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Radionuclide Accumulation by Aquatic Biota Exposed to Contaminated Water in Artificial Ecosystems Before and After Its Passage Through the Ground

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Prepared for U.S. Nuclear Regulatory Commission

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ABSTRACT

Aquatic biota (fish, clams, algae) and an aquatic emergent vascular plant were experimentally exposed to mixtures of radionuclides in three artificial streams for time periods extending up to 86 days. The experimental streams consisted of industrial water discharged directly into a leaching trench, and the same water after it had migrated through the ground for a distance of 260 meters. The third stream was river water.

After migrating through the ground, the leach water still contained measurable amounts of 60 Co and 90 Sr. These were accumulated by all classes of aquatic biota and by tomato plants grown in a quasi-hydroponic system. This experiment indicated that 50 Co and 90 Sr can be transported along with ground water flow and are available for biological uptake by aquatic biota.

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EXECUTIVE SUMMARY

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This study was designed to investigate the comparative accumulation of radionuclides from contaminated water in artificial ecosystems before and after the water's passage through the ground.

Plants and animals typically associated with small, flowing freshwater streams were experimentally exposed to flowing water from three different sources. Fish, clams, algae, and an emergent vascular plant were experimentally exposed to mixtures of radionuclides in three aqueous streams. Two streams consisted of industrial water discharged directly into a leaching trench, and the same water after it had migrated through the ground for a distance of 260 meters. The third stream was river water, which served as a background or control.

Biota exposed to river water in the control stream had very low concentrations of ⁶⁰Co, less than 3 pCi per gram dry weight (pCi/g DW). Other radionuclides were essentially unmeasurable.

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The biota exposed to trench water accumulated very high relative concentrations of ⁶⁰Co. Aquatic biota exposed to trench water also had measurable concentrations of ¹⁵⁵Eu, ¹⁴⁴Ce, ¹⁴¹Ce, ¹²⁵Sb, ¹²⁴Sb, ¹⁰³Ru, ¹⁰⁶Ru, ¹³⁷Cs, ⁹⁵Zr, ⁹⁵Nb, ⁵⁸Co, ⁵⁴Mn, ⁵⁹Fe, ⁶⁵Zn, ⁹⁰Sr, ²³⁹ ²⁴⁰Pu, and ²³⁸Pu.

Aquatic biota exposed to ground water had concentrations of ⁶⁰Co that ranged between 50 and 1200 pCi/g DW. Fish flesh had the lowest concentration of ⁶⁰Co and algae the highest. Strontium-90 was measured in the tissues of aquatic biota at concentrations ranging between 360 pCi/g DW in clam flesh to 3400 pCi/g DW in leaves and stems of Veronica. In some cases, ⁹⁰Sr was two times greater in concentration than it was in the biota exposed to trench water. The leaves and fruits of tomato plants rooted in the ground water accumulated ⁹⁰Sr at concentrations of 160 pCi in fruits and 4200 pCi in leaves.

The data indicate that ⁶⁰Co and ⁹⁰Sr migrated through the ground along with ground-water flow and were available to all classes of aquatic biota and tomato plants rooted in the water via orrt uptake, sorption, and food chain transfers.

INTRODUCTION

One of the most important missing pieces of information concerning the environmental behavior of radionuclides associated with shallow land disposal of low-level radioactive wastes is how well they are transported by ground water. Radicnuclides can appear in ground water if they are discharged into leaching trenches with sufficient water to carry them into the saturated zone.

Radionuclides moving through the ground can change valence, which in turn affects their mobility. The valence state and mobility of radionuclides in ground water has been studied by Fruchter et al. (1984). A logical extension of these hydrogeochemical studies is to determine whether radionuclides moving in ground water can be assimilated by aquatic plants and animals. Plants rooted in water can obtain radionuclides through their root systems and transport them to leaves and fruits. Aquatic animals obtain radionuclides by eating plant materials or other kinds of animals that have accumulated radionuclides.

The purpose of this investigation was to compare radionuclide uptake by similar sets of aquatic biota after exposure to artificial experimental streams with different kinds and concentrations of radionuclides originating from industrial processes and discharged to a leaching trench. One set of biota was exposed to the contaminated water prior to its entry into the leaching trench, and a second set was exposed to the same water after it had passed through about 200 m of soil and emerged as surface springs. A third set was exposed to noncontaminated water.

This report presents the results of this study.

EXPERIMENTAL EXPOSURE SYSTEMS

The experimental protocol consisted of establishing three pairs of artificial streams, each pair receiving a different source of water, i.e., river, trench, and ground water. The artificial stream system consisted of (1) a 2000-L head tank, for mixing and providing a constant head for the regulated flow of test water, and (2) a pair of troughs, 3 m long x 30 cm wide x 20 cm deep, which channelled the test water and served as flowing water habitat for biota introduced into the troughs (Figure 1). The control troughs received river water pumped directly into a head tank. Outflow from the head tank was adjusted to provide a flow of about 12 L/min through the experimental troughs. The troughs with ground water were provided with a thermostatically regulated refrigeration system so that the warmer waters could be adjusted to approximate the ambient river water temperatures. Temperature control was provided to keep growth conditions as comparable as possible among the three different systems. A graphic representation of water temperature regimes is shown in Figure 2. Trench water was diluted with 10 parts river water and mixed before it entered the

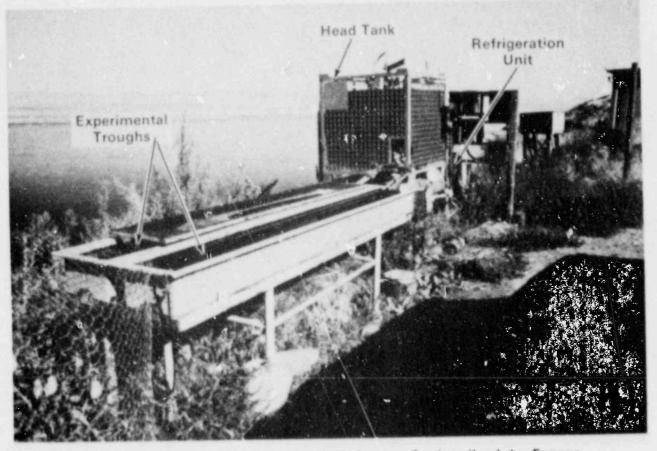
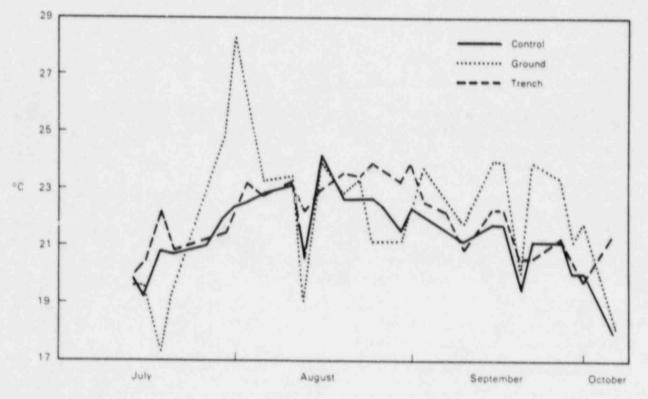


FIGURE 1. Photograph of an Experimental Stream System Used to Expose Aquatic Biota to Water Flows with Different Radionuclide Concentrations and Charge Forms experimental troughs. The dilution diminished the trough water temperature to near that of river temperature, and it reduced the concentration of radionuclides in the trough water by a factor of ten. All water sources were filtered through a $25-\mu$ filter to remove waterborne detritus particles before they entered the head tank.

Test organisms included (1) filamentous green algae (Spirogyra sp.), which naturally colonized the stones placed in the troughs as attachment substrates; (2) clams (Corbicula sp.) collected from a nearby stream and acclimated prior to their introduction into the troughs; (3) two species of juvenile fish (carp, Cyprinus carpio, and goldfish, Carassius auratus) obtained commercially and acclimated similarly as the clams; (4) an emergent aquatic plant (Veronica anagallis-aquatica), which was placed near the outlet end of each trough; and (5) hydroponically grown tomato plants (Lycocopersicon sp.), rooted in a 95-L barrel that received test water via a siphon from each trough at the control and the ground water troughs. Tomatoes were not grown in the trench water.





E 2. Water Temperature Fluctuations in Control, Ground, and Trench Water Experimental Stream Systems, July to October 1983

PROCEDURES

The test organisms were introduced to the artificial experimental streams on July 13, 1983. Two to three times per week temperatures and flow rates were adjusted, and filters replaced.

Organisms were collected from each pair of streams at the termination of the study in late September and early October. Veronica and tomatoes were collected on September 28, 1983, before the onset of a plant-killing frost; and algae, clams, and fish were collected on October 5. This procedure resulted in a test exposure of 78 and 86 days, respectively, and allowed sufficient time for the organisms to reach uptake equilibrium (Merlini and Bittel 1970; Reed 1971). Tomato roots, foliage, and fruit were collected separately for radioanalysis, as were the roots and foliage of Veronica. Clam flesh was separated from the shell.

Samples were dried to constant weight (dry weight, DW) at 50°C, homogenized by grinding, and placed for radioanalysis into either 0.1-dram polyethylene vials, 2-dram polyethylene vials, or plastic containers of 1.3 cm thickness and 5.1 cm diameter, depending on the quantity and activity levels of the samples. The most radioactive samples, as determined by screening with a Geiger-Mueller survey meter, were placed in the 0.1-dram vials, and the moderate and least radioactive samples were contained in the 2-dram and 1.3 x 5.1-cm containers, respectively. Radiochemical analyses were performed at the Pacific Northwest Laboratory.

The samples were sealed in plastic bags before the counting process. Gamma-emitting radionuclides were identified and counted using two Ge(Li) gamma-ray spectrometers having efficiencies of 16% and 19%. The counting interval varied from 10 to 30 minutes for the samples with highest activity to 100 to 1000 minutes for the least radioactive samples. The 0.1-dram or 2-dram vials containing the highest concentrations of radionuclides were counted on shelves positioned at standard distances from the Ge(Li) detectors, in order to achieve analyzer dead times of less than 5% to 10%. All of the geometries and shelf positions were calibrated using sources calibrated by the National Bureau of Standards (a ¹²⁵Sb/¹⁵²Eu mixture) prepared in each of the three sample containers.

The gamma-ray spectra were deconvoluted using a computer program that integrated the peak areas of the radionuclides of interest; performed back-ground, Compton, and decay corrections; and computed the counting error.

Plutonium-238 and -239, 240 were determined on ashed samples after the ash was dissolved with acid. The plutonium was separated from interfering radionuclides by ion exchange chromotography and then deposited on stainless steel discs. Isotopes of plutonium were measured by alpha energy analysis using ²³⁶Pu as a procedural tracer.

Strontium-90 was determined on beta counters after a lengthy series of chemical dissolutions, precipitation, filtration, and centrifugation.

Resulting radionuclide measurements were assumed valid if the counting statistical error was less than 50%.

RESULTS AND DISCUSSION

At least 18 radionuclides were measured in trench water (Fruchter et al. 1984; however, see Jenne et al. 1984). The most abundant were ^{5*}Mn, ⁹⁵Nb, ⁶⁰Co, ¹³⁷Cs, ⁵⁹Fe and ⁹⁵Zr. These same radionuclides were also measured in ground water but at greatly reduced concentrations.

The radionuclides in the trench water were mostly in particulate form (Table 1). Notable exceptions were ^{54}Mn , ^{125}Sb , and ^{137}Cs . Cationic

TABLE 1.	Percentages of Particulate and Charge Forms of Radionuclides in	1
	Trench and Ground Water, June 1983 ^(a)	

Radionuclide	% Particulate	* Cationic	& Anionic
	••••••	Trench Water	
Mn Fe	5.8 93.9	94.1 3.2	1 2,8
Co	99.1 41	8 58.5	1
Zn	56.2	43.7	1
Sr	5.8	89.1	1
Zr Nb	77.3 51.2	6.4 35.5	16.1 13.1
Ru Ru	65.1 59.9	13.4 18.5	20 20
Sb Sb	82.4 12.1	6 33	17.6 87.9
Cs	10.2	89.8	1
Ce Ce	78 77	13.7 14.6	7.8 7.9
	************************	Ground Water	
Co	1	49.1 45.7	43.3 48.3
Sr	· · · · · · · · · · · · · · · · · · ·	99.9	1
Ru Ru	1	10.4 12.6	86 82
Sb Sb	;	1	19.8 21.5

(a) Data from Fruchter et al. (1984).

charge forms usually dominated the radionuclides; a notable exception was ¹²⁵Sb. However, anionic forms tended to dominate in the ground water, with nearly equal proportions of ⁵⁸Co and ⁶⁰Co in both anionic and the cationic charge forms. Strontium-90 was mostly present in cationic form (Table 1). Radionuclides were in extremely low concentrations in river water; ⁶⁰Co at 0.0033 pCi/L, ⁹⁰Sr at 0.14 pCi/L, and ¹³⁷Cs at 0.029 pCi/L. Global fallout is the likely source of these radionuclides.

RADIONUCLIDE CONCENTRATIONS IN AQUATIC BIOTA EXPOSED TO TRENCH WATER

As expected, the bicta exposed to a continuous flow of diluted trench water accumulated a mixture of radionuclides in amounts that reflected their concentrations in the trench water (Figures 3 and 4, 1.51e 2). Differences in trophic level were not always consistent for the various radionuclides, but the most common pattern was Veronica roots > algae > Veronica foliage > goldfish > clam flesh > carp. Because carp and goldfish have similar feeding habits, it was somewhat of a surprise to find a marked difference in their accumulation of several radionuclides, including 144Ce and 125Sb. Veronica roots had the greatest concentrations of radionuclides. The levels of radionuclides accumulated by various organisms are described as follows:

Cerium-144 and -141

Cerium-144 was accumulated by all organisms. Levels of accumulation were roughly comparable for all organisms except Veronica roots, which had the greatest accumulation. The levels of accumulated 144Ce were higher than those of 141Ce.

Antimony-124 and -125

Algae and Veronica roots and foliage accumulated ¹²⁵Sb and ¹²⁴Sb, and goldfish accumulated low levels of ¹²⁴Sb. No other organism accumulated measurable amounts of either of these radionuclides.

Ruthenium-103 and -106

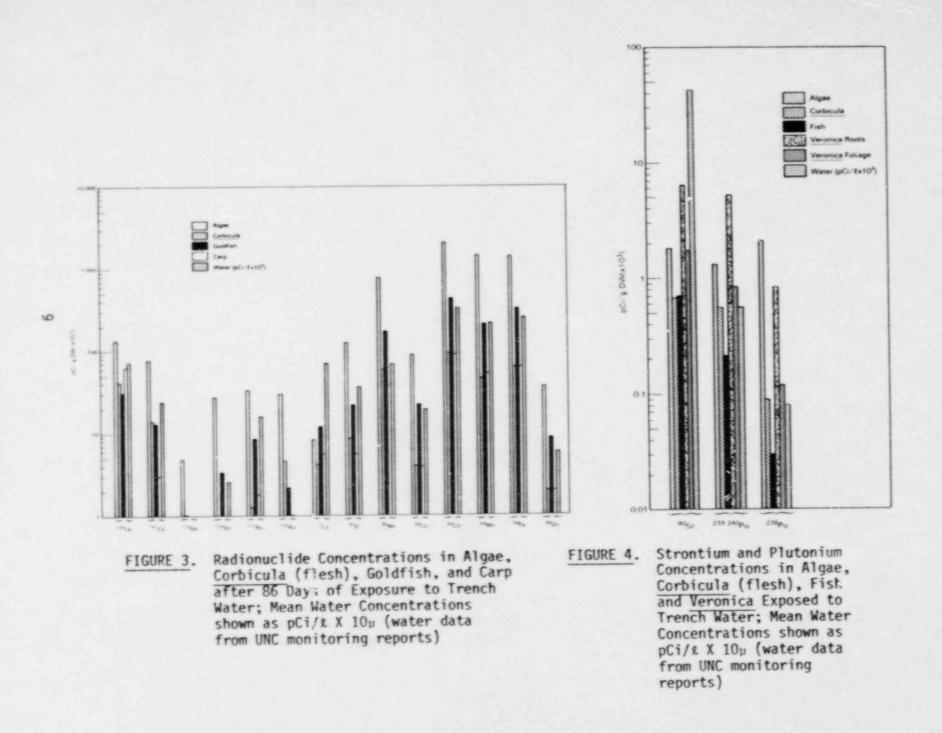
Algae and Veronica foliage accumulated comparable levels of both 103Ru and 106Ru. Clams accumulated more 106Ru than 103Ru, but the converse was observed for fish.

Cesium-137

Comparable levels of ¹³⁷Cs were accumulated for all biota except Veronica roots, which had the greatest concentration. This was the only radionuclide in which bioaccumulation by an organism of higher trophic level (goldfish) exceeded algal accumulation.

Zirconium-95 and Niobium-95

All organisms accumulated 95Zr and 95Nb. Accumulation of 95Nb by all organisms exceeded that of 95Zr by an order of magnitude except for



Radio-		Algae (a)	C	lam Fle	sh		Goldfi	sh		Carp			Veronica	Roots	Ver	onica F	oliage
nuclide	С	CW	T	C	CW	T	с	CW	т	с	GW	т	с	CW	т	с	CW	т
54 Mn		7.0	1400			45			210		0.12	51			6900	0.3	1.7	820
58 _{Co}			88			4			22			3.9			550			65
59 Fe		49	1400	20	20	61			310			61		85	7600	2		830
60 Co	2.0	1200	2000	2.5	160	91	2.0	130	440	1.6	50	87	1.6	940	11000	1.2	87	1320
65 Zn			36			2.1			9.6	2		2		14	180		0.3	22
90 95 2r		1300	1.8		360	0.66		620 ^(c)	0.69 ^(c)					2000	6.2		3400	1.7
95 Zr			130			8.5			22			5.5			770			84
95 _{Nb}			770			58			170	7.5		38		45	5100			520
103 Ru			34			2.9			8.6			1.8		50	287		1.1	12
106 Ru	3.5		30	-		4.7		8.0	2.2		·	0.95		63	90		2.7	13
124 Sb			28			1	-		3.3			1.1			140			12
125 Sh		24	4.8		1.6					1.3	0.6	0.13		46	22		4.3	
137 Cs			8.4	-		4.5			12		0.35	5.5	3.9	3	37	0.4		8.8
141 Ce			77			14			13			3			540			48
144 Ce			135			42			31	***	0.34				1140			95
155 Eu	0.6	(b)				0.41			80								1	
238 Pu			2.1		23	0.09			0.03 ^(c)					0.01	0.83			1.3
240 Pu		0.02	1.3		0.24				0.21 ^(c)					0.05	5.1			0.12

TABLE 2. Radionuclide Concentrations (pCi/g DW) in Aquatic Biota After Exposure to Control, Ground, and Trench Water (multiply all trench values by 10³)

(a) C = control, GW = ground water, T = trench.

(b) -- = unmeasurable.

0

(c) Analysis of combined goldfish and carp samples.

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0

algae; algal uptake was about five times higher. Both 95Zr and 95Nb followed the accumulation pattern of Veronica roots > algae > Veronica foliage > goldfish > clams > carp.

Cobalt-60 and -58

The biota accumulated ⁶⁰Co to the highest level of any gamma-emitting radionuclide present in the trench water. Patterns of accumulation of radioactive cobalt were similar, although ⁶⁰Co uptake exceeded ⁵⁸Co uptake by an order of magnitude.

Manganese-54, Iron-59, and Zinc-65

Levels of accumulated ⁵⁴Mn and ⁵⁹Fe were comparable; trophic-level patterns were also similar. Trophic-level accumulation of ⁶⁵Zn was similar to ⁶⁰Co, ⁵⁴Mn, and ⁵⁹Fe, but uptake was two orders of magnitude lower for algae and goldfish and one order of magnitude lower for clams, carp, and Veronica roots and foliage.

Strontium-90

Veronica roots showed the highest concentrations of 90Sr at 6200 pCi/g DW. Veronica foliage and algae accumulated similar but lower levels, as did clams and fish.

Plutonium-239 and -240

All biota accumulated 239, 240 Pu, with Veronica roots taking up the highest level, 5100 pCi/g DW, followed by algae with 1300 pCi/g DW.

Plutonium-238

Accumulation patterns for ²³⁸Pu were similar to those of ²³⁹ ²⁴⁰Pu, viz. Veronica root > algae > Veronica foliage > clams > fish.

The accumulation of specific radionuclides by aquatic biota exposed to diluted trench water was directly related to the concentration of the radionuclides in the water. It is important to recognize that the radionuclide concentrations reported here are experimentally obtained values and are unlikely to occur during waste disposal practices.

RADIONUCLIDE CONCENTRATIONS IN AQUATIC BIOTA EXPOSED TO GROUND WATER

The experimental troughs represent a habitat that is capable of supporting at least some aquatic biota. Therefore, the radionuclide concentrations obtained in our experimental streams might be attained in a real-world situation in which contaminated ground water appears at the ground surface at some distance from a disposal site and flows across the ground surface. The various organisms introduced to the experimental troughs are listed below along with their radionuclide concentrations. Algae - Algae exposed to ground water accumulated ¹²⁵Sb, ⁶⁰Co, ⁵⁴Mn, and ⁵⁹Fe (Table 2). Cobalt-60 concentrations were 3 orders of magnitude lower in the ground water compared to the trench water. Both of these values greatly exceeded the 2.3 pCi/g DW accumulated in the river water. Algae accumulated more ⁹⁰Sr than ⁶⁰Co, but very low levels of ²³⁹ ²⁴⁰Pu (Table 2). These data indicate that ⁶⁰Co and ⁹⁰Sr are available for uptake by algae after migrating through the ground from the disposal trench.

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- Clams The concentrations of ⁶⁰Co in clams exposed to ground water was 160 pCi/g DW, 600X less than in the trench water (Table 2). The only other gamma-emitting radionuclides measured in clam flesh were ⁵⁹Fe and ¹²⁵Sb. Levels of ⁹⁰Sr measured in clam flesh were the highest of any radionuclide, 360 pCi/g DW. Concentrations of ²³⁹ ²⁴⁰Pu were measurable, but in extremely low concentrations (Table 2).
- Goldfish Goldfish exposed to ground water accumulated 130 pCi
 ⁶⁰Co/g DW, 3 orders of magnitude less than for goldfish exposed to trench water (Table 2). The only other radionuclide measured in goldfish was ¹⁰⁶Ru at 8 pCi/g DW.
- Carp Carp exposed to ground water had concentrations of 50 pCi ⁵⁰Co/g DW (Table 2), again, 3 orders of magnitude less than for carp exposed to trench water. Carp also accumulated low concentrations of ¹⁴Ce, ¹²⁵Sb, ¹³⁷Cs, and ⁵⁴Mn (Table 3).
- Fish Concentrations of ⁹⁰Sr in combined carp and goldfish exposed to ground water were nearly identical to those exposed to trench, 620 and 690 pCi/g DW, respectively, (Table 2).
- Veronica Veronica roots exposed to ground water had 940 pCi ⁶⁰Co/g DW. This was about 10,000 times less than for Veronica roots exposed to trench water. Veronica roots also had measurable concentrations of ¹²⁵Sb, ¹⁰³Ru, ¹⁰⁶Ru, ⁵⁴Mn, and ⁶⁵Zn (Table 2). Strontium-90 concentrations in Veronica roots and foliage exceeded those in all other aquatic organisms (Table 2). However, plutonium concentrations were nil.

These data indicate that ⁶⁰Co and ⁹⁰Sr were available for accumulation by the different test organisms and were present in biota in concentrations greatly exceeding those of other radionuclides.

The uptake of ⁶⁰Co by the different kinds of aquatic organisms was directly related to the differences in concentration of ⁶⁰Co in the water to which they were exposed. The range of concentrations was from roughly two orders of magnitude greater for the biota exposed to spring water (as compared to river water) to five or six orders of magnitude greater for biota exposed to trench water. In terms of trophic-level comparisons, the plants--at least those portions directly exposed to the water (algae and Veronica roots)--accumulated roughly an order of magnitude more ⁶⁰Co than did the animals of higher trophic levels. The aerial stems and leaves of Veronica accumulated ⁶⁰Co at about the same concentrations as did the fish and clams.

RADIONUCLIDE CONCENTRATIONS IN TOMATO PLANTS EXPOSED TO GROUND AND RIVER

Tomato seedlings (Lycopersicon sp.) were grown in soil in screen bottomed garden plots for 2 weeks to allow the plants to establish root systems. The pots were then suspended one inch above the water level in a 95-L steel barrel. The tomato roots grew downward into the test water, thus supplying the plants with the water needed to sustain growth and source of radionuclides. A siphon was used to maintain a steady flow of stream water in each barrel for 78 days. By the end of summer, the plants had flowered and produced fruits.

The data indicate that ⁹⁰Sr and ⁶⁰Co were the radionuclides in the ground water most actively accumulated by tomato plants. Roots generally had greater concentrations of radionuclides than stems, leaves, and fruits (Table 3). The stems and leaves had greater concentrations of ⁹⁰Sr than

TABLE 3. Radionuclide Concentrations (pCi/g DW) in Tissues of Tomato Plants Rooted in Ground and River Water for 78 Days

		Ground Water	
	Tomato	Tomato	Tomato
Radionuclide	Roots	Stems and Leaves	Fruits
	(a)		
Eu		··· (b)	
	4.0	(7.5) ^(b)	**
Ce		**	
Sh	250		0.02
55	6.0		
Ce Ce Sb Sb Ru	130		0.11
Ru	440	0.6	
RU	440	0.0	0.01
55		0.2 (1.6)	3.3
Cn	3.5	0.2 (1.6)	
Zr			
Nb	**		
Co	0.8	0.4	
Co	870	0.4 (1.7)	
Mo	6.3 6.0 3.3	0.25	1.0
Fe	6.0		
7.0	3 3		0.06
Ln	1700	4200	160
Dr.	0.07	4200	
RU Cs Cr D Co M F c r U S PU	0.07		
Pu	0.01		

(a) -- = unmeasurable.

(b) Values in parentheses are concentrations measured in tomato plants exposed to river water. The three values for stems and leaves represent the only radio culides that were detected in the tomato plants grown in river water.

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did roots, 4200 pCi/g DW as compared to 1700. Fruits had 160 pCi/g DW. However, roots had greater concentrations of ⁶⁰Co than the stems and leaves, 870 and 22 pCi/g DW, respectively. Cobalt-60 was not detected in tomato fruit. Cesium-144, ¹³⁷Cs, and ⁶⁰Co were the only radionuclides detected in the stems and leaves of tomato plants grown in river water (Table 3). The radionuclide concentrations measured in tomato roots does not necessarily mean that the radionuclides were incorporated into the plant tissue. Some of the radioactivity may represent external entrapment. However, the radionuclide concentrations measured in shoots and fruit represents root uptake with transport through stems to leaves and fruit.

CONCENTRATION RATIOS

At the trench cite, the highest concentration ratios were calculated for Veronica roots, followed, generally, by algae and Veronica foliage. Lowest values were usually in carp and clams. Niobium-95 had the highest concentration ratio of all radionuclides with a value of 76000 (Table 4).

At the spring seep site concentration ratios fluctuated widely among different organisms and with different radionuclides. Strontium-90 was the radionuclide concentrated to the highest level in <u>Veronica</u> foliage (Table 4).

IMPLICATIONS FOR ENVIRONMENTAL MONITORING

Environmental monicoring programs are routinely conducted to evaluate the effectiveness of engineered control systems in keeping environmental releases of radionuclides within guidelines. Air, surface water, ground water, soil, sediments, and biota, especially those plants and animals likely to be eaten as human foods, are systematically collected and analyzed for changing concentrations of radionuclides (Till and Meyer 1983). The data of this investigation show that the migration of water through the ground filters out many kinds of radionuclides. However, some radionuclides can move and these are available for accumulation by aquatic biota. A measurement of the radionuclide concentration in a flowing water system at a single point in time may not be representative of its concentrations over a 90-day or longer growing season. Biota that live in flowing water act as selective accumulators of radionuclides over extended time periods. The magnitude of radionuclide accumulation by different kinds of aquatic biota can be influenced by the stable-element composition of the water and the chemical form and concentration of the radionuclide. For these reasons, a radiochemical analysis of water may not always be a reliable indicator of biological availability (Kevern 1964; Austin, Klett. and Kaufman 1967; Till and Meyer 1983).

The concentration ratios for ⁶⁰Co in trench water was 7000, 310, 1500, 300, 38000 and 4500 for algae, clams, goldfish, carp, Veronica roots and Veronica foliage, respectively. This compares with 10000, 1300, 1100, 420, 7800 and 730 for the same biota exposed to seep water. The concentration ratios for ⁹⁰Sr for algae, clams, fish, Veronica roots, Veronica foliage were 40, 20, 20, 150 and 40 respectively when exposed to trench water.

	Algae	Clams	Coldfish	Carp	Veronica Roots	Veronica Foliage
			TREE H WATER			
144 20	2.1	0.7	0.49	0.10	18.1	1.5
Talc.	3.0	4.1	0.50	0.12	20.8	1.8
124 Sb	10.8	0.4	1.3	0.42	53.9	4.6
103 _{Ru}	2.2	0.2	0.58	0.12	19.1	0.80
134		0.01		0.04	1.7	0.25
137	0.12	0.07	0.17	0.08	0.52	0.13
957.	3.7	0.25	0.63	0.15	22.0	2.4
95 _{Nb}	11.5	0.87	2.5	0.57	76.1	7.8
5.00	5.1	0.23	1.3	0.23	32.4	3.8
60 _{Cc}	7.0	0.31	1.5	0.30	38.0	4.5
Stom	8.3	0.26	1.2	0.30	40.6	4.8
59 F.	5.1	0.22	1.2	0.22	28.2	3.1
657-	5.7	0.33	1.6	0.31	28.5	3.5
90.	0.04	0.02	0.02*		0.15	0.04
23. J. 240 p.	0.23	0.10	0.04*		0.91	0.14
238 _{Pu}	26	1.1	0. **		10.4	1.5
54 _{Mn}	1.2	· · ·		0.02		0.28
60 _{Co}	10.0	1.3	1.1	0.42	7.8	0.73
163 _{P.1}					0.35	0.008
106Ru			0.05		0.35	0.02
1255b	0.18	0.01		0.005		0.03
137 Co				0.06	0.50	
905r		21000	36000*		117000	200000
	76000					
106 _{Ru}						
137 _{Cs}	50	**				•• ••
95 _{Nb}	50				56	
60 _{Co}				1500		
		250	200	160	160	120
	230					

TABLE 4. Concentration Ratios (pCi/g DW:pCi/ml water)1*2

¹Multiply all values by 10³. To convert CRs to met weight basis, divide above values by the following factors: algae-4.7, class-5.1, gold/ist and carp-3.6, <u>Veromica</u> roots and foliage-4.0. *Combined goldfish and carp samples.

This compares with ratios of 76, 21, 36, 117 and 200 million for the same biota exposed to spring seep water. These data indicate that all classes of biota have a capacity to accumulate ⁹⁰Sr from spring seep water.

Concentration ratios for tomatos are shown in Table 5.

TABLE 5. Concentration Ratios (pCi/g DW:pCi/ml water) for Tomato Leaves and Stems, Roots and Fruits Grown with Roots Exposed to Spring Seep Water

Radionuclide	Roots	Leaves and Stems	Fruit
⁵⁴ Mn	1100	40	
60Co	7300	180	60
103Ru	1000		1
106Ru	2500	4	
124Sb	300		
125Sb	2000		
⁹⁰ Sr	100000000	247000	9400000

ENVIRONMENTAL CONSIDERATIONS

It is well known that the accumulation of radionuclides by aquatic organisms is influenced by many factors including the stable element composition of the water, the concentrations of individual radionuclides, and the kind of organism (Kevern 1964, Austin 1967). Therefore, the concentration ratio is a somewhat specious concept, and can fluctuate widely over short periods of time depending on ambient water concentrations. Nevertheless, concentration ratios are useful in the formulation of radiological dose assessment models (Till and Meyer 1983). Poston and Klopfer (1986) have recently reviewed the extensive literature pertaining to radionuclide concentration ratios in fish. For comparison purposes, the concentration ratios calculated in this study and selected literature values from Till and Meyer (1983) are presented in Table 6. These data indicate that organisms exposed to the spring seep water had higher concentration ratios than values published in the literature for similar kinds of aquatic organisms. This suggests that 60Co and 90Sr in the spring seep water are readily available for biological uptake after migrating through the ground.

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Radionuclide	Benthic Algae	Clams	Carp/Goldfish
54Mn	2300	400	160*
⁵⁹ Fe	4800	9600*	2000*
60Co	800	790	125*
58Co	2300	1850*	125*
65Zn	16000*	4100	830*
⁹⁰ Sr	640*	320*	8.2
⁹⁵ Zr	34000*	2.6	2.0*
106Ru	1600*	1.2	19*
103Ru	1600*	3.6*	19*
124,125Sb	85		100*
134 · 137 Sb	1000	220	5600*
141,144Sb	9800*	9000	125*

TABLE 6. Radionuclide Concentration Ratios¹

¹Adapted from Till and Meyer (1983). *Generic values i.e., plants, molluscs, fish.

REFERENCES

- Austin, J. H., C. A. Klett and W. J. Kaufman. 1967. "Use of Continuous Cultures for Estimating Strontium Sorption by <u>Chlorella pyrenoidosa</u>." Int. J. Oceanol. Limnol. 1:1-27.
- Fruchter, J. S., C. E. Cowan, D. E. Robertson, D. C. Girvin, E. A. Jenne, A. P. Toste and K. H. Abel. 1984. Radionuclide Migration in Ground Water Annual Report for FY1983. NUREG/CR-3712, PNL-5040. Prepared by Pacific Northwest Laboratory for the U.S. Nuclear Regulatory Commission Washington, D.C. 122 p.
- Jenne, E. A., C. E. Cowan and D. E. Robertson. 1984. "A Comparison of Analytical Charge-Form and Equilibrium Thermodynamic Speciation of Certain Radionuclides." In Proceedings Fifth International Conference on Nuclear Methods in Environmental and Energy Research, ed. J. R. Vogt, pp. 302-310, CONF-840408, National Technical Information Service, Springfield, Virginia.
- Kevern, N. R. 1964. "Strontium and Calcium Uptake by the Green Alga, Oocystis eremosphaeria." Science 145:1445-1446.
- Merlini, M., and R. Bittel. 1970. "The Uptake, Loss, and Distribution of Radiocobalt in a Freshwater Fish Lepomis gibbosus, L.". In Actes du Symposium International de Radioecologie, pp. 399-412. Centre D'Etudes Nucleaires de Cadarache, Cadarache, France.
- Poston, T. M., and D. C. Klopfer. 1986. A Literature Review of the Concentration Ratios of Selected Radionuclides in Freshwater and Marine Fish. PNL-5484, Pacific Northwest Laboratory, Richland, Washington.
- Reed, J. R. 1971. "Uptake and Excretion of ⁶⁰Co by Black Bullheads Ictalurus melas (Rafinesque)." Health Phys. 21:835-844.
- Till, J. E., and H. R. Meyer, eds. 1983. Radiological Assessment: A <u>Textbook on Environmental Dose Analysis</u>. NUREG/CR-3332, ORNL-5968. Prepared by Oak Ridge National Laboratory for the U.S. Nuclear Regulatory Commission, Washington, D.C.

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Radionuclide Accumulation by Aquatic Biota Exposed to Contaminated Water in Artificial Ecosystems Before and After Its Passage Through the Ground	4 DATE REPORT FOMPLETED MONTH YEAR September 1987
AUTHORIS) C. E. Cushing, W. H. Rickard and D. G. Watson	MONTH TEAR February 1988
Performing Organization name and Mailing address finction Zio Cook Pacific Northwest Laboratory P.O. Box 999 Richland, Washington 99352	8 PROJECT DSK WORK UNIT NUMBER 9 FIN OF GRANT NUMBER B2437
Division of Engineering Office of Nuclear Regulatory Research U.S. Nuclear Regulatory Commission Washington, DC 20555	Technical
2 SUPPLEMENTARY NOTES	1
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CONTAMINATED WATER IN ARTIFICIAL ECOSYSTEMS BEFORE AND AFTER ITS PASSAGE THROUGH THE GROUND

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