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Technical Report to the

ATOMIC ENERGY COMMISSION

EHE-73-01

CRWR-97

Radioactivity Transport In Water ---
Final Report

THE UNIVERSITY OF TEXAS AT AUSTIN

CENTER FOR RESEARCH IN WATER RESOURCES

ENVIRONMENTAL HEALTH ENGINEERING

CIVIL ENGINEERING DEPARTMENT

MASTER

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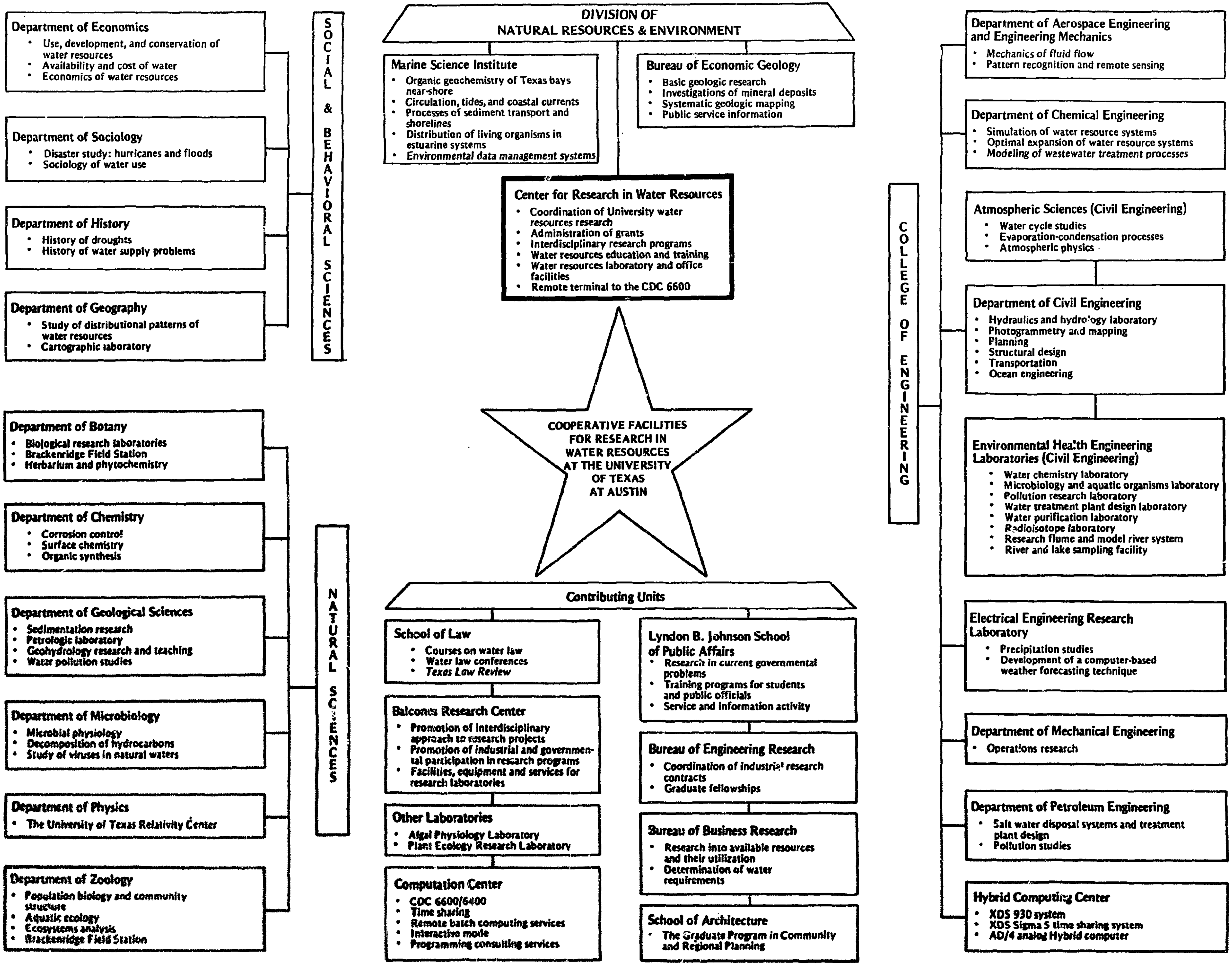

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ORO. 23

RADIOACTIVITY TRANSPORT IN WATER --
FINAL REPORT

Technical Report -- 22
to the

U. S. Atomic Energy Commission
Contract AT(11-1)-490

by

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ABSTRACT

A series of studies, "Radioactivity Transport in Water," by E.F. Gloyna and associates (1963-1972) were undertaken to obtain a more accurate concept of the transport of radionuclides in aquatic systems. Various stages of this research were completed and published previously in progress reports. These are listed in the references (1-22). These reports describe the dispersion, uptake, release, and transport of strontium, cesium, ruthenium, cobalt, and chromium. Transport data were obtained by conducting laboratory, model river, and field tests. Canal, river, and estuarine environments provided the necessary field information on hydrodynamic dispersion and on uptake and release of radionuclides by sediments. Model river data provided most of the useful uptake and release information involving special sediments, plants, and aquatic environments.

Technical Report No. 20, an evaluation report, summarized the factors affecting radionuclide transport and assessed much of the data obtained from small-scale, ecosystem experiments and model river studies. These results and the simulation techniques that developed as a result of these studies were published in Technical Report Numbers 1 through 19.

Technical Report No. 21 described the use of model rivers to study continuous release of radionuclides. The experimentally determined parameters were applied to predictive models and these relationships were checked against some observed data.

The primary objective of the work reported in Technical Report No. 22 was to evaluate the influence of various environmental factors on the accumulation and desorption of mercurials by a fresh water alga. Radioactive isotopes of mercury in the form of mercuric mercury and methyl mercury were used.

The purpose of this report is to present a final summary of the research conducted at The University of Texas at Austin on "Radioactivity Transport in Water."

CONCLUSIONS

1. Small-scale ecosystems established in aquaria, control reservoirs and flumes are useful tools for simulating stream processes and evaluating the complex factors which interact with various pollutional stresses. Response measurements of the aquatic system to conventional pollutional stresses aid in the development and verification of radionuclide transport models.

2. The following equation describes the rate of radionuclide movement in an aquatic system:

$$\frac{\partial C}{\partial t} = D_x \frac{\partial^2 C}{\partial x^2} - U \frac{\partial C}{\partial x} - \sum_{i=1}^{n} f_i k_i \left[G(C) - C_i \right]$$

The first two terms define the mixing characteristics and dilution while the third term establishes the uptake and release by the various aquatic surfaces. For dissolved wastewaters containing radionuclides, the principal mechanism of transport is hydrodynamic. However, the rate of movement is influenced by: (a) amount and type of suspended matter; (b) chemical character of the bottom sediments and aqueous system; and (c) surface concentrations on both the bed sediment and biomass.

3. Stream pollution influences the transport of radionuclides. The physiochemical characteristics of the radionuclides, bottom sediments, and sorbing surfaces change materially under environmental stress caused by various types of pollution. Pollutional stresses change the rate of uptake and release of radionuclides on surfaces and thereby increase or decrease the transport as predicted on a hydrodynamic basis alone.

4. Environmental factors and growth stimulants such as light, temperature and nutrients may exert a strong influence on initial uptake and subsequent release of certain radionuclides. As the photosynthesis/respiration ratio (P/R) exceeds one, the concentration of radionuclides in the biomass increases, while

the concentration decreases in the sediment and water. When the P/R ratio is less than one, there is a net gain of radionuclides in the sediment and water, and there is an accompanied net loss from the plants.

5. Plants, as a general rule, play an insignificant role in radionuclide transport. In cases of batch releases of radionuclides, plants will show a relatively large initial uptake, after which the specific activity of plants decreases rapidly to some equilibrium level of release.

6. Sorption of radionuclides by plant surfaces is almost completely reversible, while uptake by bed sediments may be only partially or slowly reversible. The decrease of ^{137}Cs from bed sediments in the flume was less than one percent per month. Continuous agitation of bed sediments containing ^{65}Zn, ^{58}Co, and ^{51}Cr with fresh water resulted in releases of only 5 to 10 percent of the radioactivity after a contact time of one week. However, when sediments were mixed in saline water more than 40 percent of the ^{137}Cs and ^{85}Sr was released.

7. Radionuclides are concentrated by and diffused through bed sediments. In these experiments the specific activity of the bed sediments decreased exponentially with depth. After 15 to 30 days of contact time, the specific activity of the sediments, at a depth of about 2 cm for ^{65}Zn and 14 cm for ^{137}Cs, was less than 0.1 percent of the specific activity near the surface.

8. Under continuous release of radionuclides into aquatic systems, bottom sediments and plants will concentrate radioactivity until equilibrium is reached. Under identical flow conditions plants will reach equilibrium much faster than bottom sediments.

9. Sorption isotherms can be used to evaluate the effect of radioactive isotopes of mercury on phytoplankton. Sorption and desorption, respectively, of radioactive isotopes of mercury can be described by a uniform function model and an exponential function model.

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1. INTRODUCTION

The transport of radionuclides in an aquatic environment becomes of increasing importance as the nuclear industry continues its rapid expansion. Reliable transport information must be available if useful predictions are to be made on the level of radioactivity at a particular time and location in a stream. The extent to which the environment will be affected and courses of remedial action to be taken can be determined by experimentation. However, experimentation on real rivers is constrained by the complexities of the physical, chemical, and biological characteristics of the river and magnitude of the cost involved. In contrast, a model river system offers savings in cost and time. Experimental flexibility can be provided because the parameters can be more easily established and controlled.

1.1 Objective

The principal objective of this research was the development of a mathematical model which might be used to predict the behavior and transport of selected radionuclides. The research was undertaken to provide information which would lead to the delineation of the important transport parameters and to determine the effects of specific environmental factors on transport.

1.2 Experimental Effort

Specifically, experiments included: (a) studies on the effects of community metabolism, of induced clay suspensions, and of pollution on transport; (b) investigations involving field measurements, laboratory tests, aquaria investigations, and model river (flume) experiments; and (c) the adaptation of these data to predictive transport equations. Laboratory and flume investigations were used to evaluate the uptake and release of radionuclides by clay minerals, sediments, and plants under different environmental conditions. The hydrodynamic, geochemical, and biochemical factors affecting the radionuclides transport were considered for each transport situation. The relative distribution of radionuclides in water, sediments, and biota of small-scale ecosystems (aquaria) was evaluated. Non-flowing and flowing ecosystems provided much useful information for the more detailed model river (flume) experiments. These ecosystems were converted aquaria having a volume of 50 liters.

An extensive experimental program was developed to measure the hydrodynamic, geochemical, and biological transport characteristics of the model river system. This phase of the study was designed to provide information on the transport of radionuclides resulting from instantaneous and continuous releases. The dispersion characteristics of the flume were determined through the use of dyes.

The model river consisted of a dual-channel flume, two circular concrete tanks, a pumping system, a regulating tank, two stilling basins with triangular weirs, and manometers. Each channel of the rectangular flume was 200 feet long, 1.25 feet wide, and 2 feet deep. The slope of the flume was adjustable in the range of zero to 0.006 ft/ft. The two reservoirs were 12 feet 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 water supply could be obtained from either city mains or open reservoirs which contained a viable phytoplankton population. The flows in the flume could be regulated from zero to approximately 200 liters per minute per channel, and the water depths could be controlled from zero to 2 feet. Bottom sediments were obtained from Lake Austin. All plants were common to local streams and ponds.

The prediction of radionuclide transport in an aquatic system becomes an intricate problem due to the prevailing unsteady-state conditions and the interaction of the numerous variables. Through the course of this study, a considerable amount of information was gained regarding the transport characteristics of specific radionuclides in aquatic systems. The most important parameters that influence the radionuclides transport are hydrodynamic characteristics, and sorption and desorption by the components of the ecosystem, primarily bed sediments and biomass. Hydrodynamic characteristics involve dispersion, dilution and movement of radionuclides, and they are also responsible for the transport of suspended materials and the physical movement of bed sediments. Sorption and desorption reflect the ability of sorbing surfaces, such as sediments, plants, and dead zones, to retain and release radionuclides. The fate of the radionuclides dispersed in a stream system depends on many complex factors and the hydrodynamic, geochemical, and biochemical factors all interact in establishing the rate of radionuclides transport.

Radionuclides which have been released into streams are dispersed by turbulent and molecular diffusion and by convection. Dispersion of hydraulic mixing of dissolved radionuclides greatly influence the rate of movement and the concentration of radioactivity released into a stream, but it is recognized that the interaction associated with different components of a stream produces a significant effect on the net transport of radionuclides. The uptake and release of radionuclides by the various components of the ecosystem (flume) was dependent on the type of radionuclide used, its chemical form and oxidation state, the environmental conditions such as temperature, dissolved oxygen (DO), pH, oxidation-reduction potential (ORP), salinity, presence of competing ions, mixing characteristics, and various pollutional stresses. Radioactivity released in aquatic systems is concentrated from the water phase by bed sediments, diffused into the sediments, and subsequently sorbed by sediments particles. The rate of transfer of radionuclides from the water phase to bed sediments is dependent on the contact time, the physical characteristics, and the mineralogical and chemical composition of the sediments. A useful expression of the radioactivity retained by the bed sediment is surface concentration factor K_s. The surface concentration factor, K_s, is the ratio of the radioactivity retained by the bottom sediment per square centimeter of bottom surface area to the radioactivity contained in one milliliter of water flowing in the system. The rate at which K_s values change with time, K_{st}, was calculated by the K_s value, which indicates the relative concentration of radionuclides by the sediment surface area, per unit of contact time. The amount of radionuclides retained by biomass in streams is usually negligible. However, when the biomass was abundant in the research flume, a significant amount of radionuclides was retained. Therefore, in a slow moving, enriched stream where the plants' growth is extensive, a significant portion of the released radionuclides may be immediately sorbed, and thereby influence the transport predictions.

Plants accumulate radionuclides in varying amounts depending on the type of radionuclide, the presence of stable elements, biological availability of the element, and the plant species. In the case of plants the concentration factor, K_p, is the ratio of the specific activity per gram of oven dry weight to the specific activity of radionuclide in solution. A surface concentration distribution coefficient for plants can be estimated from the concentration factor for plants and the areal density of plants. The areal density is the mass of plants present per unit area of bed sediments. The plants' sorption is rapid and mostly reversible. Once the source of radionuclides is discontinued, a gradual release at a rate dependent on the type of radionuclide and environmental conditions is observed. Experiments indicated that the uptake and release of radionuclides are related to the diurnal environmental changes.

A mathematical model of the radionuclide transport was formulated and the validity of the model was tested and verified in the research flume. Notably, the transport of radionuclides in the flume was closely related to community metabolism in an ecosystem. Toxic materials introduced into the stream will upset normal biological balance, alter the numbers and types of aquatic flora and fauna, and may cause a sudden release of stored radioactivity. An increase in velocity will scour bottom sediments and return some of the radioactivity to the water phase.

2. FIELD STUDIES

Field studies provided information on the distribution and dispersion of radioactivity in aquatic systems typical of the southwestern region. Samples of water, suspended solids, and bottom sediments from this region were collected, analyzed, and evaluated in terms of uptake and release of specific radionuclides.

2.1 Distribution of Radionuclides in Stream and Estuarine Sediments

Several field systems were used to study the uptake and release of ^{89}Sr and ^{137}Cs by naturally occurring aquatic sediments. The Guadalupe River, San Antonio Bay, Lake Austin, and Lake Falcon, all located in Texas (1) served as test units. The uptake of ^{89}Sr and ^{137}Cs by these sediments was primarily an ion exchange phenomenon since the uptake was stoichiometric and could be represented by equilibria relationships. The most important factors affecting ion exchange were: (a) cation exchange capacities of the sediments, (b) exchange capacities of the sediments, (c) concentration of competing cations in the water, and (d) mass action coefficients.

The distribution of ^{89}Sr and ^{137}Cs between a sediment and its associated water could be described by mass action equations and selectivity coefficients, providing that the concentration of Ca^{++}, Mg^{++}, Na^{+} and particularly K^{+} did not vary greatly. If ^{89}Sr and ^{137}Cs were present in trace concentrations, the equilibria could be expressed in terms of a distribution coefficient, K_d. It was noted, however, that the K_d isotherm for a particular water and sediment sample was dependent upon the concentration of the sediment in water, the concentration of the competing cations, the cation exchange capacity of the sediment, and the selectivity of the sediment for the various cations. Typical mass action coefficients as calculated from ^{85}Sr and ^{137}Cs uptake data derived from the Guadalupe River study were: $K_K^{Ca+Mg} = 0.4$ gm/ml, $K_{Na}^{Ca+Mg} = 21$ gm/ml, $K_{Ca+Mg}^{Sr} = 1.55$ ml/gm, and $K_{Ca+Mg}^{Cs} = 5.7 \times 10^6$ ml/gm.

The computed distribution coefficients for the sediment from the headwaters were K_d - Sr = 72 and K_d - Cs = 16,400, whereas the values for the sediment at the mouth of the river were K_d - Sr = 128 and K_d - Cs = 21,600.

The concentrations of ^{89}Sr and ^{137}Cs in the sediments were relatively uniform throughout the length of the fresh water reaches of the Guadalupe River, but the sediments from the San Antonio Bay contained lesser concentrations of the radionuclides than the river sediments. The sediments which were deposited in the bay released a portion of their radionuclide content as a result of an increase in competing ion concentration. The order of release for several radionuclides was ^{137}Cs > ^{89}Sr > ^{103}Ru > ^{59}Fe > ^{51}Cr. The uptake and subsequent release of radionuclides did not appear to be completely reversible. The amount of ^{137}Cs released was found to be related to the salinity of the water, and although the effects of salinity were studied only for ^{137}Cs, it was expected that an increase in salinity would also result in an increase in ^{89}Sr displacement. The uptake of the radionuclides was less in the salt water than in fresh water, and the greatest decrease in uptake occurred for ^{137}Cs. The mass action coefficients, K^{Cs}_{Ca+Mg}, K^{Cs}_{K}, and K^{Cs}_{Na}, were not constant but increased as the salinity increased. The change in uptake values might be explained on the basis that potassium fixation caused a collapse of some of the expanded mineral layers.

The monovalent cations were more inhibitive to the ^{137}Cs uptake than the divalent cations, and the order of competition was: $K^+ > Na^+ > Ca^{++} \approx Mg^{++}$. The divalent cations were more inhibitive to ^{89}Sr uptake than the monovalent ions; the order of competition was: $Ca^{++} > Mg^{++} > K^+ > Na^+$. The uptake of ^{89}Sr increased with an increase in pH, however, the uptake of ^{137}Cs was relatively insensitive to pH changes. Other radionuclides, ^{103}Ru, ^{51}Cr, ^{59}Fe, and ^{131}I, were also insensitive to pH variations.

The radionuclide distribution in the sediments was partly related to the grain size. In Lake Austin, the fine-grained fraction in the sediments increased progressively downstream; and the radionuclide distribution followed a similar pattern. In Lake Falcon, the entire length of the lake contained fine-grained bottom sediments and the radionuclides were distributed uniformly throughout the length of the lake. In Guadalupe River, 78 to 94 percent of the sediment can be classified as coarse-grained materials. In these samples, 66 to 94 percent, respectively, of ^{89}Sr and ^{137}Cs was associated with the coarse-grained fractions.

2.2 Ion Exchange Characteristics of Sediments

Contemporary sediment samples from preselected locations in the Guadalupe River drainage basin were analyzed for their mineral compositions, ion exchange capacity, and radionuclides sorption (6). The study revealed that the clay mineral sediments of the Guadalupe River and the San Antonio Bay are composed of a heterogeneous mixed-layered, clay mineral complex of poor crystallinity. Based on x-ray diffraction interpretation, a high percentage of amorphous material was present in the clay-sized sediments of the headwaters of the Guadalupe River.

Radionuclide sorption studies were made on the naturally occurring sediments and sediments from which the organic fraction had been removed. The highest percentage of ^{137}Cs and ^{89}Sr sorption occurred on sediments containing the natural organic fraction. The average ^{137}Cs sorption by samples containing organic material ranged between 91 and 93 percent versus a slightly lesser amount for treated (organic free) samples. Lower sorption values were observed for ^{89}Sr. Untreated samples removed on the average about 55 percent of the ^{89}Sr, while the treated samples removed about 45 percent of the ^{89}Sr. The highest rates of ^{89}Sr and ^{137}Cs sorption occurred during the first few minutes of contact, and the amounts of radionuclides sorbed by sediments gradually increased with time.

Ion exchange capacities of the fractionated sediments derived from the Guadalupe River and the San Antonio Bay showed variations ranging from 11 to 43 meq of barium/100 grams of sediments. Because the non-clay mineral content of these sediments ranged from 32 to 65 percent, some variations in exchange capacity for the bulk sample was expected. A recalculation of the exchange to remove the effect of the non-clay minerals increased the bulk exchange capacities, providing a range of 20 to 73 meq of barium/100 grams of clay sediments.

2.3 Dispersion of Radionuclides in Open Channel Flow

The dispersion of contaminants within a flowing system is a significant factor in the transport of radioactive wastes. Therefore, detailed longitudinal and transverse dispersion measurements were made using rhodamine B and fluorescein dyes. Typical locations for these measurements included: the Colorado River, Texas (a wide, shallow, and swift river), and the Pierce Canal, Texas (a narrow, shallow and slow moving canal) (2). The following equations provided a means for calculating the longitudinal and transverse dispersion coefficients. The concentration-time data used to test these equations were obtained from two-dimensional, tracer-plume observations.

$$\sigma_l^2 = \frac{4\beta K_1(1/2\beta) + K_0(1/2\beta)}{K_0(1/2\beta)} - \left[\frac{K_1(1/2\beta)}{K_0(1/2\beta)}\right]^2 \qquad (2\text{-}1)$$

$$\sigma_t^2 = \frac{\frac{4\beta a}{a\beta + a} K_1\left(\frac{a\beta + a}{2\beta a}\right) + K_0\left(\frac{a\beta + a}{2\beta a}\right)}{K_0\left(\frac{a\beta + a}{2\beta a}\right)} - \left[\frac{K_1\left(\frac{a\beta + a}{2\beta a}\right)}{K_0\left(\frac{a\beta + a}{2\beta a}\right)}\right]^2 \qquad (2\text{-}2)$$

where:

σ_l^2 = nondimensionalized longitudinal variance; $\sigma_l^2 = \frac{\sigma_x^2}{X}$

σ_t^2 = nondimensionalized transverse variance; $\sigma_t^2 = \frac{\sigma_y^2}{X}$

σ_x^2 = variance of concentration along x axis

σ_y^2 = variance of concentration along y axis

X = distance between the release and sampling points
β = D_x/UX
D_x = longitudinal dispersion coefficient
U = mean velocity of water
α = D_y/UX
D_y = transverse dispersion coefficient
a = $(U/v)^2$
v = speed of the boat
K_0 = modified Bessel function of the first kind
K_1 = modified Bessel function of the second kind.

The dispersion coefficient, D_x, as measured compared favorably with calculated D_x values. An empirical equation for calculating D_x follows:

$$D_x = 0.8 \exp 0.034\, U \sqrt{A} \qquad (2\text{-}3)$$

where:

A = the cross sectional area of the channel

Equation 2-3 was derived from channel data obtained in four different tests. The maximum longitudinal dispersion coefficient, as calculated using Colorado River test data, was 14.4 ft^2/sec. This value represented the total contribution of mixing, i.e. both transverse and vertical shear and turbulent diffusion. The longitudinal dispersion coefficient increased with distance from the release point, and this increase can be attributed to the increasing significance of transverse shear as the plume moves downstream. The longitudinal dispersion coefficients measured in the Pierce Canal varied between 1.0 and 1.2 ft^2/sec. These values represent the combined effects of two dimensional shear and turbulent diffusion. Transverse dispersion coefficients, as measured with dye tracers released at midstream in the Colorado River, varied between 0.50 and 0.55 ft^2/sec. The volume of tracer released influenced the measured dispersion coefficient because of the deviation from the assumed instantaneous injection and the influence of large eddies on different volumes of tracer.

Equation 2-4 represents the dispersed flow model. Thus, the long-term dilution of an instantaneous release containing radionuclides can be predicted.

$$C_{(x,t)} = \frac{M}{A\sqrt{4\pi D_x t}} \exp\left[-\frac{(x - Ut)}{4D_x t}\right] \qquad (2\text{-}4)$$

where:

M = total mass of radionuclides in water (mass per unit volume)

A = area of stream cross section

t = time

x = distance from the release point.

3. LABORATORY, AQUARIA and MODEL RIVER STUDIES

Research was performed in sequential phases, under various environmental conditions which simulate ecological systems typical of the southwestern region in the U.S. The program included laboratory, aquaria, and model river studies. The sorption and release of radionuclides by specific clay minerals were evaluated through laboratory tests. The influence of various parameters on the uptake and release of radionuclides by clay minerals, bottom sediments, and plants was investigated. The relative distributions of radionuclides (under different environmental conditions) in water, sediments, and biota of both non-flowing and flowing small-scale ecosystems were determined before more complex flume tests were undertaken. Each flume test involved studies of the transport of specific radionuclides. Dye studies preceded the release of radionuclides and these data were used to calculate the dispersion coefficients. In all flume studies the effects of community metabolism, clay suspensions, environmental effects and pollutional stresses were evaluated.

3.1 Transport of Ruthenium in an Aquatic Environment

Radioisotopes of ruthenium are present in most mixed fission products and nitrosylruthenium compounds are prevalent in reactor fuel reprocessing wastes. The mobility of nitrosylruthenium has been attributed to the fact that it tends to form anionic complexes which usually have low exchange capacities for soils. For this reason, considerable attention has also been given to the ionic nature of the various compounds studied.

3.1.1 Environmental Behavior of Ruthenium Complexes: The effects of various aquatic environments were investigated in a laboratory scale-model aquatic ecosystem (3). Variations in pH, chlorides, sulfides, and various exchange characteristics involving nitrate and nitrite complexes of nitrosylruthenium were evaluated. The distribution of nitrosylruthenium complexes in water, bottom sediments, suspended materials, and microorganisms provided considerable information which related to transport rates.

Under these test conditions over 50 percent of the ruthenium was associated with bottom muds and sediments. The high accumulation of ruthenium in bottom sediments was attributed to precipitation of insoluble nitrosylruthenium hydroxide and surface adsorption of complexed nitrosylruthenium species. Dead plant material accumulated approximately ten times as much activity per unit dry weight as did live plants. Aquatic animals exhibited a slight tendency to concentrate ruthenium.

Nitric acid derivatives of nitrosylruthenium include nitrato and nitro nitrosylruthenium complexes. Mononitrato and dinitrato nitrosylruthenium complexes are taken up by cation-exchange resins; tetranitrato, pentanitrato and nitro nitrosylruthenium complexes are taken by anion-exchange resins; and trinitrato nitrosylruthenium is not exchangeable.

In the presence of chlorides the nitrosylruthenium complexes are converted to nitrosylruthenium chlorides, the most stable of which is trichloro nitrosylruthenium. Nitrosylruthenium chlorides are likely to be of significance in marine systems because these compounds concentrate in the guts of animals. Chlorides of nitrosylruthenium are taken up by anion-exchange resins, while hydroxides of nitrosylruthenium appear to be exchangeable on either cation or anion resins but the latter group tends to form colloidal suspensions upon slight agitation. The data also showed that at pH values greater than 2.0, hydroxides replace nitrate and nitrite ligands, and the degree of replacement increases with increasing pH. It was concluded that the most stable compound at pH ranges of natural waters is nitrosylruthenium hydroxide, $RuNO(OH)_3$.

Illitic and bentonite clays with attached organic materials were found to exhibit a high exchange capacity for nitrosylruthenium compounds. Clays adsorbed only cationic species, but organic debris concentrated both cation and anion species. The exchange capacity of decaying plant debris was calculated to be in excess of 4 milliequivalents of ruthenium per gram (dry weight) of plant material. Nitrato and nitro nitrosylruthenium complexes form intermediate compounds by addition of chloride, hydroxide, sulfate, and other ligands to the nitrated nucleus.

3.1.2 Influence of Algae on Ruthenium Uptake: Chlorella pyrenoidosa was used to evaluate the impact algae might have on transport of various forms of radionuclides (4). This alga was chosen because it is commonly found in many nutrient-rich streams, and it has been a reference standard in study of other algae. Some of the environmental factors investigated involved variations in light, carbon dioxide, nutrients, plant population, and dead algal debris. The chemical form of the radionuclides was either nitro nitrosylruthenium or ruthenium chloride. The radioactive tracer was ^{106}Ru.

The investigation revealed that the uptake most likely occurred as a result of surface sorption which has many characteristics of ion exchange or a loose association that was suggestive of surface aggregation or adsorption. For example, the presence of bacteria greatly increased the uptake that is characteristic of ion exchange. In growing algal-bacterial cultures the tightly bonded uptake accounted for the total uptake after approximately 45 hours of growth. The amount of the uptake on algal debris was greater than that associated with growing algal cultures. The uptake of ruthenium chloride and nitro nitrosylruthenium, respectively, in growing algal cultures reached maximum levels of about 40 and 70 percent. Thus, these ruthenium compounds, although temporarily associated with the algae, would be transported downstream and possibly concentrate progressively as it moves through various aquatic food chains.

It was also found that the nitrosylruthenium and ruthenium chloride compounds could be washed from the algal cells with a wash solution containing chlorides (Cl^-) and nitrates (NO_3^-). The release of ruthenium chloride was not affected by an increase in either Cl^- or NO_3^- concentration, but a significant decrease in the amount of nitrosylruthenium was observed when either the NO_3^- or Cl^- concentration was increased.

3.1.3 Transport of Ruthenium Complexes: The net transport of ruthenium complexes (nitrosylruthenium and ruthenium chloride) in an aquatic environment was evaluated in both flowing and non-flowing ecosystems (9). This investigation was limited to the study of RuNO-nitro complexes in general and RuNO-dinitro complexes in particular. However, ^{137}Cs, ^{58}Co, and ^{103}Ru (chloride form) were included in a flume study for comparative purposes. The results showed that the transport of ruthenium is affected by the chemical form of ruthenium, pH, relative amount of suspended material, and the hydraulic characteristic of the stream. The net transport of nitrosylruthenium ($^{103}RuNO$) was higher than that of ruthenium chloride ($^{103}RuCl_3$)

The studies showed that the RuNO-dinitro complex $[RuNO(NO_2)_2OH(H_2O)_2]$ possesses cationic and anionic, as well as molecular characteristics. The dinitro species were anionic and molecular while the mononitro species were mainly cationic in nature. Equilibrium between the molecular, cationic, and anionic species of ruthenium complexes was represented as follows:

$$\begin{tikzcd}[column sep=small]
& \begin{matrix}\mathrm{RuNO(NO_2)_2OH(H_2O)_2} \\ \text{(neutral)}\end{matrix} \arrow[dl, shift left] \arrow[dr, shift left] & \\
\begin{matrix}\mathrm{NO_2^- + [RuNONO_2OH(H_2O)_2]^+} \\ \text{(cationic)}\end{matrix} \arrow[ur, shift left] & & \begin{matrix}\mathrm{[RuNO(NO_2)_2OH_2H_2O]^- + H^+} \\ \text{(anionic)}\end{matrix} \arrow[ul, shift left]
\end{tikzcd}$$

RuNO-nitro complexes are pH dependent. At low pH, cationic mononitro and neutral dinitro forms prevailed, while the anionic dinitro forms were prevalent at high pH. As the pH increased, hydrolysis of nitro complexes took place. The rate of hydrolysis between pH 3 and 7 was slow, but above pH 7 the rate increased exponentially. It appeared that negatively charged polymeric colloidal dispersions were formed at higher pH levels.

Interaction of RuNO-nitro complexes with five pure clay minerals and Lake Austin sediments was also studied. Sorption of these complexes by the sediments was compatible with kaolinite, attapulgite, and vermiculite clay minerals. Organic material contributed to the sorption by the sediments about 10 to 20 percent. Dolomite, one of the constituents of Lake Austin sediments, exhibited a relatively high sorption for RuNO complexes. The sorption by dolomite was believed to be due to the chemisorption of the negatively charged ruthenium colloids on the positively charged dolomite sols. The hydrogen ion concentration was also an important factor in the uptake of ^{103}Ru by clay minerals and sediments. The uptake changed in increasingly larger amounts as the pH of the system increased from pH 3 to 7. Maximum uptake occurred at pH 7 with a gradual decline beyond pH 7.

The general distribution pattern of ^{106}RuNO-nitro complexes in water, sediments, and biota of a non-flowing ecosystem agreed with previous ecosystem studies (3). The laboratory studies revealed that about 22 percent of the total ^{106}Ru was retained by plants and sediments and the remaining 78 percent remained in solution. Actual flume studies showed that 92 percent of the total ^{106}Ru released was transported out of the flume in about six hours. About 72 percent was in the solution phase, and 20 percent was associated with suspended matter. It seems evident that aquaria ecosystem studies can provide valuable information in estimating the fate of radionuclides in a more dynamic ecosystem such as a flume system. The ^{106}Ru concentration in solution decreased rapidly after initial release. At the end of 30 days, the approximate distribution of ^{106}Ru was: 85 percent in the inorganic fraction of the sediments, 5 percent in the organic fraction of sediments, 6 percent on surfaces of macroplants, 2 percent in the algal mat attached to the aquaria walls, 1 percent on the glass and about 1 percent in solution. The concentration of ^{106}Ru in dead plants was about two times as high as the concentration of ^{106}Ru in live plants. Organic matter in the sediments, as calculated on unit weight bases, sorbed approximately three times more ^{106}Ru than the inorganic constituents. The top inch of the sediment accumulated about 97 percent of ^{106}Ru. Organic matter indicated a high affinity for ^{106}RuNO-nitro complexes.

The flume studies involved three major experiments, namely, the transport of ^{103}RuNO, transport of ^{103}RuCl$_2$, and the influence of an induced clay load on the transport of ^{103}RuNO. Dye studies were made to determine the hydraulic characteristics of the system, and the results agreed fairly well with prior studies (5) (7). The time-concentration curves of ^{103}Ru displayed the same general dispersion as observed during the dye studies. Instantaneous release studies in the flume indicated that most of the ^{103}Ru remained in the dissolved phase, some became associated with suspended matter, and a small fraction was taken up by plants and bottom sediments. However, bottom sediments and biota can become large accumulators of ^{103}Ru under favorable conditions. Under a clear stream environment, ^{103}Ru showed higher sorption on the biochemical system than on the geochemical system. The above phenomenon was reversed under the induced clay conditions. On introducing kaolinite clay suspension into the flume, as much as 50 percent of the ^{103}RuNO in the water became associated with the suspended matter. The ^{103}RuNO content in the bottom sediments was increased from 1 percent to 15 percent as a result of the deposition of the suspended clay.

A small fraction of the ^{103}Ru released was removed by macroplants. The estimated uptake of ^{103}RuCl$_2$ and ^{103}RuNO, respectively, by the plant mass was only 3 percent and 5 percent. The sorption of ^{103}RuNO on plants was reduced to 2 percent in the presence of clay suspension. The concentrations of ^{103}Ru in the four dominant plant species varied as follows: Spirogyra > Myriophyllum > Chara >Potamogeton. All plant species showed a higher uptake for ^{103}RuNO than for ^{103}RuCl$_3$. The data showed that the uptake of ^{103}Ru by plants was dependent upon the plant species and the chemical form and concentration of ^{103}Ru. The concentration factors were found to be as high as 10^4 and 10^5. The data from a diurnal study conducted in a small non-flowing ecosystem indicated that the redistribution of ruthenium already in the biota and in the sediments is related to the diurnal changes in the environmental factors, such as temperature, dissolved oxygen, ORP, and pH.

3.2 Transport of ^{65}Zn in Non-flowing and Flowing Systems

The transport of ^{65}Zn through the aquatic environment of a model river system was described, and the distribution of ^{65}Zn in water, suspended solids, bottom sediments, and biota was evaluated (5). A laboratory study was also conducted to examine the uptake and release of ^{65}Zn by the components of a small, non-flowing ecosystem. Detailed laboratory tests were performed to compare the uptake and release of ^{65}Zn by Lake Austin sediment with selected reference clay minerals.

It was determined that the transport of ^{65}Zn in a flowing system is affected by the hydraulics and by the ecological characteristics of the stream. The sediment, rooted plants, and suspended fractions are of considerable importance in the uptake of ^{65}Zn. Environmental factors such as pH, dissolved oxygen, oxidation-reduction potential (ORP), and light intensity influences the amount of uptake and release. In the flowing system the greatest fraction of ^{65}Zn was associated with the solution and suspended material.

Mixing characteristics of the system were determined by dye studies. A flow of 0.847 cfm (24 liters/min) was used during the dye test. The calculated mean velocity was 0.0057 ft/sec based on an average cross sectional area of 2.5 sq ft. The dispersion coefficients varied from 0.022 to 0.059 ft^2/sec. The time concentration curves for the tracer, five feet beyond the influent, approximated a Gaussian distribution, whereas the ^{65}Zn concentration curves clearly indicated delays in transport.

The percentage of ^{65}Zn associated with the suspended material was variable and no consistent pattern of distribution was found. An average of 48 percent of ^{65}Zn was associated with suspended material. Under river conditions, the greatest fraction of ^{65}Zn could be readily transported downstream. The sediment in the non-flowing ecological system retained the greatest fraction of ^{65}Zn due to prolonged contact. It was therefore expected that ^{65}Zn in a non-flowing system such as a lake would become mainly associated with the bottom sediment. The approximate distribution of ^{65}Zn in the non-flowing system after 17 days of contact was 52 percent in the sediment, 16 percent in solution, 10 percent on the suspended material, 14 percent associated with the Myriophyllum (rooted plant), and the remainder was associated with a snail and the walls of the aquarium. Approximately 80 percent of the activity associated with the sediment was in the top 2.6 cm of the sediment.

The role of the bottom sediment in the transport of ^{65}Zn was limited by the contact, the chemical composition of the bottom sediment, and environmental factors such as pH and redox potential. The uptake and release of ^{65}Zn was pH dependent. There was an increase in the uptake of ^{65}Zn for pH levels between 5 and 10 with no uptake below pH 5, and a release of ^{65}Zn with time if the pH increased above 10. If the redox potential did not drop below -200mv, the uptake was not affected. The laboratory tests revealed that attapulgite and montmorillonite sorbed ^{65}Zn rapidly and retained it; kaolinite had a fair initial uptake but 70 to 100 percent of ^{65}Zn was released with prolonged contact. The clay fraction of the bottom sediment was the main sorbent of ^{65}Zn, and an insignificant amount of ^{65}Zn was associated with the organic material in the sediment. It was concluded that streams and lakes with bottom sediments containing clay minerals, such as attapulgite and montmorillonite, have a potential for high initial uptake and retention of ^{65}Zn if there is adequate contact and if the pH remains favorable. Also, the uptake will increase with time and will be independent of cation exchange capacity.

For rooted and macroscopic plants grown in either the non-flowing or flowing systems, there appeared to be no significant difference in the uptake of ^{65}Zn. It was determined that the transport of ^{65}Zn in a stream is initially depleted by plants because ^{65}Zn is an essential micronutrient; this association is controlled by the growth cycle of the plants. In this particular experiment the uptake of ^{65}Zn was considered to be controlled by photosynthesis rather than by hydraulic characteristics. Consequently, dissolved oxygen, pH, ORP, and light intensity can be related to the transport of ^{65}Zn. The average of the maximum percentages of the released ^{65}Zn which became associated with plants in the flume was 13 percent, and the average of the minimum percentages for the flume was 0.7 percent. On the average, the uptake of ^{65}Zn in six plant species varied as follows: Zannichella > Potamogeton > Cladophora > Spirogyra > Myriophyllum > Chara.

3.3 Transport of ^{58}Co in Non-flowing and Flowing Systems

The transport of radioactive cobalt was evaluated in the flume by measuring the depletion of ^{58}Co after an instantaneous release of $CoCl_2$ (7). The residual effects of the flume were monitored for 90 days, and the radionuclide accumulations in the bottom sediments, suspended solids, biota, and the liquid phase were investigated.

Laboratory and bench studies accompanied the flume studies. An aquarium was used to determine the relative distribution of the radioactive cobalt in various aquatic plants and sediments, and the sorption and release of the cobalt by nine selected clay minerals were studied to establish a basis for predicting cobalt uptake by river sediments of known composition.

In a flowing system, the greatest fraction of ^{58}Co at any particular distance from the inlet was in solution. Approximately 84 percent of ^{58}Co was found in the aqueous fraction of the flume. The relative transport of cobalt in an aqueous environment was defined in terms of a modified longitudinal dispersion equation:

$$C_p = \frac{2.75}{x^{0.6}} \; \frac{M}{A\sqrt{\dfrac{4\pi D_x x}{U}}} \qquad (3\text{-}1)$$

A more generalized form of Eq. 3-1 is:

$$C_p = \frac{b}{x^a} \frac{M}{A\sqrt{\frac{4\pi D_x x}{U}}} \qquad (3\text{-}2)$$

where:

C_p = the observed peak concentration of tracer in water
A = area of stream cross section
M = total mass of tracer released in test
D_x = dispersion coefficient along the x-axis
U = velocity of water
x = distance downstream for the point of release
a and b = constants for a particular stream condition.

The constants are dependent on the physical and chemical characteristics of the water, the type and form of bottom sediments, and the distribution and uptake characteristics of plants. Under the conditions of this study a flow of 0.847 cfm (24 liters/min) in the flume was kept constant during the dye release, and the average dispersion coefficient was 0.04 ft^2/sec. The calculated mean velocity was 0.0057 ft/sec based on 2.5 sq ft cross-sectional area of the flume.

The relative distribution of ^{58}Co in suspended solids was approximately 16 percent. It was postulated that the amount and distribution of radioactive cobalt on the suspended solids or in the solution would be dependent on pH, alkalinity, stable cobalt concentration and to some extent in the organic content of the solids. Three months after the release of ^{58}Co, no detectable radioactivity was found in the flume effluent, although ^{58}Co remained in the sediments and plants.

It was determined that the most important factors that influence the uptake of radioactive cobalt by bottom sediments are time of contact and pH. The uptake of ^{58}Co by bottom sediments in the flume was dependent on the mixing characteristics of the flow and the contact time. The uptake appeared to be associated with the amount of deposition. Since Lake Austin sediments were placed in the flume, detailed studies were performed with these sediments. Lake Austin sediments had a higher affinity for radioactive cobalt than any of its separate components such as calcite, dolomite, quartz, or pure clay minerals. However, the removal of the organic content of the sediments, which was approximately two percent by weight, did not influence the uptake of cobalt. Sorption of radioactive cobalt by the sediments was not completely an ion exchange phenomenon. The obtained data showed that the concentration of cobalt associated with the solid phase decreased as the concentration of the divalent ions increased. Once ^{58}Co became associated with the Lake Austin sediments, the replacement of cobalt by common ions, such as Ca^{++}, Mg^{++}, Na^{+}, K^{+}, or NH_4^{+} (with concentrations typical of those in Lake Austin) was slow or negligible. By reducing the pH below 7, most of the radioactive cobalt was removed from the sediments. Most of the cobalt in sediments appeared to be associated with carbonate and hydroxide precipitates. There was no uptake of ^{58}Co by the sediments when the pH dropped below 4, but the uptake increased with an increase in pH. However, when the pH increased above 10, ^{58}Co was again released from the sediments. The sorption of ^{58}Co by clay minerals was related to the selectivity coefficients which varied over a wide range. For example, the selectivity coefficients for radioactive cobalt were found to be 6.5 for attapulgite and 0.003 for kaolinite. The distribution coefficients, K_d, varied from 1766 for kaolinite to 6.5 for attapulgite. Preference of clays for cobalt ions over calcium ions occurred in the following order: Attapulgite > Wyoming Bentonite > Na^{+}Montmorillonite > Llano vermiculite > Illite > Kaolinite ≈ Plum Bentonite ≈ Helms Bentonite.

Aquatic plants played a considerable role in the movement and transport of ^{58}Co. Rooted and floating plants acted as biological detention reservoirs. The immediate uptake of ^{58}Co by plants appeared to be significant. After 24 hours from the time of release, approximately 30 percent of the total ^{58}Co was in the flume biomass. After three months of observation less than one percent of ^{58}Co still remained in the biomass. Although the plants released a significant part of all the sorbed radioactive cobalt after initial uptake, some of the release appeared to be related to a direct equilibrium adjustment. Additional cobalt was released due to environmental changes. The highest concentration factor, K_c = 91,000, was exhibited by Spirogyra sp. This organism was found to be prevalent during the entire experiment.

In batch releases of $^{58}CoCl_2$, a significant amount of ^{58}Co was associated with suspended material, biomass, and bottom sediments, but the greatest fraction of ^{58}Co remained in the solution. Similarly, the aquarium data indicated that after 17 days 45 percent of the activity of ^{58}Co remained in the solution, 30 percent was associated with bottom sediments, 10 percent with suspended solids, 12 percent with plants, one percent with snails, and 2 percent on the glass walls. About 80 percent of the cobalt was in the upper one cm of the bottom sediment. The uptake of ^{58}Co by Myriophyllum was also studied, and the analysis showed that leaves contained most of the cobalt.

3.4 Transport of ^{85}Sr and ^{137}Cs in an Aqueous Environment

The transport and distribution of ^{85}Sr and ^{137}Cs in a model river under induced turbidity, controlled plant growth, and active bed sediments was actively studied with the model river unit (12).

3.4.1 Transport of ^{85}Sr and ^{137}Cs in Model River Systems: The uptake and release of these radionuclides following a single release were monitored during a 120-day period. This study showed that the transport of ^{137}Cs and ^{85}Sr after an instantaneous release in the flume was primarily a function of hydrodynamic mixing; however, sorption on sediment and plant surfaces was significant. As in previous studies (5) (7) diffusion and dispersion was determined with rhodamine B. The flow was 0.847 cfm and the dispersion coefficients varied from 0.022 to 0.059 ft^2/sec. It was determined that the distribution of both ^{137}Cs and ^{85}Sr in the components of the stream were affected by such environmental factors as: oxygen content, pH, ORP, conductivity, and light intensity. Generally, the fractions of the ^{85}Sr and ^{137}Cs in the aqueous solutions were relatively larger as compared to the levels of ^{65}Zn and ^{58}Co (5) (7). The fluctuations of ^{85}Sr uptake in various stream components were found to be larger as compared to the ^{137}Cs uptake. It appeared that ^{85}Sr was more readily released and resorbed by various stream components than ^{137}Cs. The amount of ^{85}Sr and ^{137}Cs associated with bottom sediments was a function of both mixing characteristics and contact time. The uptake of these radionuclides in the flume by the sediments was dependent upon the suspended organic debris and the change of environmental factors, and increased with regard to the longitudinal distribution along the flume, the average percentage of total ^{85}Sr and ^{137}Cs added to the flume, and the depth of migration. The average values of the selectivity coefficients for the Lake Austin sediments in the flowing system were found to be about 3.47×10^{-3} gm/ml for K^{Sr}_{Ca+Mg} and about 3.35 gm/ml for K^{Cs}_{Ca+Mg}. The values of these selectivity coefficients differ from the values obtained previously (1). However, K^{Cs}_{Ca+Mg} in the flume was much greater than K^{Sr}_{Ca+Mg}, which corresponds to the same inequality previously obtained. It was postulated that the major reason for the difference was due to the presence of organic debris and the special plant population in the flume. The aquatic plants played an important role by rapid sorption of ^{85}Sr and ^{137}Cs and by causing eddy turbulence, thereby providing contact opportunity. For ^{85}Sr 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 ^{137}Cs uptake, the various species of aquatic plants exhibited the following uptake: *Cladophora sp.* > *Potamogeton sp.* > *Chara sp.* > *Spirogyra sp.* > *Myriophyllum sp.* > *Zannichellia sp.*

The net transport of ^{85}Sr and ^{137}Cs across a given section of a stream, as used in this study, may be formulated as follows:

$$F_x = F_o \exp(-kx) \qquad (3-3)$$

where:

F_x = net transport (cumulative fraction of the radionuclide discharged through a section x feet from point of release)

F_o = cumulative fraction of radionuclide discharged through the release point

k = slope of line (semilog plot)

x = distance downstream from the point of release.

The depth of penetration of ^{85}Sr and ^{137}Cs into the bottom sediments was defined in terms of a first-order reaction by:

$$C_s(d,t) = C_{so} \exp[-p(t)d] \qquad (3-4)$$

where:

C_s = concentration of radionuclides at depth d and after contact time t

C_{so} = concentration of radionuclides at interface

d = depth of penetration

$p(t)$ = penetration constant (defined by the slope of the line), which is the probability of sorption per number of radionuclides present per unit path length.

The bottom sediments were assumed to be graded homogeneously.

It was postulated that in a sluggish stream system free of turbidity and plants, the greatest fraction of ^{85}Sr and ^{137}Cs will remain in the water phase. 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.

Because of the difficulty of measurement, the depth, d, was transferred into weight of sediments (12). The Eq. 3-4 was expressed in terms of radionuclide concentration per unit weight of dried sediments (dpm/gm), and the migration of radionuclide (as shown for ^{85}Sr in the bottom sediments) was represented as:

$$C_s(w,t) = C_{so} \exp[-p'(t)w] \tag{3-5}$$

where:

$C_s(w,t)=$ radionuclide concentration per unit weight of dried sediments at the depth with w cumulative weight from the surface and after contact time t

C_{so} = saturated interface radionuclide concentration per unit weight of dried sediments

$p'(t)$ = penetration coefficient, a function of contact time, t, (the probability to be retained in the sediments per travel length of radionuclide).

The function for the penetration coefficient was, p'(t), for ^{85}Sr associated with the sediments in the model river was expressed as:

$$p'(t) = mt^{-n} \tag{3-6}$$

where:

$n = 0.0167(1.002)^{T-25}$

$m = 0.60(1.03)^{T-25}$

T = temperature, °C

t = contact time, days.

The concentration factor of ^{85}Sr by bottom sediments, K_s, in a stagnant aqueous environment was shown to be a function of contact time, t (days) and the temperature (a range of 10°C and 30°C). This funcational relationship was expressed as:

$$K_s = 21\ t^{0.55} \tag{3-7}$$

The empirical relationship expressing the dependence of the concentration factor of ^{85}Sr by <u>Vallisneria</u>, K_p, on both the contact time, t (days), and the temperature, T° C, was presented as:

$$K_p = (250 + T°C)\ t^{-0.1} \tag{3-8}$$

3.4.2 Transport of ^{85}Sr and ^{137}Cs under Induced Clay Suspensions:

The effect of induced clay suspensions on the aquatic transport of radionuclides and, in particular, the effect of an induced turbidity load on the transport of ^{85}Sr and ^{137}Cs in an aquatic environment was investigated (13). Model river experiments were conducted by simulating a slow-moving, unpolluted, turbid stream. Attapulgite and kaolinite were used to create the turbidity. Aquaria and laboratory studies preceded the model river experiments to define more specifically some of the physical and chemical characteristics.

Suspended clay in the model river system produced a significant reduction in the transport of ^{137}Cs, however, the transport of ^{85}Sr was not affected appreciably since most of the ^{85}Sr remained in solution. Modified longitudinal dispersion equations were used to describe the transport of the radionuclides in turbid streams. The transport equations were presented in terms of observed peak radionuclide concentrations with respect to time and position in the flume. The transport was approximated by the following modified longitudinal dispersion equation:

$$C_{(x,t)} = \frac{M}{A\sqrt{4\pi D_x t}} \exp\left[-\frac{(x-ut)}{4D_x t}\right] \exp(-Rt) \qquad (3\text{-}9)$$

where:

$C_{(x,t)}$ = radionuclide concentration with respect to time and distance (mass per unit volume), dpm/ml

M = total mass of tracer released, dpm

A = area of stream cross section, ft^2

D_x = longitudinal dispersion coefficient, ft^2/min

t = time, min

u = average velocity of water, ft/min

x = distance downstream from point of release, ft

R = sorption coefficient, per hour.

The observed model concentrations of ^{85}Sr and ^{137}Cs in a turbid stream typical of these studies were estimated from:

$$C_{OP} = \frac{3.53 \times 10^{-4} M}{A\sqrt{4\pi D_x (x/u)}} \exp(-ax) \qquad (3\text{-}10)$$

where:

C_{op} = observed peak radionuclide concentration in water, dpm/ml

a = transport coefficient, per ft.

Values of the transport coefficient, a, and the sorption coefficient, R, as established for ^{85}Sr and ^{137}Cs under particular experimental conditions are summarized in Table 1.

Table 1. Transport Coefficients, a(per ft), and Sorption Coefficients, R(per hour)

Radionuclide	a(ft^{-1})		R(hr^{-1})	
	Induced Suspended Clay Load			
	Attapulgite	Kaolinite	Attapulgite	Kaolinite
^{85}Sr	0.0013	----	0.055	----
^{137}Cs	0.0046	0.0055	0.262	0.138

It was pointed out that the transport coefficient, a, and the sorption coefficient, R, are dependent on the amount of turbidity in the stream, settling rate of suspended solids, sorption, nature of radionuclide, and hydrodynamic characteristics. The values of the transport coefficient, a, for ^{137}Cs were found to be about four times greater than those for ^{85}Sr in the presence of attapulgite clay suspensions. The data represented one steady-state release of ^{137}Cs and attapulgite clay, and two instantaneous releases, ^{85}Sr and ^{137}Cs with attapulgite clay and ^{137}Cs with kaolinite clay. A flow of 50 liters/min with an average velocity of 1.7 ft/min was maintained in the continuous release study conducted in a dual-channel system, and a flow of 100 liters/min with an average velocity of 1.1 ft/min was maintained during the two instant release tests. For conditions other than those described in these experiments it is necessary to reevaluate the coefficients.

It was found that the relationship between the specific activity of ^{137}Cs on suspended clay and the specific activity in water is directly dependent on the clay concentration, radionuclide concentration, and velocity, and it was expressed as:

$$S_y = b(S_w)^c \qquad (3\text{-}11)$$

where:

S_y = specific activity of radionuclide on suspended clay, dpm/gm

S_w = specific activity of radionuclide in water, dpm/ml

b,c = constants depending on the radionuclide and the type of clay suspension.

The relationship between the specific activity of ^{85}Sr on suspended clay and in water did not follow the type of reaction as described by Eq. 3-11.

The data of the model river experiments indicated that the biomass sorbed a small fraction of the total amount of radionuclides released. The uptake of ^{85}Sr and ^{137}Cs by the plant mass under attapulgite loading was estimated to be 3 percent and 2 percent respectively, and of ^{137}Cs under kaolinite loading 2 percent. The retention of ^{85}Sr and ^{137}Cs by Vallisneria (rooted plant) was insignificant. Plants as collection reservoirs for ^{85}Sr and ^{137}Cs played a minor role as compared to bottom sediments deposition, particularly for the streams with a high turbidity. The sorption-activity relationship for ^{137}Cs with both attapulgite clay and Lake Austin sediment was found to follow the typical Freundlich-type reaction. The distribution coefficient isotherms of attapulgite clay and Lake Austin sediment were not linear. It was shown from aquaria studies that the bottom sediment concentration factor, C_s, and the plant (Vallisneria) concentration factor, C_p, for ^{85}Sr are functions of contact time. The relationships were expressed as:

$$C_s = 29\ t^{-0.475} \qquad (3\text{-}12)$$

and

$$C_p = 235\ t^{-0.128} \qquad (3\text{-}13)$$

where:

t = contact time, days.

Notably, the sediment concentration factor, C_s, increased while the value of the plant concentration factor, C_p, decreased with time. Such a relationship was previously reported (12).

3.4.3 The Role of Plants in ^{85}Sr Transport: Plant uptake of radionuclides may be defined for any given plant type, biomass, surface to volume ratio, pH, and concentration of competing ions and radionuclides. For certain radionuclides (^{85}Sr), pH, competing ion concentration, and plant type are not very important in natural waters. The amount of radionuclides taken up by plants during instantaneous release is significant for a short period. Previous studies in the model river showed, on a percentage basis, that ^{65}Zn and ^{58}Co are retained for long periods of time while ^{137}Cs and ^{85}Sr are released quite rapidly. Plant uptake in continuous release appeared to be lower on a mass balance basis than in instantaneous release.

The role of plants in ^{85}Sr in aquaria and a model river was examined (14). Using the macrophyte, Vallisneria americana Michanx, it was found that almost instantaneous equilibrium was reached in solution through a rapid uptake by adsorption followed by a slower and more complete uptake by absorption. A characteristic pattern of uptake and release was observed after instantaneous releases into aquaria and at stations downstream from the point of release in a model river. Under continuous release, uptake tended toward an equilibrium level which was a function of the amount of attached algae present. The content of ^{85}Sr integrated by plants in the model river after instantaneous release was less than five percent of the total ^{85}Sr introduced. Plants in the model river contained only about five percent of the total ^{85}Sr at near equilibrium during continuous release. In aquaria a rapid transfer to plants was followed by a slower transfer to sediments, but plants released rapidly in response to equilibrium conditions while sediments continued to take up activity through penetration. It was concluded for conditions used in this study that plants played a negligible role in ^{85}Sr transport. The uptake of ^{85}Sr by plants after instantaneous release could be modeled using a sorption-desorption equation.

3.4.4 Interaction Between Flowing Water and Bed Sediment Using ^{137}Cs: The delay in the transport of radionuclides through aquatic systems caused by the interaction between flowing water and bed sediments was investigated (17). This interaction phenomenon for ^{137}Cs was observed by using a set of flumes (each 50 feet in length) and a model river (200 feet long). Four subjects have been emphasized regarding the interaction between flowing water and bed sediments, namely, the sorption of ^{137}Cs into bed sediments, the migration into sediments, the transport of bed sediments containing radionuclides, and the desorption of ^{137}Cs from the bed sediment into the flowing water.

Several continuous releases of ^{137}Cs were made during the flume study to evaluate the interaction between flowing water and bed sediment. The flow velocity ranged from 0.0160 fps to 1.60 fps. The initial radionuclide concentration was constant ranging between 196.5 cpm/ml to 288.3 cpm/ml (1755 to 2562 dpm/ml). During the first 50 minutes of injection of ^{137}Cs and when the velocity was 0.0160 fps, the values of the uptake coefficients, K_{so} [(cpm/cm^2)/^{137}Cs concentration in water], K_{st}(K_{so}/contact time, cm/sec), and K_{sd}[K_{so}/detention time (t, t^{-}), cm], respectively, were 1.29 cm, 4129 x 10^{-4} cm/sec, and 1.31 cm. For the same velocity, the values for the second injection were 0.720 cm, 2.40 x 10^{-4} cm/sec, and 0.728 cm for the uptake coefficient, K_{so}, K_{st}, and K_{sd}, respectively. During a continuous ^{137}Cs release of a 50-minute duration, with a velocity of 0.170 fps, the values of K_{so}, K_{st}, and K_{sd} were found to be 11.27 cm, 37.57 x 10^{-4} cm/sec, and 1.14 cm, respectively. A continuous release of ^{137}Cs, with velocity of 0.533 yielded the values of the uptake rates of K_{so}, K_{st}, and K_{sd} of 1.98 cm, 3.30 x 10^{-4} cm/sec, and 1.00 cm, respectively, for an average contact time of 20 minutes. A velocity of 1.60 fps resulted in the transport of bed sediments and the uptake values as described by K_{so}, K_{st}, and K_{sd} were 5.32 cm, 88.7 x 10^{-4} cm/sec, and 0.629 cm, respectively. The greatest ^{137}Cs uptake rate, K_{st}, by bed sediment was 88.7 x 10^{-4} [(cpm/cm^2)/(cpm/cm^2)/sec)]. The velocity and flow for this test was 1.60 fps and 0.345 cfs/ft. When bed sediment transport did not occur, 82 to 95 percent of the radionuclides passed through the flume in the water phase. However, using higher velocities of 1.60 fps, the portion of radionuclides in the water phase was markedly reduced. The bed sediment uptake was assumed to equal the product of the total surface area of the flume (1.36 x 10^4 cm^2) and the average uptake. Bed sediment uptake ranged from about 3.5 to 17 percent.

The relationship between the velocity of water and the uptake of ^{137}Cs by bed sediment was established. The ^{137}Cs uptake rate, K_{st}, by bed sediment increased with the mean velocity of the water. The formula which describes radionuclides migration into bed sediments is presented as follows:

$$C/C_{os} = A\left[z/(2\sqrt{D_z t}\right]^{-N} \qquad (3\text{-}14)$$

where:

C = concentration of radionuclides in the bed sediment

C_{os} = concentration of radionuclides in the surface of bed sediment

A = coefficient (non-dimensional)

Z = depth of bed sediment from the surface

D_z = diffusion coefficient of radionuclides in water

t = contact time

N = coefficient (non-dimensional).

The ratios of concentration in the bed sediment to concentration at the surface for ^{137}Cs in Lake Austin sediment are represented by:

$$C/C_{os} = 1.33 \times 10^{-5} \left(z\sqrt{t}\right)^{-1.0} \tag{3-15}$$

where:

t = time of contact, second

z = depth of bed sediment from the surface, cm.

It was indicated that the coefficient in Eq. 3-15 is applicable for Lake Austin sediment, assuming a temperature at 24°C.

Since the migration phenomenon includes sorption and decay by radionuclide disintegration, the following mathematical form is proposed:

$$\frac{\partial C}{\partial t} = D_z \frac{\partial^2 C}{\partial z^2} - S_{(c,t)} - \lambda t \tag{3-16}$$

where:

$S_{(c,t)}$ = sorption term

λ = radionuclide decay constant.

Hydraulic characteristics of the model river were compared to real river systems, Figure 1. The data show that experiments were conducted in subcritical laminar flow regions. The flow rates varied from 0.00175 to 0.347 cfs/ft. The maximum of the mean velocity was 3.20 fps. The transport rate for a velocity of 1.65 fps was 36 gm/sec-ft (0.0795 lbs/sec-ft). The sorption of ^{137}Cs by bed sediments and the desorption of this radionuclide into the water phase was determined in the model river studies. The continuous ^{137}Cs release was made in which the initial concentration was 241 dpm/ml. The velocity was 0.0263 fps with a depth of 0.832 feet. The sediment uptake rates, K_{st}, were calculated for ranges of 0 to 10, 10 to 30, and 30 to 70 feet downstream as 113×10^{-4}, 9.3×10^{-4}, and 9.5×10^{-4} cm/sec, respectively. The desorption studies were conducted using mean velocities of 0.005 fps and 0.10 fps. No radioactivity above a background

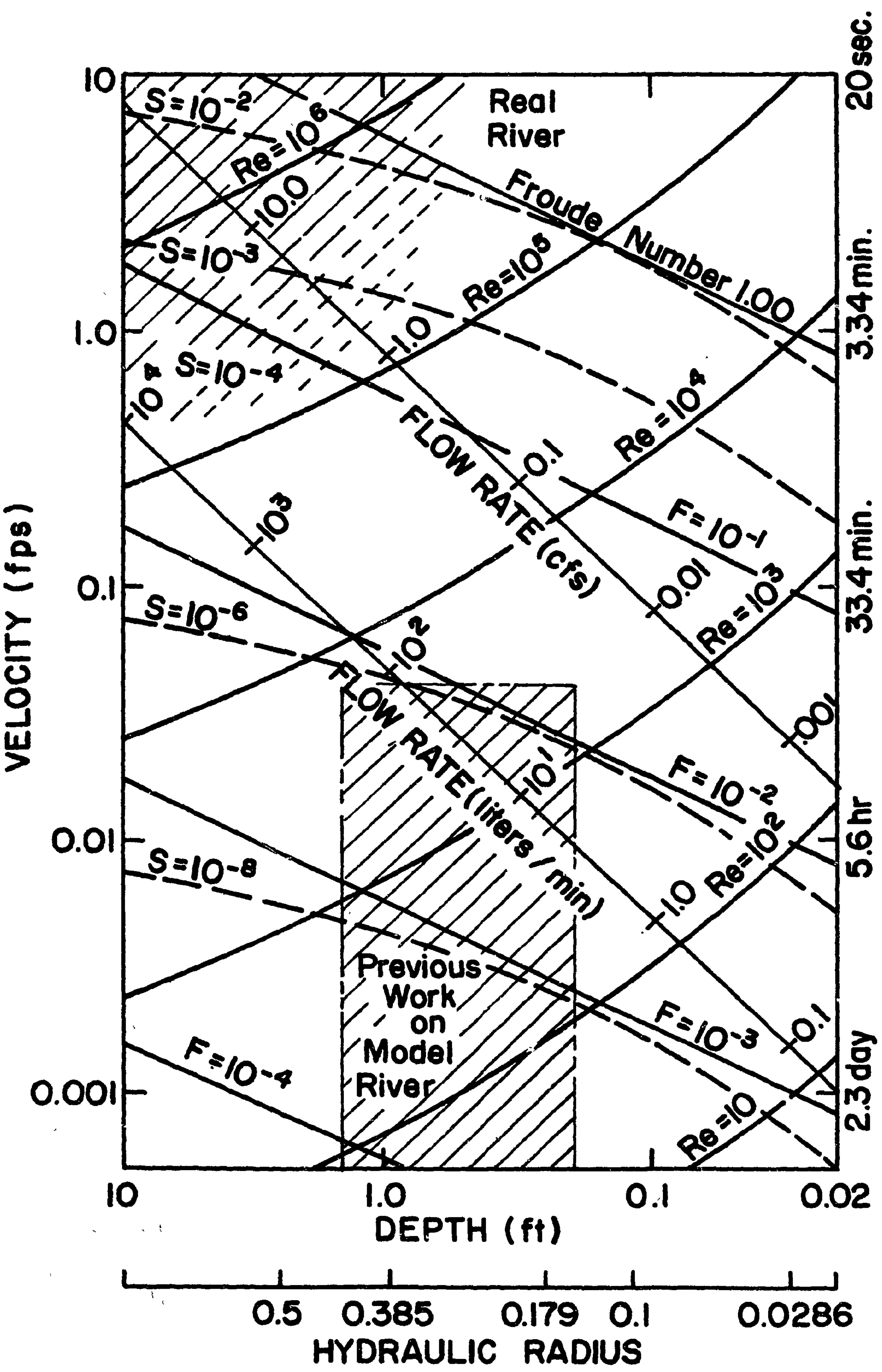

FIG. I. HYDRAULIC CHARACTERISTICS OF MODEL RIVER.

level was detected in the water phase, after a one-month period, however, 0.3 percent of ^{137}Cs was removed by rooted aquatic plants. The desorption rate of the radioactive material from the bed sediment into the water was extremely low. The decrease in radioactivity of bed sediment was less than 1.0 percent per month.

3.5 Transport of ^{51}Cr in an Aqueous Environment

The transport of both trivalent radioactive chromium, ^{51}Cr(III) and hexavalent radioactive chromium, ^{51}Cr(VI), in an aqueous environment was evaluated (11). Measurements were made of the distribution of ^{51}Cr(III) and ^{51}Cr(VI) among the solution, sediment, rooted plants (Vallisneria), and suspended material in a model river and aquaria under both non-polluted and polluted conditions. Trivalent ^{51}Cr existed in cationic form and ^{51}Cr(VI) existed in an anionic form.

In experiments conducted under non-polluted conditions, the initial oxidation state of ^{51}Cr affected its transport characteristics in the model river. Twice as much ^{51}Cr was retained in the flume when the ^{51}Cr was initially in the trivalent oxidation state, as compared to the hexavalent form. Trivalent ^{51}Cr was oxidized as it moved through the model river. The oxidation rates observed in the model river experiments ranged from 0.03 percent to 0.67 percent per minute. It was concluded that due to the differences in the sorptive tendencies of ^{51}Cr(III) and ^{51}Cr(VI), the oxidation of trivalent ^{51}Cr would tend to limit the retention of ^{51}Cr in non-polluted streams. Hexavalent ^{51}Cr was chemically reduced to a large extent upon its sorption by bottom sediments and Vallisneria in the model river; therefore, the ^{51}Cr released was mainly in the trivalent form. The observed percentage reductions in the model river experiments ranged from 52 percent to 100 percent. Even though these chemical reductions occurred, the released ^{51}Cr(III) tended to be reoxidized to ^{51}Cr(VI), thus indicating a natural mechanism that would limit the retention of ^{51}Cr in non-polluted streams.

The transport of ^{51}Cr(VI) was influenced by organic pollution. From 67 percent to 100 percent of the added ^{51}Cr(VI) was chemically reduced in the polluted environment, and as a result of the settling of suspended materials and the chemical reduction of ^{51}Cr(VI) twice as much of the added ^{51}Cr(VI) was retained in the polluted side than in the control side of the model river.

Also, the transport of ^{51}Cr(III) was influenced by inorganic turbidity (attapulgus clay). Approximately 75 percent of the added ^{51}Cr(III) was sorbed and transported by the suspended clay. Approximately 45 percent of the added clay settled in the polluted flume and consequently a greater retention of the added ^{51}Cr(III) was observed. Mass balances for the radionuclides at a given release point and a given time are presented in Table 2.

In the presence of non-polluted water at 25°C, ^{51}Cr (III) was oxidized as the pH increased. Approximately 20 percent of the added ^{51}Cr(III) was oxidized to the hexavalent state after 5 days at a pH of approximately 8.2; whereas at a pH of approximately 9.0, 32 percent was oxidized. In contrast, hexavalent ^{51}Cr was not chemically reduced after 17 days under alkaline pH conditions, however, at a pH of 4.2, approximately 73 percent of ^{51}Cr(VI) was chemically reduced after 6 days. The oxidation of trivalent ^{51}Cr(III) in non-flowing aquaria increased over the range examined; at 10°C it was 7.4 percent per day; at 24°C it was 49.5 percent per day; and at 29.5°C it was 69.1 percent per day.

Trivalent ^{51}Cr exhibited a greater affinity for attapulgus clay and Lake Austin sediment than did hexavalent ^{51}Cr. Attapulgus clay had a chemical composition of 55 percent SiO_2, 10 percent Al_2O_3, 10 percent MgO, 20 percent H_2O, and 5 percent other constituents. Lake Austin sediment was composed of 50 percent calcite, 28 percent clay, 10 percent dolomite, 10 percent quartz, and 2 percent organic matter. Laboratory studies on the sorption of ^{51}Cr(III) and ^{51}Cr(VI) by attapulgus clay and Lake Austin sediment indicated that 75 to 80 percent of ^{51}Cr(III) was sorbed by both sorbents in 1 hour, whereas only 20 to 25 percent of ^{51}Cr(VI) was sorbed after 10 days. It was postulated that ^{51}Cr(III) would be retained in river systems to a greater extent than ^{51}Cr(VI). The release of ^{51}Cr from attapulgite clay and Lake Austin sediment was slow and incomplete. After attapulgus clay and sediment were placed in fresh water, about 8 to 13 percent of the sorbed activity was released after 8 days. After 8 days, the release of ^{51}Cr(III) from Lake Austin

Table 2. Mass Balances for Natural Condition Experiments and Environmental Stress Experiments with Radioactive Chromium

Phase	Percentage in Phase (%)							
	Natural Conditions				Environmental Stress Conditions			
	Experiment 1 (a)		Experiment 2 (b)		Experiment 3 (c)		Experiment 4 (d)	
					Organic Pollution		Inorganic Pollution	
					(control)	(polluted)	(control)	(polluted)
	^{51}Cr(III)	^{51}Cr(VI)	^{51}Cr(III)	^{51}Cr(VI)	^{51}Cr(VI)	^{51}Cr(VI)	^{51}Cr(III)	^{51}Cr(III)
Total water (e)	71.4	86.3	76.0	85.7	92.6	82.0	83.1	80.5
Sediment	11.0	8.4	6.4	5.4	4.4	13.1	4.8	9.7
Vallisneria	2.9	0.8	2.7	1.9	----	----	----	----
Flume walls	14.5	4.4	15.0	6.9	3.0	4.9	12.1	9.8
Flow (l/min)	44.7	44.8	15.3	16.5	4.87	4.79	46.0	46.6

a. Mass balances made at the end of the first passage of radioactivity (instantaneous release through the flume (2 hr); percentages are based on the observed mass balance at 20 ft from the release point.

b. Mass balances made at the end of the 2 day period of continuous radionuclide release; percentages are based on the observed mass balance at 20 ft from the release point.

c. Mass balances made at the end of the 2 day period of continuous radionuclide release; percentages are based on the observed mass balance at 30 ft from the release point.

d. Mass balances made at the end of the 4 hour period of continuous radionuclide release; percentages are based on the observed mass balance at 10 hr from the release point.

e. Includes ^{51}Cr discharged from the flume as well as that present in the water phase in the flume at the time of the mass balance.

sediment was approaching an equilibrium value of approximately 4 percent. The ^{51}Cr released from Lake Austin sediment previously contacted with ^{51}Cr(III) was still in the trivalent form, however, hexavalent ^{51}Cr was chemically reduced. Approximately 70 percent of the ^{51}Cr released from Lake Austin sediment previously contacted with ^{51}Cr(VI) was in the trivalent oxidation state.

The distribution of ^{51}Cr in a non-flowing aquarium was dependent upon the initial oxidation state of the added ^{51}Cr and the temperature of the system. At a given time and temperature, the percentage of the added ^{51}Cr remaining in the water phase was greater in the ^{51}Cr(VI) systems than in the ^{51}Cr(III) systems. For the ^{51}Cr(III) systems, the effect of temperature on the percentage of ^{51}Cr remaining in the water phase at a given time in a non-flowing aquarium was related to both a higher sorption rate and a greater degree of oxidation at higher temperatures.

The percentage of ^{51}Cr associated with the sediment phase was normally higher in the systems contaminated with ^{51}Cr(III) as compared to the ^{51}Cr(VI) systems. The sorption rates and the penetrations were greater at higher temperatures. At a given time and temperature, the percentage of ^{51}Cr associated with Vallisneria was normally greater when the chromium was in the trivalent state.

3.6 Structure and Metabolism of a Lotic Community

The transport of radionuclides is related to the lotic community (8). The animals in the flume ecosystem were limited in species diversity but existed in relatively large numbers. Twelve species were identified. The numbers varied from 6 per square meter for the least abundant (Dugesia tigrina) to over 10,000 per square meter for the most abundant (Lymnaea chmella). The number of animals decreased as summer temperatures increased. Five genera of macroplants and 33 genera of algae were found to be present in the flume, and 22 species of algae were found in the water storage reservoir. During the experiment, the water temperature ranged between 16.0°C to 36.2°C; dissolved oxygen ranged between 0 and 22 mg/l; and the pH varied between 7.2 and 10.0. Observations and calculations were made to determine total solar input, available light for photosynthesis, oxygen and carbon dioxide photosynthesis, oxygen and carbon dioxide respiration, efficiency of the ecosystem, photosynthetic/respiration ratio, and photosynthetic and respiratory quotients. Solar energy input ranged between 67.8 langleys and 544.3 langleys, and about 63 percent of the total solar energy was computed to be photosynthetically usable. Oxygen metabolism ranged between 2.8 and 15.3 gm/m^2 per day for photosynthesis and between 7.5 and 15.3 gm/m^2 per day for community respiration. Carbon dioxide metabolism ranged between 4.0 and 21.3 gm/m^2 per day for photosynthesis and between 3.7 and 21.5 gm/m^2 per day for community respiration. The photosynthetic/respiration (P/R) ratios for oxygen metabolism ranged between 0.3 and 1.1, with an average of 0.66. The carbon dioxide metabolism ratios varied from 0.75 and 1.65, with an average of 1.03. The conversion of solar energy to protoplasmic energy ranged between 0.3 to 4.8 percent for oxygen metabolism and between 0.3 and 6.9 percent for carbon dioxide metabolism, with averages of 1.5 and 1.34, respectively. The highest efficiencies occurred on cloudy days when light intensity was relatively low. With the exception of cloudy days, the percentage of the efficiency decreased as the summer progressed. Photosynthetic quotients (PQ) ranged between 0.4 and 2.9, with an average of 1.36, and respiratory quotients (RQ) ranged between 0.2 and 1.1, with an average of 0.54.

The relationship of photosynthesis to respiration appeared to be an important factor in the uptake and release of ^{65}Zn and ^{58}Co. There was a net decrease in concentration of radionuclides in the plant biomass when respiration exceeded photosynthesis.

3.7 Influence of Organic Pollution on Radionuclide Transport

Organic pollution in streams may influence the uptake and release of radionuclides. Organic pollutants, together with their resulting oxygen demand, usually change the local environmental conditions. The effect of organic pollution on the transport of ^{51}Cr was previously described (11). The organic pollution caused a reducing environment, resulting in a reduction of radioactive chromium and sorption of the chromium by suspended matter. The influence of organic pollution (continuous release of dextrose and raw sewage) on the transport of ^{85}Sr was also reported (12). The ^{85}Sr was accumulated in the suspended debris, causing ^{85}Sr to be more concentrated in the bottom sediments of the polluted channel. The organic pollution increased the sorptive capacity of sediments. Quantitatively, the ^{85}Sr uptake by sediments was found to increase by 100 dpm/core per 1.0 mg/l of chemical oxygen demand added.

3.8 Environmental Factors in Oxygen Distribution in a Model River System

The effect of organic pollution on radionuclide transport, especially as related to the dissolved oxygen in streams, and its distribution in a model river (flume) was investigated (16). The objective of this study was to develop a mathematical model. Various parameters which describe the distribution of dissolved oxygen in a stream, namely, the reaeration rate constant, the biological rate constant, and photosynthesis-respiration rates of benthal systems were measured. The characteristics of the spatial and temporal variation of the oxygen concentration in the flume water were determined.

Reaeration rate constants, k_2', were determined in the model river under two different conditions. In one case, the reaeration rate was 0.18 per day (base 10) with a water depth of one foot, a flow rate of 1.7 in/min, and an average water temperature of 28°C. In another case, the reaeration rate was 0.23 per day with a water depth of one foot, a flow rate of 3.3 in/min and an average water temperature of 26°C. These values fall within the range of those obtained in natural streams. In the determination of reaeration rate constants, the oxygen concentration of the influent was maintained at a constant level by the use of sodium sulfite. The biochemical oxygen demand

(BOD_5) of the feed water was 1.3 mg/l. The measured biological oxidation rate constant, k_1', in the model river was 0.006 per day (base 10) with an inlet sugar concentration of 200 mg/l. In an experiment conducted to obtain the oxidation rate constant for soluble BOD, the flow rate was 5 l/min, and the water depth was 12 inches. Cane sugar was used as a pollutant, and the average BOD_5 along the flume was 137 mg/l. The retention period of water in the flume was one day, the same as in the case of the first reaeration experiment. It was postulated that the reason for the small value of k_1 in the model river was probably due to lack of nutrients and oxidizing organisms. Photosynthetic oxygen production, at points 30 feet and 50 feet from the inlet, respectively, was 20 lbs/acre/day and 22 lbs/acre/day. The areal respiration rate was 20 lbs/acre/day at both stations. It was concluded that various photosynthesis-respiration relationships can be measured in the model river with suitable accuracy.

The roles of organic pollution and dissolved oxygen in radionuclide transport were investigated (18). The ecological environment of a river system was simulated. The diurnal variations of downstream sags of dissolved oxygen (DO) were determined under different organic loads and environmental conditions. Powdered milk was used as a source of pollution.

The sources and sinks of dissolved oxygen are indicated in Figure 2. Considering the balance in the control volume bounded by ΔX and the cross sectional area, A, and assuming the D_L and A are constant, the rate of change of dissolved oxygen concentration, (C), at a point (or volume) fixed, may be represented by the following equation:

$$\frac{\partial C}{\partial t} = -V\frac{\partial C}{\partial x} + D_L\frac{\partial^2 C}{\partial x^2} + Re - De + P - R \qquad (3\text{-}17)$$

In a steady-state system $\frac{\partial C}{\partial t} = 0$, but in an unsteady-state system, i.e., diurnal variations in dissolved oxygen, $\frac{\partial C}{\partial t} \neq 0$, except for two or more instances in a 24-hour period. The term $V\frac{\partial C}{\partial x}$ represents the convective

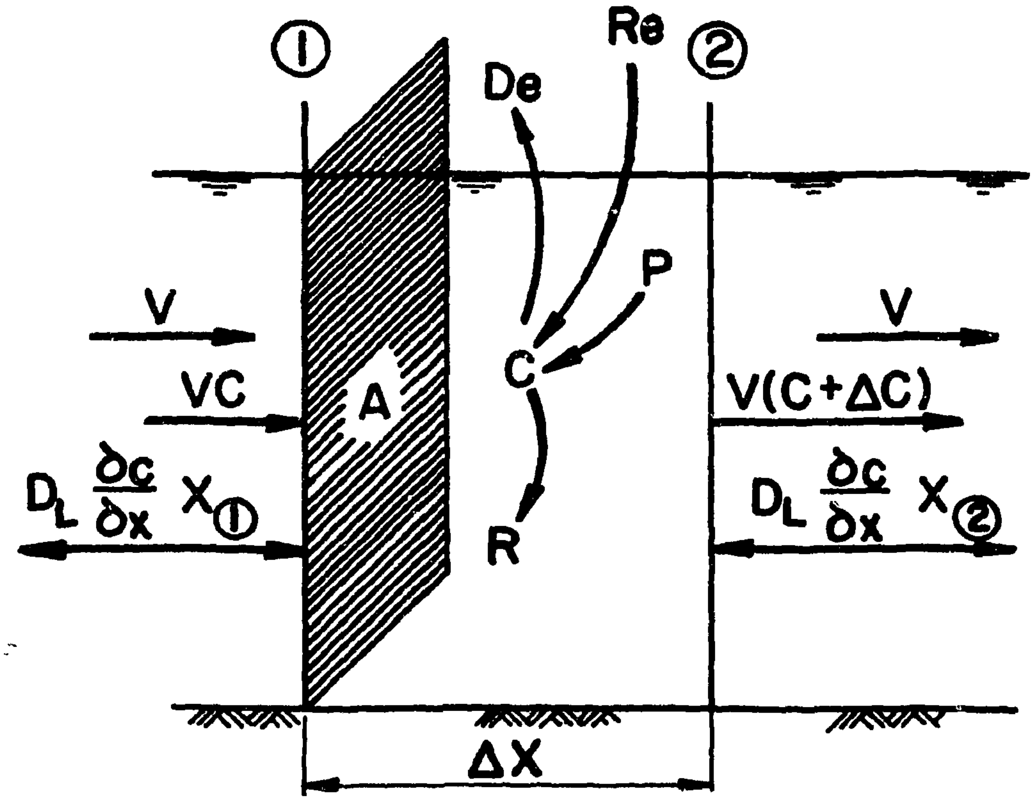

FIG. 2. ELEMENTAL CONTROL VOLUME IN RIVER.

Definitions for Fig. 2

1. C = Dissolved Oxygen (D.O.) Concentration, mg/l
2. P = Photosythesis, production of Oxygen, mg/l/hr
3. R = Respiration, reduction of Oxygen, mg/l/hr
4. V = Velocity of Flow
5. Re = Transfer of Oxygen from Atmosphere into Water
6. De = Transfer of Oxygen from Water to Atmosphere, mg/l/hr
7. D_L = Longitudinal Dispersion Coefficient
8. $\left.\frac{\delta c}{\delta x}\right|_{①}$ = Rate of Change of D.O. along River at ①
9. $\left.\frac{\delta c}{\delta x}\right|_{②}$ = Rate of Change of D.O. along River at ②
10. ΔC = Change in D.O. over Length Δx
11. A = Cross-sectional Area of River
12. Δx = Length of Control Volume
13. AΔx = Volume

Assumptions for Fig 2

1. Steady, Uniform Flow
2. Constant Longitudinal Dispersion Coefficient, D_L

change of dissolved oxygen into and out of the control volume. The term $D_L \frac{\partial^2 C}{\partial x^2}$ indicates the rate of exchange into or out of the control volume due to molecular and turbulent diffusions, and mixings due to transverse velocity gradients in the flow. In this study, the term $D_L \frac{\partial^2 C}{\partial x^2}$ was assumed to be negligible. Reaeration, Re, and deaeration, De, represent the volumetric rate of the exchange of oxygen through the stream surface, and both are usually represented by a single term, namely K_2' $(C_s - C)$. The term K_2' is a constant relating to the hydraulic properties of the stream, and C_s is the saturation value for DO. The K_2' was assumed to have the same oxygen transfer in either direction $(C < C_s;\ C > C_s)$ as determined by the relationship of C_s and C. The reaeration rate constant, K_2', was determined to be between 0.06 and 0.14/hr, and it was probably more affected by wind than turbulence. Measurements were made to evaluate DO, physical reaeration, pH, carbon, biochemical oxygen demand (BOD), chemical oxygen demand (COD), and oxygen loss by bubbles. It was postulated that the bubble loss could be considered as a loss of oxygen produced by photosynthesis; however, it was also noted that no mathematical representation of bubble loss is available for inclusion in Eq. (3-17)

Photosynthesis (P) and respiration (R) rates were evaluated. Respiration rates were calculated using BOD reduction data and benthic respirometer measurements. With these observations it was possible to calculate (P-R) and the ratio of the net photosynthesis to nighttime respiration (P_n/R_{nt}). These ratios were calculated for different points throughout the length of the flume.

When diurnal variations are important, a form of Eq. 3-17 should be used. The (P-R) variable should be established along with transport, dispersion, and reaeration. The (P-R) input might be utilized more effectively through the use of decaying full-wave perturbation employing a value R as determined from BOD, COD, or carbon reduction data. The (P-R) for the flume system [as a function of time, t(hr), and the distance from the pollution source, X, ft] is expressed as follows:

$$(P-R) = 5.92 \exp[-0.0947\ (t-8)]\ \sin\frac{\pi}{8}\ (t-8) - \frac{60.3 - 0.2X}{45} \qquad (3\text{-}18)$$

where:

$8 < t < 24$.

The light period was assumed to be 16 hours. Numerical solutions of Eq. 3-18 can be developed assuming (P-R) is a function of time, K_2', V, D_L, and the initial condition for C is a function of X.

3.9 Continuous Release of Radionuclides in Small-Scale Ecosystems

The continuous release of radionuclides in the model river system was reported (21). The control channel of the model river system contained a typical bottom sediment and received potable water. The second channel, in addition to bottom sediment, contained a community of rooted aquatic plants and received water rich in phytoplankton. Both channels were subjected to a continuous release of ^{134}Cs and ^{85}Sr for periods up to thirty-five days. The distribution of the radionuclides in the bottom sediment, plants, algae, and water was determined, and a profile of radioactivity or areal distribution of ^{134}Cs and ^{85}Sr was established for the flume sediment. Under continuous release of radionuclides, bottom sediment and plants concentrated radioactivity until a quasi-equilibrium was reached. Plants reached equilibrium much faster than the bottom sediments. Uptake equilibrium for plants was reached in less than 36 hours, as compared to about 30 days for the bottom sediment. Radionuclides were concentrated in bottom sediments or aquatic plants at a rate directly proportional to the difference between the saturation concentration (concentration at equilibrium) and the concentration in sediments or plants at a given point in time. After saturation was reached at the sediment surface, radionuclide uptake by the sediment was equal to the radioactivity loss by the surface as a result of a slow migration process into the sediment. The concentration of radionuclides in the sediment was affected by sediment movement, and the areal distribution of the radionuclides was not uniform. It was expected that under real river conditions the area distribution will not be uniform and some areas of bottom sediment will never reach saturation while other areas will be repositories of contaminated sediment. Under continuous release conditions the extraction of radionuclides from the water phase on a mass balance basis had a minor effect on the specific activity in the water. The amount of activity retained in the bottom sediment during the first 22 days of release was approximately 10 percent of the released activity.

In a continuous release of radionuclides, wherein the specific activity remained relatively constant in the water phase, the correlation between ^{134}Cs and ^{85}Sr concentrations in bed sediments may be expressed by:

$$[C_{so} \text{ for } ^{134}C_s] = 26.2\,[C_{so} \text{ for } ^{85}\text{Sr}]^{0.66} \qquad (3\text{-}19)$$

where:

C_{so} = concentration at surface, μμc/cm^2.

The relationship between the retention of ^{134}Cs and ^{85}Sr by plants may be defined by the following equation:

$$[Cp \text{ for } ^{134}\text{Cs}] = 6.18\,[Cp \text{ for } ^{85}\text{Sr}]^{0.76} \qquad (3\text{-}20)$$

where:

Cp = retention of radionuclides by plants, μμc/gm of dry weight.

The linear correlation coefficient was 0.86. The above equations are only applicable for the stated experimental conditions. The specific activity on the plants after release was discontinued may be expressed as follows:

$$Cp = A\ 10^{-Bt} \qquad (3\text{-}21)$$

where:

t = time after discontinuance of the radionuclide source, days

for ^{134}Cs: $A = 47.9$; $B = 0.061$

and

for ^{85}Sr: $A = 18.4$; $B = 0.121$.

The surface concentration factor, K_s, of the bottom sediments which were derived from Lake Austin, ranged from 250 to 300 ml/cm^2 for ^{134}Cs, and from 70 to 90 ml/cm^2 for ^{85}Sr. The bottom sediments of Lake Austin had higher affinity for ^{134}Cs than ^{85}Sr, but the opposite was true in the case of aquatic plants. The concentration factors, K_p, for ^{134}Cs and ^{85}Sr for the predominant species of plants were determined over a 21-day period following initiation of continuous release. From the 2nd to 21st day the concentration factor, K_p, varied between 220 ml/gm and 580 ml/gm with an average of 400 ml/gm for ^{134}Cs, and from 430 to 860 ml/gm with an average of 600 ml/gm for ^{85}Sr. Suspended plankton contributed to the surface concentration of radionuclides on bed sediments. Plankton, unlike rooted plants, exhibited a higher concentration factor for ^{134}Cs than for ^{85}Sr. Assuming an average amount of suspended phytoplankton concentration of 20 mg/l, the values of the concentration factor, K_a for ^{134}Cs under the maximum and minimum conditions, were 13,000 ml/gm and 4000 ml/gm, respectively. In the case of ^{85}Sr the value for K_p ranged from approximately 200 ml/gm to 700 ml/gm, averaging 400 ml/gm. The uptake of ^{134}Cs by suspended phytoplankton in the flume varied from 12 μμc/liter to about 50 μμc/liter depending upon the time of day. The percentage of activity related to ^{134}Cs in the phytoplankton averaged between 8.9 and 27.9 percent of the total activity in the water. The percentage of the activity related to ^{85}Sr in the suspended phytoplankton as compared to water, averaged between 0.4 and 1.5 percent. The uptake of ^{134}Cs by suspended phytoplankton followed a pattern similar to the diurnal cycle and photosynthetic oxygen production:

$$Y = A' + B'X \tag{3-22}$$

where:

Y = change in rate of concentration of ^{134}Cs, µµc/liter/hr
X = rate of photosynthetic oxygen production, mg/l/hr
A' & B' = constants.

The average values of the constants A' and B' were -0.42 and +5.57, respectively. The correlation coefficient was 0.953. The amount of ^{85}Sr uptake associated with suspended phytoplankton varied from 0.2 µµc/liter to 0.8 µµc/liter; however, no cyclical diurnal relationship was noted between photosynthetic oxygen production and the uptake of ^{85}Sr. The rate of net photosynthetic oxygen production was determined as:

$$(P-R) = -\frac{\Delta D}{\Delta t} - 0.052\,\bar{D} + 0.046 \tag{3-23}$$

where:

(P-R) = net photosynthetic oxygen production rate during time period Δt, mg/l/hr
$\frac{\Delta D}{\Delta t}$ = change of oxygen deficit during time period Δt, mg/l
$\bar{D}$ = average oxygen deficit during time period Δt, mg/l.

The biochemical oxygen demand of the water entering the flume was less than 2 mg/l and therefore negligible. In the above equation values for the reaeration rate constant (base e), and sediment oxygen demand were determined to be 0.052 hr^{-1} and 0.046 mg/l/hr, respectively.

3.10 Accumulation of Mercurials in an Algal Environment

The sorption and desorption of mercurials were investigated (22) as auxiliary input to the overall radionuclide evaluation problem. The biological accumulation and release of inorganic mercury (mercuric chloride, $^{203}HgCl_2$) and organic mercury (methylmercuric chloride, $CH_3{}^{203}HgCl$) were studied through the use of a green alga, Chlorella pyrenoidosa.

It was determined that a sorption isotherm could be used to evaluate the toxic effects of mercury on Chlorella pyrenoidosa. The total sorption isotherm was classified into three stages:

$$\text{Stage I,} \quad \text{where } n < 1, \text{ and } K = \frac{x/m}{C} = kC^{\frac{1}{n}-1} \qquad (3\text{-}24)$$

$$\text{Stage II,} \quad \text{where } n = 1, \text{ and } K = \frac{x/m}{C} = k \qquad (3\text{-}25)$$

$$\text{Stage III,} \quad \text{where } n > 1, \text{ and } K = \frac{x/m}{C} = kC^{\frac{1}{n}-1} \qquad (3\text{-}26)$$

where:

n = a parameter of the Freundlich isotherm
K = the concentration factor
x = the amount of a radionuclide absorbed by plant
m = the mass of plant
k = a coefficient
C = specific activity of water at equilibrium.

Three mass transfer models employing a uniform function, an exponential function, and a power function were proposed to describe the kinetics of sorption and desorption, Table 3.

The sorption and desorption kinetics of mercuric mercury by Chlorella were expressed by uniform functions:

Table 3. Mass Transfer Models for Three Types of Sorption and Desorption Kinetics

Form of the Function	Cp(t)	t_s	K_2	$\hat{K}_2$
Exponential				
Sorption				
$\frac{\partial Cp}{\partial t} = K_2[G(c)-Cp]$	$G(c)[1-\exp(-K_2 t)]$ (a)	$4.60/K_2$ (b)	$-\frac{1}{t}\ln[1-\frac{Cp}{G(c)}]$	
Desorption				
$\frac{\partial Cp}{\partial t} = -\hat{K}_2[Cp-G(c)]$	$G(c)[1+(n-1)\exp(-K_2 t)]$ (c)	$-\frac{1}{\hat{K}_2}\ln[\frac{0.01}{(n-1)}]$ (d)		$-\frac{1}{t}\ln\left\{\frac{1}{(n-1)}[\frac{Cp}{G(c)}-1]\right\}$
Uniform				
Sorption				
$\frac{\partial Cp}{\partial t} = \frac{K_2}{t}$	$K_2 \ln t/t_s + G(c)$ (e)	$\frac{1}{t}\exp\left\{\frac{1}{K_2}[G(c)-Cp]\right\}$	$\frac{Cp-G(c)}{\ln t/t_s}$	
Desorption				
$\frac{\partial Cp}{\partial t} = -\frac{\hat{K}_2}{t}$ [redacted]	$K_2 \ln t_s/t + G(c)$ (e)	$t\exp\left\{\frac{1}{\hat{K}_2}[Cp-G(c)]\right\}$		$\frac{Cp-G(c)}{\ln t_s/t}$
Power				
Sorption				
$\frac{\partial Cp}{\partial \ln t} = K_2[G(c)-Cp]$	$G(c)[1-t^{-K_2}]$ (a)	$[0.01]^{-\frac{1}{K_2}}$ (b)	$-\frac{1}{\ln t}\ln[1-\frac{Cp}{G(c)}]$	
Desorption				
$\frac{\partial Cp}{\partial \ln t} = -\hat{K}_2[Cp-G(c)]$	$G(c)[1+(n-1)t^{-\hat{K}_2}]$ (c)	$[\frac{1.01}{(n-1)}]^{-\frac{1}{\hat{K}_2}}$ (d)		$-\frac{1}{\ln t}\ln\left\{\frac{1}{(n-1)}[\frac{Cp}{G(c)}-1]\right\}$

a) If $Cp = 0$, when $t = 0$
b) If the quasi-steady level is assumed to reach 0.99 G(c)
c) If $Cp = nG(c)$, when $t = 0$
d) If the quasi-steady level is assumed to reach 1.01 G(c,
e) If $Cp = G(c)$ at the steady level, when $t = ts$

and where

Cp = Specific activity of plant at time t
K_2 = Sorption mass transfer coefficient
G(c) = Steady-state level of radionuclide in plant
C = Specific [illegible]

Form of the Function	Cp(t)	t_s	K_2	$\hat{K}_2$
Exponential				
Sorption				
$\frac{\partial Cp}{\partial t} = K_2[G(c)-Cp]$	$G(c)[1-\exp(-K_2 t)]$ (a)	$4.60/K_2$ (b)	$-\frac{1}{t}\ln[1-\frac{Cp}{G(c)}]$	
Desorption				
$\frac{\partial Cp}{\partial t} = -\hat{K}_2[Cp-G(c)]$	$G(c)[1+(n-1)\exp(-K_2 t)]$ (c)	$-\frac{1}{\hat{K}_2}\ln[\frac{0.01}{(n-1)}]$ (d)		$-\frac{1}{t}\ln\left\{\frac{1}{(n-1)}[\frac{Cp}{G(c)}-1]\right\}$
Uniform				
Sorption				
$\frac{\partial Cp}{\partial t} = \frac{K_2}{t}$	$K_2 \ln t/t_s + G(c)$ (e)	$\frac{1}{t}\exp\left\{\frac{1}{K_2}[G(c)-Cp]\right\}$	$\frac{Cp-G(c)}{\ln t/t_s}$	
Desorption				
$\frac{\partial Cp}{\partial t} = -\frac{\hat{K}_2}{t}$	$K_2 \ln t_s/t + G(c)$ (e)	$t\exp\left\{\frac{1}{\hat{K}_2}[Cp-G(c)]\right\}$		$\frac{Cp-G(c)}{\ln t_s/t}$
Power				
Sorption				
$\frac{\partial Cp}{\partial \ln t} = K_2[G(c)-Cp]$	$G(c)[1-t^{-K_2}]$ (a)	$[0.01]^{-\frac{1}{K_2}}$ (b)	$-\frac{1}{\ln t}\ln[1-\frac{Cp}{G(c)}]$	
Desorption				
$\frac{\partial Cp}{\partial \ln t} = -\hat{K}_2[Cp-G(c)]$	$G(c)[1+(n-1)t^{-\hat{K}_2}]$ (c)	$[\frac{1.01}{(n-1)}]^{-\frac{1}{\hat{K}_2}}$ (d)		$-\frac{1}{\ln t}\ln\left\{\frac{1}{(n-1)}[\frac{Cp}{G(c)}-1]\right\}$

2

a) If Cp = 0, when t = 0
b) If the quasi-steady level is assumed to reach 0.99 G(c)
c) If Cp = nG(c), when t = 0
d) If the quasi-steady level is assumed to reach 1.01 G(c)
e) If Cp = G(c) at the steady level, when t = ts

and where

Cp = Specific activity of plant at time t
K_2 = Sorption mass transfer coefficient
G(c) = Steady-state level of radionuclide in plant
C = Specific activity of water at equilibrium
K_2 = Desorption mass transfer coefficient
ts = the time to reach the steady-state level

(a) Sorption: $Cp(t) = (0.930) \ln t/10 + 7.50$ (3-27)

(b) Desorption: $Cp(t) = (0.965) \ln 6/t + 13.40$ (3-28)

The sorption and desorption kinetics of methyl mercury by *Chlorella* were expressed by exponential functions:

(a) Sorption: $Cp(t) = (21.2)(1 - e^{-0.570t})$ (3-29)

(b) Desorption: $Cp(t) = (10.8)(1 + 0.12e^{-0.374t})$ (3-30)

These mass transfer models may be applied to forecast levels of concentration and the extent of desorption from algae.

The environmental factors which influence the biological accumulation of mercuric mercury in *Chlorella* were examined and the results were as follows:

(a) Light intensity did not have an important influence on the uptake of mercuric mercury by the algae until the saturation level was reached. However, after the saturation was reached, the algal growth rate exerted a significant influence on uptake. When the algae increased in population, a decrease in the specific activity was observed.

(b) The specific activity on the algae and the concentration factor decreased with an increase in the algal population.

(c) The toxic effect of trace elements on algal growth was dependent upon the concentration of trace elements and algae.

(d) The effect of pH on uptake was considerable. The lower the pH, the higher the uptake of mercuric chloride by *Chlorella*. The change in pH from 5.6 to 8.2 decreased the specific activity by about 50 percent and decreased the concentration factor by about 78 percent.

(e) Magnesium in concentrations varying from 2.47 mg/l to 100 mg/l exhibited no significant influence on the uptake of mercuric chloride by the algae. However, when the concentration of calcium was increased, the uptake of mercuric chloride by algae increased.

(f) An increase of the uptake of mercuric chloride by the algae was observed when the calcium concentration increased. The range of calcium concentration used was between 4.5 ppm and 450 ppm. It should be noted that most concentrations of magnesium and calcium in fresh water lie within the above ranges.

(g) The release of mercuric mercury from the algae due to washing with water and EDTA solution was very small. EDTA solution did not enhance the release of mercury, but it was a good elimination agent for zinc and cobalt.

(h) Desorption of mercuric mercury from the algae was small, and there was no apparent effect of light intensity on desorption.

(i) Methionine exhibited better desorption capability than the other two chelating agents, namely citric acid and EDTA. After 10 hours of contact 39 and 27 percent, respectively, of the original mercury was desorbed by methionine and EDTA.

The influence of the environmental factors on biological accumulation of methyl mercury in Chlorella was examined and the results of the investigation were as follows:

(a) Methyl mercury was accumulated in the algae more efficiently than mercuric mercury, so that relatively small portions of methyl mercury remained in the solution phase. After 10 hours, 60 percent of the initial methyl mercury remained in the culture, and 87 percent of the remaining methyl mercury was associated with the algae. After completion of the uptake, methyl mercury associated with the algal phase decreased gradually with time. Since methyl mercury in the solution phase also decreased gradually, it was postulated that the loss of methyl mercury from the algal phase was due to volatilization.

(b) After completion of the uptake process of methyl mercury by algae, the algal growth rate exerted an influence on the uptake and influenced the volatilization of methyl mercury. Therefore, it was concluded that the uptake of methyl mercury by algae as expressed by the concentration factor is inadequate, because a large portion of methyl mercury escapes from the system and the remaining methyl mercury appears to become associated with algae.

(c) Light intensity did not have an influence on the uptake process of methyl mercury by algae.

(d) A similarity existed between the toxicity exhibited by methylmercuric chloride and mercuric mercury to Chlorella. If the initial algal concentration was small, methylmercuric chloride was toxic to algae at low concentrations. If the initial concentration increased, relatively higher concentrations of methylmercuric chloride were required to reach toxic levels.

(e) The specific activity on algae decreased as the algal density increased.

(f) The specific activity on algae increased moderately with an increase in pH.

(g) There was no influence of sodium and potassium on the uptake of methyl mercury by the test algae. The sodium and potassium concentrations ranged between 30 ppm and 400 ppm, and 9.6 ppm and 56.7 ppm, respectively.

(h) Release of methyl mercury from the algae was small and 63.7 percent of methyl mercury remained after washing with 500 ml of demineralized water, and 70 percent of methyl mercury remained after washing with the same amount of the EDTA solution. Fifty-four percent of the methyl mercury remained after washing with 10 ml of Na_2S solution, but no further removal of methyl mercury was noted when additional amounts of Na_2S solution were used.

(i) During desorption of methyl mercury from Chlorella the total activities in the culture and the activities associated with algae remained constant in the case of the basic culture medium. In the case of desorption in the methionine medium, both the total activities and the activities associated with the algae decrease with time.

In summary, the influence of the environmental factors could be classified as follows: the light intensity did not influence sorption and desorption of mercurials but pH influenced the sorption of mercurials; the sorption of mercuric mercury was influenced by calcium ions but was not influenced by magnesium ions; sodium and potassium ions did not influence the sorption of methyl mercury by the alga; and citric acid and EDTA solutions did not exert their chelating abilities on the desorption of mercuric mercury but DL-methionine solution exerted a greater chelation effect as compared to the above two solutions.

This research investigated the sorption and desorption of mercurials by Chlorella pyrenoidosa in particular, but an analysis of this investigation embodied an evaluation of uptake of radionuclides by plants. Radionuclides as well as trace elements are taken up by plants via an adsorption reaction which is followed by an absorption reaction. However, the uptake of radionuclides by plants may be classified as physical adsorption, chemisorption on the surface of plant, and absorption. The process that prevails depends upon the type of radionuclide, the type of plant, the physiological stage of of plant, the reaction period, the temperature, and other environmental factors.

The results of this investigation showed the effect of algal density on the accumulation of mercurials by Chlorella. An increase of algal biomass resulted in a decrease in the equilibrium concentration and the amount of radionuclide concentrated by the test alga. The influence of the algal biomass on the accumulation of mercurials is important especially when considering the transport of mercurials through the food chain of aquatic ecosystems. The transport pattern of mercurials through the food chain may vary considerably. In general, in an oligotrophic ecosystem the ecological efficiencies are high and the mercury accumulation in top carnivores (fish) is relatively high compared with a eutrophic type of ecosystem.

The toxicity of mercuric chloride and methyl mercuric chloride to Chlorella pyrenoidosa was related to algal density. Thus, sorption isotherms may be applied to the evaluation of the emergency release of toxic mercurials.

4. MATHEMATICAL SIMULATION

4.1 Mathematical Model for Transport of Radionuclides

A mathematical model describing the transport and distribution of radionuclides in the environment was developed and presented previously (12). A transport function was derived by applying the sorption and desorption concepts to the mass balance principle. Homogeneous distribution of various solid media and constant inflow of water were assumed. The validity of the model was examined by comparing the predicted ^{85}Sr concentration with the result of experimental work. Model river studies were conducted using a steady-state influx of ^{85}Sr and an instantaneous release of ^{85}Sr and dye. Laboratory studies provided data on the sorption and desorption of ^{85}Sr by various aquatic components. Predictions based on the sorption-desorption model were found to be comparable with experimental data derived from the model river studies.

The overall transport model may be given by Eq. 4-1.

$$\frac{\partial C_j}{\partial t} = \frac{\partial}{\partial x}\left(E_x \frac{\partial C_j}{\partial x}\right) + \frac{\partial}{\partial y}\left(E_y \frac{\partial C_j}{\partial y}\right) + \frac{\partial}{\partial z}\left(E_z \frac{\partial C_j}{\partial z}\right) \qquad (4\text{-}1)$$

$$+ D_m \nabla^2 C_j - U \frac{\partial C_j}{\partial x} + \sum_{i=1}^{n} \alpha_i k_{ij}\left[C_{ij} - g_j(C_j)\right]$$

$$+ k_{dj} \frac{P}{A} (C_{dj} - C_j)$$

and

$$\frac{\partial C_{dj}}{\partial t} = k_{dj} \frac{P}{A_{dj}} (C_j - C_{dj}) \qquad (4\text{-}2)$$

$$\frac{\partial C_{ij}}{\partial t} = k_{ij}\left[g_j(C_j) - C_{ij}\right] \qquad (4\text{-}3)$$

where:

C_j = concentration of j^{th} radionuclide in water
α_i = total weight of i^{th} sorbent in unit volume
C_{ij} = the concentration of j^{th} radionuclide in i^{th} sorbent
$g_j(C_j)$ = equilibrium concentration function
k_{ij} = mass transfer function for j^{th} radionuclide associated with i^{th} sorbent
C_{dj} = concentration of j^{th} radionuclide in the dead zone
k_{dj} = mass transfer coefficient for j^{th} radionuclide in the dead zone
P = wetted contact length
A_{dj} = dead zone area
D_m = molecular diffusivity
E_x = turbulent diffusivity along x direction
E_y = turbulent diffusivity along y direction
E_z = turbulent diffusivity along z direction
i = index for sorbent; $i = 1, 2, \ldots, n$
j = index for radionuclide; $j = 1, 2, \ldots, m$

Each type of radionuclide was considered separately, and the detention caused by the dead zone was included. The dependent variable of radionuclide concentration and various chemical, physical, and biological complexities was expressed as the term, k_{ij}. The transport function, considering instantaneous injection of radionuclides into a model river system which has a bottom sediment exhibiting a high adsorption capacity, may be expressed by Eq. 4-4.

$$C_{(x,t)} = \frac{Mx}{2AU\sqrt{\pi D_x}} \exp\left(\frac{Ux}{2D_x} - k_1 t\right) N_{(x,t)} \qquad (4\text{-}4)$$

where:

$$N_{(x,t)} = \int_0^t I_1\left[2\sqrt{dk_1 K_s (t-\tau)\tau}\right]\sqrt{\frac{dk_1 K_s}{t-\tau}}\,\frac{1}{\tau}\,\exp-\left(\frac{x^2}{4D_x\tau} - B_\tau\right)d\tau$$

B = $\frac{U^2}{4D_x} + dK_s - k_1$

d = $\frac{k_1}{Ha}$

H = depth of water

a = sampling area of the sediment

k_1 = mass transfer coefficient of the radionuclide through the interface of the flowing water and bottom sediments:

$$k_1 = -\frac{1}{t} \ln \frac{K_s\,(C_w) - M}{K_s(C_w)} \qquad (4\text{-}5)$$

where:

K_s = equilibrium distribution coefficient of the sediments

I_1 = the modified Bessel function of the first order of the first kind

M = the total radionuclides in a constant sampling area (sample core of 0.6 sq in x 6 in)

D_x = longitudinal dispersion coefficient

C_w = radionuclide concentration in water

U = average velocity of flow

τ = the variable for time varying between 0 to t.

It is also possible to write Eq. 4-6.

$$M_{(x,t)} = k_1\, K_s \int_0^t \exp\left[-k_1(t - \lambda)\right] C_{(x,\lambda)} d\lambda \qquad (4\text{-}6)$$

where:

λ = dummy variable.

Since the integrand in Eq. 4-4 can not be integrated analytically, numerical integration is used.

The sorption-desorption model was also applied to plants. The transport function of radionuclides instantaneously injected into a model river system containing selected aquatic plants may be expressed by Eq. 4-7.

$$C_{(x,t)} = \frac{Mx}{2AU\sqrt{\pi D_x}} \exp\left(\frac{Ux}{2D_x} - K_2 t\right) N'_{(x,t)} \qquad (4\text{-}7)$$

where:

$$N'_{(x,t)} = \int_0^t I_1\left[2\sqrt{m_b K_2^2 K_c (t-\tau)\tau}\right] \sqrt{\frac{m_b K_2^2 K_c}{t-\tau}\frac{1}{\tau}} \exp - \left(\frac{x^2}{4D_x\tau} + B'\tau\right) d\tau$$

$B' = \frac{U^2}{4D_x} + m_b K_2 K_c - K_2$

m_b = biomass per unit volume

K_2 = mass transfer coefficient through the interface of water and aquatic plants

K_c = the equilibrium concentration factor of the aquatic plant for the radionuclide released.

Thus, the basic transport equation can be written, Eq. 4-8:

$$\frac{\partial C}{\partial x} = D_x \frac{\partial^2 C}{\partial x^2} - \bar{U}\frac{\partial C}{\partial x} - \sum_{i=1}^{n} f_i k_i \left[g_i(C) - C_i\right] \qquad (4\text{-}8)$$

where:

C = concentration of a particular radionuclide in the flowing stream at any point (x) and time (t)

D_x = longitudinal dispersion coefficient

$\bar{U}$ = average velocity of flow

f_i = mass of surface area of the ith sorbent affecting a unit volume of the flow zones

k_i = transfer rate coefficient for phase i

$g_i(C)$ = a transfer function relating to the concentration of the activity in water to the equilibrium level of activity in phase i

C_i = specific activity in the ith position of the n-sorption phases.

The first two terms in Eq. 4-8 define the mixing characteristics and dilution, while the third term describes uptake and release. In some cases g(C) may be represented by the Freundlich isotherm form, $g(C) = KC^n$, where K and n are constants pertaining to a given radionuclide and sorption phase. For uptake of most radionuclides n = 1.0 and K is approximately equal to K_d. A general relationship between the specific activity in sediments and plants as compared to the specific activity in the water is represented by Eq. 4-9.

$$\frac{\partial C_i}{\partial t} = k_i \left[g_i (C) - C_i \right] \qquad (4\text{-}9)$$

The activity level in sediments or plants was assumed to be linear, and it was represented as $g(C) = K_s C_w$ for sediments, and $g(C) = K_p C_w$ for plants. Therefore, the mass transfer coefficients for sediments and plants may be expressed as follows:

$$k_1 = \frac{1}{t} \ln \frac{K_s (C_w) - C_s}{K_s (C_w)} \qquad (4\text{-}10)$$

$$k_2 = \frac{1}{t} \ln \frac{K_p (C_w) - C_p}{K_p (C_w)} \qquad (4\text{-}11)$$

where:

k_1 & k_2 = mass transfer coefficients for sediments and plants $(t)^{-1}$, respectively

K_s = surface concentration coefficient of radionuclides by bed sediments at equilibrium

K_p = concentration coefficient of radionuclides by plants at equilibrium

C_w = specific activity of water at equilibrium
C_s = specific activity of sediments at time t
C_p = specific activity of plants at time t.

The mass transfer rate coefficient, k_1, was found to be dependent on turbulence. The k_1 values for ^{85}Sr uptake by sediments were found to be 0.0045 hr^{-1} and 0.011 hr^{-1} for flow conditions having Reynolds numbers of 2440 and 3700, respectively. Based on the average water temperature of 25°C, the equilibrium distribution coefficient, K_s, for ^{85}Sr was estimated to be 140 dpm/core/cpm/ml. The transfer rate coefficients for plants were much higher than those for bottom sediments (21). The transfer rate coefficients of plants for ^{134}Cs and ^{85}Sr were 0.0293 hr^{-1} and 0.0378^{hr-1}, respectively. In the case of sediments, the transfer rate coefficients for ^{134}Cs varied between 0.0014 hr^{-1} and 0.0018 hr^{-1} and for ^{85}Sr varied between 0.0010 hr^{-1} and 0.0013 hr^{-1}. Solutions for the basic transport model, Eq. 4-8, were developed and verified in the model river through the use of dye studies and the release of radionuclides.

The longitudinal mixing in the model river was approximated (12). The relationship between Sherwoods number and Reynolds number can be shown as follows:

$$S_h = 3.78(R_e)^{-0.8} \qquad (4\text{-}12)$$

where:

S_h = Sherwoods number
R_e = Reynolds number, $R_e = \frac{UR}{\theta} = 2{,}500$ to $10{,}000$
R = hydraulic radius.

The slope and the cross section of the model river were constant, and it was assumed that the only variable was the average velocity. Thus, the dispersion coefficient in the model river may be represented by Eq. 4-13.

$$D_x = 29(\overline{U})^{0.9} \quad (4\text{-}13)$$

where:

D_x = dispersion coefficient, ft^2/sec

$\overline{U}$ = average velocity, ft/sec

Longitudinal dispersion coefficients, D_x, were calculated (20) for velocities ranging from 0.33 to 3.30 ft/min, Eq. 4-14. Time-concentration curves developed with dye studies (5,7,9,12,14), under various flow conditions in the flume, provided the basic information.

$$D_x = 3.26\,(\overline{U})^{0.607} \quad (4\text{-}14)$$

4.2 Numerical Solutions of Radionuclide Transport Equations

A numerical dispersion model was developed to predict the net transport of radionuclides in water and uptake by other components of ecosystems (14). The transport of radionuclides in flowing aquatic systems was predicted by an explicit-difference numerical solution derived from the following transport equation:

$$\frac{\partial C}{\partial t} = D_L \frac{\partial^2 C}{\partial x^2} - \overline{U}\frac{\partial C}{\partial x} \quad (4\text{-}15)$$

where:

D_L = dispersion coefficient

$\overline{U}$ = average velocity of flow.

Since the dispersion equation is written as a function of some time, T, and some distance, L, by taking $\Delta X = L/J$ (J being an integer) and $\Delta t = T/N$ (N being an integer) as increments of the variables x and t, the set of points in the x,t - plane was presented as a net and was given by $x = j\Delta x$ and $t = n\Delta t$, where $j = 0, 1, 2, \ldots J$ and $n = 0, 1, 2, \ldots, N$. The concentration at each mesh point was approximated by $U(j\Delta x, n\Delta t)$ which was denoted by U_j^n. Using the explicit method in which derivatives were approximated by expanding U_j^n in the Taylor series, the numerical solution was expressed as:

$$U_j^{n+1} = A\,U_{j-1}^n + BU_j^n + CU_{j-1}^n \tag{4-16}$$

where:

$$A = \frac{D_L\,\Delta t}{(\Delta x)^2} - \frac{\overline{U}\Delta t}{2\Delta x}$$

$$B = 1 - \frac{2D_L\Delta t}{(\Delta x)^2}$$

$$C = \frac{D_L\,\Delta t}{(\Delta x)^2} + \frac{\overline{U}\Delta t}{2\Delta x}\,.$$

Two stability conditions were derived from the coefficients, A, B, and C:

$$\frac{\Delta t}{\Delta x}\,\overline{U} \leq 1\ ;\ \frac{D_L\,\Delta t}{(\Delta x)^2} \leq 1/2$$

An assumption was made that convergence occurs as Δx, $\Delta t \rightarrow 0$, but it was pointed out that $\Delta t/\Delta x$ is inversely proportional to $\overline{U}$. Therefore, for large velocities, extremely small mesh sizes should be used. The error obtained in the numerical solution in the dispersion equation is dependent not only on Δx and Δt but on the size of D_L and $\overline{U}$. Probably only a portion of concentration-time curves, instantaneous releases, should be used to calculate the dispersion coefficient and the radionuclide-cloud velocity. The solutions based on best-fit approximations can be obtained when the cut-off point is taken to be that interval in which the average concentration is just less than one-tenth of the observed peak concentration. Dispersion coefficients, using the two station method, allowed accurate prediction of the concentration-time curves from which they were calculated. The numerical solution of the dispersion model predicted transport curves of ^{85}Sr after instantaneous release with less than 3 percent error. Continuous release curves of ^{85}Sr were also modeled with the same numerical solution. It was emphasized that the previously presented method (12) overpredicted the dispersion coefficient and underestimated the tracer cloud velocity.

The dispersion equation with a sorption term for plants may be expressed by Eq. 4-17.

$$\frac{\partial C}{\partial t} = D_L \frac{\partial^2 C}{\partial x^2} - \bar{U} \frac{\partial C}{\partial x} + m_b k_2 \left[C_p - K_c C\right] \qquad (4\text{-}17)$$

where:

C_p = radionuclide concentration in plants
k_2 = mass transfer coefficient
K_c = function relating plant activity to that in water
m_b = biomass of plants.

A numerical solution to the dispersion equation, including a plant sorption term can be written as Eq. 4-18.

$$U_j^{n+1} = AU_{j+1}^n + BU_j^n + CU_{j-1}^n + k_2 m_b \Delta t \left[W_j^n - K_c U_j^n\right] \qquad (4\text{-}18)$$

where the constants A, B, and c are given in Eq. 4-16. W_j^n is an approximation of sorption and may be calculated:

$$W_j^n = W_j^{n-1} + k_2 \Delta t \left[K_c U_j^{n-1} - W_j^{n-1}\right] \qquad (4\text{-}19)$$

Equation 4-19 is a numerical solution for the sorption-desorption equation developed previously (12). The stability conditions may be presented as:

$$\frac{2D_L\Delta t}{(\Delta x)^2} - K_c k_2 m_b \Delta t \leq 1; \frac{\Delta t}{\Delta x}\bar{U} \leq 1 \qquad (4\text{-}20)$$

Plant uptake was modeled using the sorption model. Predicted curves provided reasonable approximations to the observed data. Estimates of the mass transfer coefficient, k_2, and the equilibrium concentration factor, K_c, were developed for each system since attached algae markedly affected these values. The sorption model is:

$$k_2 = -(1/t) \ln [(K_c C - C_p)/K_c C] \qquad (4\text{-}21)$$

where:

C = the concentration of ^{85}Sr in water

C_p = the specific activity of ^{85}Sr in plants.

Values for k_2 in the model river ranged from 0.0126 hr^{-1} to 0.0322 hr^{-1} with an average of 0.022 hr^{-1}. Values for K_c ranged from 260 dpm/gm dry wt/dpm/ml in aquaria studies to 680 dpm/gm/dpm/ml in the model river studies. Attached algae raised the latter figure to about 3000 dpm/gm/dpm/ml. Two solutions were obtained using the Runge-Kutta method. Values for K_c from the aquaria studies (260 dpm/gm/dpm/ml) and k_2 of 0.03 hr^{-1} from the continuous release experiment were used in the equation to obtain one solution. A second solution was obtained by using an equation which in the term C^n, n had the value of 1.12. The analytical solution of the dispersion equation with the values for D_L of 1.0 ft^2/min and $\bar{U}$ of 0.52 fpm was used as the non-linear input for C.

It was concluded that numerical solutions for the dispersion equation and for the dispersion equation with a sorption term for plants hold more promise than analytical solutions because of the ease of setting boundaries and initial conditions.

4.3 Simulation of Radionuclide Transport

A simulation model for the transport of radionuclides in rivers was developed (14). The model included stochastic, temporal-flow variation; deterministic longitudinal-flow variation; three-dimensional, channel-geometry variation of time and distance; generalized boundary sorption; boundary erosion; and flooding. The transport model was written in Fortran IV computer language and possessed five hydraulic subroutines. All hydraulic relationships used in simulation were Fortran subroutines, except for the routing procedure. It was pointed out that the Lagrangian routing technique employed is particularly applicable to variable and instantaneous radionuclides releases if some type of intersegment mixing is incorporated into the model. The main program of the model contained the sorption-desorption and scour-deposition operations. The benthal system (sorption) in the model represented the combined sorption of all immobile components, and a single distribution coefficient characterized the system.

The important input data for the model were the mean river flow rate, the temporal flow variation scheme, the longitudinal variation of flow, and sorption parameters. Hydrologic statistical variations were studied and the effects of variations in radioactive half-life, critical scour velocity, surface concentration distribution coefficient, mass transfer coefficient, and sorption reversibility were also investigated. Different parametric values were assumed to demonstrate the effect of each parameter on the system. The standard output from the model was the monthly transport and storage data for stations at 10, 100, and 1000 miles below the waste source, plus total storage on the flood plain above the 1000-mile station. In parametric studies, the standard physio-chemical parameters were: (a) a nondecaying nuclide released at a constant rate of 0.01 curie per day; (b) totally reversible sorption by the benthal system characterized by a mass transfer coefficient of 1.0 per day; and (c) a surface concentration distribution coefficient of 100 feet. Rivers of erratic flow, gradually varying flow, and non-varying flow were studied. By simple modifications of the model, special cases including two sources of radionuclide discharge and a reservoir with a high trap efficiency were demonstrated.

Considerable insight has been gained into the behavior of radionuclides in a river system using this simulation model. Reasonable results were obtained with the model using prototype data for river flow rates, channel shape, and benthal system composition. In simulating large rivers, the model indicated that benthal sorption had little effect on transport of long-lived radionuclides on a long-term basis. However, a significant effect was observed on the transport of short-lived radionuclides. The model further indicated that rapid releases of radionuclides from the benthal system can not increase concentrations of radionuclides in a river's maximum permissible levels. Flow variability had minor effects on the transport of long-lived radionuclides on a long-term basis, but major effects for short-time periods. Greater transport of short-lived nuclides was observed in more variable rivers due to the effects of high flows. The model predicted surface concentration of long-lived radionuclides deposited on the flood plain to be insignificant on a hazard basis, although as much as a curie of radionuclides was deposited over a distance of one thousand miles by a single flood.

Variations in the mass transfer coefficients exhibited insignificant effects on the transport of radionuclides through the river system, but variations in surface concentration distribution coefficients were influential on transport and storage. The degree of reversibility of benthal system sorption exhibited little effect on transport or storage on either an annual or long-term basis when substantial erosion of the system occurred at flows above the mean annual flow. However, irreversible sorption caused higher storage maxima which were maintained for two years in the benthal systems which were more resistant to erosion. Studies on continuous sedimentation zones indicated that significant quantities of long-lived radionuclides may accumulate therein. The model indicated that large reservoirs which trap all suspended solids from the incoming flow may accumulate long-lived radionuclides at a rate as high as half the waste release rate. The model demonstrated the possible environmental effects of radioactive wastes discharged to rivers with low concentrations over several decades. This study has demonstrated techniques for modeling the transport of radionuclides on a long-distance and long-time basis. Detailed modeling for short-time and distance transport can be accomplished with adaptations of mathematical models presented previously (12) (14).

A general model, in a form suitable for simulation on a digital computer, for routing a radionuclide through a model river (flume) was developed (19). The study was restricted to flume data, however, it was also kept in mind that the model would eventually be applied to an actual river system. Therefore, the basic capabilities of this simulation model were summarized as follows:

(a) it distributes radionuclide activity by advective and dispersional mechanisms along with the longitudinal axis of the system;

(b) it may be discretized into any number of segments (vertical planes normal to the longitudinal axis) as may be appropriate to describe the spatial variations in radionuclide activity;

(c) it is capable of treating instantaneous, continuous, or time-varying releases of radioactive activity;

(d) it provides for a temporal description of radionuclide activity throughout the system; and,

(e) it provides for sorption and desorption by both plants and bottom sediments.

Verification of the mathematical model was conducted in three phases. In the first phase the dispersion characteristics of the flume were verified using the dye-release data presented in (14). In the second phase the bottom sediments sorption and desorption data presented in (12) were incorporated into the model. The final phase was concerned with the inclusion of sorption and desorption by aquatic plants into the model using the data presented in (14). Both the above used ^{85}Sr as the radionuclide. The results, using sorption terms for bottom sediments and plants showed good agreement between the predicted and observed curves. The transport equation with sorption terms for both bottom sediments and plants was also used in the modeling of continuous releases of radionuclides. Applications of the model to a real river system and its limitations, as well as the data requirements for a normal river simulation problem were analyzed.

The numerical solutions for the basic transport model, Eq. 4-8, were used to predict the dye time-concentration curves and the transport of the continuous releases of ^{134}Cs and ^{85}Sr in the flume, and proved to be satisfactory (21).

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