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BIOACCUMULATION AND DISTRIBUTION OF ^{95m}Tc IN AN EXPERIMENTAL FRESHWATER POND*

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Abstract

BIOACCUMULATION AND DISTRIBUTION OF ^{95m}Tc IN AN EXPERIMENTAL FRESHWATER POND.

An acute release of ^{95m}Tc was made to a small experimental freshwater pond to determine the behaviour of technetium in a freshwater ecosystem. The objectives of the study were (1) to determine the distribution of ^{95m}C in the components of the ecosystem and (2) to determine the concentration in freshwater biota. Prior to the release of ^{95m}Tc, the pond was stocked with aquatic macrophytes, fish, and invertebrates. All components of the pond were sampled for a period of 37 days. Analyses of filtered and unfiltered water samples showed that ^{95m}Tc did not sorb significantly to particulates suspended in the water but remained dissolved. Sediments accumulated ^{95m}Tc slowly as the experiment progressed. In the biota, periphyton accumulated ^{95m}Tc rapidly, reaching the highest concentration (3482 dis/min per g dry wt) 4 hours after the release and maintaining a relatively high concentration throughout the experiment. Fish and invertebrates accumulated ^{95m}Tc gradually. Elimination studies and tissue analyses showed that a large percentage of the body burden was in the digestive system of all fish, suggesting that fish were accumulating ^{95m}Tc through the food chain. Biological half-lives determined from elimination studies for carp *(Cyprinus carpio)*, mosquitofish *(Gambusia affinis)*, and snails *(Helisoma* sp.) were 2.5, 4.3, and 21.3 days, respectively. Calculated concentration factors for the same species were 11 for carp, 75 for mosquitofish, and 121 for snails. The estimated size of the biomass components in the ecosystem in descending order were: periphyton, macrophytes, invertebrates, fish, and algae. Based on biomass estimates and concentrations of the ^{95m}Tc in the aquatic biota, approximately 1% of the ^{95m}Tc accumulated in the biota. Thus, most of the technetium released into a freshwater pond ecosystem remained dissolved in the water with only a small percentage accumulating in the biota and sediments.

1. INTRODUCTION

The sources of technetium and its behavior in the environment have been reviewed by Wildung et al. [1]. Technetium in the environment is primarily the result of fallout from nuclear weapons testing, releases from enrichment facilities and nuclear waste operations, and disposal after pharmaceutical use. Although ^{99}Tc which is found in fallout has a

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long half-life (2.15×10^5 a) and a fission yield approximately equivalent to ^{90}Sr or ^{137}Cs, few measurements have been made of its concentrations in the environment.

With the exception of data reported from the present studies [2], very little data exist on the behavior of technetium in freshwater biota; however, some data are available for marine biota. The whole-body concentration factor for ^{99}Tc in the red abalone, Haliotis rufescens, ranged from 135 to 295 [3]. A comparison was made of the concentration of ^{99}Tc and ^{037}Cs in the crab (Cancer pagurus), the mussel (Mytilus edulis), and the brown algae (Fucus vesiculosus) collected from the Irish Sea near Windscale [4]. Concentration factors in excess of 10^4 have been reported for some species of marine algae [5].

In this study, ^{95m}Tc was released to a small experimental freshwater pond to determine the distribution of ^{95m}Tc in the freshwater ecosystem and its concentration in freshwater biota.

2. MATERIALS AND METHODS

An acute release of ^{95m}Tc (γ emitter, $T_{1/2}$ = 61 d, as NH_4TcO_4) was made to a small experimental freshwater pond. Thereafter, four water samples (two samples near the surface and two near the bottom) were collected daily for the first 10 d and at least five times weekly for the remainder of the study. Samples were analyzed in an Armac Auto-Gamma spectrometer to determine the concentration of ^{95m}Tc in nonfiltered water; a subsample was filtered through an 0.45-μm Nucleopore filter.

Wellwater (alkalinity, 122 mg/ltr and hardness, 125 mg/ltr), was used to replace evaporation and drainage losses; rainfall dilution was monitored. The pH of the pond water ranged from 8.0 to 8.3, the dissolved oxygen from 7 to 11 mg/ltr and the temperature from 24 to 30°C.

The pond was stocked with aquatic macrophytes, fish, and macroinvertebrates 112 d prior to the release of ^{95m}Tc. The macrophytes consisted of cattails (Typha latifolia), Ceratophyllum demersum, Myriophyllum spicatum, and Elodea canadensis. Three species of fish (mosquitofish, Gambusia affini; bluegill, Leopomis macrochirus; and carp, Cyprinus carpio) were released into the pond. The major macroinvertebrates introduced into the pond were a species of snail (Helisoma sp.) and a species of crayfish (Orconectes sp.). Corbicula minlensis, asiatic clam, were placed in metal baskets and kept in the pond throughout the experiment.

The fish population in the pond at the time the ^{95m}Tc was released consisted of approximately 37 carp ranging from 6 to 13 cm in length, 23 bluegill ranging from 6 to 6.5 cm, and more than 50 mosquitofish

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ranging from 0.5 cm to 4.5 cm. To facilitate the capture of the fish, ten individuals of each species were captured from the pond and placed in a 100-ltr holding cage that rested on the bottom of the pond. Five fish were removed 4 h after the initial release and live counted to determine the whole body concentration of ^{95m}Tc. The fish, which were returned to the cage, were analyzed daily for their radioactive content for the first 4 d and weekly thereafter until completion of the experiment. Fish from the pond were also captured at various intervals for whole body analysis to compare with the caged fish.

One hundred adult snails were originally stocked in the pond. Ten snails were collected by grab sampling 4 h after the ^{95m}Tc was released. The whole-body concentration of ^{95m}Tc was determined for each snail. The snails were weighed and numbers were painted on their shells for identification before they were returned to the pond. Five snails were collected daily for the next 3 d and weekly for the remainder of the experiment. Before analyzing the snails in a gamma spectrometer, the algae was washed from their shells.

The four species of macrophytes which developed in the pond and the algae (Spyrogyra) were collected by grab sampling. The plants were separated according to species and washed twice before ^{95m}Tc determination.

Periphyton was sampled from 5- to 10-cm plastic slides which had been placed in the pond 30 d before the release. Twenty slides were suspended near the surface of the water and 20 were placed on the bottom of the pond. Two slides were sampled from each location 4 h after the ^{95m}Tc was released, daily for the next 3 d, and weekly thereafter for the remainder of the experiment. Sediment was sampled by taking cores 2.2 cm in diameter from each quadrant of the pond. Samples were dried at 100°C for 24 h to determine the dry weight and ashed at 450°C to determine the organic content.

Macrophytes and periphyton were also sampled (stratified random procedure) at the end of the experiment to estimate their biomass. A similar procedure was used to sample the periphyton that had developed on the sides of the pond.

3. RESULTS

3.1. Water

A large percentage of the $^{95m}TcO_4$ released into the experimental pond remained dissolved in the water during the experimental period. The concentration of ^{95m}Tc decreased from 91.5 dpm/mltr 1 h after the

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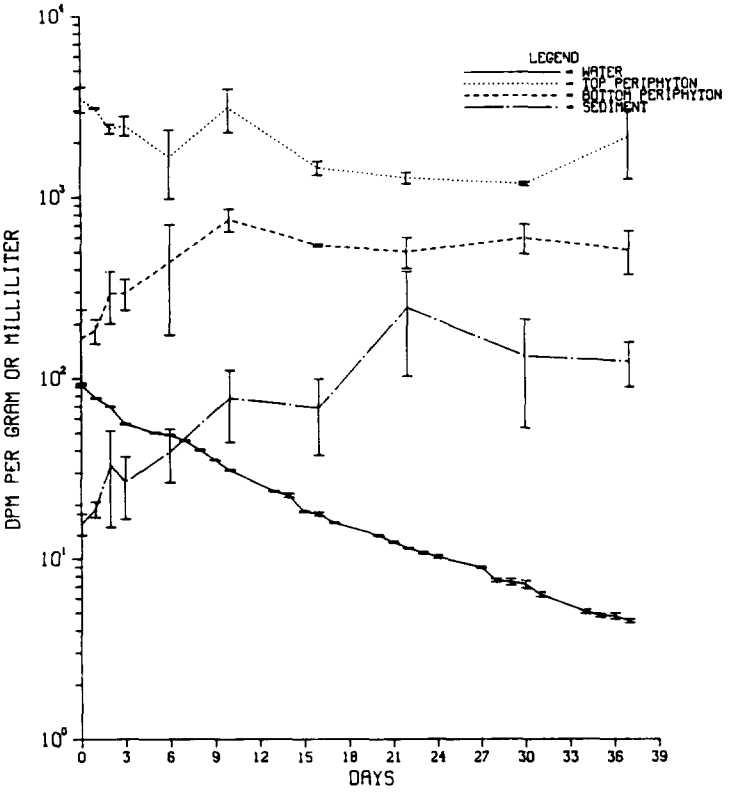

FIG. 1. Mean concentrations of ^{95m}Tc (± 1 SE) for pondwater (dpm/ml), periphyton and sediments (dpm/g dry wt) in an experimental freshwater pond. Values are given for periphyton collected from slides near the surface of the water and from slides on the sediment.

release to 4.5 dpm/mltr 37 d later (Fig. 1).[1] Filtering the water removed less than 5% of the ^{95m}Tc; thus, it appeared that ^{95m}Tc released in the pertechnetate form does not adsorb to the particulate material in the pond water.

In addition to the decay of the isotope (6.98 x 10^8 dpm to 4.59 x 10^8 dpm on day 37), the activity in the water decreased because of dilution by rainfall and inflow of water. Drainage and seepage from the pond rather than sediment sorption accounted for most of the loss of ^{95m}Tc from the system. Accumulation by biota and sediment accounted for only a small fraction of the ^{95m}Tc that was released into the pond.

[1] 60 dpm (dis/min) = 1 Bq.

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3.2. Sediment

The concentration of ^{95m}Tc in the pond sediment increased gradually from 15.6 ± 4.3 dpm/g to 245.4 ± 285.8 dpm/g dry wt on day 22 (Fig. 1). At first, ^{95m}Tc in the sediment was uniformly distributed, but, as the experiment progressed, more variability was observed in the samples. The organic content of the sediment was relatively uniform ranging from 3.5 to 5.8% and was not correlated with ^{95m}Tc content.

3.3. Periphyton

Periphyton growing on plastic slides near the surface of the water accumulated ^{95m}Tc rapidly and maintained a relatively high concentration for 37 d. Four hours after ^{95m}Tc was released to the pond, periphyton had accumulated 3481.8 dpm/g dry wt - a concentration higher than that for the other biota. Periphyton growing on similar slides lying on the sediment (bottom) did not accumulate ^{95m}Tc as rapidly as the surface (top) slides (Fig. 1). The accumulation of ^{95m}Tc on the bottom slides increased gradually, whereas on the surface slides there was a rapid uptake by periphyton, followed by a gradual decrease.

The percent (19.2 ± 1.0) of the organic content from the top slides was approximately four times greater than the percent (5.6 ± 1.0) organic content on the bottom slides. However, the concentration of ^{95m}Tc in the periphyton of the surface slides ranged from 21 times greater 4 h after the release to 2.5 times greater on day 24 of the experiment. The percentage of the organic content of the bottom slides was approximately the same as the organic content of the sediments; however, the concentration ranged from 2 to 10 times greater than in sediments. Apparently ^{95m}Tc on the bottom slides was accumulated in living material growing on the slides or in material that had incorporated ^{95m}Tc and settled to the bottom. This could also be true for the surface sediments which gradually increased in concentration of ^{95m}Tc during the experiment.

3.4. Macrophytes and algae

Three submergent macrophytes (Ceratophyllum, Myriophyllum, and Elodea) and one emergent macrophyte (cattail) were collected from the pond and analyzed for ^{95m}Tc. Only Myriophyllum and Ceratophyllum were sampled throughout the experiment. The mean concentrations for the two species are shown in Fig. 2. Single grab samples of Elodea were collected on five dates. The concentrations of ^{95m}Tc in Elodea were similar to those of the other macrophytes for the same day.

After only 4 h the macrophytes (including roots, stem, and leaves) had accumulated a significant amount of ^{95m}Tc. This initial accumulation could be the result of surface adsorption; however, the macrophytes continued to accumulate ^{95m}Tc for the next 10 to 20 d.

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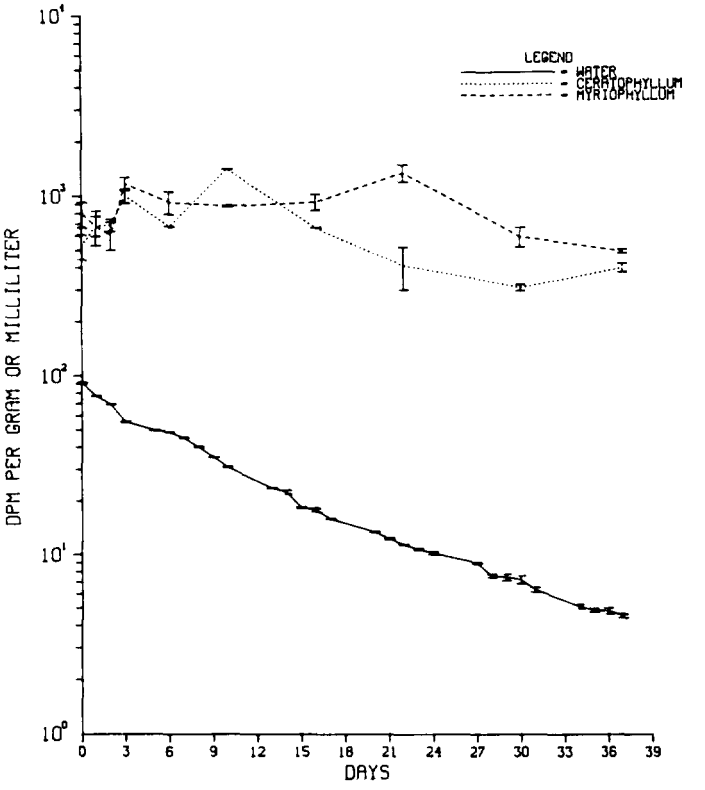

FIG.2. Mean concentrations of ^{95m}Tc *(± 1 SE) in aquatic macrophytes* (Ceratophyllum demersum *and* Myriophyllum spicatum*) (dpm/g dry wt), and pondwater (dpm/ml) from an experimental freshwater pond.*

The roots, stems, and leaves of cattails were analyzed separately. The roots accumulated ^{95m}Tc at a rapid rate which was quickly translocated to the leaves. After 4 h of exposure, the leaves contained 101.5 dpm/g dry wt, while the roots contained 461.3 dpm/g dry wt.

On day six of the experiment and for the remainder of the sampling period, green algae (Spirogyra) were collected for analyses. The concentration of ^{95m}Tc in the algae ranged from 662 dpm/g dry wt on day six to 3045 dpm/g on day 22.

3.5. Macroinvertebrates

The macroinvertebrates introduced into the pond were clams, crayfish, and snails. Accumulations of ^{95m}Tc by clams and snails are shown in Fig. 3. Whole-body concentrations in the filter-feeding clam

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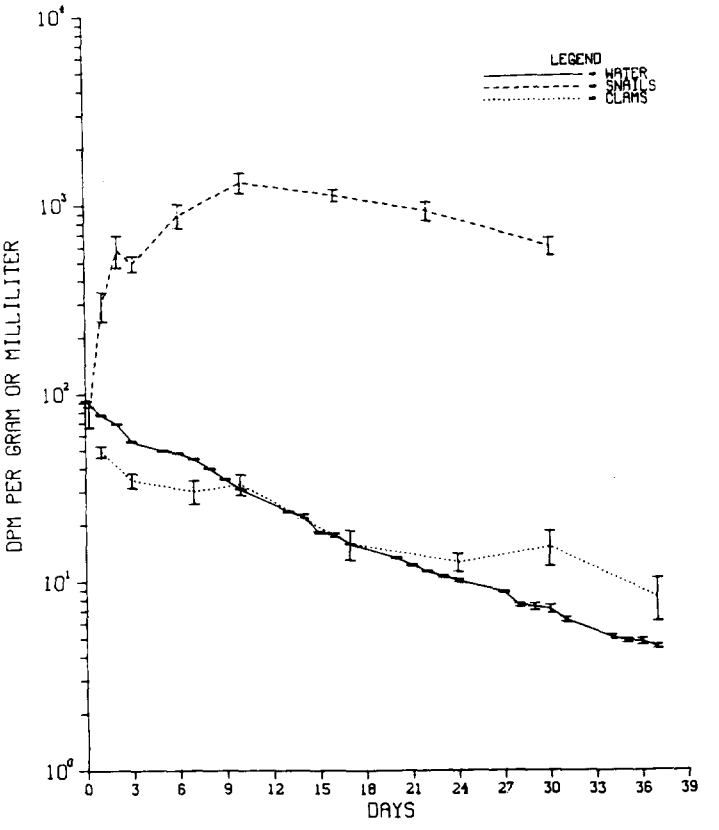

FIG.3. Mean concentrations of ^{95m}Tc *(± 1 SE) in the clam* (Corbicula manilensis) *and snail* (Helisoma *sp.) (dpm/g whole-body fresh wt) and in pondwater (dpm/ml) from an experimental freshwater pond.*

reached approximately the same concentration as the water and closely paralleled the concentrations in the water. The accumulation of ^{95m}Tc by snails was much greater than that in the clam and appeared to be more closely related to the concentration of in periphyton, their food source, than to the concentration of ^{95m}Tc in the water.

On day 7 of the experiment, three snails and four clams were sacrificed to determine the concentration of ^{95m}Tc in the soft tissue and shell. The concentration in the soft tissue of the snail at that time was approximately six times that in the shell. The soft tissue, which included the digestive system and constituted approximately 46% of the weight of the snail, contained 82% of the ^{95m}Tc. The concentration of ^{95m}Tc in the soft tissue of the clam was greater than that in the shell by a factor of 4. The soft tissue of the clam averaged 38% of the body weight and contained approximately 70% of the activity.

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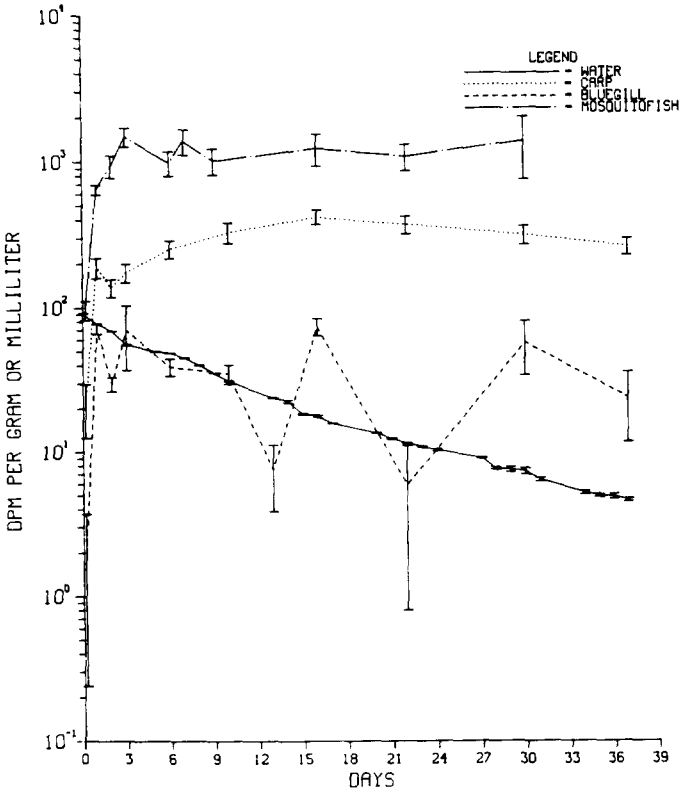

FIG.4. Mean concentrations of ^{95m}Tc *(± 1 SE) in bluegill* (Lepomis macrochirus), *carp* (Cyprinus carpio), *and mosquitofish* (Gambusia affinis) *(dpm/g fresh wt of whole fish) from an experimental freshwater pond.*

3.6. Fish

The accumulation of ^{95m}Tc by bluegill, carp, and mosquitofish is shown in Fig. 4. The concentration was greatest in the mosquitofish and lowest in bluegill.

An analysis of tissue and the gastrointestinal (G.I.) tract (Table I) from the three species of fish showed that the G.I. tract contained a large fraction of the ^{95m}Tc. For example, the G.I. tract of the mosquitofish constituted about 8% of the body weight but contained an average of 37% of the total body activity. Mosquitofish, which had the highest concentration in their G.I. tracts, also had the highest whole body concentration of ^{95m}Tc. Contents of the digestive systems could account for the variability in the concentrations observed in the whole-body

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TABLE I. MEAN CONCENTRATIONS OF ^{95m}Tc (dpm/g fresh wt) IN THREE SPECIES OF FISH AFTER SEVEN DAYS OF EXPOSURE IN AN EXPERIMENTAL POND

Tissue	Bluegill[a]	SD	Carp[b]	SD	Mosquitofish[b]	SD
Muscle	6.3	3.0	49.2	25.1	323.0[c]	94.3
Bone	26.8	37.9	91.4	20.0		
Gills	8.9	12.5	102.1	18.4	1595.5	185.0
Skins and scales	21.7	2.3	68.5	8.4	1317.4	354.5
G.I. tract	211.8	14.9	1525.5	711.5	4576.4	1483.6

[a] n = 2.

[b] n = 3.

[c] Bone and muscle.

analysis of live fish. These observations suggest that the accumulation of ^{95m}Tc by fish was related to their feeding habits. The source of food for mosquitofish can be closely related to the periphyton. Carp, a bottom-feeding fish, accumulated ^{95m}Tc gradually as the sediment concentration increased. Bluegill, an omnivore, had the lowest concentration of the three species tested and showed considerable variability in whole body concentrations. These observations can be related to an opportunistic feeding habit.

3.7. Uptake and elimination of ^{95m}Tc by fish and snails

Uptake and elimination of ^{95m}Tc by carp, mosquitofish, and snails are shown in Fig. 5. The uptake data represent the mean concentration of ^{95m}Tc measured in these three species of biota collected from the experimental pond. The elimination data represent the mean concentrations measured in carp, mosquitofish, and snails after they had been removed from the experimental pond and placed in a holding cage in an adjacent control pond. A Marquardt nonlinear least-squares technique [6] was used to fit the data to curves to determine the uptake and elimination rates.

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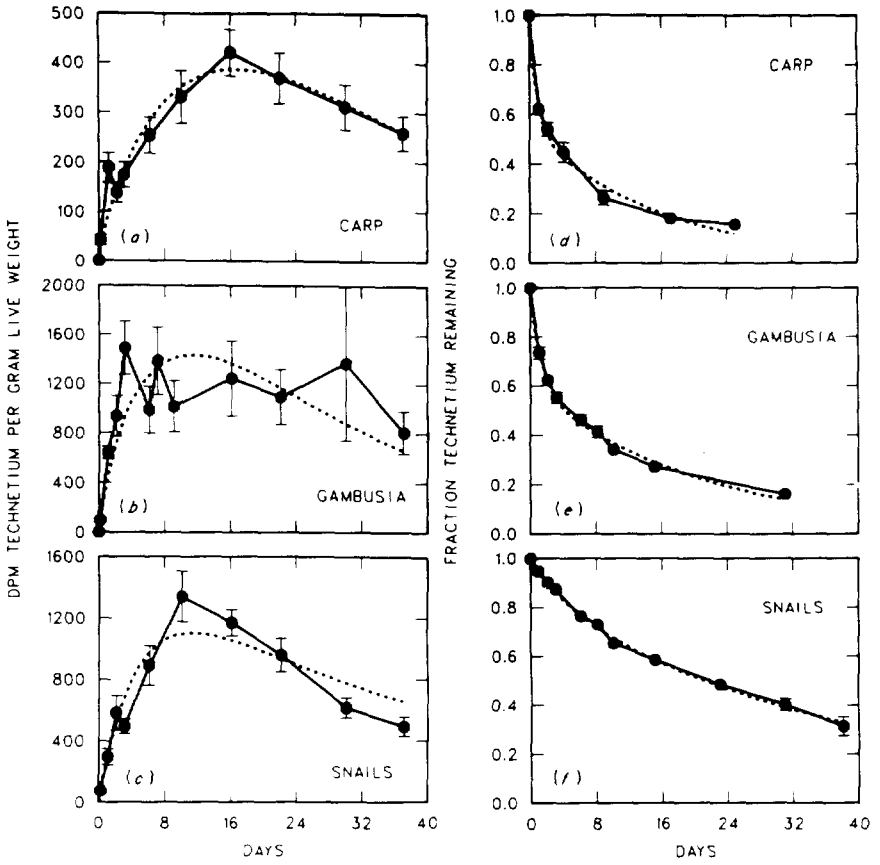

FIG.5. Uptake of ^{95m}Tc *is shown for (a) carp, (b) mosquitofish, and (c) snails in an experimental freshwater pond that recieved an acute release of* ^{95m}Tc. *Elimination of* ^{95m}Tc *is shown for (d) carp, (e) mosquitofish, and (f) snails after they were removed from the pond. The solid line connects data points representing the mean concentration of* ^{95m}Tc *(± 1 SE). The data points were used with a Marquardt nonlinear least-squares technique to determine the uptake and elimination rates of the isotope.*

Carp, mosquitofish, and snails reached their maximum concentrations of ^{95m}Tc in 10 to 14 d. The effective biological half-lives of ^{95m}Tc for carp, mosquitofish, and snails were 2.5, 4.3, and 21.3 d, respectively. When corrected for physical decay of the isotope, the biological half-life for the same organisms would be 2.6, 5.0 and 38.8 d.

3.8. Distribution of ^{95m}Tc in the components of the pond ecosystem

After termination of the experiment, the pond was destructively sampled to determine the biomass of the macrophytes and periphyton.

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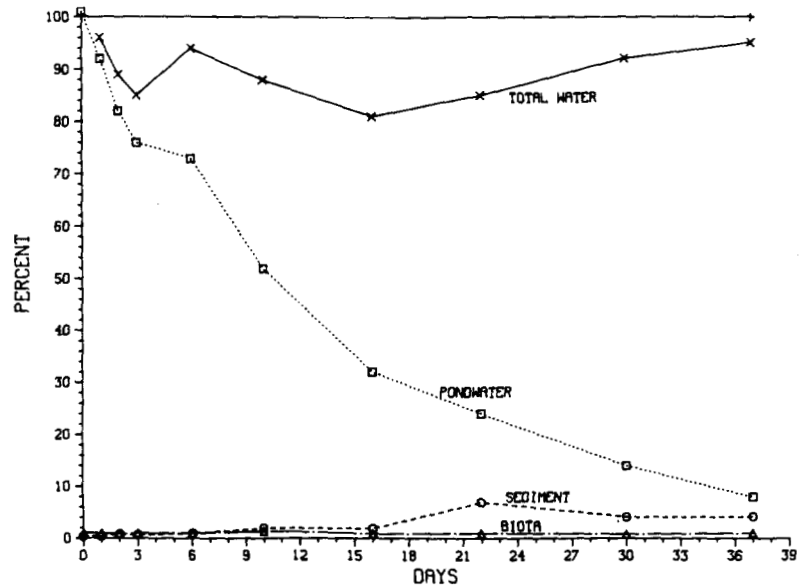

FIG.6. The distribution of ^{95m}Tc is shown for the components of a freshwater pond ecosystem after an acute release of the isotope. The percentage in each component of the pond represents the total amount of ^{95m}Tc in that component at a particular sampling time which was then divided by the amount of ^{95m}Tc released after correction for physical decay. The percentages of ^{95m}Tc in the total water represent the sum of the ^{95m}Tc in the pondwater and in the water lost by drainage or seepage.

The biomass of the fish and macroinvertebrates populations was estimated from the number of individuals released into the pond and the range of weights accumulated during the routine sampling of the pond. During the experiment, growth of individual organisms was insignificant; therefore, the same biomass was used for each compartment throughout the experiment.

The distribution of ^{95m}Tc in the various components of the pond ecosystem for each sampling period is shown in Fig.6. The biomass of the aquatic biota was assumed to be constant throughout the experiment. The amount of water in the pond fluctuated with inflow of water and rainfall and subsequent seepage from the pond. In Fig. 6, 100% represents the amount of ^{95m}Tc that was released into the pond and the quantity at each sampling period after physical decay of the isotope. The percentage of ^{95m}Tc in water represents the amount of ^{95m}Tc in the pond water in addition to the percentage lost by drainage and seepage. The percentage in the sediment and in the biota represents the amount of ^{95m}Tc related to the total amount released. The relationships between the concentrations in the biota and pond water are shown in Figs. 1-5.

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4. DISCUSSION

Under the range of environmental conditions given for the experimental pond in this study, one would predict that most of the technetium released in the form of a pertechnetate to a freshwater system would remain in the water. Only a small percentage of the total amount released would accumulate in the biota and sediments. Thirty-seven days after the release of ^{95m}Tc to the experimental pond, less than 1% of the total amount remained in the biota, while approximately 4% was in the sediments and 8% was in the pond water. The remainder had been transported from the system by drainage and seepage. Thus, it appears that, if technetium was released to a freshwater system with a rapid turnover, most of the technetium would be transported through the system in the water column.

Periphyton accumulated ^{95m}Tc rapidly and maintained a relatively high concentration throughout the experiment. Apparently technetium accumulates in rapidly growing organisms that have a large surface to volume ratio. Spies [3] attributed the relatively high concentration of ^{99}Tc in the shell of the red abalones to the epifauna and periphyton growing on the shell. Birks [5] reported that some marine algae concentrated ^{99}Tc to more than 10^4 times the concentration in water. In field studies in the Irish Sea, Pentreath et al. [4] confirmed these high levels of accumulation of ^{99}Tc in brown algae, Fucus vesiculoous. Although steady-state conditions were not achieved in the experimental pond, the greatest accumulation of ^{95m}Tc by the bluegreen alga, Spirogyra, occurred on day 22. The concentration in the algae (dry wt) was 270 times the concentration in water. Aquatic macrophytes are similar to terrestrial plants in their uptake and translocation of technetium. Gast [7] observed the translocation of ^{99}Tc to aboveground tissue in plants grown in soil watered with a solution containing ^{99}Tc pertechnetate. In the experimental pond, cattails had only their roots and lower stems submerged in water; however, after only 4 h of exposure, ^{95m}Tc was incorporated into the cattails and translocated to the above-water leaves where the concentration ratio (dpm g^{-1} dry wt/dpm/mL^{-1} water) was approximately 1. Although there was a continual decrease in the concentration of ^{95m}Tc in the water, both the submergent and emergent macrophytes continued to accumulate ^{95m}Tc, with the highest concentration ratio (60) occurring on day 22 of the experiment.

The distribution of ^{95m}Tc in the tissue of freshwater invertebrates (snails and clams) was similar to the distribution of ^{99}Tc in a marine invertebrate reported by Spies [3]. His report showed the concentration was highest in the soft tissue of the red abalone. In snails and clams from the experimental pond, the concentration of ^{95m}Tc in the soft tissue was greater than in the shell by factors of 2.5 and 4.0, respectively.

Because the release of ^{95m}Tc to the experimental pond was an acute release and the concentration in the water gradually decreased,

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equilibrium conditions in the biota would not be expected. However, the uptake and elimination data for fish and snails in Fig. 5 was used in a mathematical model [2] to estimate the steady-state body burden of these organisms in the experimental pond. Steady-state concentrations for carp, mosquitofish, and snails (whole body basis including gut-contents) were calculated to be 11, 75, and 121 times those in ambient water, respectively.

These values, although conservative, may be compared with the concentration factors listed in the U. S. Regulatory Guide [8] that are used for calculating the potential dose to man through the freshwater aquatic food chain. The value for carp was less than the concentration factor of 15 listed for fish in the regulatory guide, but the concentration factor for mosquitofish was approximately five times the regulatory guide value. The calculated value for snails, which was approximately 25 times the value of 5 given in the regulatory guide for invertebrates, showed the greatest deviation.

The values in the regulatory guide are for the edible portion of the fish and invertebrates, while the calculated concentration factors are for whole organisms. A more realistic comparison may be obtained by extrapolating the slow component of the elimination curves in Fig. 5 to time zero to obtain the fraction of the body burden not associated with the G.I. tract contents. Concentration factors for carp, mosquitofish, and snails calculated for this fraction of the organism were 6, 44, and 102, respectively. These estimates may still be conservative, especially for invertebrates, because the shell which is not edible is included in the estimates. However, the concentration factors for mosquitofish and snails still exceed the regulatory guide values.

It must be remembered that the Regulatory Guide concentration factors are default values based on the behavior of iodine in the environment, while the calculated values in this study are based on some of the first data available on the concentration of technetium in freshwater biota.

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