that of Cs^{137} , 0.66 Mev. Because of pair production and backscattering, contributions from the higher energy isotopes of Co^{58} and Zn^{65} were made to the photopeak values of Cs^{137} and Sr^{85} . The photopeak of the background usually fell below an energy level of 0.3 Mev. Contributions from higher energy sources were calculated with the aid of a CDC 1604 computer. These coefficients are listed in Table 4-1.

In order to check the drift of the gamma spectrometer, a Co^{60} standard was used. The Co^{60} peaks of 1.172 Mev and 1.333 Mev were adjusted to fall in channels 150 and 170, respectively. As a consequence, the photopeaks of Sr^{85} and Cs^{137} were positioned in channels 66 and 85, respectively. The half-photopeak channel is the channel with one-half of the counts of the maximum channel in the photopeak. By summing up the counts of all channels between the two, half-photopeak channels of each isotope, the total counts for that

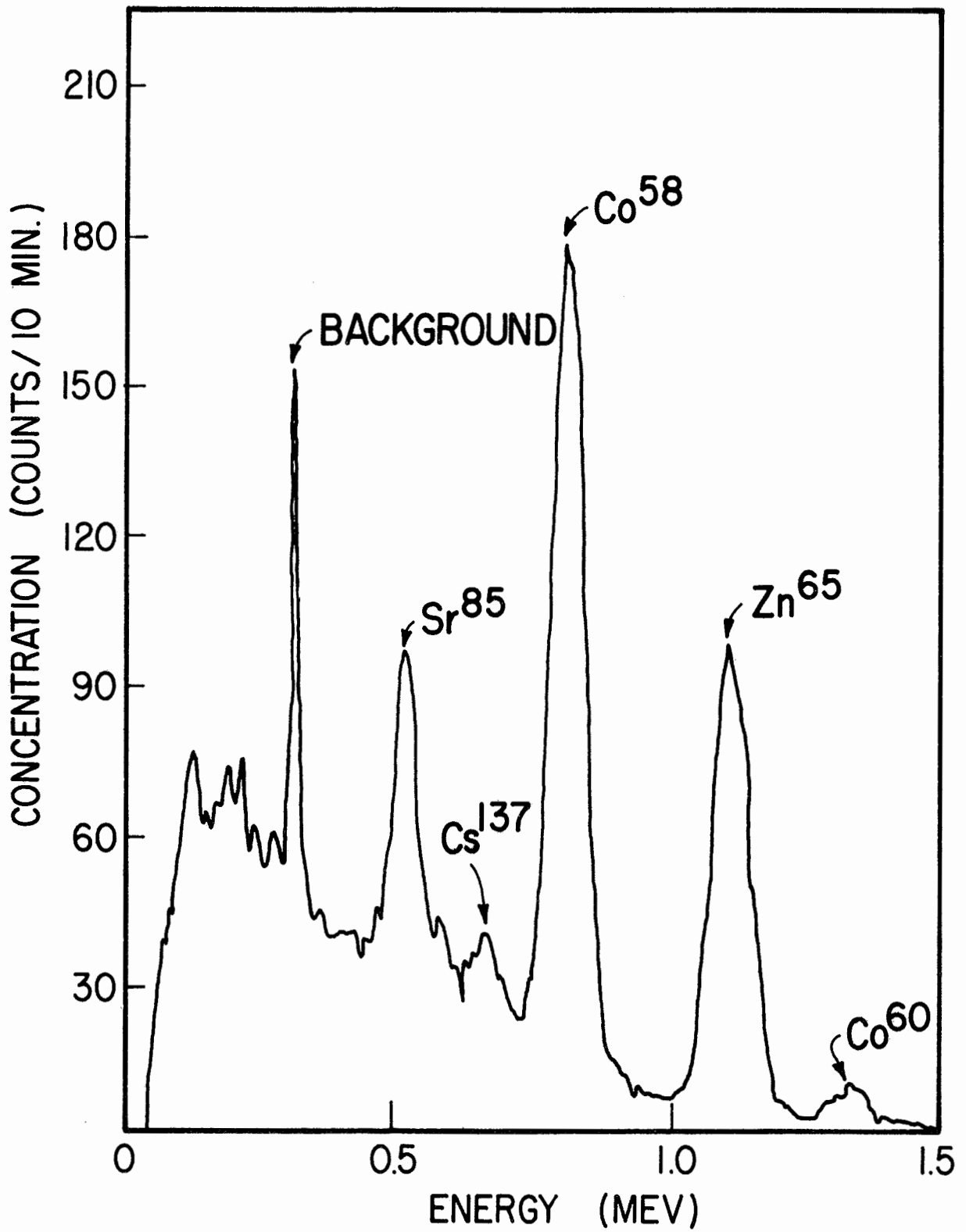


FIG. 4 - I. SPECTRUM FOR GAMMA SPECTROMETER

isotope in the spectrum were obtained.

A series of standards of various known concentrations of Sr^{85} and Cs^{137} were used to calibrate the spectrum. After correction for background, the counting efficiency was 15.4 percent for Sr^{85} and 17.5 percent for Cs^{137} . Calibrated with standards of each radionuclide used in this study, the channel range for the summation of counts was from channel 64 to channel 71 for Sr^{85} and from channel 82 to channel 90 for Cs^{137} .

Table 4-1

Contribution Coefficients

Contributing Isotopes	Influenced Isotopes				
	Sr^{85}	Cs^{137}	Co^{58}	Zn^{65}	Co^{60}
Sr^{85}	1.00000	0.0066	0.00738	0.00843	0.00659
Cs^{137}	0.03651	1.0000	0.00637	0.00101	0.00111
Co^{58}	0.27400	0.0070	1.00000	0.16000	0.04100
Zn^{65}	0.15800	0.1120	0.16010	1.00000	0.02610
Co^{60}	0.21700	0.2526	0.31810	0.60000	1.00000

Because of the necessity for the constant terms used in the simultaneous equations used in solving for the true counts for each isotope, the areas between the half-photopeaks under the spectrum for each of the photopeaks for Co^{58} , Zn^{65} , and Co^{60} were required. By analyzing various concentrations of standards for each isotope, the channel range was determined from channels

100 to 109 for Co^{58} , from 137 to 148 for Zn^{65} and from 164 to 175 for Co^{60} (2nd peak).

Data Handling

The computer program was written for the general use of gamma spectrometer data. The program can be used to analyze 50 unknowns simultaneously provided the proper coefficients for the simultaneous equations are available.

In order to save computer time, the input data were punched on Friden perforated tape after counting. On the paper tape, the binary number system was used and each channel was furnished with space for six digits. This information was then condensed on magnetic tape for reading into the computer.

Each paper tape usually contained 20 spectra which were grouped according to the counting date, the sample content, and the counting time. The background spectrum of the same counting date was attached to the beginning of each roll.

The Subroutine Program CON(IABC) was designed to change the binary number system on the magnetic tape into digital numbers. CON(IABC) was also programmed to pick out and eliminate erroneously punched spectra.

Mathematical Interpretation of the Program

In order to give a clearer interpretation, five linear equations were set to solve the true counts of each isotope involved in the flume study. The solutions were obtained by use of Gauss's method of elimination with row pivoting and back substitution. The matrix condition parameter was chosen to be 0.000001. The contribution coefficients used in these linear algebraic equations are shown in Table 4-1. The elements in the first row

were for Sr^{85} , the 2nd row for Cs^{137} , 3rd row for Co^{58} , 4th row for Zn^{65} , and 5th row for Co^{60} . Each element in the column vector was the sum of the counts under the spectrum between the two half-photopeak channels. The elements in the column vector were, from top to bottom in order, Sr^{85} , Cs^{137} , Co^{58} , Zn^{65} , and Co^{60} .

The coefficients may be arranged as follows:

a = the coefficients in the simultaneous equations

and

b = the constant terms in the equations.

$$A = \begin{vmatrix} a_{11} & a_{12} & a_{13} & a_{14} & a_{15} \\ a_{21} & a_{22} & a_{23} & a_{24} & a_{25} \\ a_{31} & a_{32} & a_{33} & a_{34} & a_{35} \\ a_{41} & a_{42} & a_{43} & a_{44} & a_{45} \\ a_{51} & a_{52} & a_{53} & a_{54} & a_{55} \end{vmatrix} \quad B = \begin{vmatrix} b_1 \\ b_2 \\ b_3 \\ b_4 \\ b_5 \end{vmatrix}$$

Matrix A was triangularized by adding multiples of one row to another row and subtracting the value of $\frac{a_{i1}}{a_{11}}$ (a_{ij}) from row i where,

$$i = 2, 3, 4, 5$$

$$j = 1, 2, 3, 4, 5.$$

Then the elements in column 1 below the principal diagonal were eliminated. By the same process of pivoting, the elements below the diagonal in columns 2, 3, and 4 were also eliminated.

The process of addition of multiples of rows is also applied to vector B. Finally, a new matrix F is formed by combining the triangularized matrix A and vector B as follows:

$$F = \begin{vmatrix} f_{11} & f_{12} & f_{13} & f_{14} & f_{15} & f_{16} \\ 0 & f_{22} & f_{23} & f_{24} & f_{25} & f_{26} \\ 0 & 0 & f_{33} & f_{34} & f_{35} & f_{36} \\ 0 & 0 & 0 & f_{44} & f_{45} & f_{46} \\ 0 & 0 & 0 & 0 & f_{55} & f_{56} \end{vmatrix}$$

Back substitution was then used where,

$$X_i = \frac{f_{i6} - S}{f_{ii}} \quad i = 1, 2, 3, 4, 5.$$

and when

$$\begin{aligned} i &= 5, & S &= 0 \text{ and when} \\ i &\neq 5, & S &= \sum_{k=i+1}^5 f_{ik} X_k \end{aligned}$$

According to the solutions from simultaneous operations, the disintegrations per minute for each isotope in the sample were calculated by the following formula:

$$DPM = (X_i) \exp(0.693 t/Tr)/E/M/Ab,$$

where

DPM = Disintegration per minute of a certain isotope for the content in planchet,

X_i = Solutions of simultaneous equations,

t = Time interval between the counting date and date of release (April 24, 1964) in days,

Tr = Half-life of radioisotope decay in days,

E = Efficiency of gamma spectrometer, calibrated by internal standards,

- M = Counting time of sample in minutes, and
 Ab = Self-absorption coefficient of each sample.

Arguments in the Program

- 1) N - Number of isotopes to be analyzed on each spectrum
- 2) MMN - Number of rolls of paper tape to be analyzed in the batch
- 3) I - Order for isotopes to be analyzed, such as 1, 2, 3, . . .
- 4) KJJ - Channel number of lower limit for summation range of each isotope derived from standards
- 5) KJI - Channel numbers for photopeak of each isotope
- 6) KII - Channel number of upper limit for the summation range of each isotope derived from standards
- 7) ISO - Name of each isotope, such as SR (85), CS (137)
- 8) EFF - Efficiency of counting by the RCL Gamma Spectrometer for the specific isotope
- 9) DECAY - Half-life of specific isotope in days
- 10) FRAC (I,J) - Contribution coefficient on a specific isotope I, from another isotope J
- 11) CNN - Counting time for background in minutes
- 12) AMM - Counting time for samples in minutes
- 13) DAYS - Number of days between release date and counting date
- 14) BOX - Arbitrary catalog number to identify the roll of paper tape
- 15) ABS - Self-absorption coefficient for typical sample
- 16) JJJ - Net counts of each channel after stripping out the corresponding background counts

- 17) IRLM - Order number of spectra in each paper tape roll
- 18) IJK - Total summed counts between the two summation limits
- 19) A - Total summed counts between the two summation limits
in real numbers
- 20) IX2 - Check to determine if spectrometer has drifted excessively
- 21) IDELT¹ - Number of channels between two summation limits derived
from standard of specific isotope
- 22) IDELT² - Number of channels between two summation limits
figured from sample spectrum
- 23) F - The matrix array for the coefficients and constant term
of simultaneous equations
- 24) LKL - Error flag (fixed-point)
1 indicates no error
2 indicates that the equations are inconsistent or
dependent
- 25) IKI - Channel number of photopeak of certain isotope in
spectrum
- 26) COUNT - Solutions of simultaneous equations
- 27) DIS - Radioactivity of specific isotope in the sample in
disintegrations per minute
- 28) I2 - Counts for half-peak-value

The printout and Fortran flow chart are provided in Appendixes A and B.

Analyses

Generally, as Sr^{85} and Cs^{137} are transported downstream the radio-nuclides are transferred from the aqueous phase to the bottom sediments and plants and then back into solution.

Analyses of Water Samples: Although the aquatic plants in the flume sorbed some of the Sr^{85} and Cs^{137} during the first day following release, the fraction of Sr^{85} and Cs^{137} in the aqueous phase remained as high as 88 percent and 93 percent, respectively. The relative distributions of Sr^{85} and Cs^{137} in suspended solids were 12 percent and 14 percent, respectively.

Time-Concentration Relationships: The relative concentrations of Cs^{137} and Sr^{85} in the aqueous phase of the flume at stations 5, 50, 100, 150, and 195 ft from release point are presented in Figs. 4-2 and 4-3. The average value was derived from sample data obtained from the three different levels at each section. The data show that the amount of Cs^{137} and Sr^{85} along the length of the flume decreased very rapidly after the first day following release. During the following three months, the concentrations of Cs^{137} in the aqueous phase fluctuated between 0.5 to 4.0 dpm/ml. The concentrations of Sr^{85} fluctuated between 0.3 and 8.0 dpm/ml.

Since the rapid change in the concentration of both Cs^{137} and Sr^{85} in the aqueous fraction occurred during the first two days following release, a more detailed study was made of the vertical distribution of the radio-activity during this period. The results from this investigation are shown in Figs. 4-4 and 4-5. It appears that the areas under the curves differ from one station to another. For Sr^{85} the areas under the curves for stations 50, 100, 150, and 195, respectively, were 43, 29, 25, and 21 percent of the area derived from the data collected at station 5, i.e., five feet

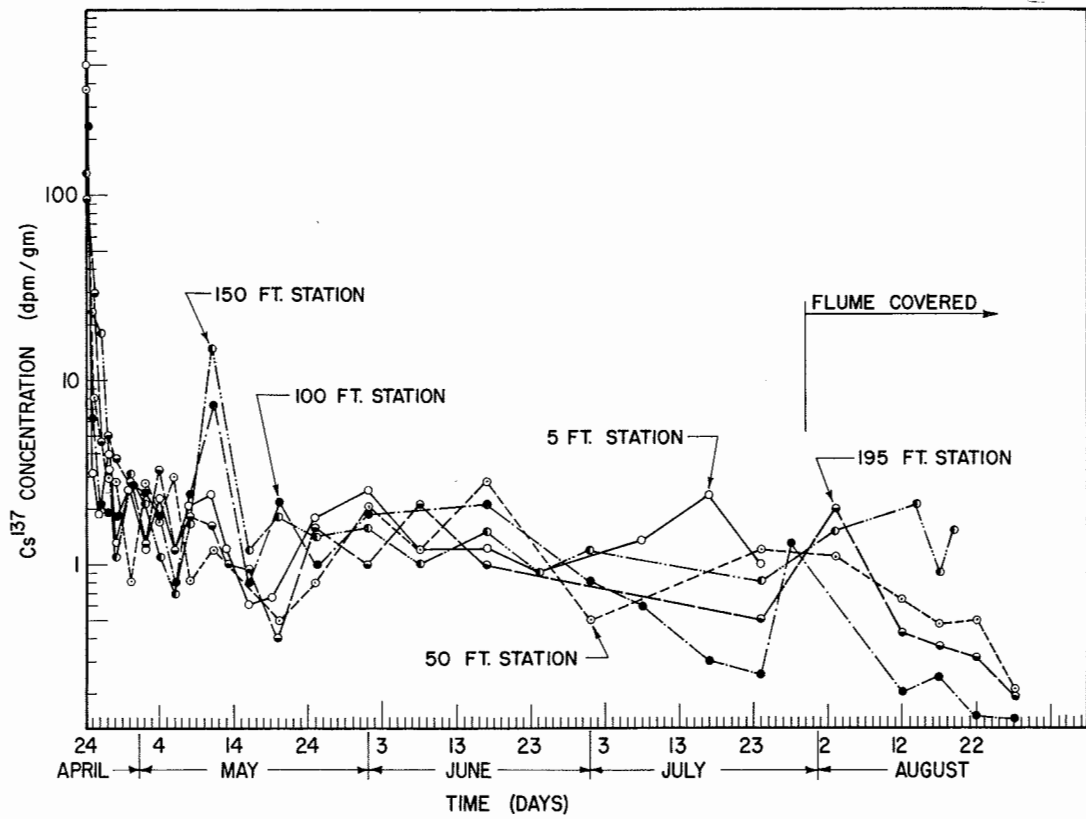


FIG. 4-2. LONGITUDINAL DISTRIBUTION OF Cs^{137} (WATER)

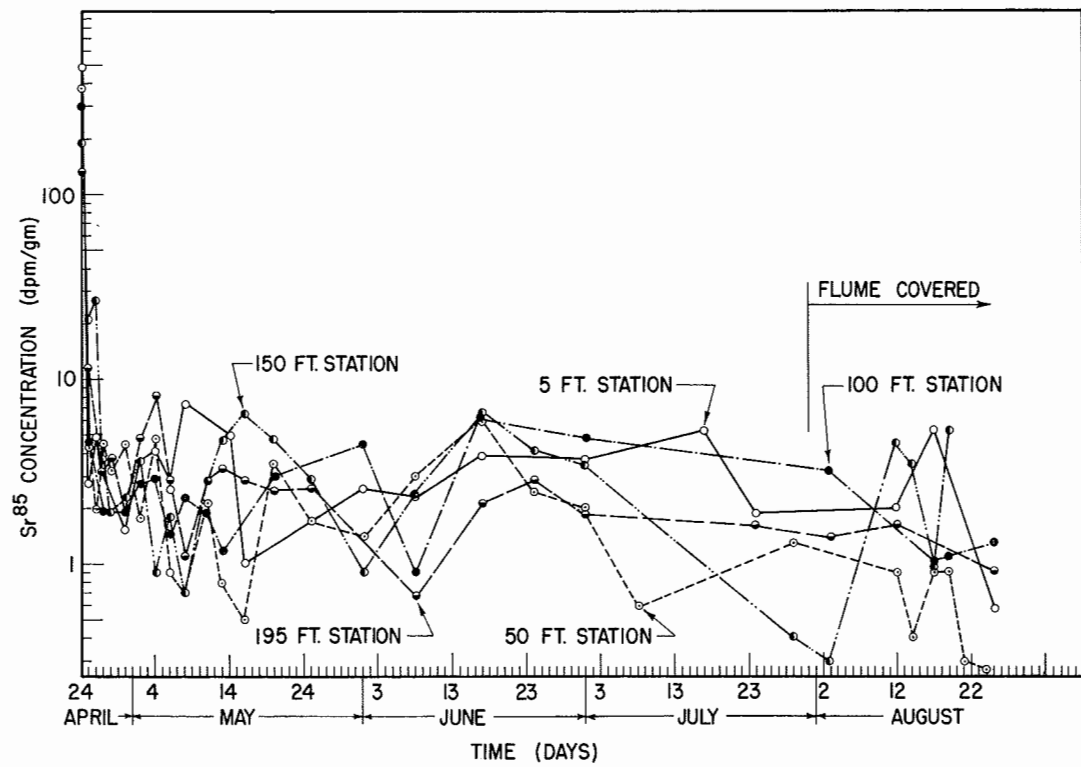


FIG. 4-3. LONGITUDINAL DISTRIBUTION OF Sr^{85} (WATER)

from the entrance. For Cs^{137} , the areas under the curves developed at the 50-, 100-, 150-, and 195-ft stations respectively, were 52, 40, 38, and 36 percent of the area under the curve for station 5. Notably this percentage decreased along with increased distance from the release point. This decrease in percentage characterizes the existence of sorption of the radionuclides by the aquatic plants and sediments that were present between the stations.

From the time-concentration curves and the discharge through the flume, an estimation of the total mass of Cs^{137} and Sr^{85} passing each station at any time can be computed. The cumulative quantities of Cs^{137} and Sr^{85} passing each station during the first week following the release are shown in Tables 4-2 and 4-3.

Table 4-2
Longitudinal Transport of Cs^{137} in the Flume Water

Days After Release	Percent of the Total Released Cs^{137} Carried in the Water				
	5-ft Station	50-ft Station	100-ft Station	150-ft Station	195-ft Station
1	61.2	44.7	32.0	25.7	18.4
2	65.0	49.2	36.3	31.1	25.6
3	69.3	55.1	40.6	35.4	28.0
4	72.4	58.6	44.2	39.7	30.8
5	76.6	61.4	48.4	41.2	34.4
6	80.2	64.1	51.0	44.6	36.6
7	84.0	67.1	54.0	47.1	38.0

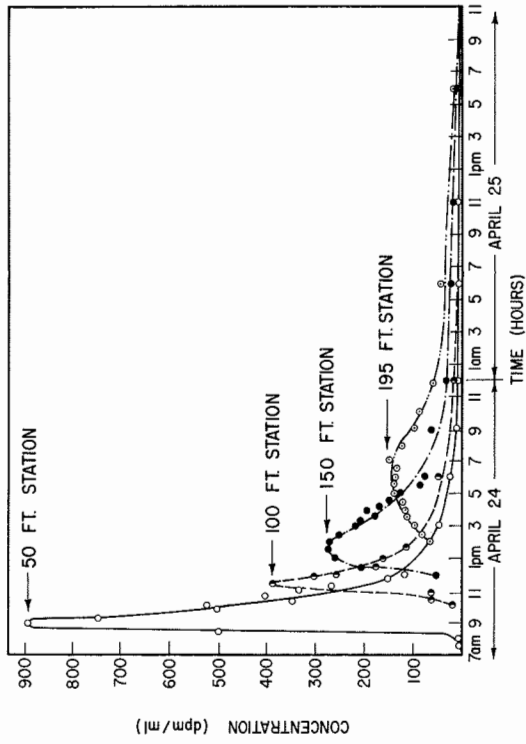


FIG. 4-4. VERTICAL DISTRIBUTION OF Cs^{137} (WATER)

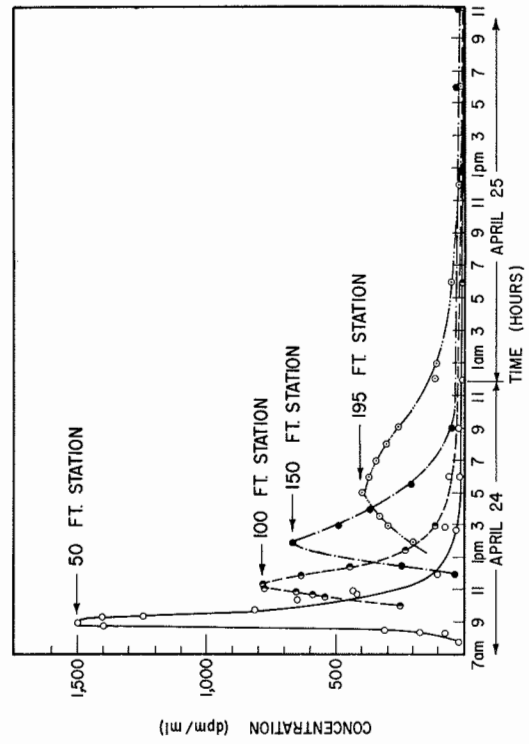


FIG. 4-5. VERTICAL DISTRIBUTION OF Sr^{85} (WATER)

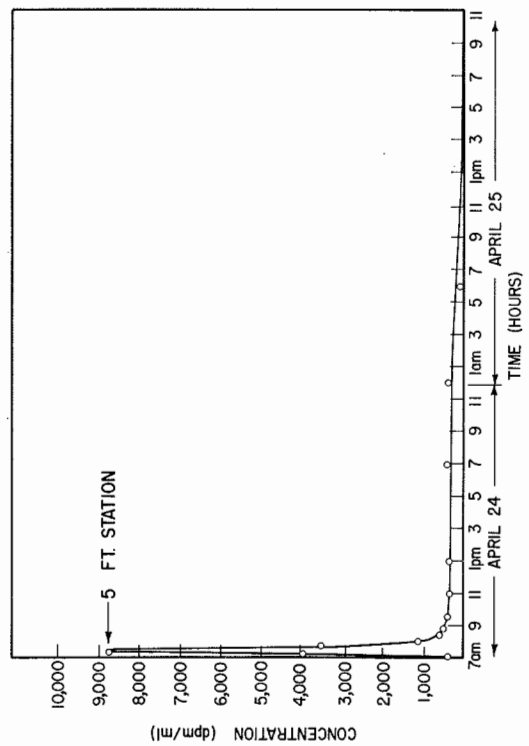
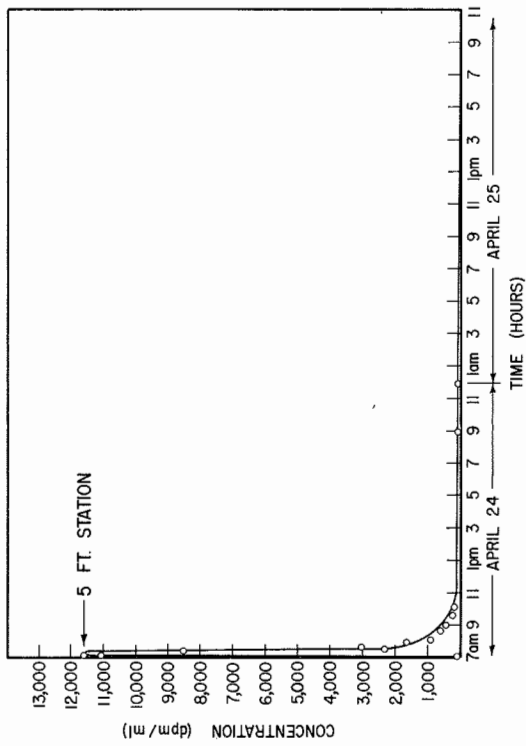


Table 4-3
 Longitudinal Transport of Sr⁸⁵ in the Flume Water

Days After Release	Percent of the Total Sr ⁸⁵ Released Carried in the Water				
	5-ft Station	50-ft Station	100-ft Station	150-ft Station	195-ft Station
1	66.3	57.7	41.4	32.2	24.1
2	74.5	63.2	44.1	37.7	29.0
3	78.2	66.4	67.9	40.9	33.1
4	80.6	69.7	50.6	43.9	34.9
5	83.1	72.9	53.2	46.4	36.8
6	84.7	74.6	56.1	49.2	38.6
7	86.2	77.4	59.4	51.4	40.2

From Tables 4-2 and 4-3, it can be seen that more than 38 percent of Cs¹³⁷ and 33 percent of Sr⁸⁵ released in the flume remained within the first 5 feet of the flume after one day. This is quite different from the results shown in the dye release experiment which showed that only 10 to 20 percent of the dye remained. The considerable decrease of both Cs¹³⁷ and Sr⁸⁵ passing from one station to the next was mainly a result of the uptake of radionuclides by bottom sediments and plants existing between the stations.

Peak Concentrations: The peak concentrations of Cs¹³⁷ at the 5-, 50-, 100-, 150-, and 195-ft stations were found to be 12, 100, 886, 388, 274, and 129 dpm/ml, respectively. It appears that for both Cs¹³⁷ and Sr⁸⁵, there was a large difference between the peak concentrations at the 5-ft and 50-ft stations. This is caused by the radionuclides at the 5-ft station not having

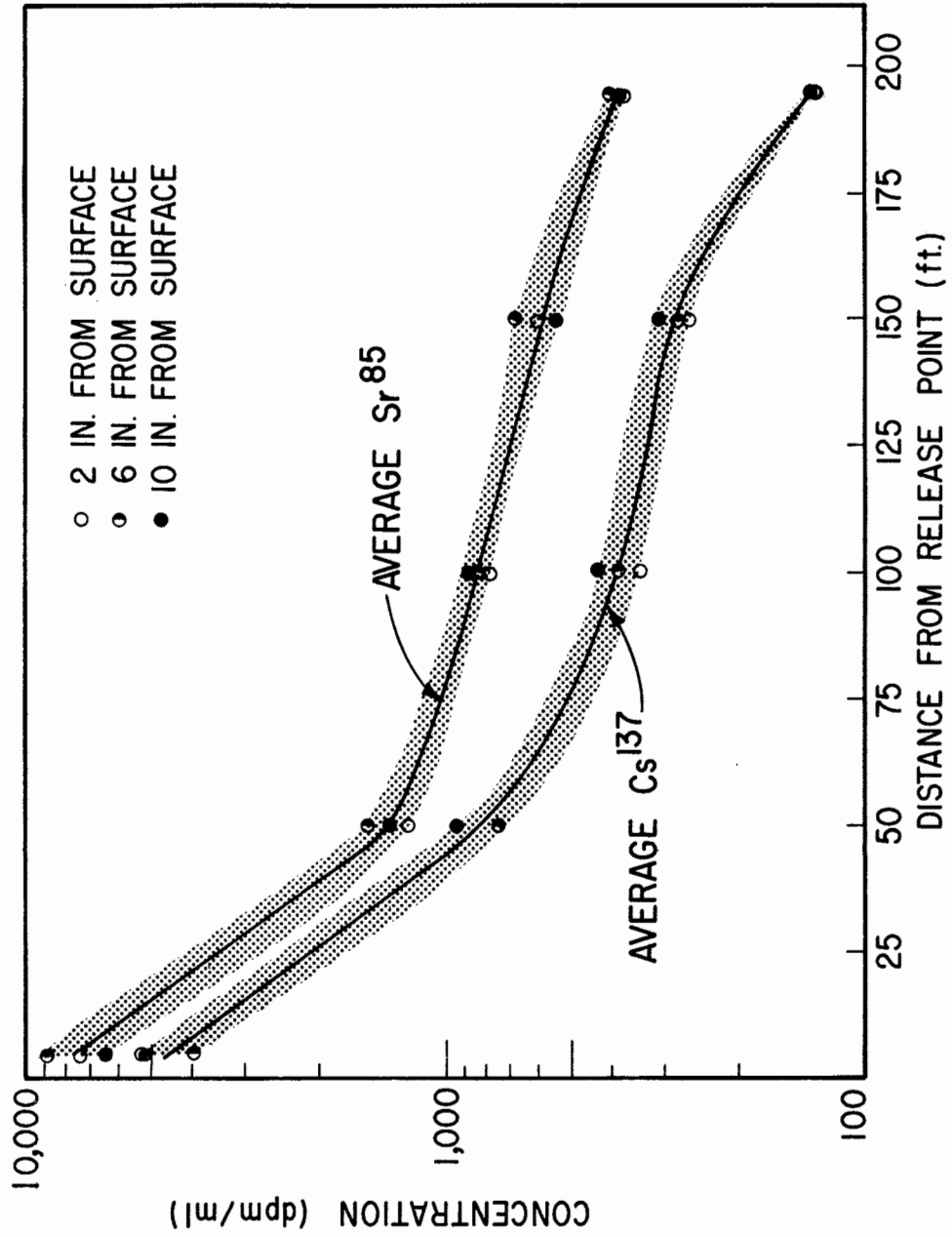


FIG. 4-6. MAXIMUM CONCENTRATION OF Sr⁸⁵ AND Cs¹³⁷ ALONG THE FLUME

diffused appreciably. The travel time and the average velocity of the peaks of Cs^{137} and Sr^{85} are shown in Tables 4-4 and 4-5. The travel time was derived from the time which had elapsed after release until the occurrence of the peak concentration. The theoretical travel time for plug flow is 690 minutes for 195 feet of flume length.

The peak values of Sr^{85} and Cs^{137} in solution at each station were correlated with distance of travel in Fig. 4-6. There is a significant change in slopes of the curves for both Cs^{137} and Sr^{85} . Therefore, the transverse dispersion across the section of the flume was not complete as the peaks passed the 5-ft station. The peak concentrations for different levels in the vertical section of the flume were found to occur at different times for both Cs^{137} and Sr^{85} . This indicates that the dispersion through the cross section of the flume was not homogeneous.

Table 4-4

Movement of Cs^{137} Peak Concentration in the Flume Water

Station (ft)	Travel Time From Release Point (min)	Peak Concentration (dpm/ml)	Average Velocity of Peak (ft/min)
50	135	886	0.37
100	285	388	0.35
150	420	274	0.36
195	645	129	0.31

Table 4-5
 Movement of Sr⁸⁵ Peak Concentration in the Flume Water

Station (ft)	Travel Time From Release Point (min)	Peak Concentration (dpm/ml)	Average Velocity of Peak (ft/min)
50	150	1,500	0.33
100	270	700	0.37
150	235	600	0.34
195	600	395	0.33

Vertical Dispersion: The vertical dispersion of Cs¹³⁷ and Sr⁸⁵ are plotted in Figs. 4-7 and 4-8. The data represent the levels of radionuclides at 2, 6, and 10 inches of depth and at the 5-ft, 100-ft, and 195-ft stations. It is recognized that the radionuclide concentrations were similar although somewhat cyclic. This similarity suggests that complete longitudinal mixing was accomplished, probably by the plants.

It is observed that the peak concentrations of both Cs¹³⁷ and Sr⁸⁵ did not occur at the same time. This indicates that the movement of the radionuclides was highly affected by the presence of the plants. Also, it is noted that the vertical dispersion across the flume was not homogeneous.

Mass Curve: The total amounts of Cs¹³⁷ and Sr⁸⁵ discharging through the flume were obtained by the total accumulated products of concentrations and flow discharges at the 195-ft station. As cited in Table 4-2, the Cs¹³⁷ leaving the flume after one day was 10.4 percent of the total Cs¹³⁷ released,

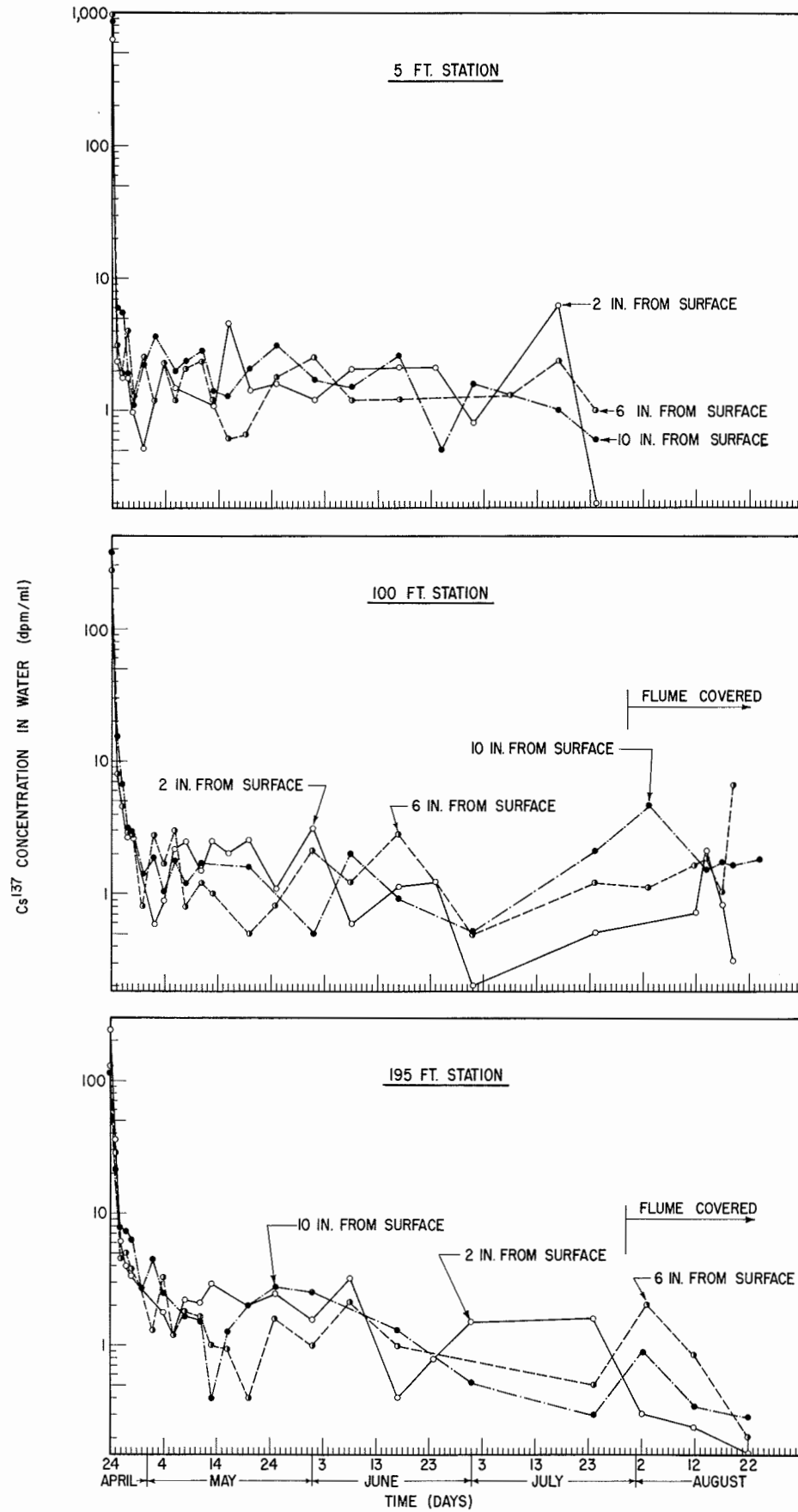


FIG. 4-7. VERTICAL DISTRIBUTION OF Cs^{137} (WATER)

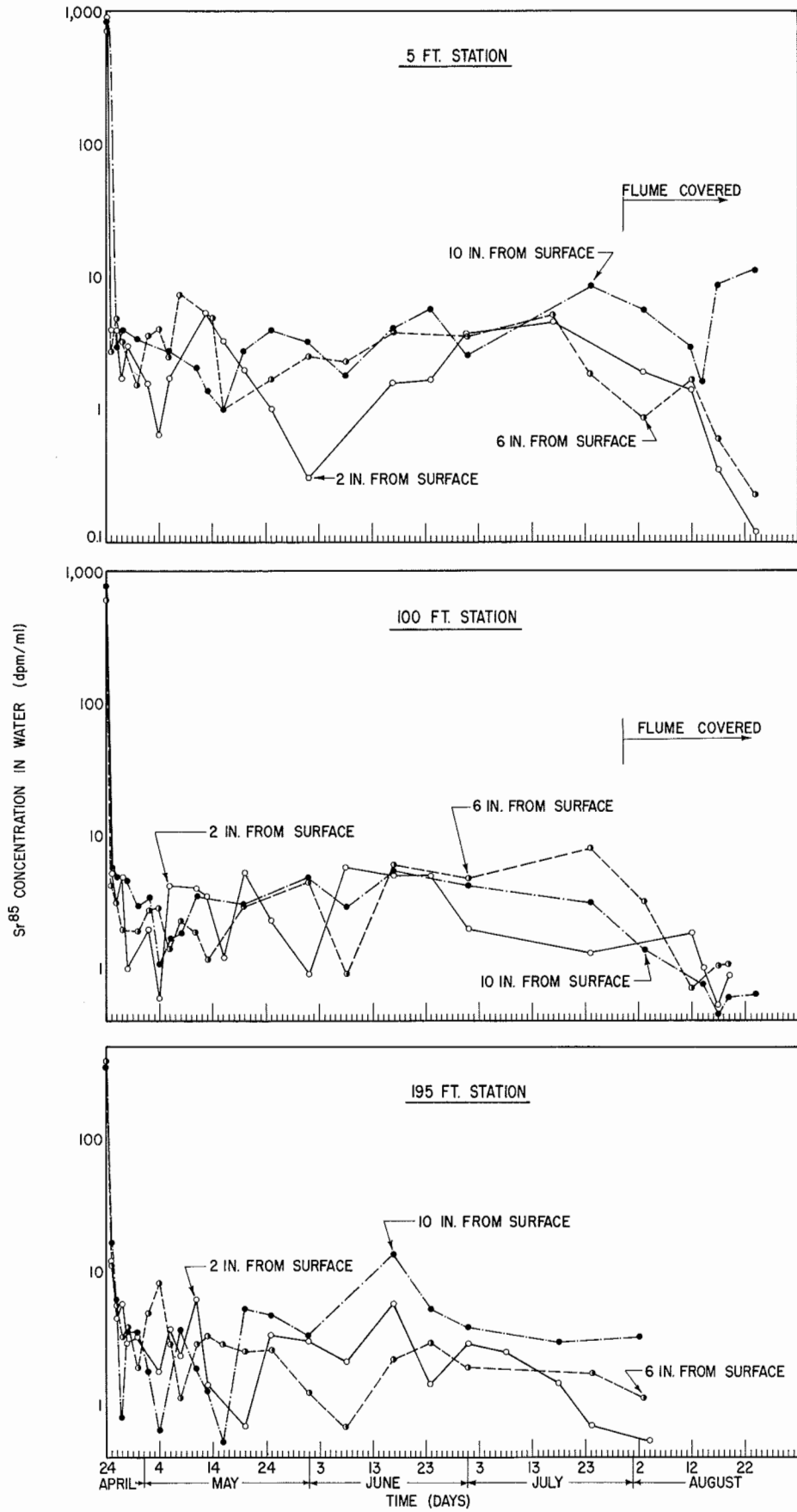


FIG. 4-8. VERTICAL DISTRIBUTION OF Sr⁸⁵ (WATER)

as compared with 38.0 percent after one week. Also, as shown in Table 4-3, the Sr^{85} discharged through the flume was 24 and 40 percent, respectively, of the total released after one day and one week of elapsed time. It appears that the amount of Sr^{85} sorbed by the aquatic plants and bottom sediments was less than that of Cs^{137} . In Figs. 4-9 and 4-10, the accumulated percentage of Cs^{137} and Sr^{85} discharged through the flume was correlated with the time following the release of radionuclides. After three months of continuous discharge, the total Cs^{137} leaving the flume was 88 percent of the initial amount released, whereas 93 percent of the total Sr^{85} passed through the flume after three months. The remaining 12 percent of Cs^{137} and 7 percent of Sr^{85} remained in the bottom sediments and aquatic plants.

Suspended Materials: The average percentage of Sr^{85} and Cs^{137} associated with the suspended material was 12 and 14 percent, respectively, Tables 4-6 and 4-7. As the total concentration of radionuclides in water solution was lower than 10 dpm/ml, the fraction sorbed on suspended solids became undetectable. The results presented in Tables 4-5 and 4-6 were derived from the data collected during the first week following the release of radionuclides.

After the first day of release, the Sr^{85} concentration in solution varied between 390 dpm/ml and 27 dpm/ml at the 195-ft station. Thus, the fraction of Sr^{85} associated with suspended solids varied between 2340 dpm/mg and 145 dpm/mg. For Cs^{137} , the fraction in water varied between 128 dpm/ml and 35 dpm/ml, but following the release on the first day the Cs^{137} associated with suspended solids at the 195-ft station ranged between 895 dpm/mg and 246 dpm/mg. The concentrations of Sr^{85} associated with suspended solids exhibited greater variation than those of Cs^{137} .

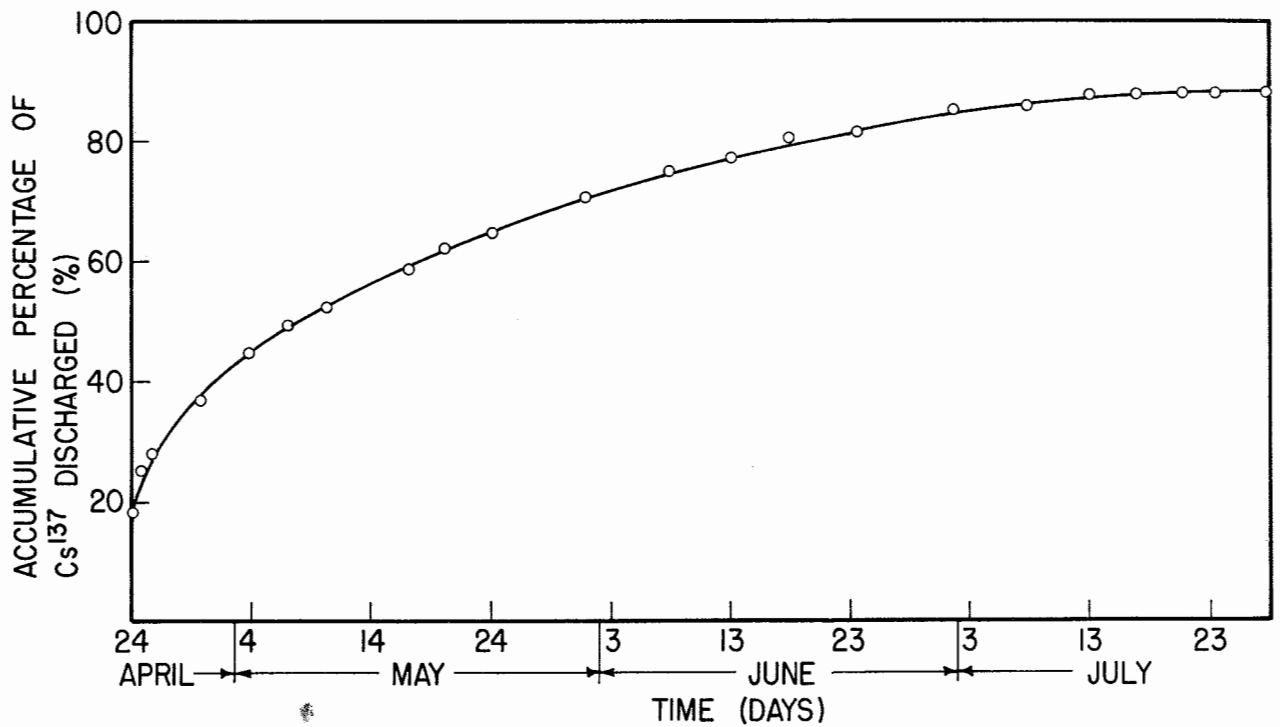


FIG. 4-9. Cs¹³⁷ DISCHARGED THROUGH THE FLUME

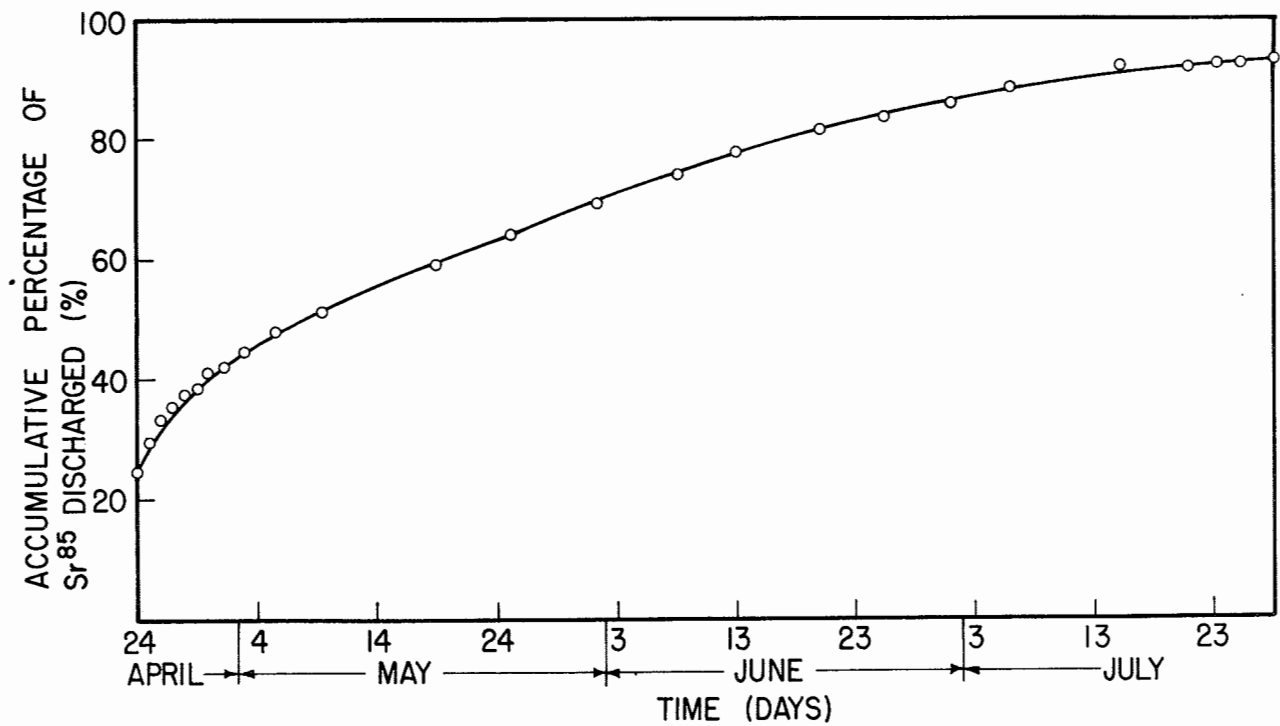


FIG. 4-10. Sr⁸⁵ DISCHARGED THROUGH THE FLUME

Table 4-6
 Fraction of Sr⁸⁵ Associated with
 Suspended Solids in Flume Water

Stations (ft)	Fraction Sr ⁸⁵ Sorbed on Suspended Solids			Average
	2 in. from Surface	5 in. from Surface	10 in. from Surface	
5	0.16	0.15	--	0.16
50	0.05	0.08	0.14	0.09
100	--	0.15	0.07	0.11
150	0.04	0.06	0.26	0.12
195	0.08	0.07	0.09	0.08

Table 4-7
 Fraction of Cs¹³⁷ Associated with
 Suspended Solids in Flume Water

Stations (ft)	Fraction Cs ¹³⁷ Sorbed on Suspended Solids			Average
	2 in. from Surface	5 in. from Surface	10 in. from Surface	
5	0.20	0.12	--	0.16
50	0.14	0.11	0.16	0.13
100	--	0.21	0.13	0.17
150	--	0.08	0.13	0.11
195	0.10	0.09	0.17	0.12

The suspended solids content in the flume water varied from 6 mg/l to 54 mg/l as shown in Fig. 4-11. The components of the suspended solids were primarily organic suspensions consisting of algae, etc. It can be seen in Fig. 4-11 that the suspended solids concentration fluctuated along the mean value of 20 mg/l until July. At this time, the concentration of suspended solids suddenly increased, reflecting a change in the biomass. The volatile fraction of the suspended solids varied between 70 and 90 percent.

It appears that the concentration of both Cs^{137} and Sr^{85} in the aqueous solution finally decreased to almost undetectable levels after three months of continuous discharge even though there was about 12 percent of Cs^{137} and 7 percent of Sr^{85} remaining in the flume. Actually, the concentration of Cs^{137} and Sr^{85} was very low one month after release

Analyses of Bottom Sediments Samples: The amount of Cs^{137} and Sr^{85} associated with bottom sediments in a function of both the mixing characteristics and contact time. The radionuclide uptake in the flume by the sediments increased with regard to: (1) the longitudinal distribution along the flume, (2) the average percentage of total Cs^{137} and Sr^{85} added in the flume, and (3) the depth of migration. The uptake was dependent upon the suspended organic debris and the changes of environmental factors; i.e., pH, ORP, temperature, etc.

Longitudinal Distribution of Sr^{85} and Cs^{137} : As shown in Figs. 4-12 and 4-13, the Cs^{137} in the bottom sediments varied between 1000 dpm/gm and 25 dpm/gm, while the Sr^{85} varied between 1,600 dpm/gm and 13 dpm/gm. This suggests that the uptake of Sr^{85} by bottom sediments was more sensitive to environmental changes than Cs^{137} . Generally, the concentration of both Cs^{137} and Sr^{85} in the bottom sediments increased with time until a peak

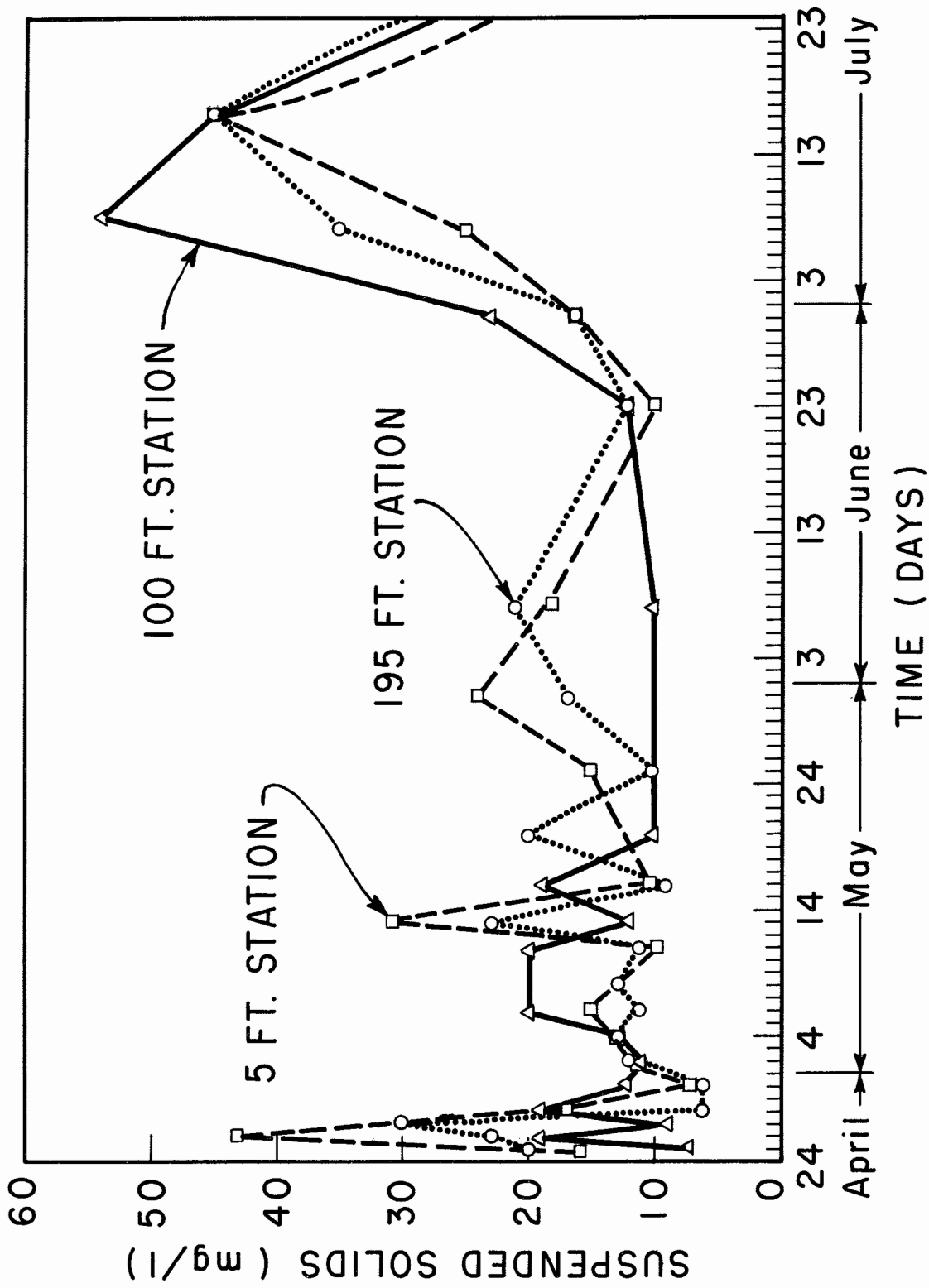


FIG. 4-11. DISTRIBUTION OF SUSPENDED SOLIDS IN THE FLUME

value was reached and then a rapid drop occurred. There was a strong tendency to maintain equilibrium concentration. The peak concentrations of Sr^{85} at different stations occurred within the first week following the release but the peak concentrations of Cs^{137} at different stations occurred at different times, varying over a period of more than two months. The Sr^{85} associated with the sediments was displaced more readily than Cs^{137} . Figures 4-12 and 4-13 also show that the concentration of both Sr^{85} and Cs^{137} in the bottom sediments increased appreciably during June and July. During this period there was a considerable change in the ecosystem.

Average Concentration: The average concentrations for Sr^{85} and Cs^{137} were derived from the data at all sampling stations for the same sampling time. The weight factor used for each station was 1, 2, 2, 2, and 1, respectively, for the 5, 50, 100, 150, and 195-ft stations. As shown in Fig. 4-14, the fluctuation of the average concentration of Sr^{85} was much more than that of Cs^{137} . The variation of Sr^{85} content was also greater than that of Cs^{137} . These differences imply that the interchangeability of Sr^{85} between the attached phase and the liquid phase was higher than that of Cs^{137} .

Percentage of Total Sorbed Sediments: The percentages of Sr^{85} and Cs^{137} sorbed on or in the bottom sediments are presented in Figs. 4-15 and 4-16. The average percentage of Cs^{137} associated with bottom sediments increased to its peak value of about 24 percent on May 2 and then decreased to 3 percent on June 1. After June 10 the percentage of Cs^{137} associated with sediments increased again to 18 percent and by July 3 decreased to the lowest value of 2.6 percent. The peak percentage of Sr^{85} of 17 percent occurred on April 27; the percentage fluctuated until a value of 0.5 percent

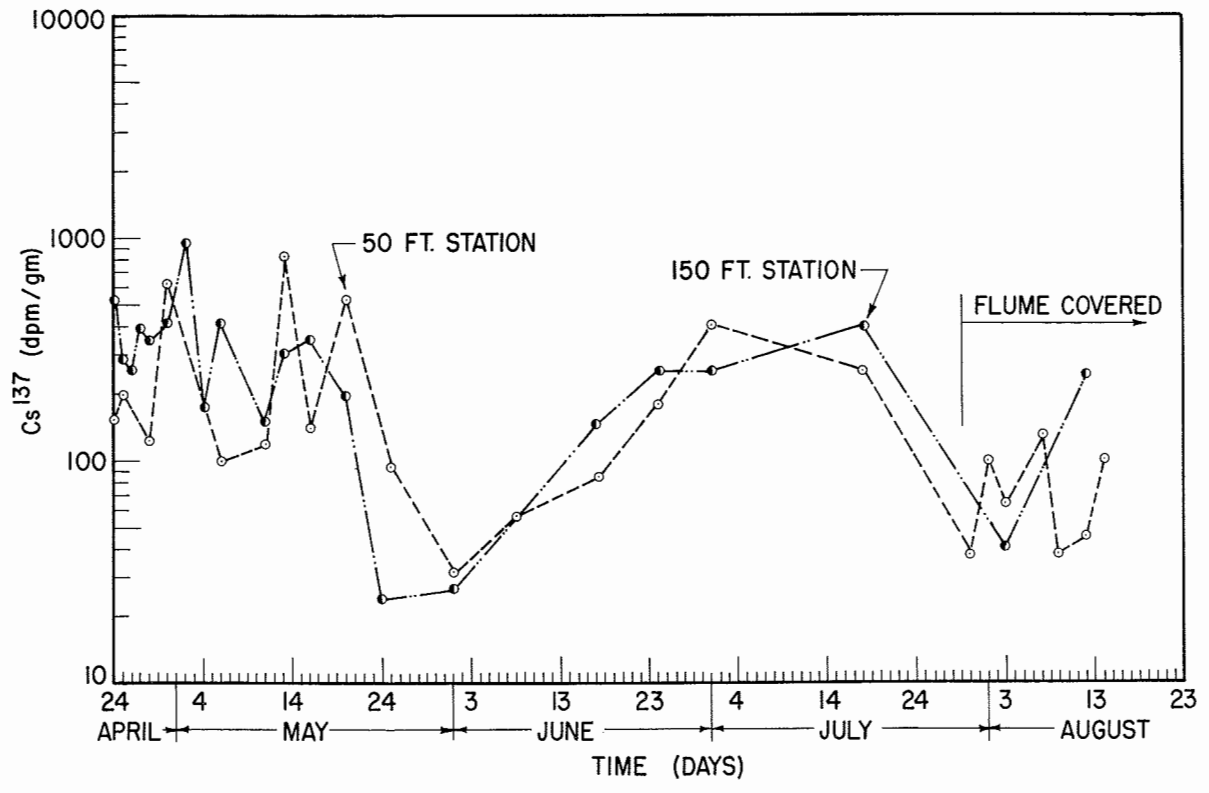
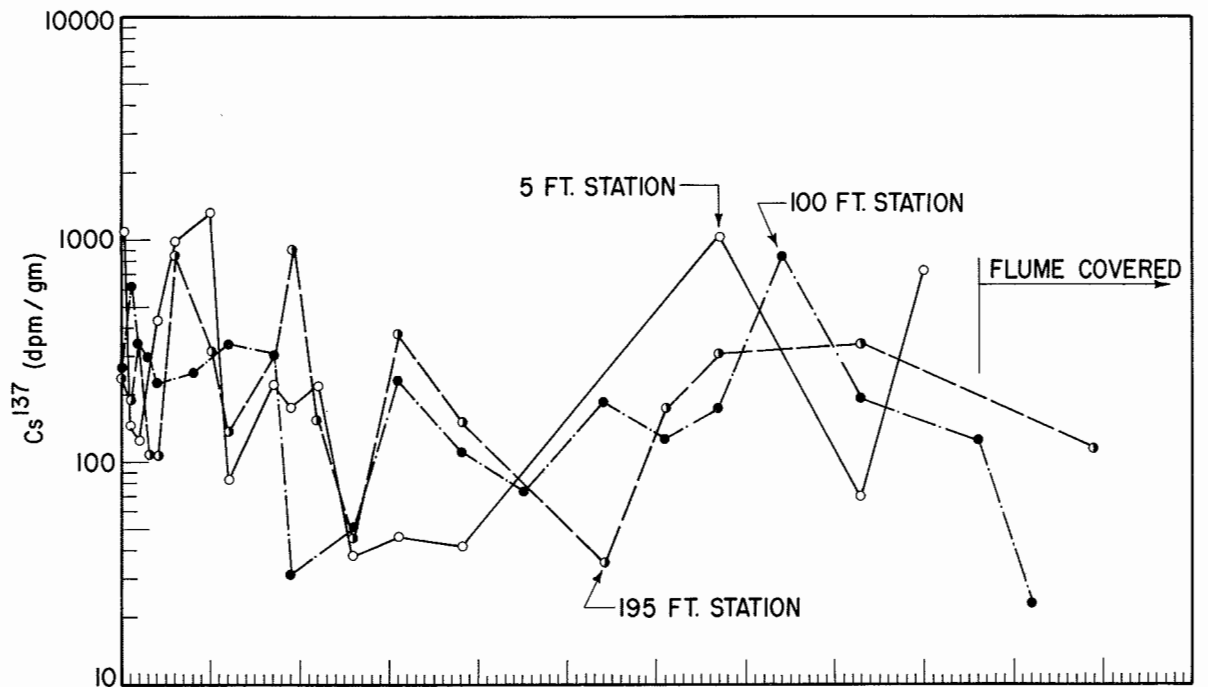


FIG. 4-12. SORPTION OF Cs^{137} BY BOTTOM SEDIMENTS IN THE FLUME

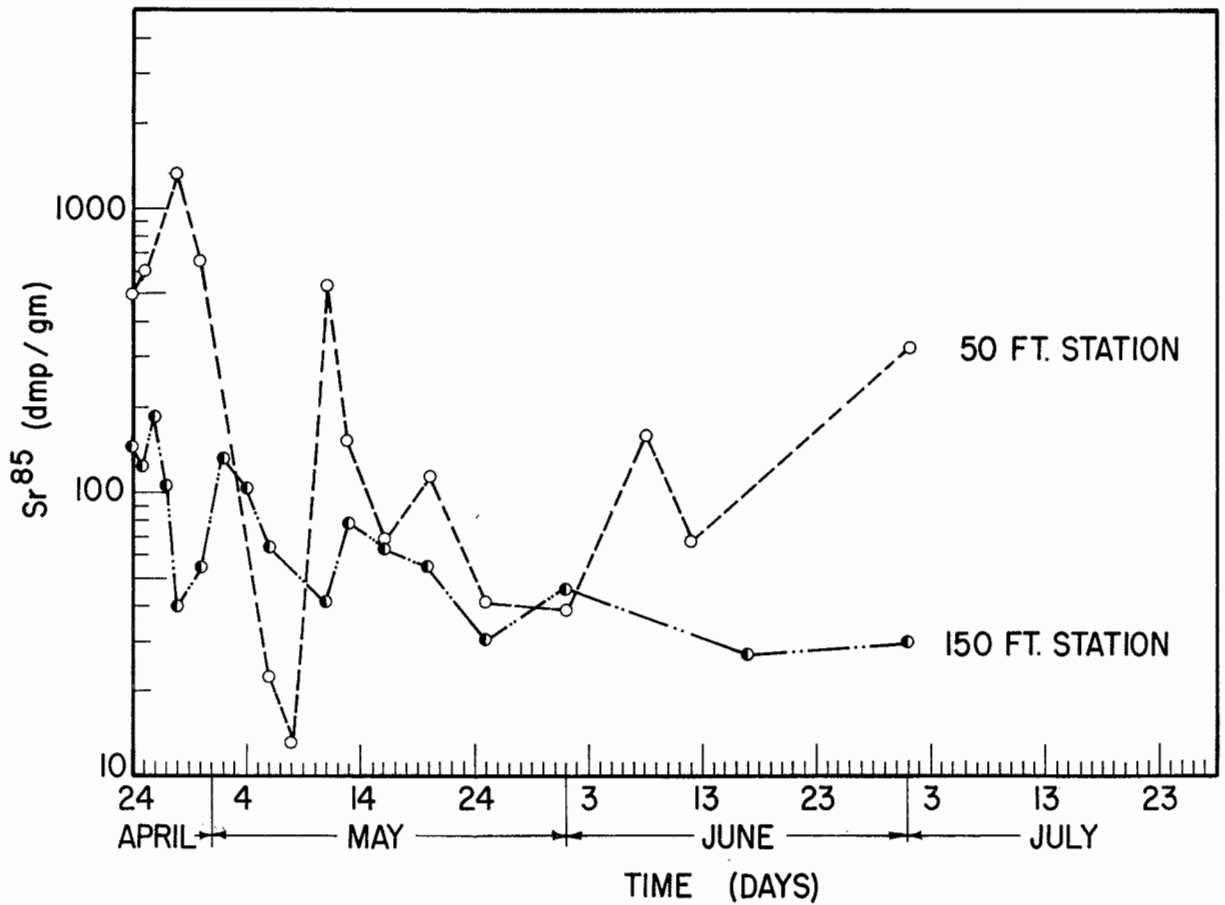
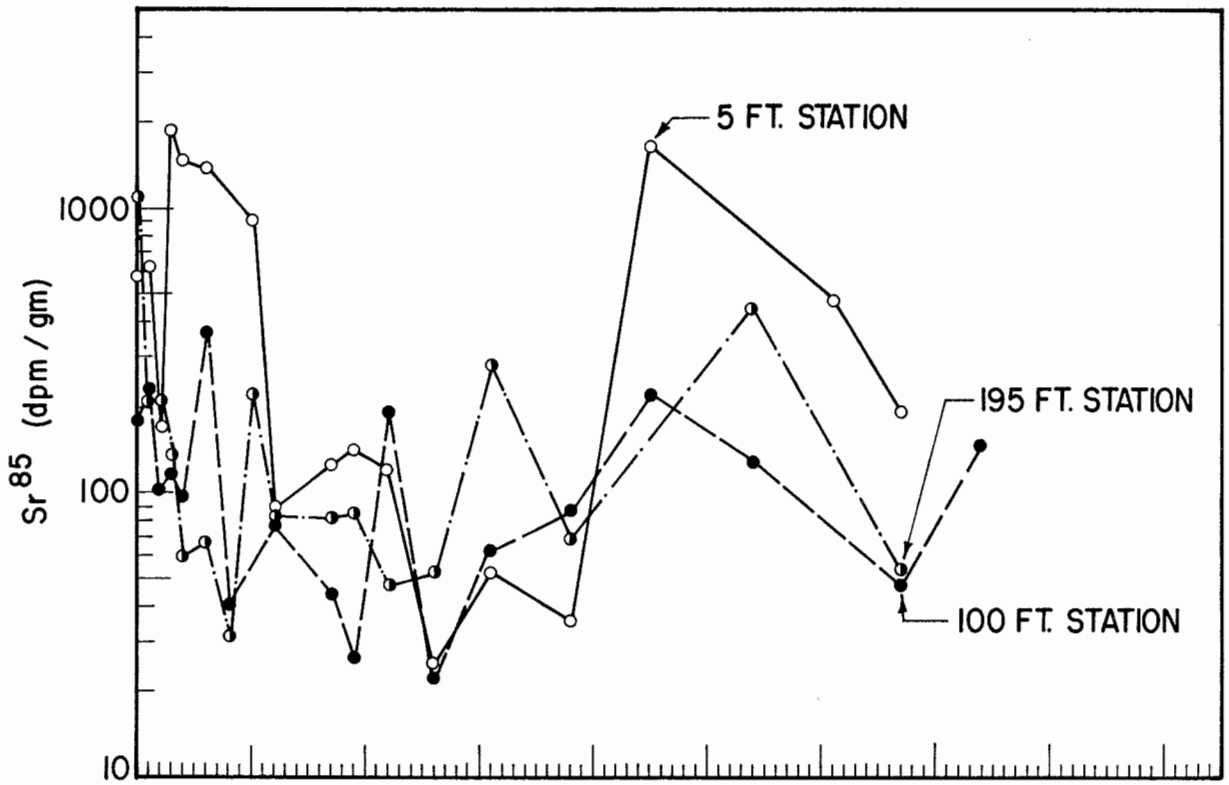


FIG. 4-13. SORPTION OF Sr^{85} BY BOTTOM SEDIMENTS IN THE FLUME

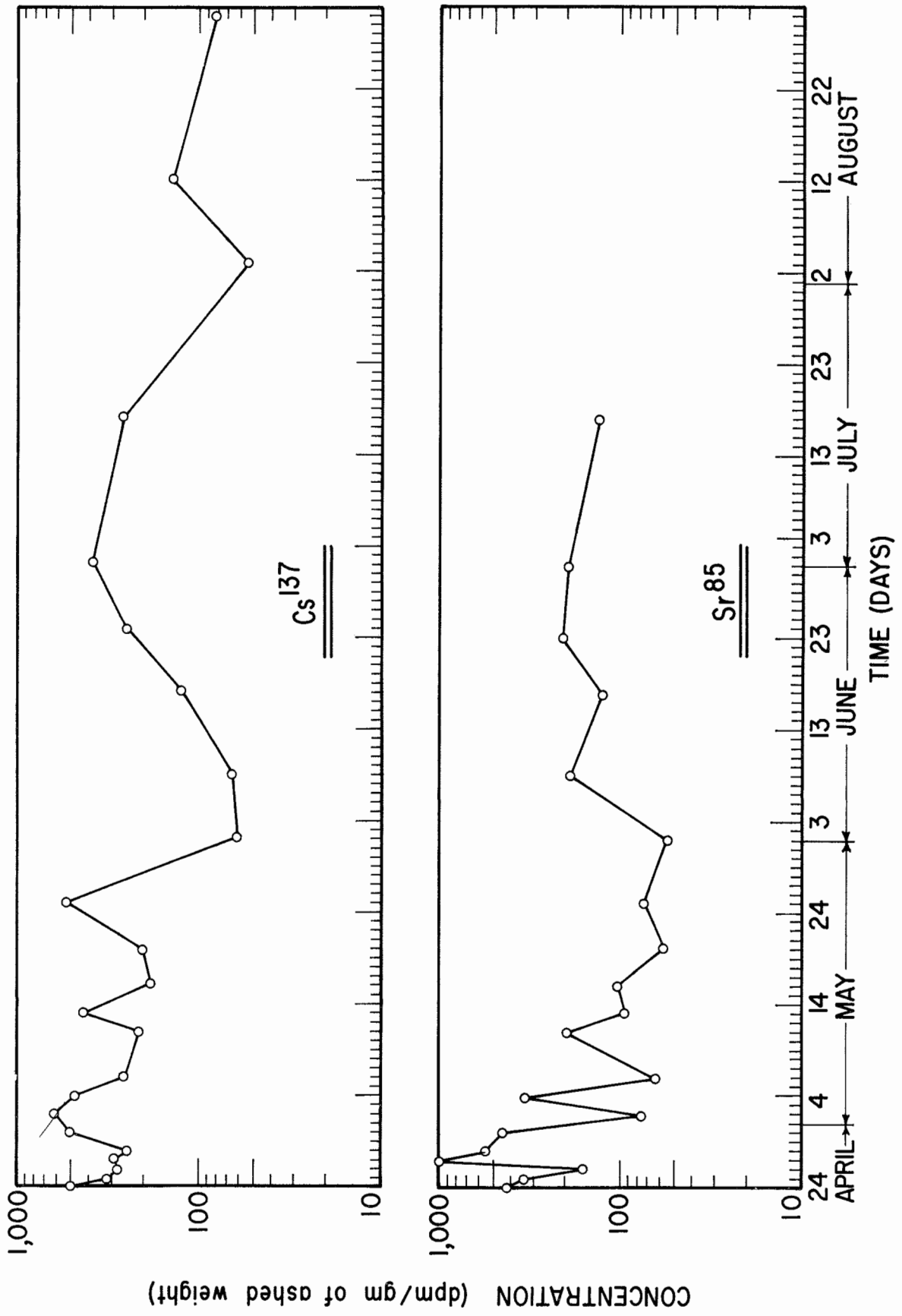


FIG. 4-14 AVERAGE DISTRIBUTION IN BOTTOM SEDIMENTS

was reached on June 1. The percentage of Sr^{85} increased to about 11 percent by June 23 but decreased to 2.5 percent by July 25. The minimum concentration of Sr^{85} was 2.2 percent, that of Cs^{137} 2.7 percent. Some of the radionuclides which were absorbed probably were held in the interlayer positions of the clay minerals in the sediments. The appearance of the peak percentage values of both Sr^{85} and Cs^{137} at the beginning of July was caused by the re-uptake of radionuclides which were released from dead plants.

Migration of the Radionuclides: The depth at which the Sr^{85} and Cs^{137} penetrated into the sediments was dependent upon the amount of radionuclides present and time of contact, Figs. 4-17 and 4-18 and Table 4-8. It can be seen that more than 65 percent of the Cs^{137} and 70 percent of the Sr^{85} in the sediments were in the top inch.

The uptake of Sr^{85} and Cs^{137} by bottom sediments is related to the concentration of Sr^{85} and Cs^{137} at the interface of the flume water and bottom sediments, the elapsed time, the plant growth rate, and a number of environmental factors. However, considering the pH range of 6.8 to 7.5, the uptake of Sr^{85} and Cs^{137} by the bottom sediments should not change appreciably. This is especially true if the final sorbed radionuclide concentrations are considered to be taken up mainly by ion exchange. However, because of the settled organic debris from dead plants and the transport of released radionuclides from dying plants by water, the concentrations of Sr^{85} and Cs^{137} associated with bottom sediments changed significantly. Thus, the uptake of Sr^{85} and Cs^{137} by the bottom sediments was affected by environmental changes.

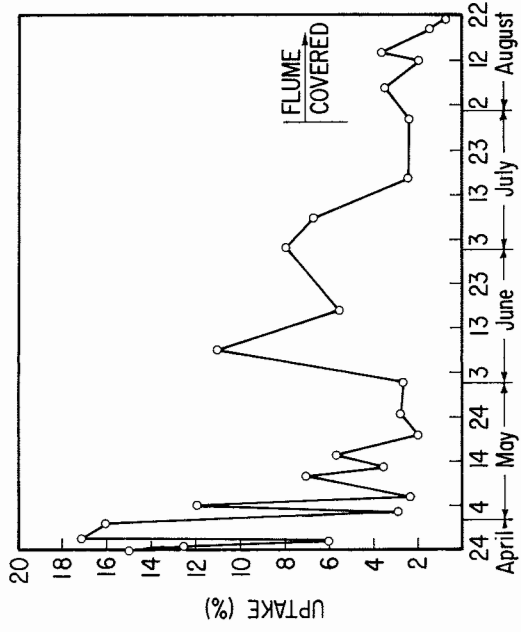


FIG. 4-16. Sr⁸⁵ SORBED BY BOTTOM SEDIMENTS

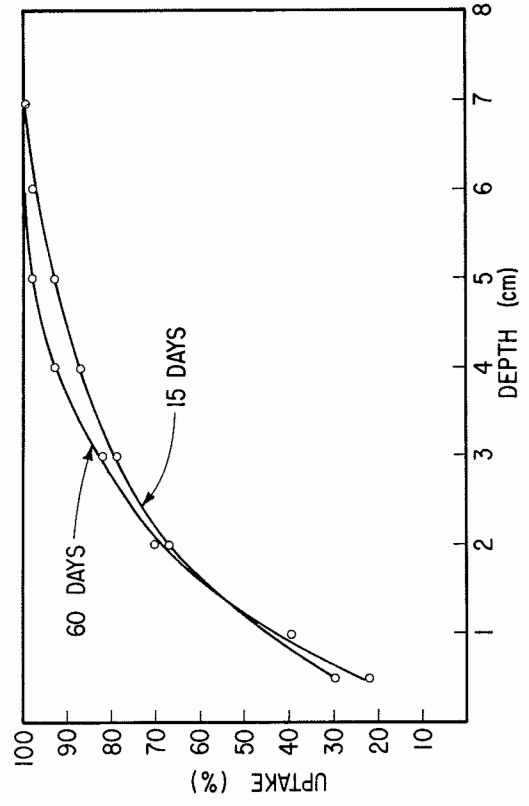


FIG. 4-18. PENETRATION OF Sr⁸⁵

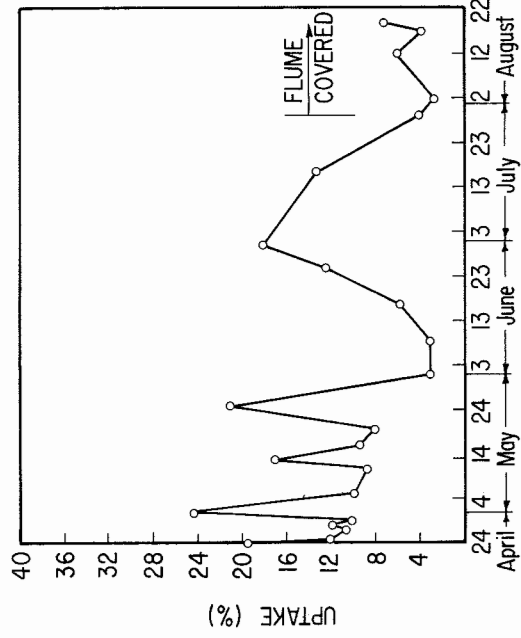


FIG. 4-15. Cs¹³⁷ SORBED BY BOTTOM SEDIMENTS

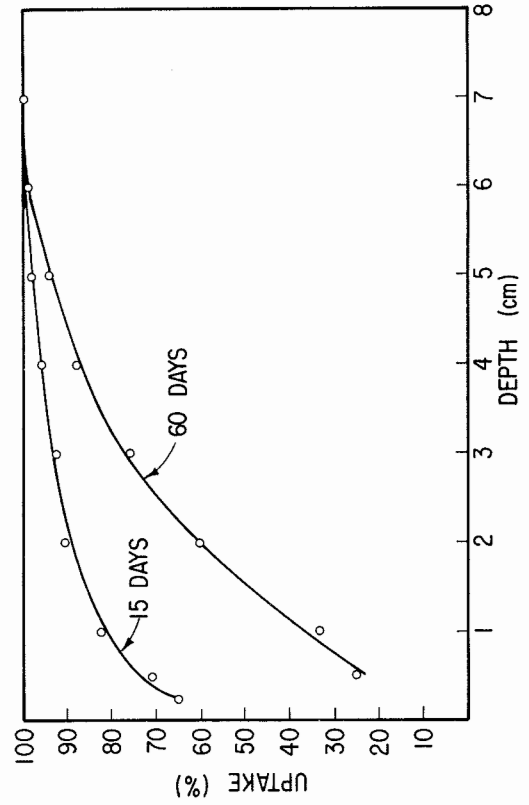


FIG. 4-17. PENETRATION OF Cs¹³⁷

Table 4-8
 Sr^{85} and Cs^{137} Penetrating into the Bottom Sediments

Depth (cm)	15 Days Following Release		60 Days Following Release	
	Sr^{85} (%)	Cs^{137} (%)	Sr^{85} (%)	Cs^{137} (%)
$\frac{1}{4}$	-	65	-	-
$\frac{1}{2}$	29	71	22	25
1	39	82	39	33
2	67.5	90	70	60
3	79	93	82	76
4	87	96	93	88
5	93	98	98	94
6	98	99	100	99
7	100	99.5		100

Analyses of Plant Samples: The aquatic plants played an important role, by rapid sorption of radionuclides on the plant surfaces and by causing eddy turbulence, thereby providing contact opportunity. The immediate uptake of Cs^{137} by all rooted and floating plants was about 20 percent of the total Cs^{137} added in the flume, that of Sr^{85} about 14 percent. The radionuclides sorbed during the first day were released back into the water very rapidly. This indicates that the sorption was mainly a surface reaction. During the study, it was revealed that the radionuclides sorbed on a certain species of the plants were transferred to other species after the original species died.

Distribution of Cs¹³⁷ and Sr⁸⁵ in Aquatic Plants: The studies showed that there were six predominant species of macroplants and algae present in the flume at the beginning of the study. These were Myriophyllum sp., Potamogeton sp., Zannichellia sp., Chara sp., Cladophora sp., and Spirogyra sp. The longitudinal distributions of Cs¹³⁷ and Sr⁸⁵ associated with plants are shown in Figs. 4-19 and 4-20. It appears that the maximum uptake by the aquatic plants occurred on the first day following the release of radionuclides for all the species. The concentration of Cs¹³⁷ and Sr⁸⁵ sorbed on the plants was found to decrease with an increasing distance from the release point during the first two days following the release. However, after one week following the release, the concentrations of Sr⁸⁵ and Cs¹³⁷ sorbed on plants were approximately the same for samples collected at all stations. In June, both the Sr⁸⁵ and Cs¹³⁷ uptake by aquatic plants increased considerably. This was believed to be caused by the death of large amounts of Potamogeton, Zannichellia, and Chara.

Change of Ecological System: The effects of die-off of certain plants and the increase in uptake of Cs¹³⁷ and Sr⁸⁵ by the remaining plants can be seen in Fig. 4-21. The ORP reading dropped from 400 mv to 100 mv; the pH value dropped from 9 to 7.5; the DO dropped from 8 mg/l to 2 mg/l; and the conductivity dropped from 1,500 mho/cm to 1,000 mho/cm. Accordingly, there was a concomitant decrease in the biomass. Samples from nine different stations showed that the average biomass, ashed weight, decreased from 13 gm/sq.ft to 8 gm/sq.ft.

Percentage of Radionuclides Sorbed by Plants: The total uptake of both Cs¹³⁷ and Sr⁸⁵ by all aquatic plants was estimated by the product of the average uptake of all species per weight and the weight of the biomass.

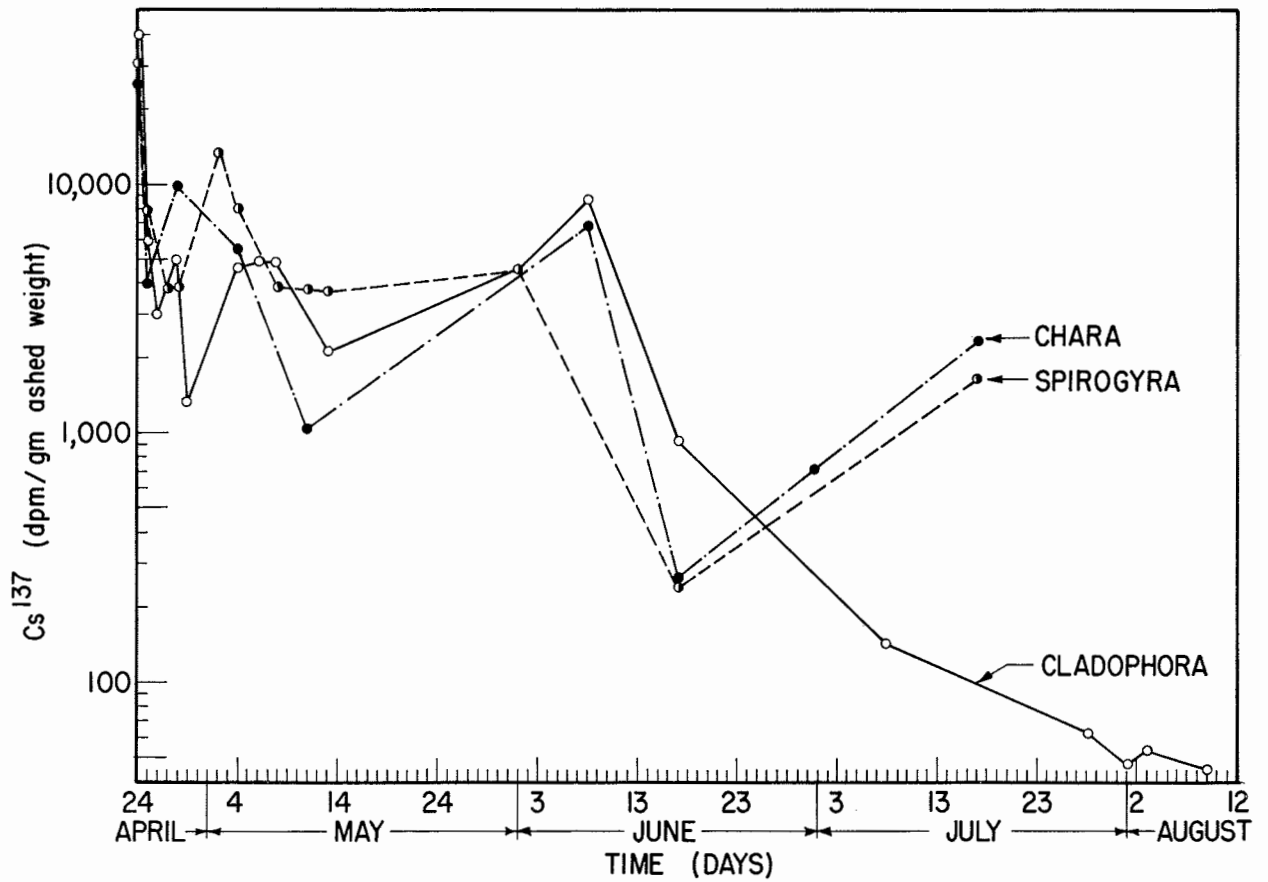
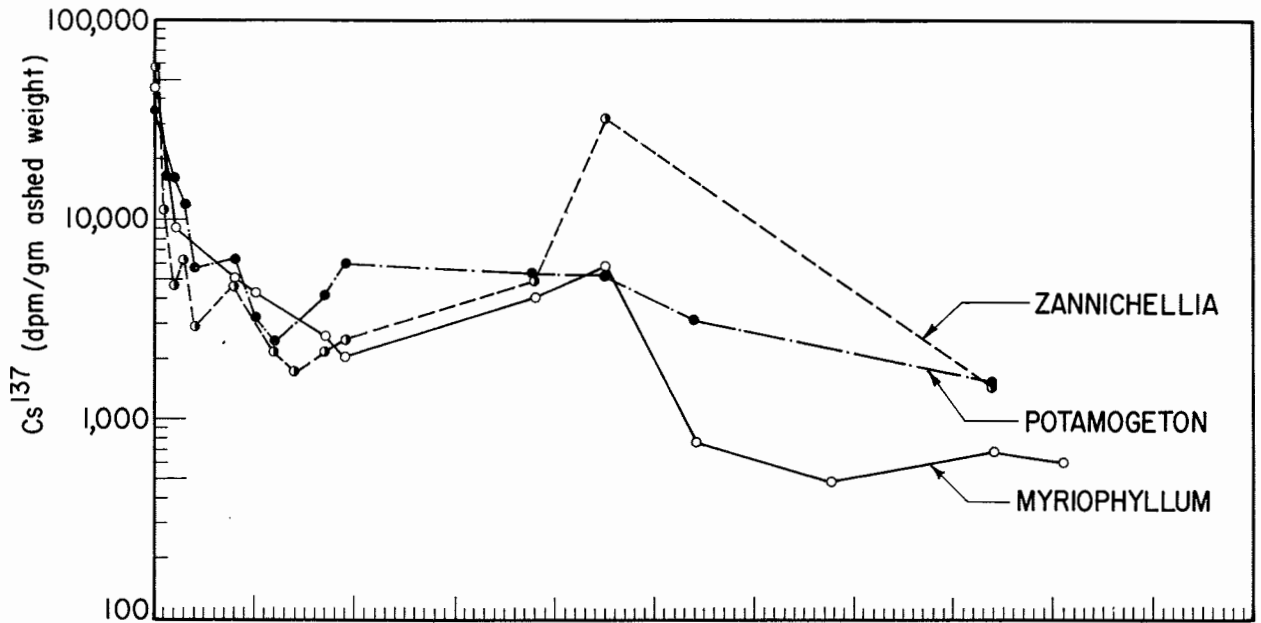


FIG. 4-19. DISTRIBUTION OF Cs¹³⁷ BETWEEN VARIOUS PLANT SPECIES

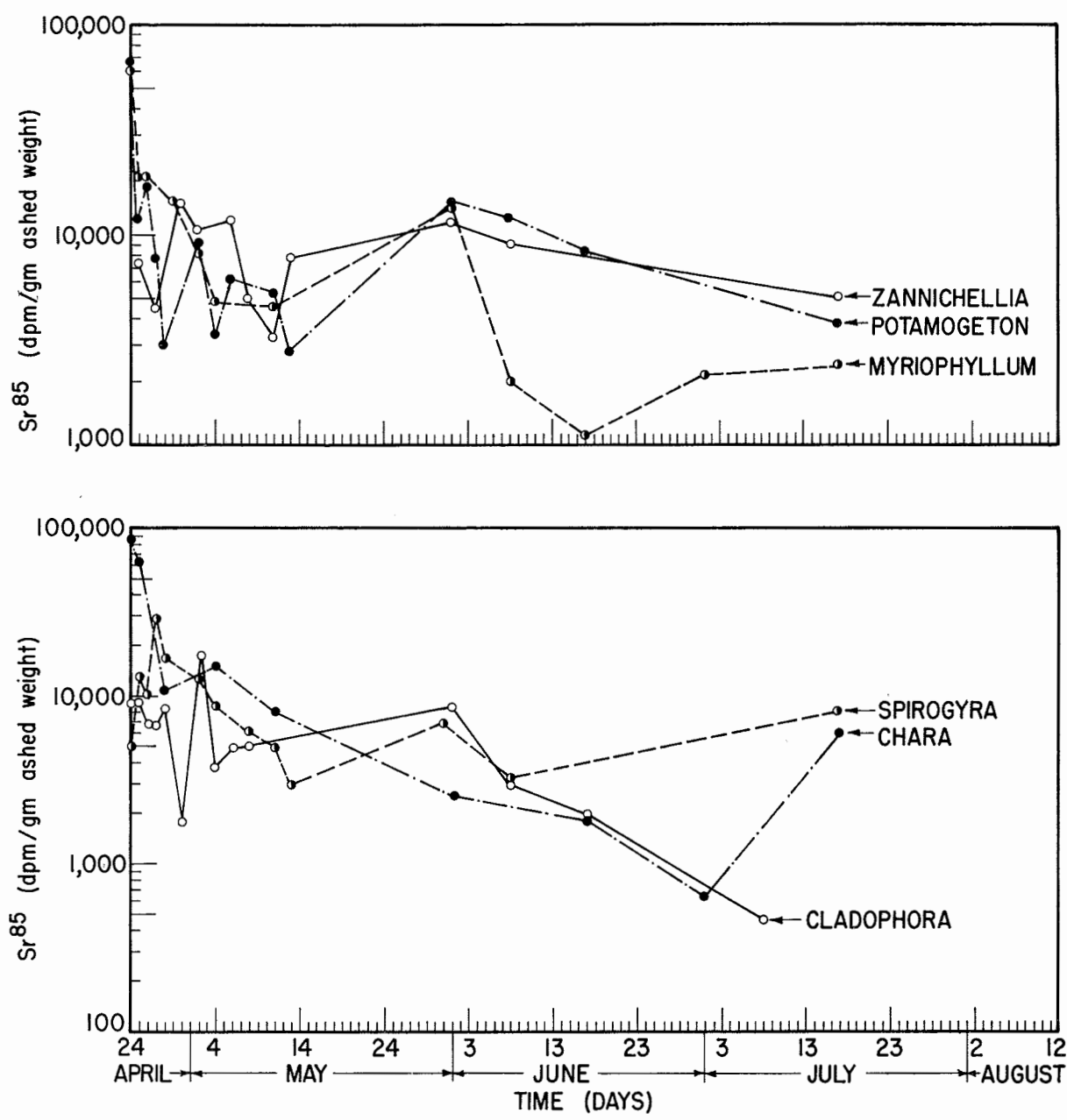


FIG. 4-20. DISTRIBUTION OF Sr⁸⁵ BETWEEN VARIOUS PLANT SPECIES

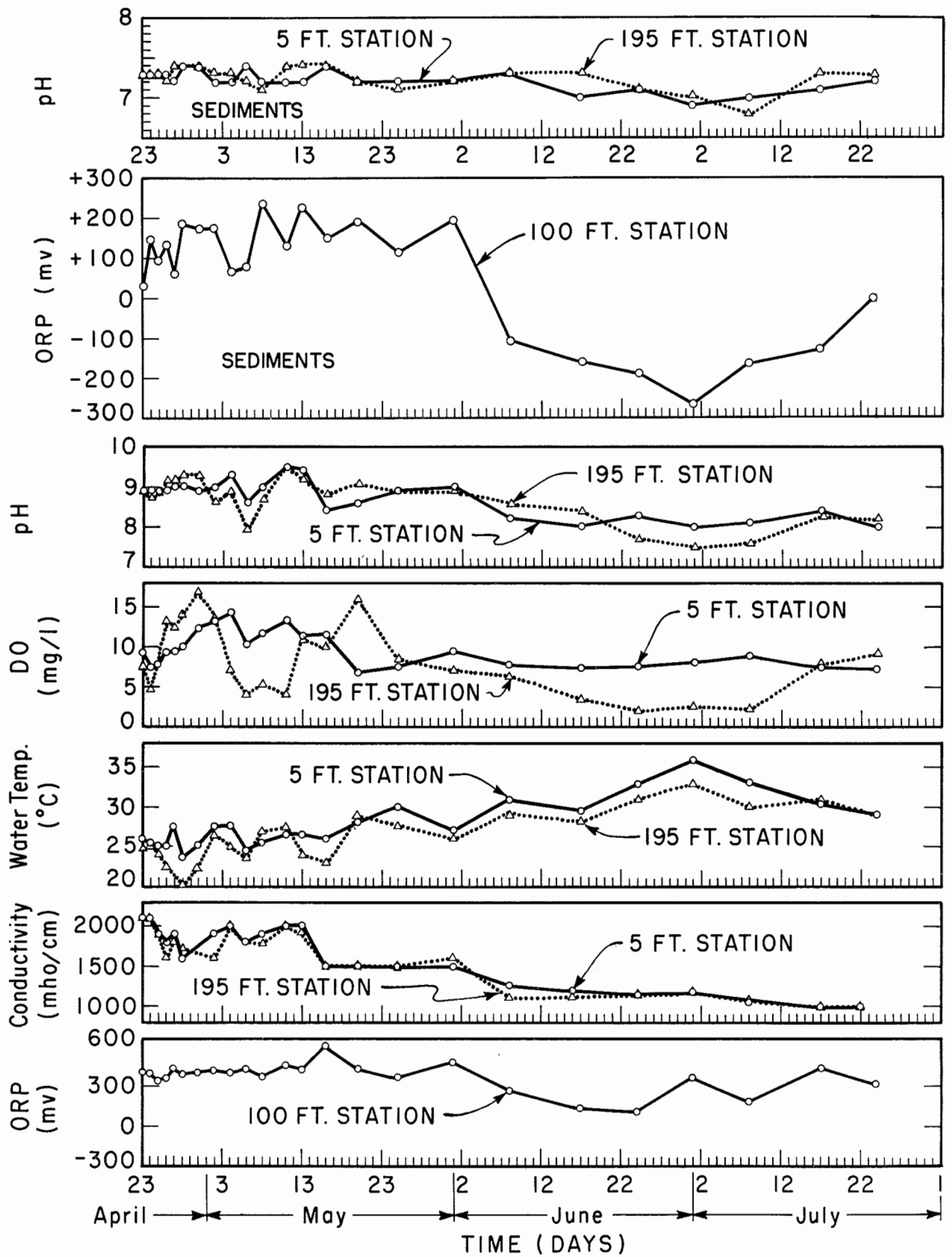


FIG. 4-21. ENVIRONMENTAL FACTORS

The percentages of Sr^{85} and Cs^{137} sorbed by plants are shown in Figs. 4-22 and 4-23. It appears that the percentage of Cs^{137} sorbed by the plants was higher than that of Sr^{85} . During the period from May 10 to June 10, the increase in percentage of both Cs^{137} and Sr^{85} sorbed on plants was believed to result from the normal adjustment back to equilibrium. During this period, the percentage of radionuclides associated with bottom sediments decreased significantly as shown in Figs. 4-15 and 4-16.

Concentration Factors: The ratio of the radioactivity in one gm of ashed plant weight to the radioactivity in one ml of the surrounding water is defined as the concentration factor, K_c . Cladophora exhibited the highest K_c and Zannichellia the lowest for uptake of Cs^{137} . Conversely, Spirogyra exhibited the highest K_c and Cladophora the lowest for Sr^{85} . In Figs. 4-24 and 4-25, the K_c values are shown for Sr^{85} and Cs^{137} . It can be seen that the K_c values changed with time.

For macroplants, Potamogeton exhibited the highest K_c value for both Sr^{85} and Cs^{137} . Among the algae group, Cladophora exhibited the highest K_c value for the uptake of Cs^{137} , Spirogyra for Sr^{85} , Table 4-9.

Plants Washed by Citric Acid: Plants were washed with citric acid to determine the degree of sorption. The Sr^{85} sorbed on the surface of various plants in the flume was estimated to vary from 12 to 37 percent, Cs^{137} from 15 to 46 percent, Table 4-10.

Analysis of Release Study: On July 30, 1964 the first 105 ft of the flume was covered with plywood in an attempt to produce an anaerobic condition that would kill all the plants in the first half of flume. By August 11, all the aquatic plants in the first 100 ft of the flume died and settled to the bottom. Since the concentration of both Cs^{137} and Sr^{85} in the

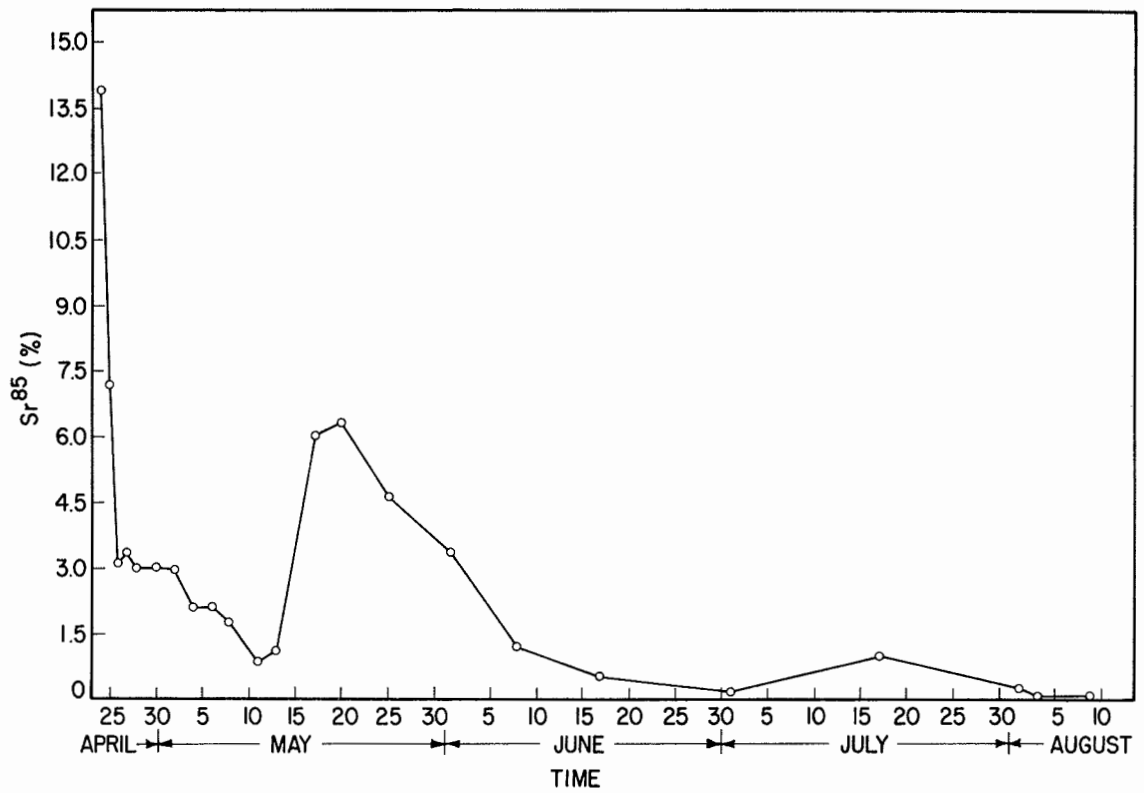


FIG. 4-22. Sr⁸⁵ SORBED BY AQUATIC PLANTS

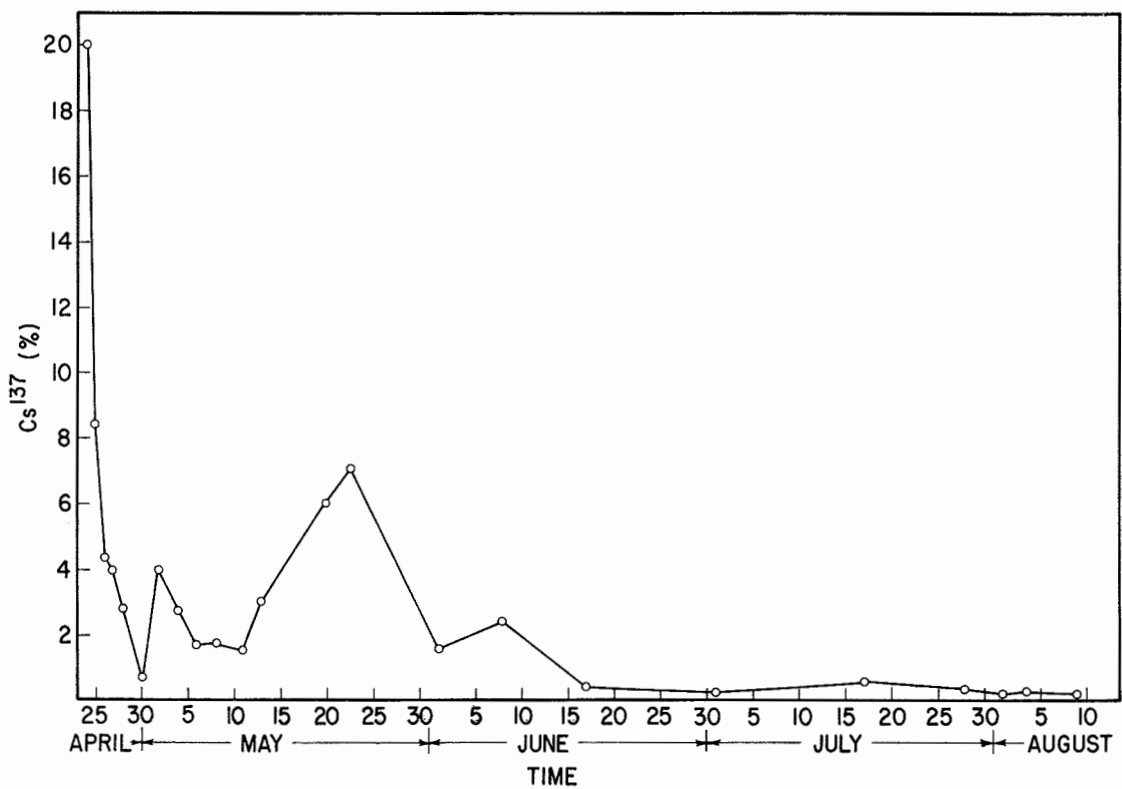


FIG. 4-23. Cs¹³⁷ SORBED BY AQUATIC PLANTS

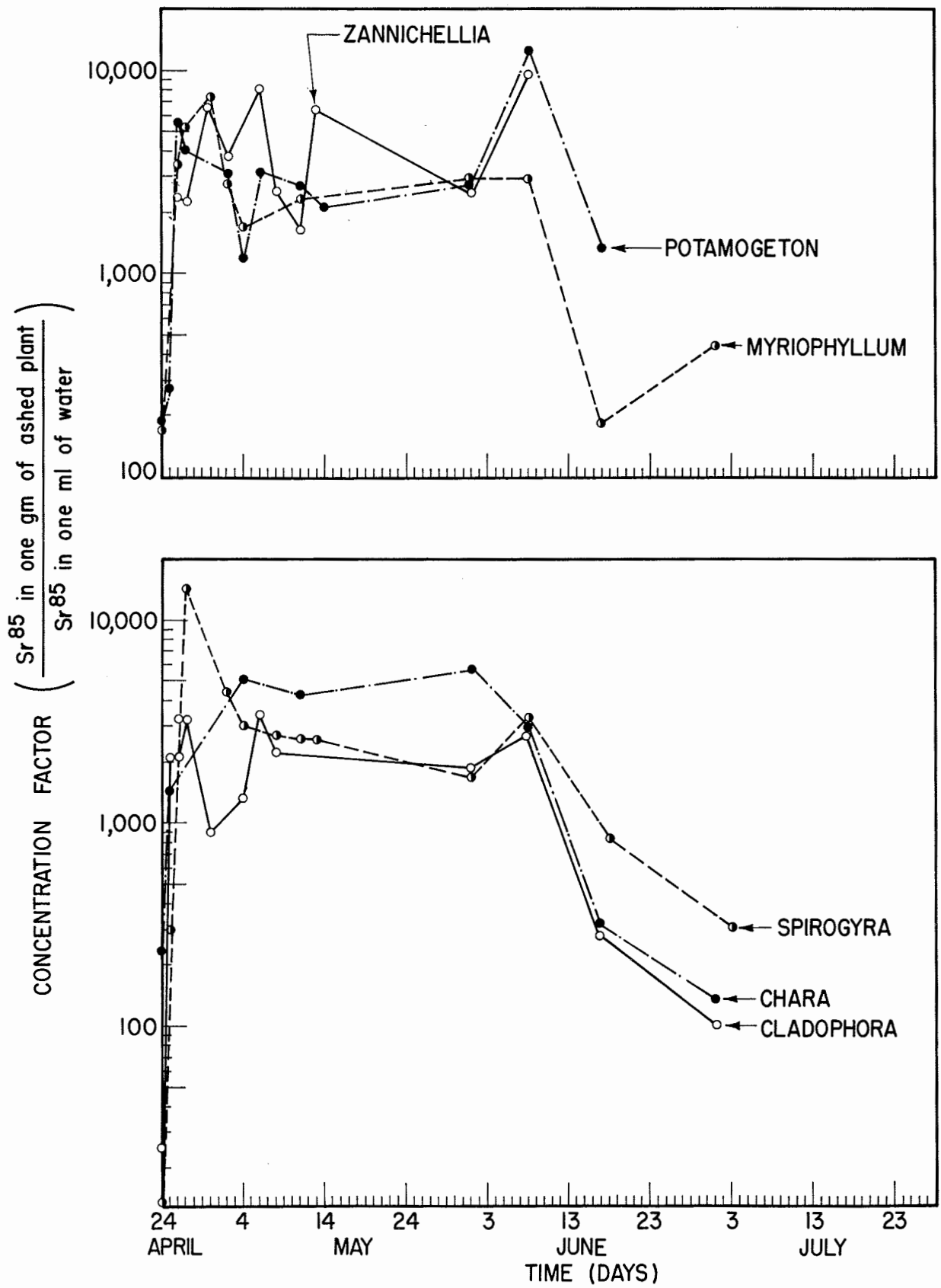


FIG. 4-24. CONCENTRATION OF Sr^{85} BY AQUATIC PLANTS

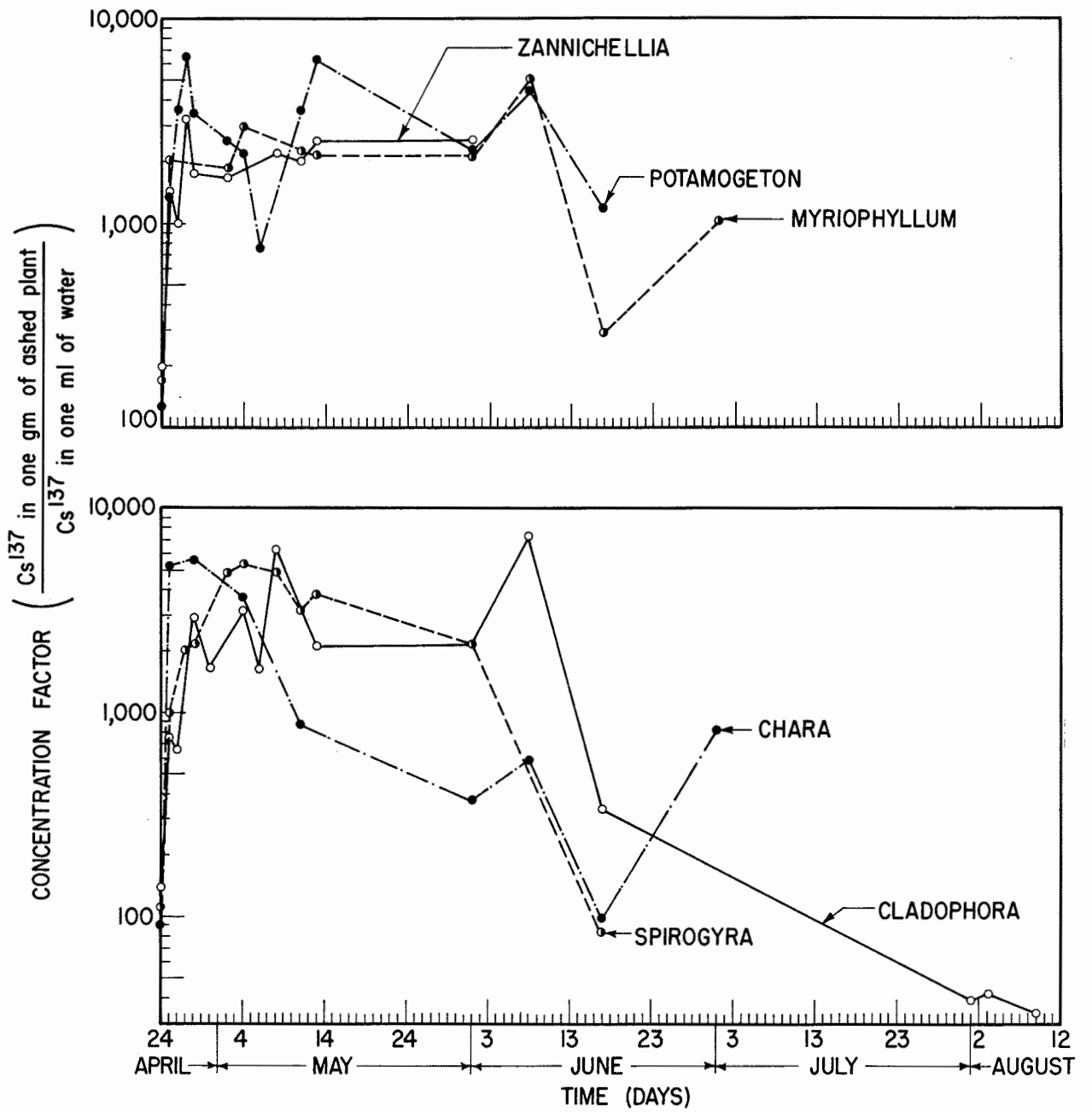


FIG. 4-25. CONCENTRATION OF Cs¹³⁷ BY AQUATIC PLANTS

plants before the flume was covered was very low, there was no significant increase of Cs¹³⁷ and Sr⁸⁵ in aqueous solutions as shown in Figs. 4-8, 4-9, 4-1, and 4-2. In Figs. 4-7 and 4-8 there were a few values that increased suddenly because of the additional suspended debris. However, in the sediment samples, there were only small increases as shown in Figs. 4-15 and 4-16. These changes were mainly caused by the settled organic debris containing radionuclides. There was a difference between the data derived from the covered area and that of the uncovered area, Fig. 4-26. ORP readings in the sediments were mostly negative, which showed that an anaerobic condition existed. The DO values dropped during the day from 8 to 2 mg/l after the flume was covered. By August 17, the flow was stopped but the distribution of radionuclides in the various components of the flume system did not change significantly. The DO values, however, dropped from 4 to 0 mg/l as shown in Fig. 4-26.

Table 4-9
Maximum Concentration Factor, K_c , of Sr⁸⁵ and Cs¹³⁷
for Freshwater Plants in the Flume

Plant	Cs ¹³⁷	Maximum K_c	Sr ⁸⁵
<u>Macroplants</u>			
1. <u>Zannichellia</u> sp.	3,200		8,060
2. <u>Potamogeton</u> sp.	6,500		12,400
3. <u>Myriophyllum</u> sp.	5,005		7,450
<u>Algae</u>			
1. <u>Spirogyra</u> sp.	5,300		14,500
2. <u>Cladophora</u> sp.	7,250		3,400
*3. <u>Chara</u> sp.	5,600		5,700

*Macroalgae

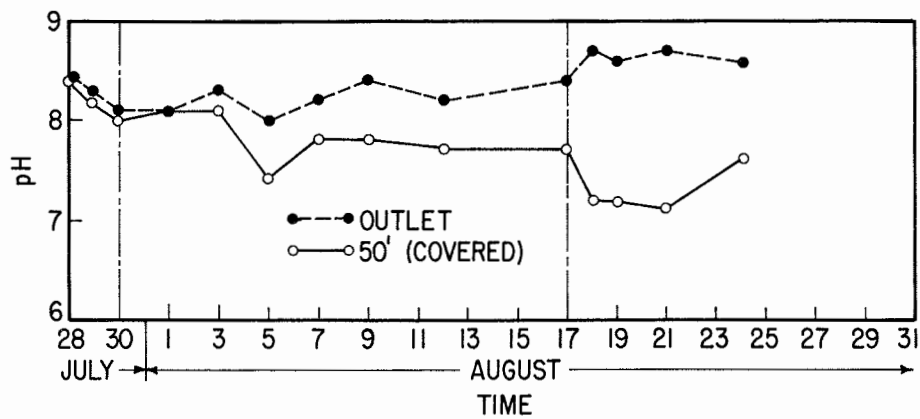
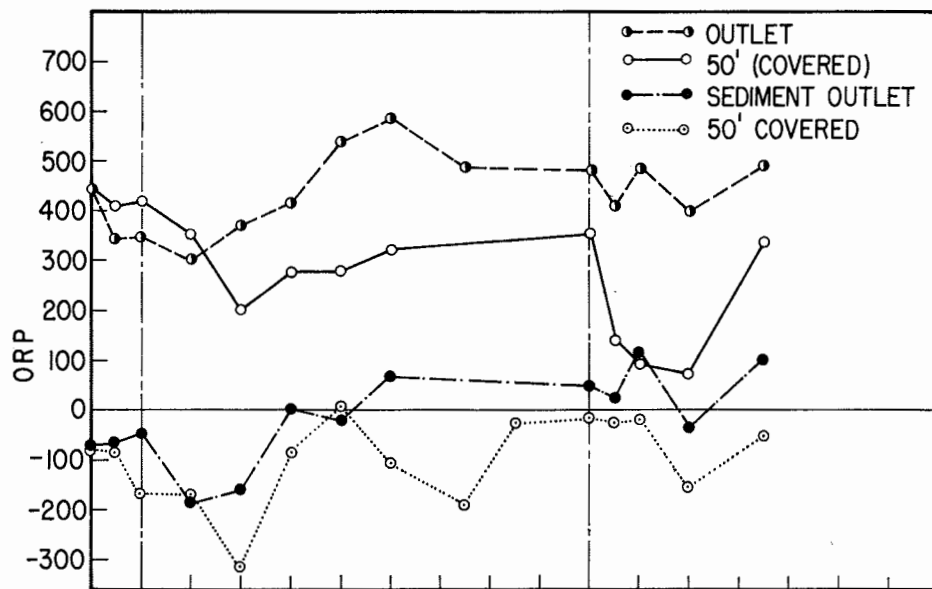
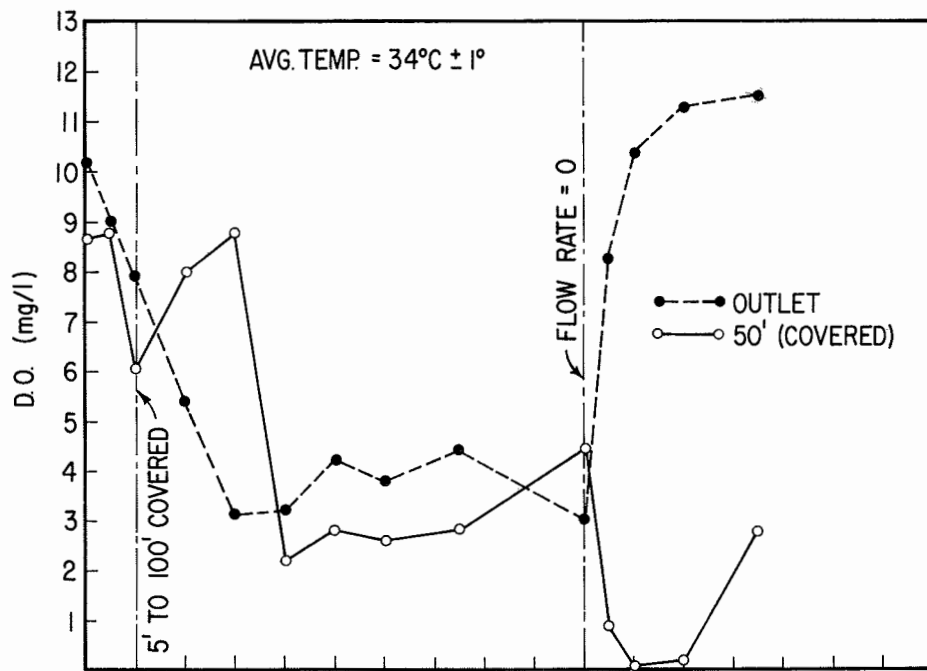


FIG. 4-26. ENVIROMENTAL FACTORS FOR RELEASE STUDY

Table 4-10
 Release of Sr⁸⁵ and Cs¹³⁷ from Aquatic Plants
 after Washing with Citric Acid

Plant	Release of Cs ¹³⁷ (%)	Release of Sr ⁸⁵ (%)
<u>Macroplants</u>		
1. <u>Zannichellia</u> sp.	14	37
2. <u>Potamogeton</u> sp.	46	20
3. <u>Myriophyllum</u> sp.	15	29
<u>Algae</u>		
1. <u>Spirogyra</u> sp.	27	23
2. <u>Cladophora</u> sp.	44	12
3. <u>Chara</u> sp.	36	19

CHAPTER V

DISCUSSION

This study showed that the transport of Cs^{137} and Sr^{85} after an instantaneous release in the flume system was primarily a function of hydrodynamic mixing; however, sorption reactions were significant. Sorptive materials included the bottom sediments, aquatic plants, and the suspended solids in the water. It was determined that the distributions of both Cs^{137} and Sr^{85} in the components of the stream were affected by such environmental factors as measured in pH, dissolved oxygen content, oxidation-reduction potential, conductivity, and light intensity. Generally, the fractions of the Sr^{85} and Cs^{137} in the aqueous solutions were larger than those of Co^{58} and Zn^{65} . These latter radionuclides are discussed in separate reports. (22,41) It was also realized that variations in uptake of Sr^{85} were greater than those of Cs^{137} .

Dispersion of Cs^{137} and Sr^{85}

In comparing the radionuclides and dye data, it was found that 80 percent of the total Rhodamine B discharged passed the 5-ft station during the first 24 hours. The amounts of Sr^{85} and Cs^{137} which passed the same distance were 66 and 61 percent, respectively. It was found that the areas under the concentration curves for the 5-ft station in Figs. 4-4 and 4-5 were greater than the corresponding areas for downstream stations. The area ratios are presented in Table 5-1 and represent the area for the 5-ft station. It appears that the difference of the area ratio between the 5-ft and 50-ft station was much larger than that between other stations. Moreover, the slope for the plot of maximum concentration along the flume

as shown in Fig. 4-6 changed significantly at about the 50-ft station.

It appears that the transverse dispersion across the flume was not complete until the plume of radionuclides reached the 50-ft station.

Table 5-1
Normalized Areas Under the Time-Concentration Curves

Isotope	Stations (Distance from Inlet in ft)				
	5	50	100	150	195
Sr ⁸⁵	1.0	0.43	0.29	0.25	0.21
Cs ¹³⁷	1.0	0.50	0.40	0.38	0.36

Estimation of Sr⁸⁵ and Cs¹³⁷ in Aqueous Solution

It was found that a linear relationship exists between the cumulative fraction value and the distance from the release point as shown in Figs. 5-1 and 5-2. These curves are plots of the data presented in Tables 4-1 and 4-2. The general formula is shown in Eq. 5-1.

$$\log F_x = \log F_0 + k x, \quad (5-1)$$

where

F_x = the cumulative fraction of the radionuclides discharged through a section x feet from point of release,

F_0 = the cumulative fraction of total radionuclides discharged through the release point after release, and

k = slope of the straight line of the plot. The k value should be negative.

The fraction of total release passing through a certain section after a given time period can be determined graphically. As cited in Figs. 5-1

and 5-2, the lines representing a certain time period following release can be approximated by the following characteristics:

- (1) No intersection between two different lines representing two different times after release.
- (2) The longer the time after release, the smaller the slope of the straight line.
- (3) The longer the time after release, the less the difference of F_o values per time interval.

Also, by knowing the total release and the fractional value as determined graphically, the concentration of radionuclide associated with the water at a particular time and place can be calculated.

Estimation of Sr⁸⁵ and Cs¹³⁷ in Sediments

The amount of radionuclides penetrating a unit depth of sediments is dependent upon the total concentration of radionuclides present. It must be assumed that the bottom sediments are graded homogeneously.

$$N = N_o e^{-ph} \quad (5-2)$$

where

N = concentration of radionuclides present at depth

N_o = concentration of radionuclides present at the interface

p = penetration constant, which is the probability of sorption per number of radionuclides present per unit path length

The first-order reaction plots are presented in Figs. 5-3 and 5-4. The slope of the line is the penetration constant, p . In each plot, it was found that the point for the first centimeter of depth was always lower than expected on the basis of a straight-line plot. The reduced value occurs

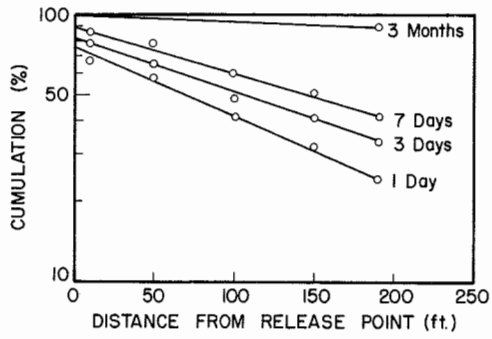


FIG. 5-1. LONGITUDINAL DISTRIBUTION OF Sr⁸⁵

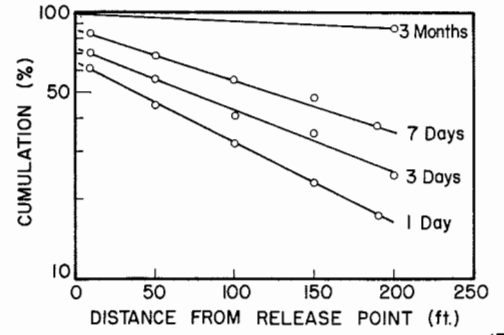


FIG. 5-2. LONGITUDINAL DISTRIBUTION OF Cs¹³⁷

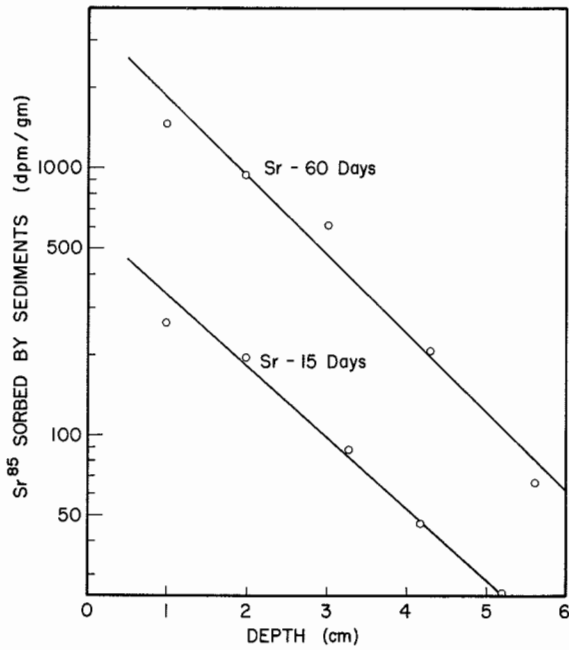


FIG. 5-3. PENETRATION OF Sr⁸⁵ INTO BOTTOM SEDIMENTS.

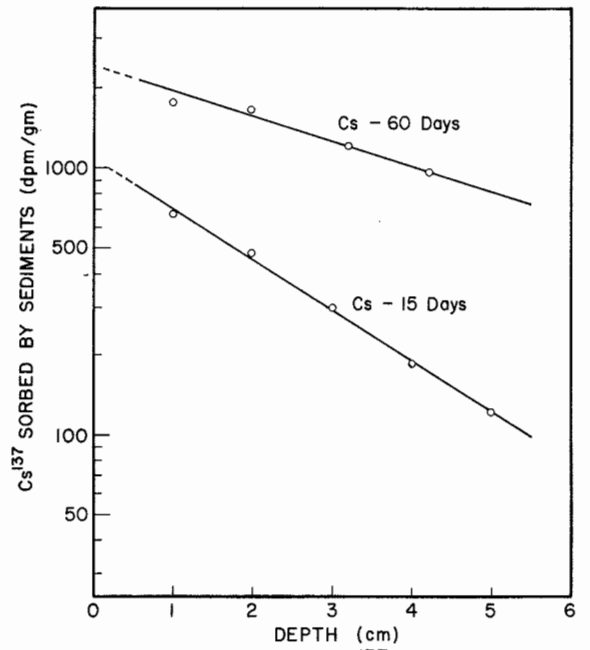


FIG. 5-4. PENETRATION OF Cs¹³⁷ INTO BOTTOM SEDIMENTS.

because of the continuous adjustment of the equilibrium between the radionuclides in solid phase and the radionuclides in liquid phase, an adjustment that is initiated by the flowing water body. However, the concentration of radionuclides in any depth can be calculated with the known value of p by Eq. (5-2).

Selectivity Coefficients for Sr^{85} and Cs^{137}

The selectivity coefficients, $K_{\text{Ca+Mg}}^{\text{Sr}}$ and $K_{\text{Ca+Mg}}^{\text{Cs}}$, are presented to characterize the uptake of Sr^{85} and Cs^{137} by bottom sediments. Gloyna and Reynolds⁽⁴⁰⁾ found that almost the entire cation exchange capacity of Lake Austin sediments is occupied by calcium and magnesium. Thus, the concentration of cations in solid phase, $q_{\text{Ca+Mg}}$, was 0.35 meq/gm which was also the cation exchange capacity of the bottom sediments in the flume. The concentration of cations, $C_{\text{Ca+Mg}}$, was 0.017 meq/ml as shown in Table 3-1. The concentrations of Sr^{85} and Cs^{137} in solid phase, q_{Sr} and q_{Cs} , and in liquid phase, C_{Sr} and C_{Cs} , were the concentrations of Sr^{85} and Cs^{137} associated with bottom sediments and water solution, respectively. The values for $K_{\text{Ca+Mg}}^{\text{Sr}}$ and $K_{\text{Ca+Mg}}^{\text{Cs}}$ were calculated as follows:

Table 5-2
Selectivity Coefficients for Sr^{85} and Cs^{137} Equilibrium in the Flume

Stations from Entrance (ft)	Average $K_{\text{Ca+Mg}}^{\text{Sr}}$	Average $K_{\text{Ca+Mg}}^{\text{Cs}}$ (gm/ml)
5	7.40×10^{-3}	9.90
50	2.58×10^{-3}	2.50
100	7.86×10^{-3}	1.75
150	1.71×10^{-3}	0.60
195	3.80×10^{-3}	2.26

As shown in Table 5-2, the average value of K_{Ca+Mg}^{Sr} and K_{Ca+Mg}^{Cs} were found to be 3.47×10^{-3} and 3.350 gm/ml , respectively. The values of K_{Ca+Mg}^{Sr} and K_{Ca+Mg}^{Cs} differ greatly from the value of 1.55 and $5.7 \times 10^6 \text{ gm/ml}$ that were obtained by Gloyna and Reynolds. However, K_{Ca+Mg}^{Cs} in the flume was much greater than K_{Ca+Mg}^{Sr} . This corresponds to the same inequality obtained by Gloyna and Reynolds. The major reason for the difference between the value obtained by Gloyna and Reynolds and that from the flume study may be due to the presence of organic debris and the special plant population in the flume.

Sr⁸⁵ and Cs¹³⁷ in Biota

For Sr⁸⁵ uptake, the various species of aquatic plants exhibited different concentration factors, K_c , as follows:

Spirogyra sp. > Potamogeton sp. > Zannichellia sp. > Myriophyllum sp. > Chara sp. > Cladophora sp.

For Cs¹³⁷ uptake, the various species of aquatic plants exhibited the following uptake order:

Cladophora sp. > Potamogeton sp. > Chara sp. > Spirogyra sp. > Myriophyllum sp. > Zannichellia sp.

BIBLIOGRAPHY

1. Ames, L. L., Effect of Base Cation on the Cesium Kinetics of Clinoptilolite, U.S.A.E.C. Report HW-SA-2452, General Electric Co., Hanford Atomic Products Operation, Richland, Wash. (Feb. 1962).
2. Ames, L. L., Kinetics of Cesium Reactions with Some Inorganic Cation Exchange Materials, U.S.A.E.C. Report HW-SA-2343, General Electric Co., Hanford Atomic Products Operation, Richland, Wash. (Dec. 1961).
3. Bowen, V. T. and Ketchum, "Biological Factors Determining the Distribution of Radioisotopes in the Sea," 2nd U.N. International Conference of Peaceful Use of Atomic Energy (September 1-15, 1958).
4. Boegly, W. J., Jr., et al., "Disposal in Natural Salt Formations," Health Physics Division Annual Progress Report for Period Ending July 31, 1962 USAFC ORNL, 13-17 (Nov. 12, 1962).
5. Brey, W. S., Principles of Physical Chemistry, Appleton-Century-Crofts, Inc., New York, 245 (1958).
6. Burkholder, Paul P., "Radioactivity in Some Aquatic Plants," *Nature*, 198, No. 4880, 601-603 (May 11, 1963).
7. Carroll, D., "Ion Exchange in Clays and Other Minerals," *Bulletin of the Geological Society of America*, 70, 749-780 (June 1959).
8. Christensen, C. W., et al., "Soil Absorption of Radioactive Wastes at Los Alamos," *J. Sew. and Ind. Wastes*, 30, 1478-1489 (Dec. 1958).
9. Daniels, F., Outlines of Physical Chemistry, John Wiley and Sons, Inc., New York, 255 (1948).
10. Dickel, G., "New Possibilities for Isotope Separation by Means of Ion Exchanges," *Nuc. Sci. Abst.*, 10, 4, 229 (Feb. 29, 1956). Original article in *Z. Elektrochem*, 54, 354-357 (1956).
11. The Disposal of Radioactive Waste on Land, Natl. Acad. of Sci., Natl. Research Council, Div. of Earth Sci. (1956).
12. Eisenbud, M., Environmental Radioactivity, McGraw-Hill Book Company, Inc., 113, (1963).
13. Foster, R. F., "The Need for Biological Monitoring of Radioactive Waste Streams," *Nuc. Engin. and Sci. Conf.*, Paper V-6 (Apr. 6-9, 1959).
14. Fujita, et al., "The Behavior of Sr⁸⁵ in a Normal Man Following a Single Injection," *Health Physics*, 9, 407-15 (April, 1963).

15. Ginell, et al. "Ultimate Disposal of Radioactive Waste," Nuc. Sci. Abst., 2, 3, 121 (Feb. 15, 1955). Original article in Nucleonics, 12, 12, 14-18 (Dec. 1954).
16. Grim, R. E., Clay Mineralogy, McGraw-Hill Book Co., New York, 27 (1953).
17. Hatch, L. P., et al., "Processes for High Level Radioactive Waste Disposal," Internat. Conf. for the Peaceful Uses of Atomic Energy, Paper No. A/Conf. I/P/553 (July 7, 1955).
18. Hayes, F. R. and Phillips, J. E., "Radiophosphorous Equilibrium Mud, Plants, and Bacteria under Oxidized and Reduced Conditions," J. Limn. and Oceanog., 3, 4, 459-475 (Oct. 1948).
19. Helfferich, F., Ion Exchange, McGraw Hill Book Co., Inc., New York, 6 (1962).
20. Jacobs, D. G. and Tamura, T., "Improving Cesium Selectivity of Bentonites by Heat Treatment," Health Physics, 5, 149-154 (1961).
21. Jacobs, D. G., et al., "Soil Column Studies," Health Physics Division Annual Progress Report for Period Ending July 31, 1960, U.S.A.E.C., ORNL, 78-95 (Oct. 24, 1960).
22. Yousef, Y. A. and Gloyna, E. F., The Transport of Co⁵⁸ in An Aqueous Environment, Tech. Report - 7 AEC Contract AT(11-1)-490 (Dec. 1, 1964).
23. Jacob, D. G. and Sealand, O. M. "Liquid Injection into Permissible Formation," Health Physics Division, ORNL-3347 (July 31, 1962).
24. Kennedy, V. S., Personal Communication, U.S. Department of Interior, Geological Survey, Denver (Sept. 8, 1960).
25. Krumholz, L. A., et al., "Etiological Factors Involved in the Uptake, Accumulation, and Loss of Radionuclides by Aquatic Organisms," The Effects of Atomic Radiation on Oceanography and Fisheries, Natl. Acad. of Sci. - Natl. Research Council, Pub. No. 551, Washington, D. C., 77 (1957).
26. Krumholz, L. A., et al., "Accumulation and Retention of Radioactivity from Fission Products and Other Radiomaterials by Fresh-water Organisms," The Effects of Atomic Radiation on Oceanography and Fisheries, Natl. Acad. of Sci. - Natl. Research Council, Pub. No. 551, Washington, D. C., 91 (1957).
27. Lacy, W. J., "Decontamination of Radioactively Contaminated Water by Slurrying with Clay," Ind. and Engin. Chem., 46, 5, 1061 (May 1954).
28. Lowman, F. G., "Marine Biological Investigations at Eniwetok Test Site," Proceedings on Disposal of Radioactive Wastes, International Atomic Energy Agency, Vienna (1960).

29. Loven, A. W., A Study of Ion Exchange Properties of Natural and Heated Montmorillonite, Doctoral Dissertation, University of North Carolina (1962).
30. McBain, J. W., Colloid Science, D.C. Heath and Co., 450 (1950).
31. McHenry, J. R., Adsorption and Retention of Cesium by Soils of the Hanford Project, U.S.A.E.C. HW31011 (Mar. 2, 1954).
32. McHenry, J. R., Adsorption and Retention of Strontium by Soils of the Hanford Project, U.S.A.E.C. HW34499 (Feb. 1, 1955).
33. Mercer, B. W., Jr., The Removal of Cesium and Strontium from Condensate Wastes with Clinoptilolite, U.S.A.E.C. Report HW-66276, General Electric Co., Hanford Atomic Products Operation, Richland, Wash. (July 29, 1960).
34. P. J. Morton, et al., Status Report No. 2 on Clinch River Study, ORNL, 34 (Apr. 13, 1962).
35. Morton, et al., Status Report No. 4 on Clinch River Study, ORNL, 25-32 (Sept. 25, 1963).
36. Parker, F. L., et al., Health Physics Division Annual Progress Report, U.S.A.E.C., ORNL, 2994, 57 (July 31, 1960).
37. Pendleton, R. C. and Hanson, W. C., "Absorption of Cesium-137 by Components of an Aquatic Community," Second U.N. Internat. Conf. on the Peaceful uses of Atomic Energy, Paper No. A/Conf.15/P/392 (June 1958).
38. Patterson, C.C. and Gloyna, E. F., The Dispersion of Radionuclides in Open Channel Flow, Tech. Report - 2, U.S.A.E.C. Contract, AT(11-1)-490 (June 1, 1960).
39. Raymond, H., Uptake of Strontium by Roots of Zea Mayo, Plant Physiology, 38, No. 2, 180-184 (March 1963).
40. Reynolds, T. and Gloyna, E. F., Radioactivity Transport in Water-- Treatment of Strontium and Cesium by Stream and Estuarine Sediments, Tech. Report 1, U.S.A.E.C., Contract No. AT(11-1)-490, 7, The University of Texas (1963).
41. Rowe, D. R. and Gloyna, E. F., Radioactivity Transport in Water-- Transport of Zn⁶⁵ in an Aqueous Environment, Tech Report 4, U.S.A.E.C., Contract AT(11-1)-490, The University of Texas (1964).
42. Robinson, B. P., "Ion-Exchange Minerals and Disposal of Radioactive Wastes-A Survey of Literature," Geological Survey Water-Supply Paper 1616, U.S. Geological Survey (1962).

43. Sayre, W. W., et al., Progress Report: Dispersion and Concentration of Radioactive Wastes by Streams, U.S.G.S. CER59ARC26, Denver, 35 (1959).
44. Story, A. H. and Gloyna, E. F., Radioactivity Transport in Water-- Environmental Behavior of Nitrosylruthenium, Technical Report 3, U.S.A.E.C., Contract AT(11-1)-490, The University of Texas (1963).
45. Straub, C. P. and Krieger, H. L., "Removal of Radioisotopes from Waste Solutions--Soil Suspension Studies," Paper presented at the 8th Industrial Waste Conference, Purdue University, Lafayette, Indiana (May 6, 1953).
46. Struxness, E. G., et al., "Clinch River Studies," Health Physics Division Annual Progress Report for Period Ending July 31, 1960, U.S.A.E.C., ORNL, 48 (Oct. 24, 1960).
47. Sverdrup, H. V., Oceanography for Meteorologists, Prentice-Hall, Inc., New York, 9 (1943).
48. Tamura, T., et al., Ion Exchange of Minerals, Health Physics Division, ORNL (July 31, 1962).
49. Taylor, Sir G. I., "Diffusion by Continuous Movements," Proc London Mathematical Soc., Ser 2, 20, 196-212 (1921).
50. Thomas, H. C. and Frysinger, G. R., "Adsorption Studies on Clay Minerals. VII. Yttrium - Cesium and Cerium (III) - Cesium on Montmorillonite," Jour. of Physical Chem., 64, 224-228 (1960).
51. Thomas, I. E., "Dispersion in Open Channel Flow," Doctoral Dissertation, Northwestern University (1958).
52. Thomas, H. C. and Tamers, M. A., "Ion-Exchange Properties of Kaolinite Slurries," Jour. of Physical Chem., 64, 29-32 (1960).
53. Wahlberg, J. S. and Fishman, M. J., "Adsorption of Cesium on Clay Minerals," Bulletin 1140.A, U.S. Geological Survey (1962).
54. Western, F., "Problems of Radioactive Waste Disposal," Nuc. Sci. Abst., 1, 7 (Sept. 30, 1948). Original article in Nucleonics, 3, 2, 43-49 (Aug. 1948).
55. Wallace, D. L., et al., Disposal by Hydraulic Fractioning, Health Physics Division, ORNL, (July 31, 1962).

APPENDIX

Print out GAMA2

```

-FTN,L,E,P.
  PROGRAM GAMA2
C    PROGRAM BY C. S. SHIH  SEPT. 1964
  COMMON/MM/III(300)
  DIMENSION JJJ(300),IJK(8),A(8),KII(8),KJI(8),  FRAC(8,8),F(8,9),
  *COUNT(8),KJJ(8),EFF(8),ISO(8),IX2(8),IX1(8),IKI(8), EV(8),DIS(8),
  *IBACK(300),ABS(8),DECAY(8)
  1  FORMAT( 5X,22HPAPER TAPE ROLL NUMBER  F6.2 )
  2  FORMAT(/50X,15,6F10.2/      )
  3  FORMAT(2X,3HJOB 5X,7HISOTOPE 5X,12HPOTOPEAK AT 5X,12HUPWARD SHIFT
  *5X,15HDISCENTGRATIONS
  */1X,6HNUMBER 14X,14HCHANNEL NUMBER 4X,12HOF PHOTOPEAK 8X,
  * ,10HPER MINUTE///)
  4  FORMAT(8I10)
  5  FORMAT(4I5,A8,2F8.3,5F6.4)
  6  FORMAT(//10X,4I10)
  7  FORMAT(6F10.3)
  8  FORMAT(2X,I4,6X,A8,7X,I5,12X,I5,5X,F15.3)
777  FORMAT(///55X,5F12.2)
888  FORMAT(8F10.2)
999  FORMAT(1H1 )
  CALL TIME
  READ 4,N,MMN
  M = N + 1
  DO 10 K = 1,N
10  READ 5,I,KJJ(I),KJI(I),KII(I),ISO(I),EFF(I),DECAY(I),
  *(FRAC(I,J),J=1,N)
42  IF(MMN)299,299,43
43  READ 888,CNN,AMM,DAYS,BOX
  READ 7, (ABS(J),J=1,N)
  PRINT 999
  PRINT 1,BOX
  PRINT 3
  IRLM = 0
  CALL CON(IABC)
  IF(IABC)199,44,199
44  DO 45 I = 1,256
45  IBACK(I) = III(I)
46  CONTINUE
  IRLM = IRLM + 1
  CALL CON(IABC)
  IF(IABC)199,47,199
47  DO 50 I = 1,256
  JJJ(I)=III(I)-(IBACK(I)*AMM)/CNN
  IF(JJJ(I))48,50,50
48  JJJ(I) = 0
50  CONTINUE
  DO 20 IX = 1,N
  IJK(IX) = 0
  IX2(IX) = 0
  IDELT1 = KII(IX) -KJJ(IX)
  KI = KJI(IX)
101  KJ = KI - 1
  IF(JJJ(KI)-JJJ(KJ))11,12,12
  11  KI = KJ

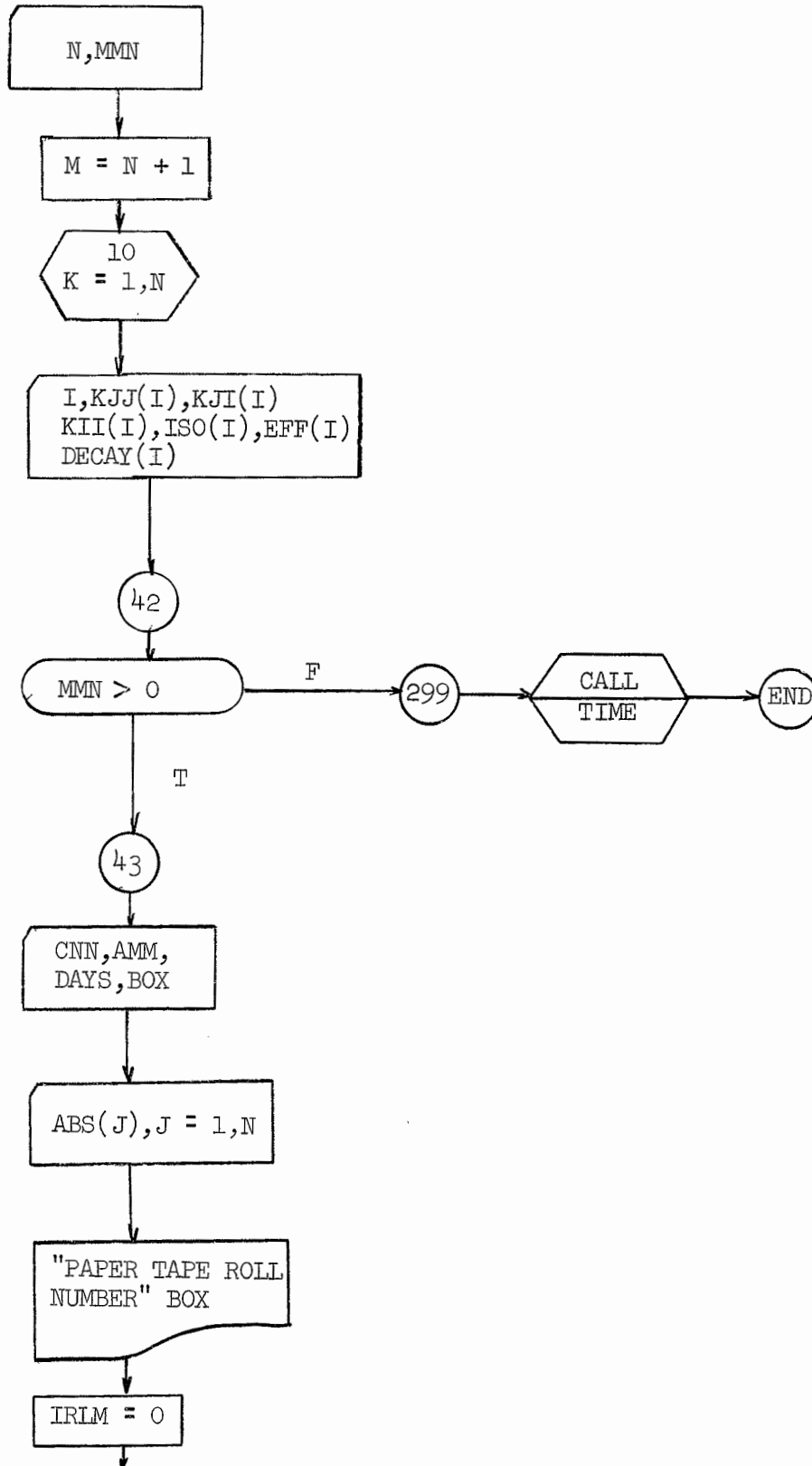
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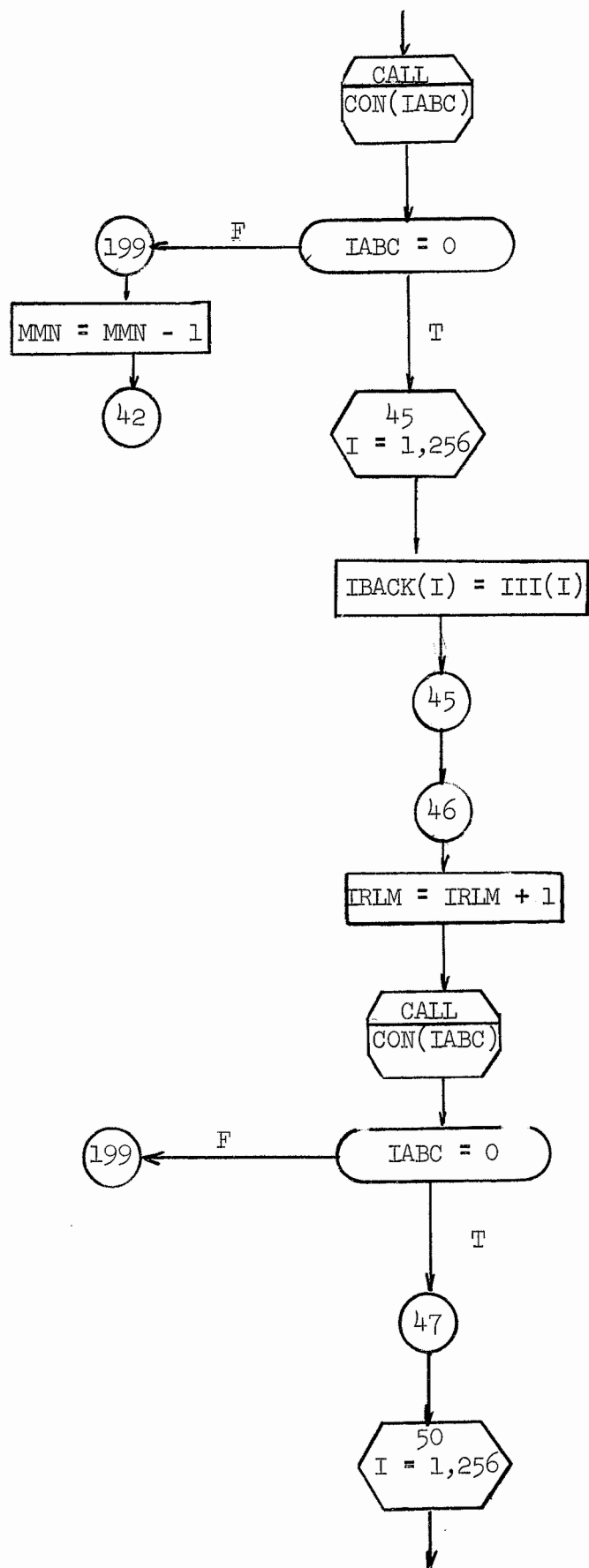
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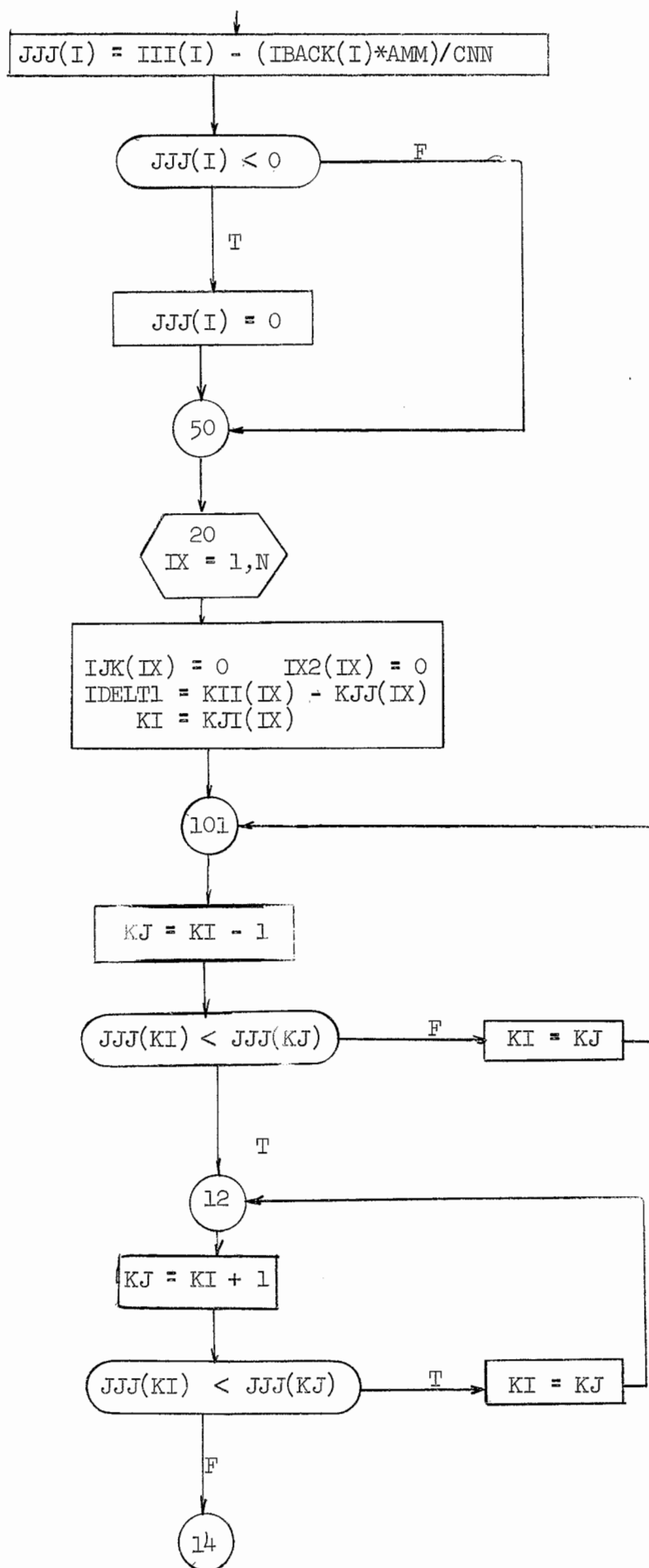
      GO TO 101
12   KJ = KI + 1
      IF(JJJ(KI)-JJJ(KJ))13,14,14
13   KI = KJ
      GO TO 12
14   I = 0
      IX2(IX) = KI - KJI(IX)
145  IF(ABSF(IX2(IX)).LT.5)196,156
156  I2 = JJJ(KI)/2
15   K = KI - I
      IF(JJJ(K)-I2)17,17,16
16   I = I + 1
      GO TO 15
17   J = K
      I = 0
18   K = KI + I
      IF(JJJ(K)-I2)195,195,19
19   I = I + 1
      GO TO 18
195  IDELT2 = K - J
      IF(IDELT2-IDELT1-2)197,197,399
399  IF(IDELT2-IDELT1+2)197,197,196
196  CONTINUE
      J = KJJ(IX)
      K = KII(IX)
      KI = KJI(IX)
197  IKI(IX) = KI
      DO 20 IA=J,K
          IJK(IX) = IJK(IX) + JJJ(IA)
20   A(IX) = IJK(IX)
      DO 23 I=1,N
          DO 23 J=1,M
              IF(J-M)21,22,22
21   F(I,J) = FRAC(J,I)
      GO TO 23
22   F(I,J) = A(I)
23   CONTINUE
      PRINT 777,(A(K),K=1,N)
      CALL GAUSS2(N,1,.000001,F,COUNT,LKL)
      PRINT 2,LKL,(COUNT(I),I=1,N)
      NR = N - 1
      DO 24 I = 1,NR
          DIS(I) = COUNT(I)*EXPF(DAYS *.693/DECAY(I))/(EFF(I)*AMM*ABS(I))
24   PRINT 8, IRLM , ISO(I), IKI(I), IX2(I), DIS(I)
      GO TO 46
199  MMN = MMN - 1
      GO TO 42
299  CONTINUE
      CALL TIME
      PRINT 999
      END

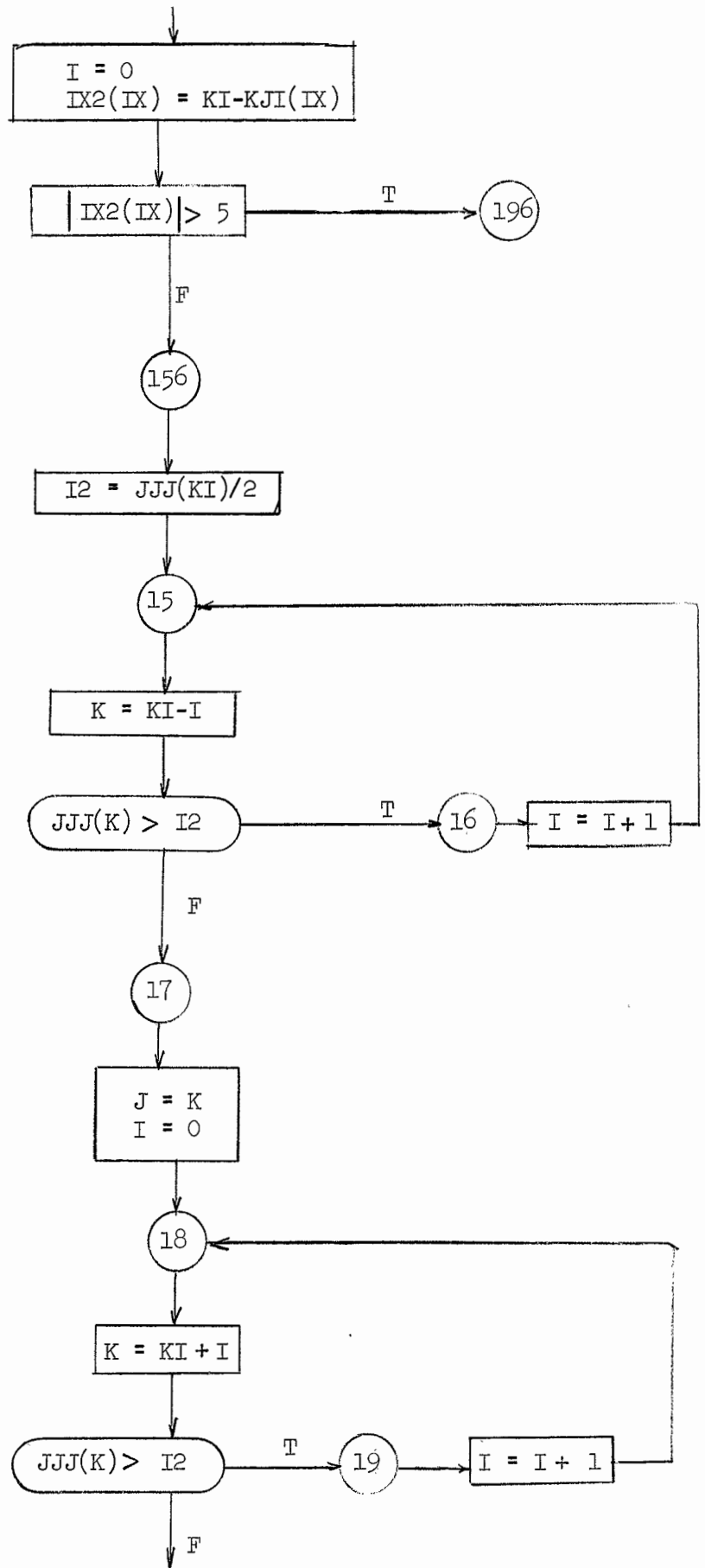
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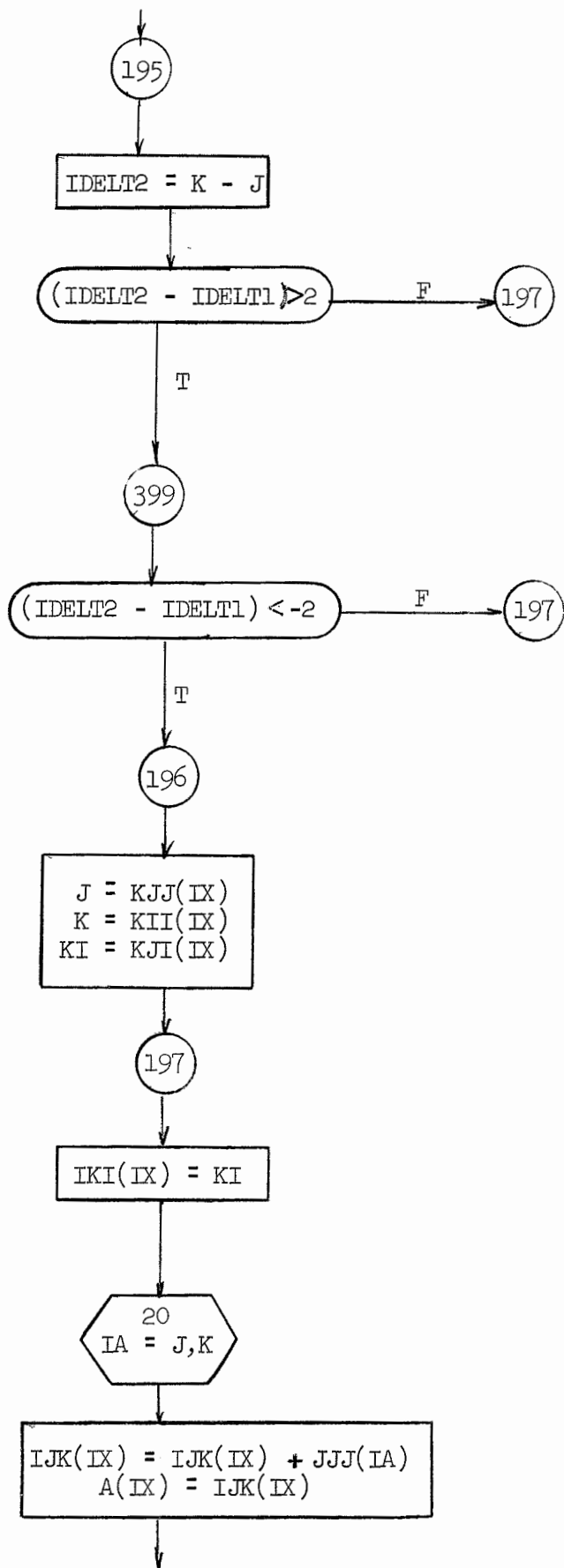
Flow chart in Fortran

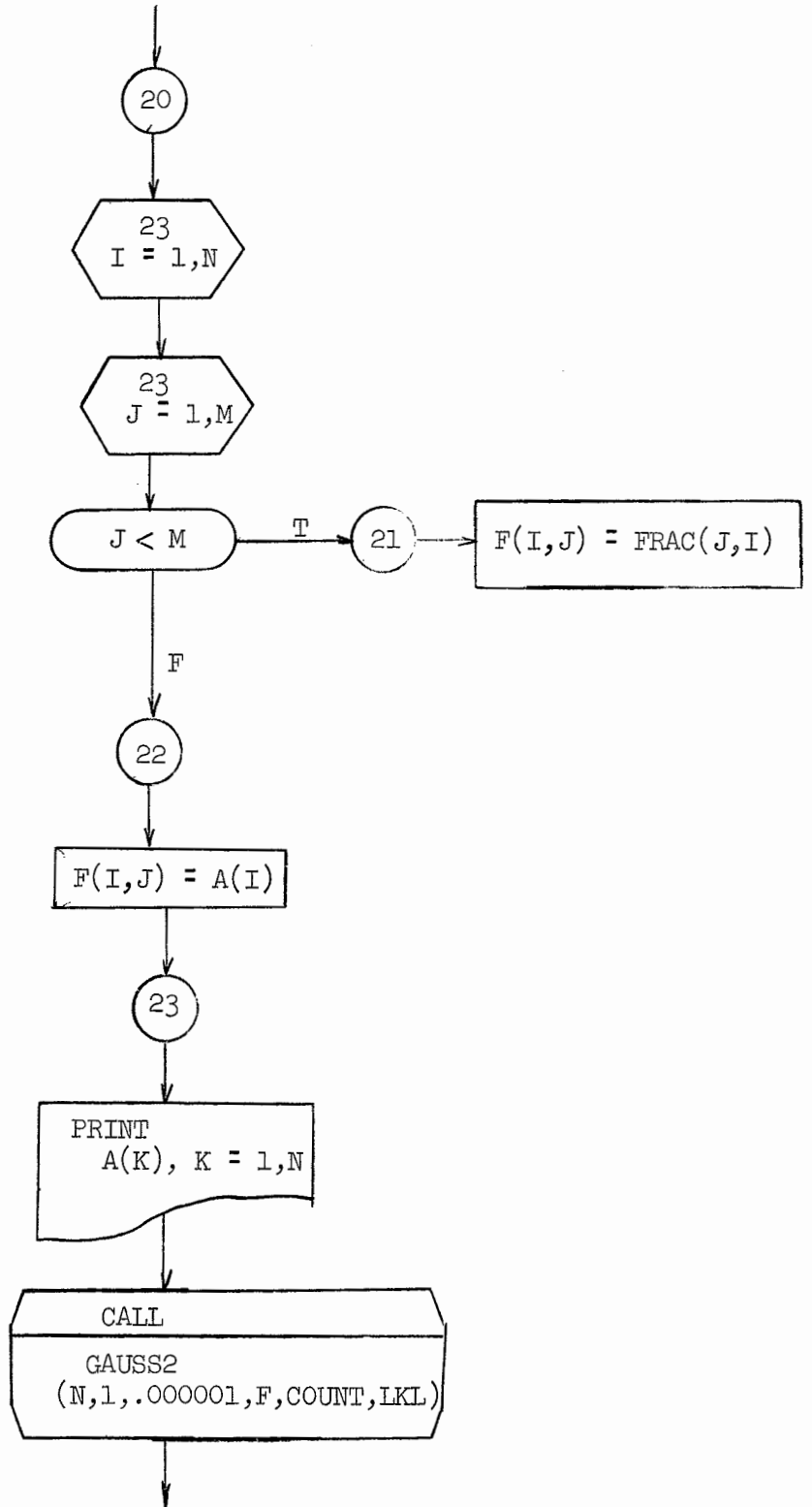


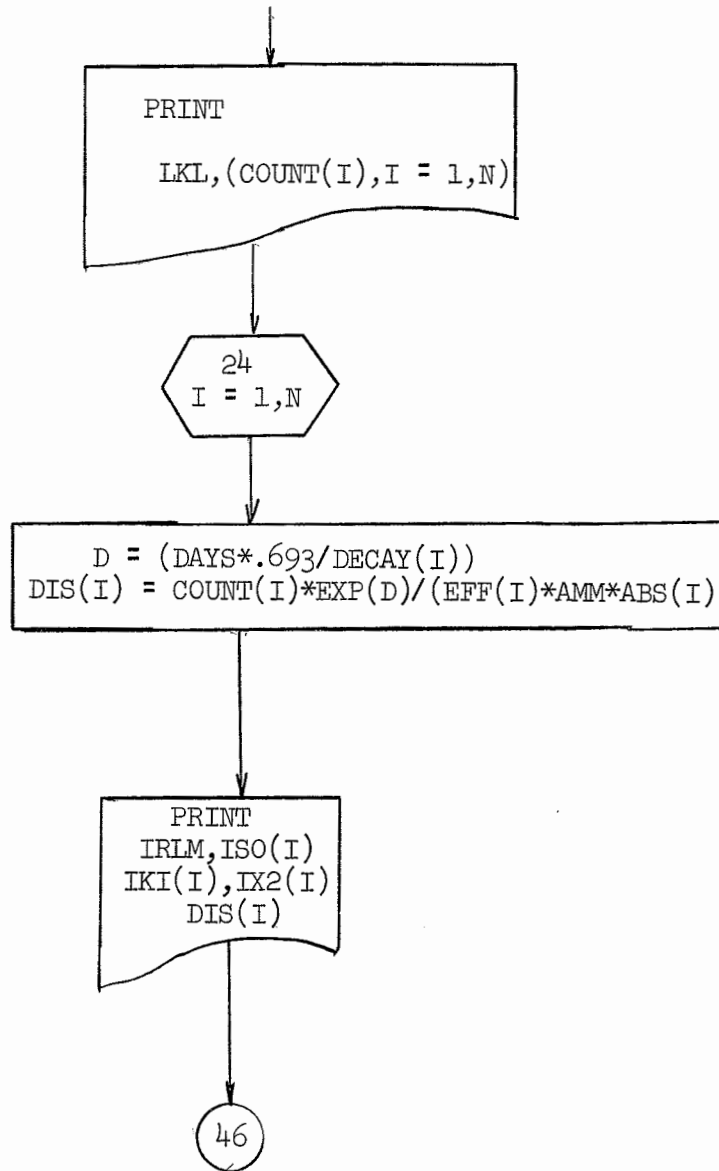












APPENDIX C

Print out Subroutine CON(IABC) and GAUSS2

```

SUBROUTINE GAUSS2(N,M,EP,A,X,KER)
DIMENSION A(8,9),X(1,8)
NPM=N+M
10 DO 34 L=1,N
    KP=0
    Z=0.0
    DO 12 K=L,N
        IF(Z-ABSF(A(K,L)))11,12,12
11 Z=ABSF(A(K,L))
        KP=K
12 CONTINUE
        IF(L-KP)13,20,20
13 DO 14 J=L,NPM
        Z=A(L,J)
        A(L,J)=A(KP,J)
14 A(KP,J)=Z
20 IF(ABSF(A(L,L))-EP)50,50,30
30 IF(L-N)31,40,40
31 LP1=L+1
    DO 34 K=LP1,N
        IF(A(K,L))32,34,32
32 RATIO=A(K,L)/A(L,L)
        DO 33 J=LP1,NPM
33 A(K,J)=A(K,J)-RATIO*A(L,J)
34 CONTINUE
40 DO 43 I=1,N
    II=N+1-I
    DO 43 J=1,M
        JPN=J+N
        S=0.0
        IF(II-N)41,43,43
41 IIP1=II+1
        DO 42 K=IIP1,N
42 S=S+A(II,K)*X(K,J)
43 X(II,J)=(A(II,JPN)-S)/A(II,II)
        KER=1
        GO TO 75
50 KER=2
75 CONTINUE
END

```

```

SUBROUTINE CON(IABC)
COMMON/MM/JJJ(300)
DIMENSION II(4000)
1  FORMAT(10I10)
2  FORMAT(5X,12HBAD SPECTRUM  )
   PIE = 3.1415926536
   IABC = 0
   DO 5 I=1,4000
5  II(I) = 0
   DO 6 I = 1,300
6  JJJ(I) = 0
11  J = 1
   L = 1
   K = J + 49
   BUFFER IN (20,2)(II(J),II(K))
12  IF(UNIT,20)12,13,104,13
13  IF(II(L))17,14,17
14  L = L + 1
   IF(L-50)13,13,11
15  CONTINUE
   J = J + 50
   K = K + 50
   BUFFER IN (20,2)(II(J),II(K))
16  IF(UNIT,20)16,17,104,17
17  L = 0
   IF(K-4000)18,18,100
18  L = L + 1
   IL = L
   IF(L- K)19,19,15
185 L = L + 1
   IF(L- K)19,19,15
19  IF(II(L))20,18,20
20  IF(II(L)-16)185,21,185
21  L = L + 1
   IF(L- K)215,215,15
215 IF(II(L)-16)22,21,22
22  IF(II(L))23,18,23
23  IY = L $      I = 0
29  J = J + 50
   K = K + 50
   L = L + 50
   BUFFER IN (20,2)(II(J),II(K))
   IF(K-4000)30,30,100
30  IF(UNIT,20)30,31,104,31
31  IF(II(K))29,33,29
33  IF(L-IL-1750)200,335,335
335 L = IY
34  L = L + 1
   IF (II(L).EQ.0)37,35
35  IF(II(L)- 16)34,36,34
36  I = I + 1

```

```

I6 = (II(L-1)-1)/2
I5 = (II(L-2)-1)*5
I4 = (II(L-3)-1)*50
I3 = (II(L-4)-1)*500
I2 = (II(L-5)-1)*5000
I1 = (II(L-6)-1)*50000
JJJ(I) = I1+I2+I3+I4+I5+I6
GO TO 34
37 L = L + 1
   IF(II(L).EQ.0)40,35
40 IF(200.LT.I.AND.I.LT.260)41,42
41 RETURN
42 IF(L.LT.200)34,43
43 PRINT 2 $          L1 = L - 25 $          L2 = L + 24
   PRINT 1,L,K,I,(II(N),N=L1,L2)
   RETURN
100 PRINT 2
    PRINT 1,L,K,I
101 IY = IY + 1
    IF(II(IY).EQ.0)102,101
102 L1 = IY - 80 $          L2 = IY + 60
    PRINT 1,L,K,I,(II(N),N=L1,L2)
500 L = 0
    J = 1
    K = 50
    BUFFER IN (20,2)(II(J),II(K))
503 IF(UNIT,20)503,501,104,501
501 IF(II(K).EQ.0)502,500
502 RETURN
200 PRINT 2
    PRINT 1,L,K,I
    RETURN
104 CONTINUE
    IABC = 1
    RETURN
    END
    END
    FINIS

```