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Behavior of Technetium in Freshwater Environments*

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ABSTRACT

Behavior of Technetium in Freshwater Environments*

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Currently the major sources of ^{99}Tc released to the environment in the United States are gaseous diffusion plants that enrich reprocessed nuclear fuel. At the present time, these plants are located on freshwater systems. Most of the ^{99}Tc that escapes these facilities is released with the liquid effluents. Information on the behavior of technetium in aquatic ecosystems is limited; most data on the uptake of technetium by aquatic organisms are restricted to marine environments. This study provides information on the behavior and fate of technetium in freshwater ecosystems to improve the data base available for assessing technetium's potential radiological impact to humans.

In a previous study, ^{95m}Tc, as a pertechnetate, was released to a small, experimental, freshwater pond, and the concentrations were determined in biotic and abiotic components of the pond ecosystem. A simple mathematical model was developed to predict the concentration of ^{95m}Tc in fish and snails. Results from this study indicated that uptake through the food chain was an important source of technetium to the higher trophic levels (i.e., fish). In the current study, an experimental pond was spiked with ^{95m}Tc in the pertechnetate form, and the concentrations of ^{95m}Tc were measured in the lower trophic levels. Emphasis was placed on measuring the concentration of ^{95m}Tc in zooplankton, benthic invertebrates, and the aquatic macrophyte *Elodea canadensis*. Fish were excluded from the pond to allow the development of a large zooplankton population. The concentration of ^{95m}Tc in water decreased from 0.75 Bq/mL 1 h after the pond was spiked, to 0.21 Bq/mL at 20 d. Throughout the experiment, at least 98% of the ^{95m}Tc in the water was in the dissolved fraction (0.4 μm). Zooplankton accumulated ^{95m}Tc rapidly, having concentration factors (Bq/g sample wet wt. divided by Bq/g water) ranging from 3 at 4 h to 36 at 20 d. Concentration factors ranged from 3 to 8 for benthic insects and from 1 to 62 for the aquatic macrophyte.

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INTRODUCTION

Currently the major sources of ^{99}Tc released to the environment in the United States are gaseous diffusion plants that enrich reprocessed nuclear fuel. At present these facilities are located on freshwater streams into which liquid effluents are released. Of the total amount of ^{99}Tc that escapes these facilities more than 90% may enter the liquid pathway [1]. Information on the behavior of technetium in aquatic ecosystems is limited; most data on the uptake of technetium by aquatic organisms are restricted to marine environments.

Most of the information available on technetium in freshwater ecosystems is based on data from an experimental pond study with ^{95m}Tc [2,3]. Concentration factors of 11, 75, and 121 were reported for carp (Cyprinus carpio), mosquitofish (Gambusia affinis), and snails (Helisoma sp.), respectively [2]. Recently, concentration factors were reported for other freshwater organisms based on laboratory studies with ^{95m}Tc. The freshwater mussel (Margaritifera margaritifera) had a whole-body concentration factor of 0.9; the newt (Taricha granulosa) had a whole body concentration factor of 11 [4]. In similar studies with the crayfish Pacifactacus lemusculus and the snail Juga silicula, whole-body concentration factors were 1.6 and 41, respectively [5].

In previous pond studies [2] periphyton and macrophytes accumulated technetium rapidly and contributed to the concentration of ^{95m}Tc in higher trophic levels via the food chain pathway. Concentrations in zooplankton and aquatic insects were not measured because these populations were kept at low densities by fish predation.

One objective of the current study was to determine the concentration of ^{95m}Tc in zooplankton, aquatic insects, and other lower-trophic-level components of an experimental pond. These data were incorporated with data from previous experimental pond studies to obtain a better understanding of the bioaccumulation of technetium in freshwater organisms. A second objective was to estimate variability by comparing the bioaccumulation of technetium in freshwater organisms with that of marine organisms.

MATERIALS AND METHODS

An acute release of ^{95m}Tc in the form of a pertechnate was made to a small, experimental, freshwater pond. The experimental pond, described previously [3], was approximately 4 m on each side and had a maximum depth of approximately 1 m. The pond was lined with a plastic (hypolon) to prevent seepage. The ecosystem in the pond was established by covering the bottom with 15 cm of sediment taken from a nearby pond and then filling it with spring water. The system was then allowed to develop naturally for approximately one year before it was spiked with ^{95m}Tc. Fish were excluded from the pond to allow for the development of denser zooplankton populations. Only one species of macrophyte (Elodea candensis) developed in this pond, whereas at least three species developed in earlier experimental pond studies.

Sampling techniques for biotic and abiotic components of the pond were similar to those described previously [3]. Collections were made at one and four h after the release of ^{95m}Tc to the pond and at days 1, 2, 3, 6, 13, and 20. The following components of the pond were sampled: water, sediments, macrophytes, invertebrates, periphyton, algae, and zooplankton. Zooplankton was collected using a plankton net (mesh size 153 μm). Samples were separated according to size by passing the zooplankton through a series of sieves (size 500, 250, 150, and 45 μm). The dominant species in the different size groups were then identified. Samples were oven dried and weighed, and the activity of ^{95m}Tc was measured in a gamma spectrometer. Aquatic insects, both mature and immature, were collected using a long-handled net. The sampling for each collection period was limited to prevent undue disturbance of the pond; therefore, only two families of insects were present for each sampling period.

RESULTS

One hour after the release of ^{95m}Tc to the pond, the water contained 0.75 ± 0.10 Bq/mL. The concentration decreased approximately 20% to 0.58 ± 0.06 Bq/mL after four h and then gradually over the next 20 d to 0.21 ± 0.14 Bq/mL (Fig. 1). Less than 2% remained on the particulate matter when the water was filtered through a 0.4 μm Nuclepore filter; thus, most of the ^{95m}Tc remained dissolved in the water.

The mean (n = 4) concentrations of ^{95m}Tc in sediment core samples are also shown in Fig. 1. The sediment did not accumulate ^{95m}Tc rapidly but continued to accumulate it throughout the experiment, with the highest concentrations occurring on day 20. The percentage of organic matter in the sediment samples averaged 7.7% and ranged from 4.7 to 14.5%; however, in this short-term experiment, the concentration of ^{95m}Tc in the sediment was not directly related to the percentage of organic matter in the sample.

Zooplankton accumulated ^{95m}Tc rapidly. One hour after ^{95m}Tc was released to the pond, zooplankton contained 31 Bq/g dry weight. The concentration of ^{95m}Tc in zooplankton and the concentration factor based on dry weight (Bq/g zooplankton/Bq/g water) are given in Fig. 2. A dry weight to wet weight ratio of 1:12:5 was determined for the zooplankton in the experimental pond.

Diaptomus reighardi was the dominant zooplankton species in the 500- to 250- m size classes and made up approximately 95% of the biomass within these size classes. Ostracods (40%), *Diaptomus* (30%), and *Difflugia* (30%) were the dominant taxa in the 150- μm -size class. A considerable amount of variability, but no consistent relationship, was observed in the concentrations of ^{95m}Tc among the different size classes of zooplankton. Data on the different size classes were combined and the mean concentrations are shown in Fig. 2.

Periphyton was not sampled until day 3 of the experiment. At this time the periphyton scraped from the sides of the pond contained

326.9 ± 20.8 Bq/g dry weight. A dry weight to wet weight ratio of 1:13.8 was determined for the periphyton in the experimental pond. By day 13 the concentration of ^{95m}Tc in periphyton had decreased to 138.3 ± 80.3 Bq/g.

Aquatic insects, both mature and immature stages concentrated ^{95m}Tc by factors of 3 to 8. Five orders of insects were collected from the experimental pond: Coleoptera, Diptera, Hemiptera, Odonata, and Trichoptera. The concentrations of ^{95m}Tc were measured in nine families of insects belonging to these five orders (Fig. 3). Only adult backswimmers (Notonectidae) and dragonfly nymphs (Libellulidae) were collected consistently because of the restrictions on sampling to prevent undue disturbance of the pond system. The highest mean concentration of ^{95m}Tc, 4.7 ± 1.4 Bq/g wet wt., was observed in Libellulidae (dragonfly) nymphs at day 2. Concentrations of ^{95m}Tc for the other families of insects are also shown in Fig. 3. Most of these samples were composite samples made up of several insects. Although the number of insects was limited, concentrations in all the insects were within the range obtained for adult Notonectidae and the benthic, predaceous nymph belonging to the family Libellulidae. Concentrations in the nymph appeared to be slightly higher than those in the adult.

The snail (Helisoma sp.) and the larva (tadpole) of the bullfrog (Rana catesbeiana) represent organisms at the secondary trophic level in the experimental pond. The snails and larvae accumulated ^{95m}Tc gradually and reached their highest concentrations of 33.5 ± 22.5 and 18.8 ± 3.2 Bq/g, respectively at day 20.

DISCUSSION

In the experimental pond, organisms are exposed not only to the ^{95m}Tc in the water but also obtain ^{95m}Tc via their natural food chain pathway. As a result, concentration (bioaccumulation) factors obtained from pond studies would be more realistic than concentration factors obtained from direct uptake studies conducted in the laboratory.

Since the experiment was conducted for a period of only 20 d following an acute release of ^{95m}Tc to the pond, organisms that have a relatively long biological half-life for technetium would not be expected to reach equilibrium conditions. A continiuous decrease in the concentration of ^{95m}Tc in the water would also prevent equilibrium conditions from being attained; however, in this experiment, ^{95m}Tc in the form of a pertechnetate remained dissolved in the water and decreased by less than a factor of three over a 20-d period. Thus, near-equilibrium conditions should be attained for organisms such as phytoplankton, zooplankton, and algae, which have relatively short biological half-lives and short life spans.

In the present study, the concentration factor was determined by dividing the highest average concentration of ^{95m}Tc in the organisms in a particular sampling period by the time-averaged water concentration. These concentration factors are probably near- equilibrium concentration factors for most organisms in the pond. For example, in a previous study [2] an equilibrium concentration factor of 121 was calculated for the snail (Helisoma sp.). This number is in relatively good agreement with the concentration factor of 75 that was measured in the present experiment.

The initial decrease of ^{95m}Tc in pond water was accompanied by a concomitant increase in organisms composing the lower trophic levels. Zooplankton and periphyton accumulated ^{95m}Tc very rapidly. After 1 h of exposure zooplankton accumulated ^{95m}Tc by a factor of 3.4. Although the concentration of ^{95m}Tc in water gradually decreased for the next 20 d, the concentration in zooplankton continued to increase with the highest concentration factor, 36, occurring on day 20. Apparently the initial uptake of ^{95m}Tc by zooplankton was directly from water. but as the experiment proceeded, ^{95m}Tc was probably also taken up through the food chain.

The concentration factor for periphyton decreased with time, reflecting the decrease in water concentration. In an earlier study [3], periphyton taken from slides had accumulated ^{95m}Tc by a factor of three after 4 h of exposure. Thus, we concluded that zooplankton and periphyton accumulate technetium very rapidly directly from water and that technetium has a short residence time in periphyton.

Information on the early uptake of technetium by freshwater algae is unavailable, but in an earlier pond study [3] the green algae *Spirogyra* had a maximum concentration factor of 10 after 22 d of exposure. The initial uptake of ^{95m}Tc by *Spirogyra* would probably be similar to that by periphyton.

Accumulation of ^{95m}Tc by *Elodea canadensis* was similar to the results obtained previously [3]. For the first 4 h of exposure *Elodea* contained approximately the same concentration of ^{95m}Tc as did the water, but after 1 d *Elodea* had accumulated ^{95m}Tc by a factor of three. Accumulation of ^{95m}Tc continued throughout the experiment with the maximum concentration factor, 62, occurring on day 20. This concentration factor is near the upper end of the range of concentration ratios reported for terrestrial plant (1). While ^{95m}Tc was increasing in the sediments, *Elodea* was probably obtaining ^{95m}Tc from the sediment as well as from the water.

Organisms grazing on periphyton accumulated ^{95m}Tc for the duration of the experiment. The snail (*Helisoma* sp.) and the larva of the bullfrog (*Rana catesbeiana*) have similar uptake patterns (Fig. 4). The highest concentrations of ^{95m}Tc were observed in these organisms. After 20 d of exposure, snails and frog larvae had accumulated ^{95m}Tc by a factor of 75 and 42, respectively. The concentration in snails is relatively close to the equilibrium concentration factor calculated by Blaylock *et al*. [2]. It is apparent that the food chain of these organisms contributes significantly to their body burden of ^{95m}Tc.

In Table I a comparison of the concentration factors in marine and freshwater organisms was made. With the exception of two laboratory studies, data on freshwater organisms were from experimental pond studies. In most cases, the data for marine organisms were obtained from laboratory studies that excluded the process of food chain uptake. Brown algae and lobster have the highest concentration factors in the marine ecosystem, their values exceeding any of the concentrations found in freshwater organisms. In the freshwater system, snails and macrophytes have the highest concentration factors. Mosquitofish have a much higher concentration factor than any of the other freshwater fish or any marine fish. In comparison, the carp has a concentration factor similar to that of marine fish. However, despite differences between the concentration factors in freshwater and marine organisms, there is good agreement in the values obtained for similar groups of organisms.

CONCLUSIONS

Technetium in the form of a pertechnetate released into an experimental freshwater pond had a very low affinity for particulate matter and sediments. Most of the technetium remained dissolved in the water column. Technetium was accumulated rapidly from the water by periphyton, zooplankton, and algae. Other organisms, such as snails and mosquitofish, accumulated technetium slowly but attained some of the highest whole-body concentrations. Aquatic insects concentrated technetium by factors ranging from 3 to 8 times greater than the concentration in water. Macrophytes very rapidly reached tissue concentrations of technetium equivalent to the water concentrations and continued to accumulate technetium until the concentration factor was equal to the upper range of those reported for terrestrial plants.

A comparison of concentration factors for marine and freshwater organisms shows that, with the exception of brown algae and lobsters, the bioaccumulation of technetium by similar organisms is in relatively good agreement despite marked differences in environmental and experimental conditions.

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TABLE I COMPARISON OF TECHNETIUM CONCENTRATION FACTORS (CF) FOR FRESHWATER AND MARINE ORGANISMS

Freshwater	CF*	Marine	CF
Periphyton		Phytoplankton	
Slide samples	40	Heterocapsa pygmaca [6]	17
		mixed flagellates [7]	1
		phytoflagellates [8]	1
Algae		Algae	
green algae	1-10	brown algae	250-2500
		red and green algae	1-20
		[9,10,11,12,13,14]	
Macrophytes			
Elodea canadensis	61		
Zooplankton		Zooplankton	
Diaptomus reighardi	36	Artemia salina [15]	3
Difflugia		euphausiid [16]	1
Ostracods			
Other Crustaceans		Other Crustaceans	
		shrimp crab, isopod	2-12
Crayfish		amphipods [7,8]	
Pacifastacus lemusculus [5]	1.6	Lobster [8,10,17]	1000-1400
Cambarus	30	Hormarus gammarus	
Molluscs		Molluscs	
Margaritifera		Mytilus edulus	2
margaritifera [4]	0.9	Mytilus galloprovinviallis	
Corbicula manilensis	1	Mytilus californianus	
		Crassostrea gigas [8,10,18]	
Snails		Red abalone	
Helisoma sp.	121	Haliotis rufescens	
		[14,19]	100-200
Juga silicula [5]	41	Gastropod	
		Aporrhais pespelicana [20]	3
Aquatic insects	3-8		
Notonectidae			
Belostomatidae			
Chironmidae			
Dytisidae			
Tricoptera			
Odonata			
Amphibian			
Taricha granulosa [4]	11		
Fishes		Fishes	
Cyprinus carpio	11	Pleuronectes platessa [11]	9
Gambusia affinis	75	Raja clavata [17]	8
		Blennius pholis [10]	2

*Freshwater data except those that have specific references were obtained from experimental ponds.

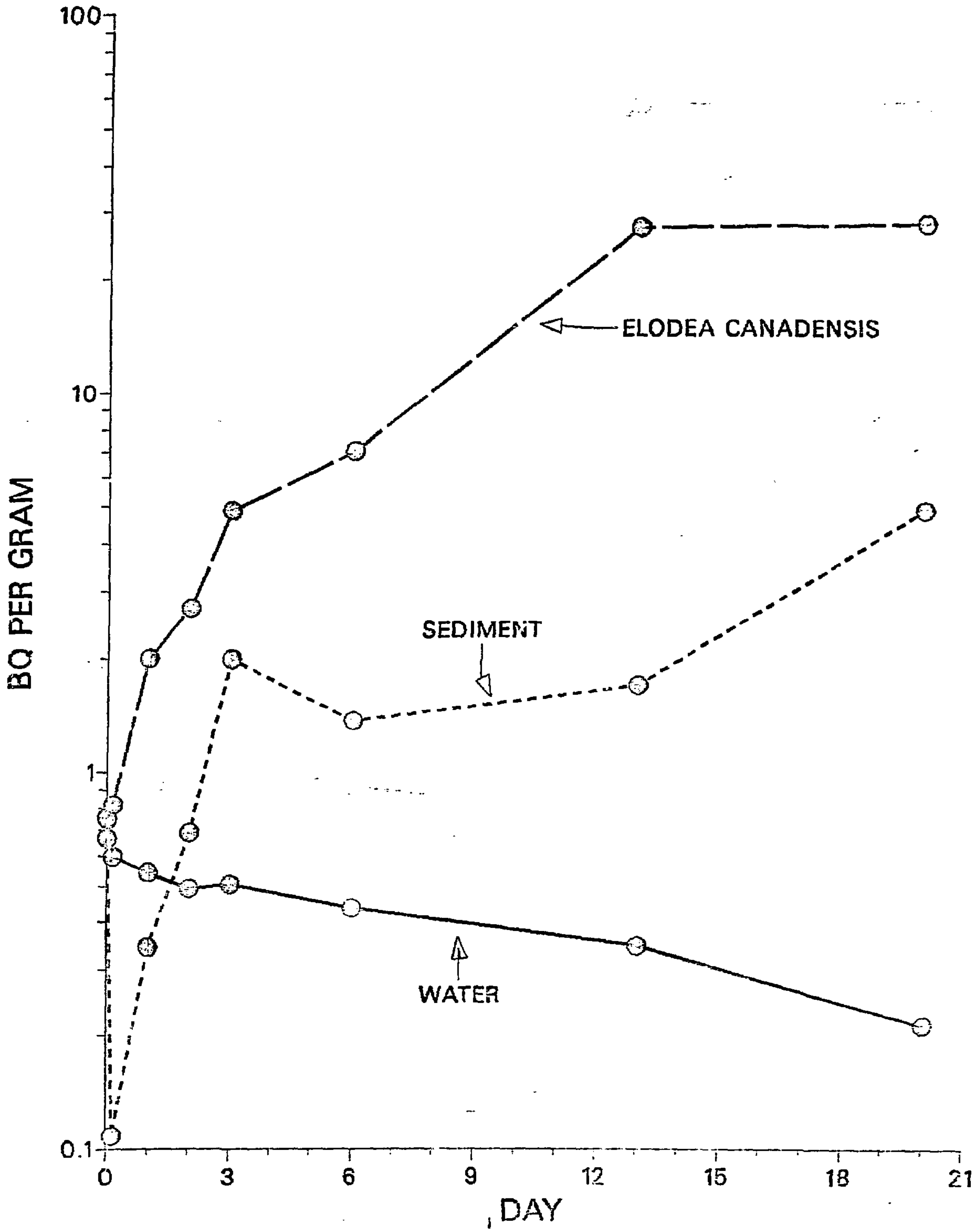
FIGURE LEGENDS

Fig. 1. The concentration of ^{95m}Tc in water (Bq/mL), sediments (Bq/g dry wt.) and Elodea canadensis (Bq/g wet wt.) are shown over time after an acute release of $^{95m}TcO_4^-$ to an experimental freshwater pond.

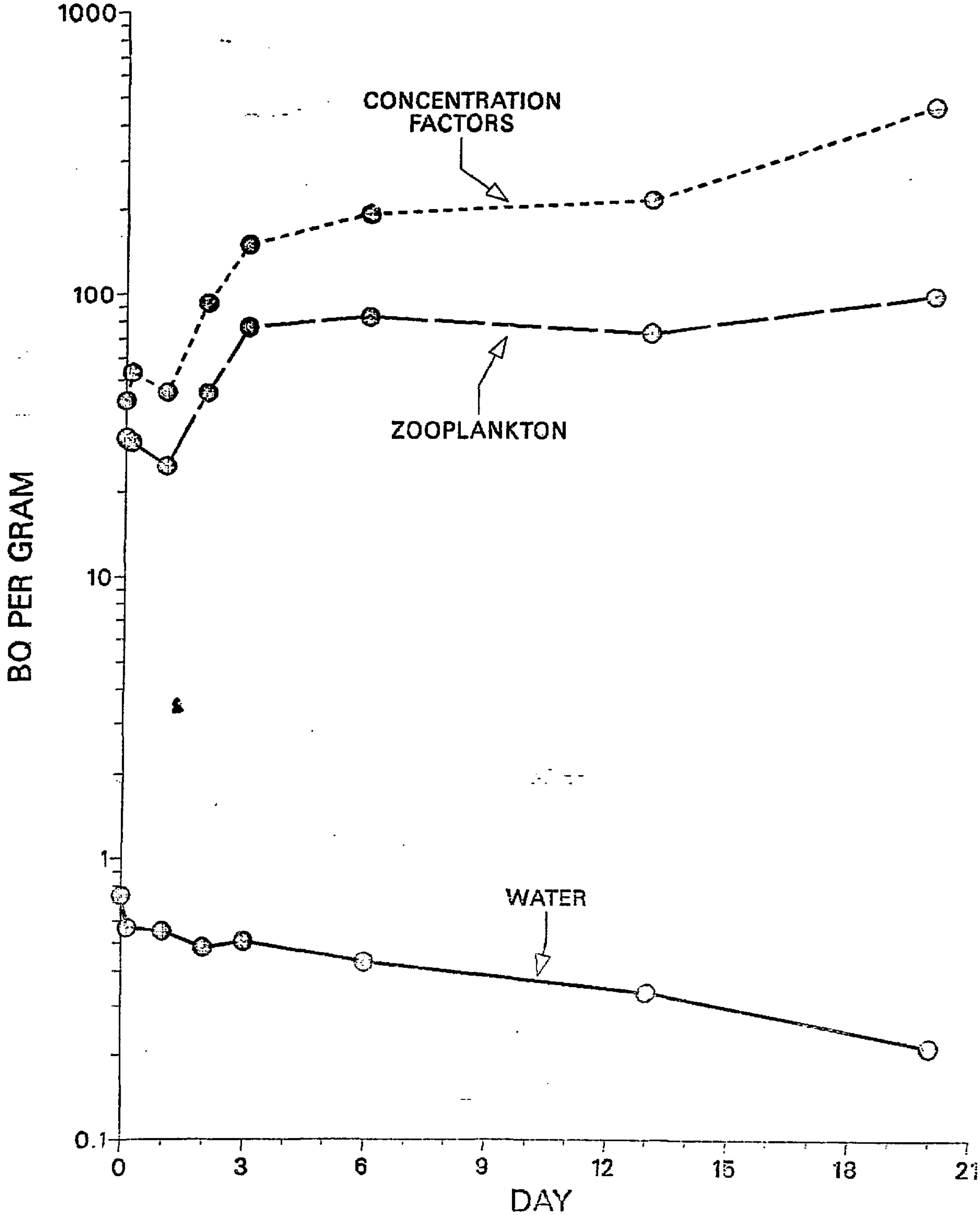
Fig. 2. The concentration of ^{95m}Tc in zooplankton (Bq/g dry wt.), water (Bq/mL), and concentration factors (Bq/g dry wt. zooplankton/Bq/mL water) are shown for different sampling periods after an acute release of $^{95m}TcO_4^-$ to an experimental freshwater pond. The wet weight to dry weight ratio for zooplankton in the experimental pond was 1:12.5.

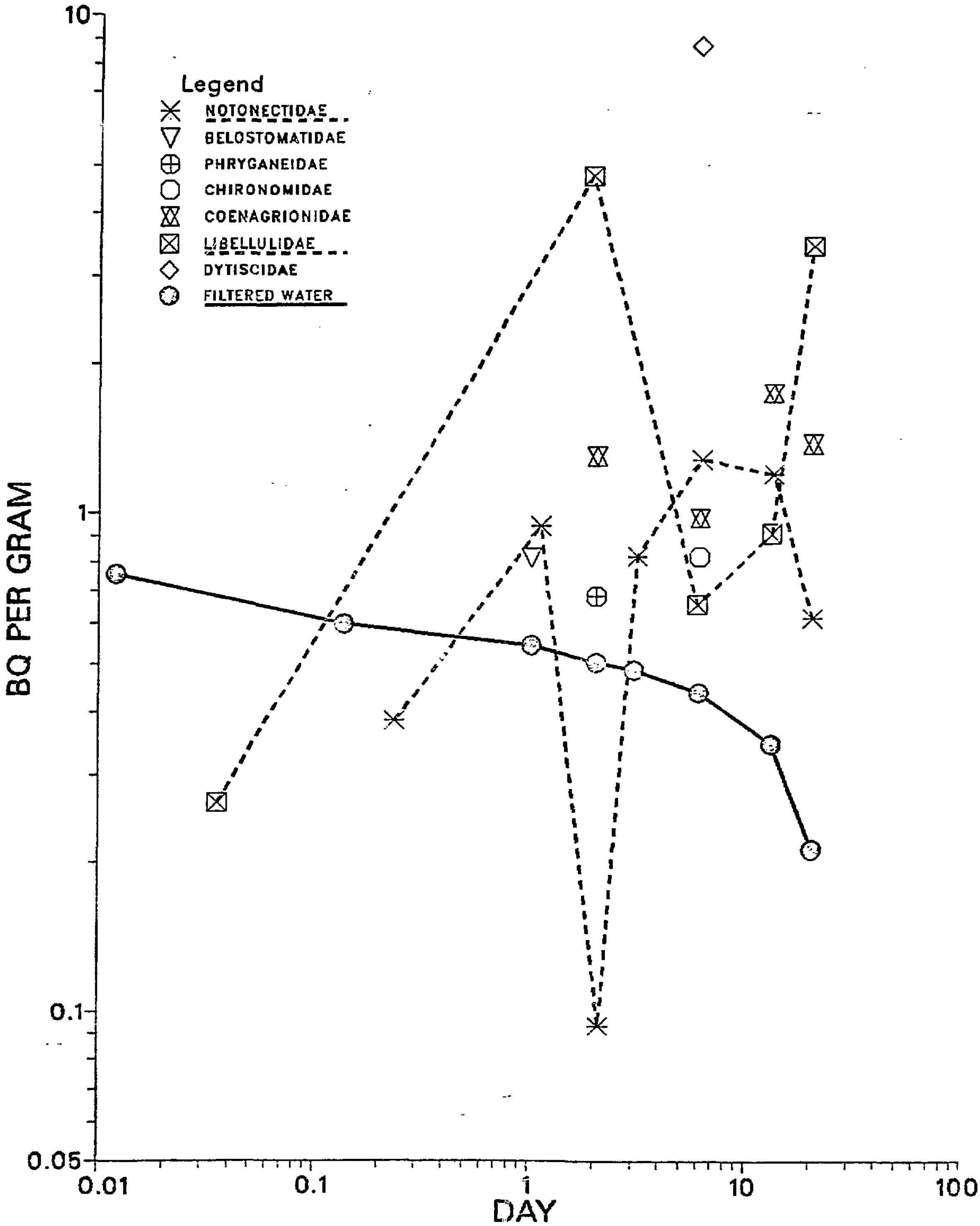
Fig. 3. Concentrations of ^{95m}Tc are shown in seven families of mature and immature insects after an acute release of $^{95m}TcO_4^-$ to an experimental freshwater pond. The adult backswimmer (Notonectidae) and the predaceous dragonfly nymph (Libellulidae) were collected the most consistently. The range of concentrations of ^{95m}Tc observed in these two families of insects included most of the concentrations measured for all the other families. In most cases, the samples were composite samples of insects.

Fig. 4. The concentrations of ^{95m}Tc in the snail (Helisoma sp.), the larva of the frog (Rana catesbeiana) in Bq/g wet wt., and water (Bq/g) are shown after an acute release of $^{95m}TcO_4^-$ to an experimental freshwater pond.



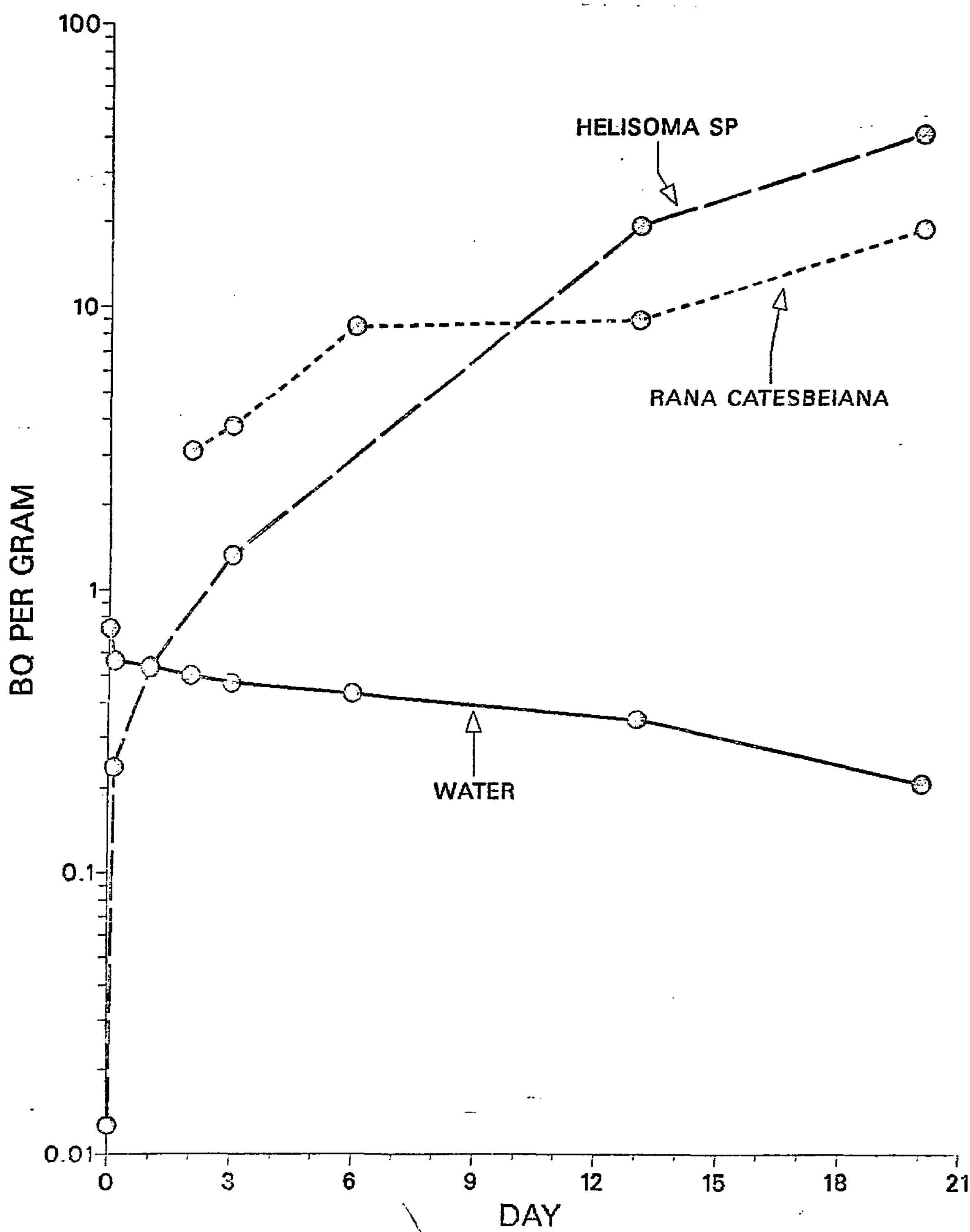
100
10
1
0.1
BQ PER GRAM
ELODEA CANADENSIS
SEDIMENT
WATER
0
3
6
9
12
15
18
21
DAY


CONCENTRATION FACTORS
ZOOPLANKTON
WATER
BQ PER GRAM
DAY
1000
100
10
1
0.1
0
3
6
9
12
15
18
21



Legend
NOTONECTIDAE
BELOSTOMATIDAE
PHRYGANEIDAE
CHIRONOMIDAE
COENAGRIONIDAE
LIBELLULIDAE
DYTISCIDAE
FILTERED WATER
BQ PER GRAM
10
1
0.1
0.05
0.01
0.1
1
10
100
DAY

Fig. 3


HELISOMA SP
RANA CATESBEIANA
WATER
BQ PER GRAM
100
10
1
0.1
0.01
0
3
6
9
12
15
18
21
DAY