

BLACK WILLOW (SALIX NIGRA MARSH.) AS A BIOACCUMULATOR OF  
RADIOACTIVE POLLUTANTS IN A FRESH WATER ECOSYSTEM

A Thesis

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## ABSTRACT

A search was made for a suitable bioaccumulator to serve as a dependable monitor for radioactive pollution in the lower Mississippi River. Small Black Willow shoots (Salix nigra Marsh.) were rooted in a man-made outdoor stream. The rootlets produced from the shoots were exposed in separate experiments to a known concentration of a typical fission product ( $^{140}\text{La}$ ) and a 4d transition metal ( $^{56}\text{Mn}$ , which mimics  $^{54}\text{Mn}$ , a typical long lived activation product in reactor effluents) in suitable compounds at known flow rates to determine if significant biological uptake occurred. Radiotracers and instrumental neutron activation analysis provided evidence that both elements were concentrated at the roots. Manganese, as evidenced by  $^{56}\text{Mn}$ , was concentrated in the rootlets by a factor as high as 56,000 over simultaneously sampled water. Similarly, stable La measured by  $^{140}\text{La}$  reached a maximum concentration of 130,000 over simultaneously sampled water. The coefficient of accumulation is the ratio of the  $\mu\text{g}$  of the tracer/g dry weight of root to the  $\mu\text{g}$  of the tracer/ml of water which was simultaneously sampled. Seven hours after introduction, the coefficient of accumulation for Mn was 29,000. The coefficient of accumulation for La was 25,000 after 8 hours. Data suggest that the willow may serve as a reliable monitor of radiopollutants in fresh water ecosystems where traditional monitors such as clams are unavailable.

## CHAPTER I

### Introduction

The need for a dependable pollution indicator in the lower Mississippi River is apparent in view of the ongoing construction of nuclear power plants on the river. The Mississippi River is not only a source of water for area inhabitants but also a source of nutrients for organisms in a commercial fishing area. Therefore, it is important to be certain that the river does not acquire concentrations of radioactive contaminants that would be detrimental to aquatic organisms and humans.

Monitoring of radioactivity would be most useful if accomplished on a cumulative exposure basis rather than by means of spot measurements, as evidenced by analysis of weekly or monthly water samples (Williams, 1976). A continuous monitor is superior because the concentration of pollutants varies with time, and samples taken at intervals may not reflect the average concentration for the period of interest (Thomas, 1973).

Organisms may use pollutants as a source of nutrients for growth (Bartsch, 1948). The low activity which will be introduced into the river coupled with the great dilution factor probably will produce no gross effect on the biota of the river (Waterford Steam Electric Station Preliminary Safety Analysis Report, 1974). But the organisms could provide both qualitative and quantitative measurements of the activity introduced by concentrating the elements available in the river water.

A bioaccumulator is an organism which tends to concentrate materials of interest over that found in the surrounding medium. Several organisms endemic to the lower Mississippi River were investigated as possible

bioaccumulators of radioactive pollutants. Oligochaete worms, caddisflies, and Asian clams were all three rejected because of a lack of sufficient numbers of specimens after several sampling attempts. Mayfly larvae, although abundant, were also unacceptable because the nymphs develop into adults which are capable of flight (Ross, 1956). This makes it difficult to be certain that adequate numbers of larvae could be collected on any particular day.

The Black Willow (Salix nigra Marsh.) occupies virtually the entire undeveloped shoreline in the middle and lower Mississippi River. This species is usually restricted to the lower parts of the floodplain and is considered a pioneer species in floodplain forests (Crites and Ebinger, 1969). The Black Willow quickly establishes itself in the mud along the river's edge, and during this study roots were observed awash in the river water even when the water was at a 20 year low.

A literature search including the Water Resources Research Catalog (1974 & 1975) and the Bibliography of Agriculture (1969-1976) revealed no research aimed at utilizing roots as bioaccumulators of radioactive pollutants. Several studies are exploring the feasibility of using roots and whole plants as a means of reducing salts from waste water in tertiary treatment schemes.

A study was done on the absorption of fission products by plants (Rediske et al., 1955). The rare earths were considered the most important fission product group. Included in the report were some fundamental assumptions of importance to a study dealing with bioaccumulation. Rediske found that plants make no distinction between isotopes

of the same element as far as absorption is concerned. Also, plants will tend to absorb elements of similar chemical characteristics, like the lanthanides, actinides, or the various periods, in a similar manner. Rediske defined concentration factor as the ratio of the concentration of fission products in the leaves to the concentration of fission products in the root environment. The concentration factor varies according to plant species, plant organ, carrier concentration, and the pH of the nutrient environment.

In preliminary examinations, Rediske found in nearly all cases that different species of the same genus have concentration factors which vary by no more than one order of magnitude. In general, the concentration factor for a fission product increases as the concentration of the stable isotopes of that element is increased in the nutrient substrate. In all cases, the concentration factor increases as the pH of the nutrient environment decreases (increased acidity).

Rediske found that leaves of plants had a concentration factor for the elements studied as great as or greater than any other organs. Roots had a concentration factor about the same or slightly less than the leaves. Therefore, these two organs provide for a maximum indication. One additional point of interest was that by reducing the specific activity of the fission products, within practical limits, uptake is not decreased. On the contrary, absorption may even be increased.

Solutions of radioelements and their compounds may be true solutions or they may be in the form of a radiocolloid. These radiocolloids readily adsorb on glass surfaces causing error when determining the amount of radioisotope present. This effect is primarily of concern

when dealing with short half-life radioisotopes (Chase and Rabinowitz, 1967; Gardner and Ely, 1967). To avoid this situation, activation polyvials were used and the samples were counted in the same containers in which they were collected.

According to Price (1972) there are three possible mechanisms leading to the observed increase in plant uptake with time: the formation of organic complexes or chelates (especially for heavy metals), a buildup of radionuclide concentration at root surface, or the slow but continual uptake by perennial plants. Radionuclides in aquatic environments tend to accumulate within the system even though the trend is toward continual recirculation of micronutrients in the ecosystem.

Manganese-56 was chosen to represent radioactive wastes which may be incorporated into the river ecosystem as products of corrosion of reactor hardware. It has a short half-life of 2.58 hours which is of primary importance in terms of safety and not contaminating the stream with activity for any length of time. This allows for additional introduction of the same radioisotope in subsequent experimentation without interference. Although the short half-life causes no potential hazard to the environment,  $^{56}\text{Mn}$  does mimic the longer lived  $^{54}\text{Mn}$  (312.5 day half-life) which is produced by  $^{55}\text{Mn}(n,2n)^{54}\text{Mn}$ . Also taken into consideration is the 100 per cent occurrence of stable precursor  $^{55}\text{Mn}$ , radiotracer capabilities, and easily resolvable peaks in a gamma ray spectrum.

Lanthanum-140 was chosen as a representative of fission fragments of the heavy group having mass numbers ranging from 125 to 155. A sesquioxide ( $\text{R}_2\text{O}_3$ ) is formed by each of the rare earth elements, all

of which are stable toward oxidation in air at room temperature. All of the sesquioxides are soluble in acids, but they do not dissolve in water. However, they can combine with water to form hydrates of the oxides (Yost et al., 1947). The isotope  $^{140}\text{La}$ 's short half-life of 40.23 hours, 99.91 per cent natural occurrence of stable precursor  $^{139}\text{La}$ , radiotracer capabilities, and easily resolvable peaks in a gamma ray spectrum made it a reasonable choice for experimentation.

All of these radiopollutants emit beta particles and characteristic gamma radiation in the process of forming a stable species. Therefore, a need to be able to monitor releases of these radioelements to assure that reasonable limits are met does exist, and a reliable, sensitive monitoring system is extremely desirable.

## CHAPTER II

### Materials and Methods

#### Experimental Stream

The study was carried out at a man-made stream located on the edge of Corporation Canal, a water drainage system for the Louisiana State University Campus in Baton Rouge, Louisiana. The stream is constructed of wood lined with fiberglass and resin. It measures 8 meters by 1 meter with baffles alternately attached to the sides which causes a serpentine flow of water down the length of the stream (Fig. 2-1). The water can be pumped by two different modes, either being drawn from a reservoir at the bottom of the stream to be recirculated in a closed system or being drawn from Corporation Canal in a "once through" mode. The stream volume, including the reservoir, is approximately 1200 liters. This volume could be varied considerably by altering the location, number, and height of dams placed in the stream.

Willow shoots measuring from 15 to 30 cm were first collected at St. Francisville, Louisiana along the Mississippi River and placed in loose gravel through which water was flowing in the artificial stream. In approximately three weeks, elaborate root and leaf systems developed (Fig. 2-2). Additional shoots and saplings were collected along the Mississippi River at Baton Rouge and along Corporation Canal, all of which were placed in the stream and used along with the others for root and leaf sampling in the experiment.

#### Manganese Run #1

The possibility of the Black Willow serving as a bioaccumulator of

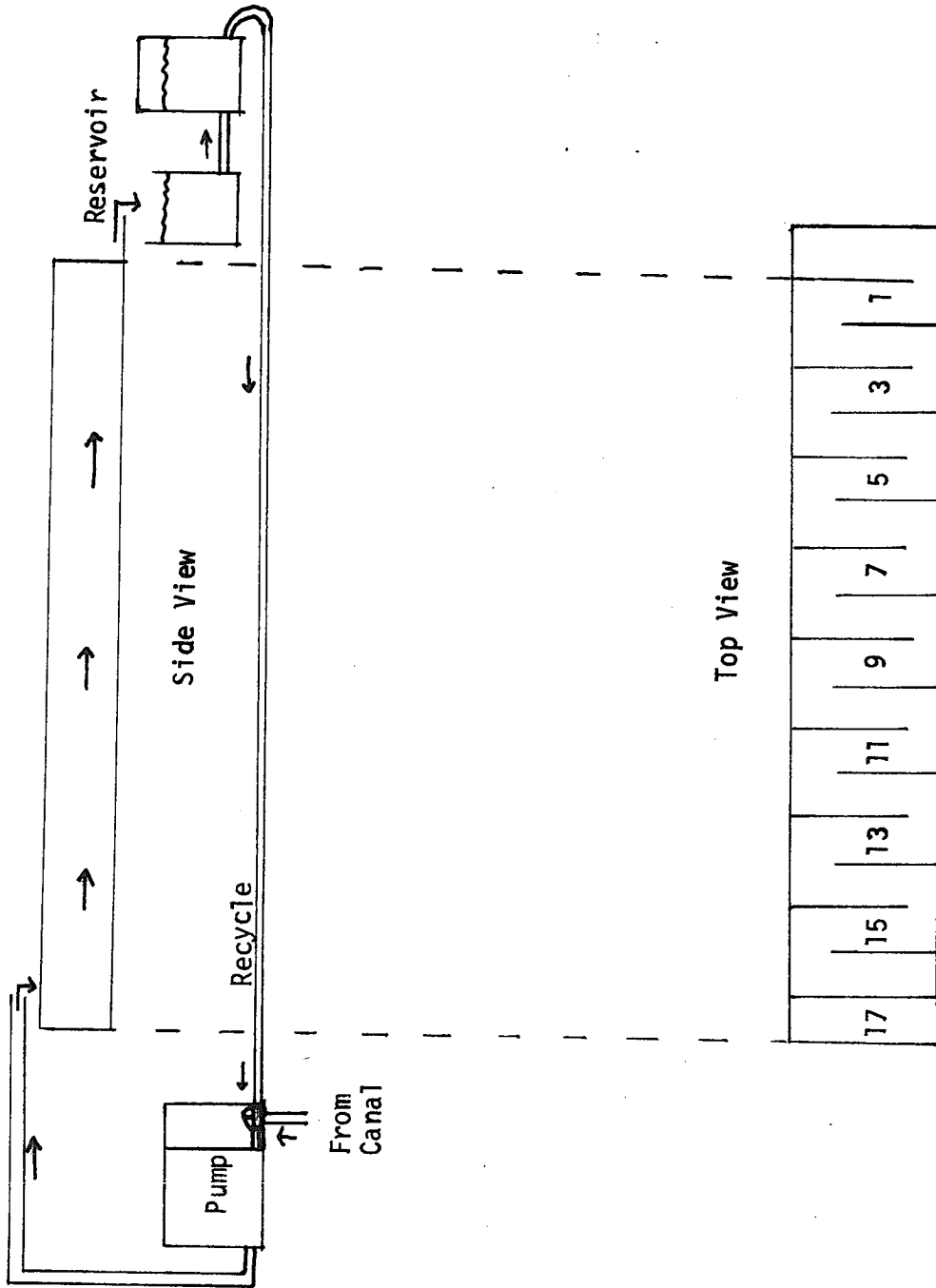


FIGURE 2-1

Side and top view of simulated stream.

corrosion products was examined first. A trial activation was conducted using the formula,  $A = N\phi\sigma(1 - e^{-\lambda t})e^{-\lambda t'}$  (Lapp and Andrews, 1972), to estimate the amount of target material needed to obtain the desired activity. Using this information, 5 g of Mn in 15.39 g of  $\text{MnSO}_4 \cdot \text{H}_2\text{O}$  dissolved in 70 ml of distilled water was irradiated with 20 mg of  $^{252}\text{Cf}$  which emits  $10^7$  n/(sec-g). The irradiation was done in the Californium Irradiation Facility at the Nuclear Science Center, Louisiana State University. Six encapsulated  $^{252}\text{Cf}$  sources were placed in a holder made specifically for this experiment (Fig. 2-3).

Since  $^{55}\text{Mn}$  is 100 per cent in isotopic abundance, the  $^{252}\text{Cf}$  neutrons produce  $^{56}\text{Mn}$  by the reaction  $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$ . Manganese-56 emits gammas of 0.847 MeV (99%), 1.811 MeV (29%), and 2.11 MeV (15%). The 0.847 MeV gamma was used in comparing the samples after both radioactive introduction and instrumental neutron activation analysis (INAA). For the first run, 1.85 mCi of  $^{55}\text{Mn}$  in 5 g of Mn was introduced into trough 16 (Fig.2-1) in a single pulse and allowed to recirculate in the stream by way of the pump. Equilibrium was established since the water was flowing at a rate of 53 l/min for a circulation time of approximately 20 minutes, leading to a calculated concentration of 4.1  $\mu\text{g Mn/ml}$ . The water temperature was 31°C. Samples of both water and roots were taken from trough 8 at regular intervals. The samples were collected in two dram polyethylene activation vials and taken to the Nuclear Science Center where they were counted on a 3" x 3" NaI(Tl) well detector (Harshaw Chemical Co.) attached to a multichannel analyzer. Samples continued to be collected until the gamma counts revealed that the activity was insignificant.

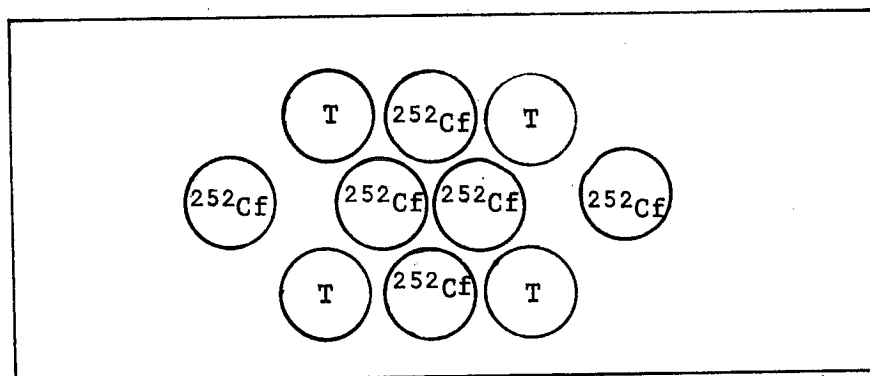


FIGURE 2-3

Holder for activation of four containers of liquid tracer. The sum of the six  $^{252}\text{Cf}$  sources is 20 mg.  
T = tracer.

For two weeks following the introduction of the Mn, a water and root sample were taken each day for INAA in order to extend the scope of the run.

#### Manganese Run #2

In the second experiment, 5 g of Mn in the same chemical form was dissolved in 100 ml of distilled water and activated as before. The temperature of the stream water was 27°C, and the water was pumped continuously from Corporation Canal and allowed to drain back into the canal at a point much below the withdrawal point. The flow was adjusted to 10 l/min. A total of 1.82 mCi was introduced into trough 12 (Fig. 2-1) with a Holter peristaltic pump at a rate of 1.2 ml/min, producing a calculated concentration of 5.6 µg Mn/ml. Samples were taken until the activity in the water was indistinguishable from background.

#### Lanthanum Run #1

To examine the fate of a typical fission product, 1 g of La in 1.17 g of La<sub>2</sub>O<sub>3</sub> was dissolved in 7 ml of concentrated HCl and was diluted in 65 ml of distilled water (72 ml total volume). This solution was then activated by <sup>252</sup>Cf as was done in the Mn runs. With an isotopic abundance of 99.91 per cent, <sup>139</sup>La activates according to the following reaction: <sup>139</sup>La(n,γ)<sup>140</sup>La. Lanthanum-140 emits gammas of 0.329 MeV (20%), 0.487 MeV (40%), 0.815 MeV (19%), 0.923 MeV (10%), 1.596 MeV (96%), and 2.53 MeV (3%). Both the 0.487 MeV and the 1.596 MeV gammas were used in the calculations.

The flow rate of the stream was regulated to an average of

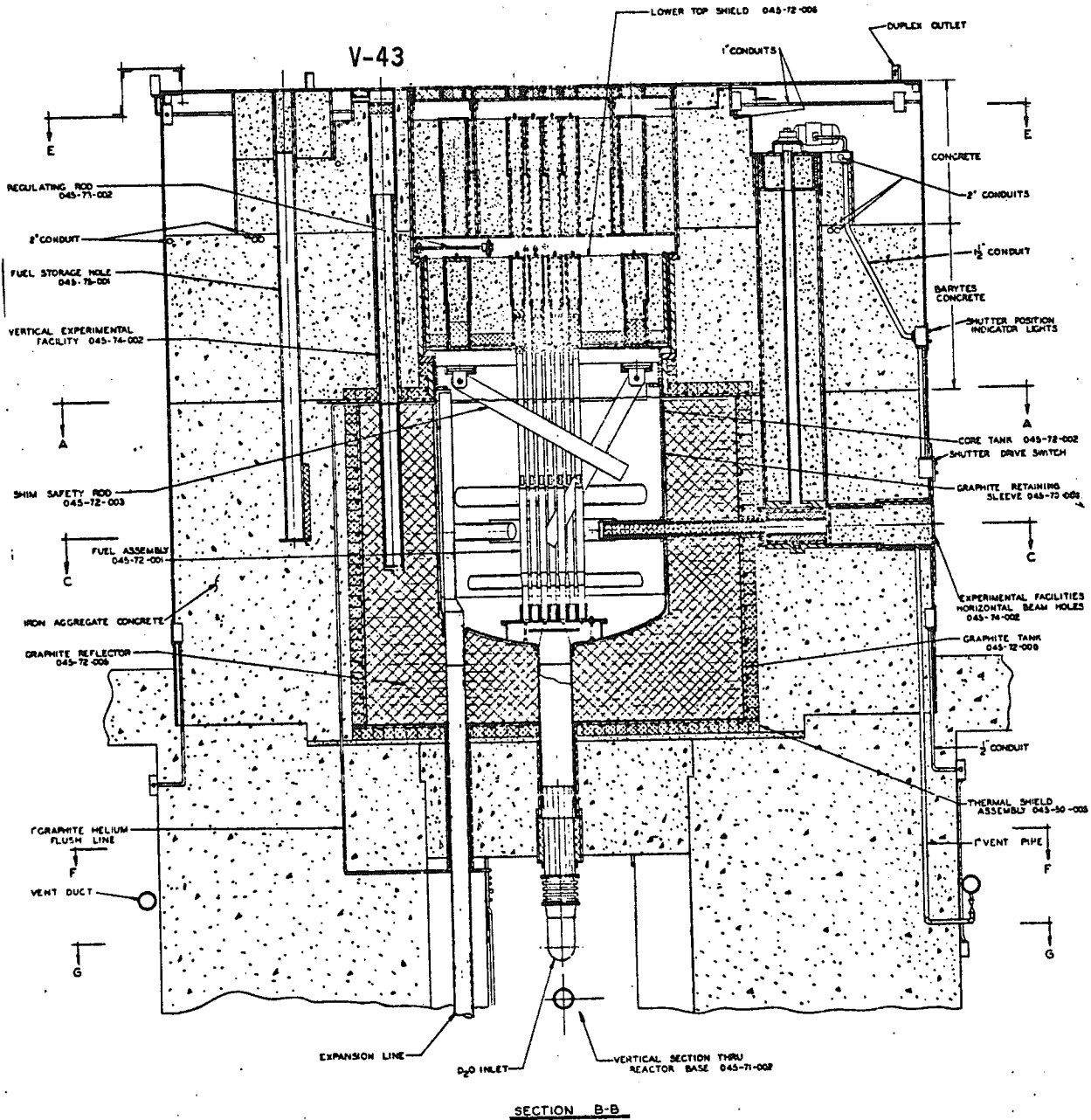
25 l/min for La run #1 and 10 l/min for La run #2. The water temperature was recorded as 27°C for run 1 and 30°C for run 2. In both runs, the water was pumped from Corporation Canal and not allowed to recirculate. In La run #1, the La was introduced in the amount of 22.1  $\mu\text{Ci}$  with a peristaltic pump at a rate of 1.2 ml/min, producing a calculated concentration of 0.67  $\mu\text{g La/ml}$ . Samples of water and roots were taken from trough 8. After 2 hours, sampling was discontinued because of low activity.

#### Lanthanum Run #2

In the second La run, 0.033 g of La in 0.102 g of  $\text{La}(\text{NO}_3)_3 \cdot 6 \text{H}_2\text{O}$  was activated at the Georgia Institute of Technology in Atlanta, Georgia. Irradiation was for 30 minutes at a flux of  $10^{13}$  n/cm<sup>2</sup>-sec in Position V-43 (Fig. 2-4). Upon receipt at Baton Rouge, the compound was dissolved in 100 ml of distilled water. The 0.45 mCi solution was dripped into trough 11 (Fig. 2-1) by means of a peristaltic pump at a rate of 0.7 ml/min, producing a calculated concentration of 0.023  $\mu\text{g La/ml}$ . The pH of the water in the stream was 6.7. Water and root samples were taken from trough 8 at regular intervals until the photopeaks for  $^{140}\text{La}$  gammas in the water samples were indistinguishable from background.

#### Instrumental Neutron Activation Analysis

Selected samples from the first Mn and first La runs were taken to Georgia Tech to be activated in the Georgia Tech Research Reactor for INAA. Samples from the #2 runs were not considered because of artificial contamination by the first run of each element. Nitrogen gas was introduced into the activation vials containing roots to reduce



Georgia Tech Research Reactor, Georgia Institute of Technology, Atlanta

FIGURE 2-4

Vertical section B-B through reactor showing Position V-43 where samples were irradiated.

the interference from  $^{40}\text{Ar}(n,\gamma)^{41}\text{Ar}$ . The Mn samples were irradiated in Position V-43 at 250 kw for 2 hours at a flux of  $4.5 \times 10^{12}$  n/cm<sup>2</sup>-sec. The Mn samples, including standards, were then counted on a 2040 channel pulse height analyzer coupled to a 7% Ge(Li) detector (Canberra Industries) and a HP 9100 B programmable calculator for data reduction. The Peak Picker program (Walker, 1972) was used to locate and integrate the peaks. The  $\mu\text{g}$  amounts of Mn were determined and compared to the values obtained in the first run.

The La samples were activated in the reactor and shipped to Baton Rouge for counting. Since no significant quantitative data was obtained from the first La run due to low production of  $^{140}\text{La}$ , the information made possible by neutron activation could only be compared with the second La run.

#### Data Acquisition and Reduction

In all four runs, standards were made from the activated solution and were each counted on the Harshaw well crystal mentioned previously and a Ge(Li) gamma spectrometer (Canberra Industries) located in the Nuclear Science Center, both of which were connected to multichannel analyzers. The absolute efficiencies of both detectors were determined using a  $^{137}\text{Cs}$  standard from the National Bureau of Standards and a Nuclear-Chicago  $^{137}\text{Cs}$  tubular well source. The values obtained were used in calculating the amount of activity introduced into the stream in each of the four runs.

For each water and root sample, the net area of the photopeak of the particular radioisotope introduced was calculated using the Covell method (Covell, 1959). This value was multiplied by a decay factor

to take into account the elapsed time from initial administration of the radionuclide to the counting time. This corrected value of cpm was then divided by the sample volume or weight in order to obtain the counts/ml and counts/g. The root samples were assessed on a dry weight basis since dry weight provides a more accurate comparison due to differences in the water content of root wood and variable wetness of the roots. Using the known quantity of Mn or La in each standard, the  $\mu\text{g}$  quantities in each sample was determined. Finally, a coefficient of accumulation (Gileva, 1960) was determined by dividing the  $\mu\text{g}$  of the tracer/g dry weight of root by the  $\mu\text{g}$  of the tracer/ml of water which was simultaneously sampled. Long counts were made of selected root samples several days after each experiment to be certain there was no contribution to the peaks from any long lived radionuclides. None were found.

#### Radiation Safety

The entire experimental procedure was monitored with portable Eberline E-120 survey meters having HP-190 probes. The standards were made in a vented hood shielded with lead bricks. Transfer of the activated solution to the stream was done in a 40 pound steel carrying pig. The area surrounding the stream was roped off and marked as a radiation area. When the activity was being introduced with the peristaltic pump, the solution was shielded with lead, and the area of introduction was avoided as much as possible. Although the total volume of activated solution measured over 50 mr/hr at the surface, the stock solution was divided into four equal parts. At no time was

more than one-half of the total volume at the site of introduction at one time. The dose received by personnel was minimal due to adequate shielding, minimal exposure time, and maximum distance between the point of introduction and the sampling area. The maximum reading obtained at the water surface in the stream was 5 mr/hr when the activity was introduced via peristaltic pump.

## CHAPTER III

### Discussion of Results

#### Manganese Run #1

In the first Mn run, 1.85 mCi was introduced into the stream (Table 3-1). The entire volume of 70 ml was introduced at one time as a pulse. The water samples reflect this by showing a drop from 9.9  $\mu\text{g Mn/ml}$  in trough 8 immediately after introduction to 1.8  $\mu\text{g Mn/ml}$  in the next sample taken 30 minutes later (Fig. 3-1). This quantity continued to drop and leveled off at less than 1  $\mu\text{g Mn/ml}$  after 6 hours.

In all four runs, the calculated and actual concentrations of the elements introduced showed reasonable agreement (Table 3-1). The differences in the calculated versus the actual concentrations can be accounted for by the lack of thorough dilution of the element and the variable flow rate of the stream caused by pumping irregularities.

The amount of Mn in the root samples was found to increase with time. On a gram dry weight basis, the sample taken immediately after introduction had only 750  $\mu\text{g Mn/g}$  (Fig. 3-2). This value increased to over 1400  $\mu\text{g Mn/g}$  after 2 hours. A maximum of over 4000  $\mu\text{g Mn/g}$  was obtained after 10 hours.

The coefficient of accumulation was less than 100 after 30 minutes (Fig. 3-3). After 2 hours, the coefficient of accumulation had climbed to 900, peaking at over 5000 at 7 hours. The coefficient remained at a value of approximately 2300 for the remainder of the run. These findings were reinforced by samples taken each day for two weeks

TABLE 3-1

## INTRODUCTION OF RADIOISOTOPES

Mn Run #1	Mn Run #2	La Run #1	La Run #2	Administration Method	Stream Flow	g of element introduced	Calculated Concentration* $\mu\text{g/ml}$	Actual Concentration (max. value) $\mu\text{g/ml}$	$\mu\text{Ci}$ at beginning of experiment
16				Pulse	Recycle	5	4.1	9.9	1850
	17			84 minute infusion	Once through	5	5.6	13.6	1820
		12		58 minute infusion	Once through	1	0.67	0.53	22
			11	142 minute infusion	Once through	0.033	0.023	0.092	450

\* Assuming full dilution in the water passing the root sampling site (trough 8).



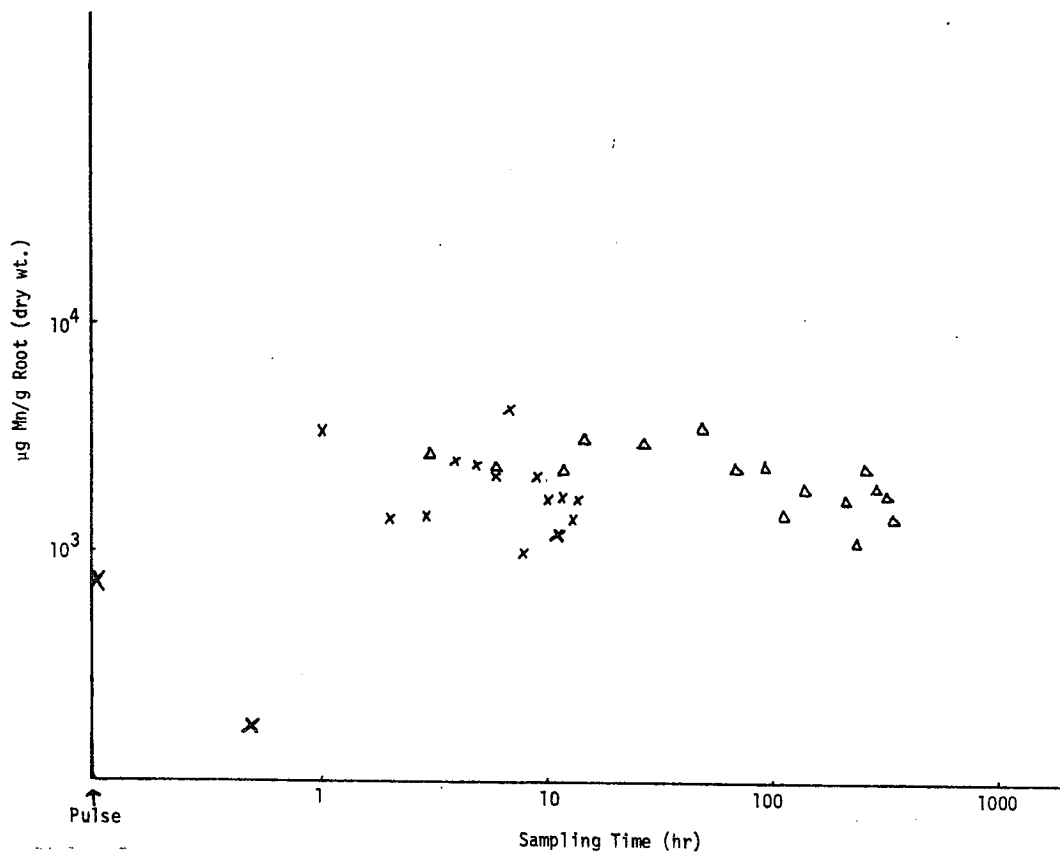


FIGURE 3-2

Mn run #1. Plot of the log of  $\mu\text{g Mn/g}$  root versus the log of sampling time. x = Radioactive introduction.  $\Delta$  = INAA. Arrow indicates pulse introduction.

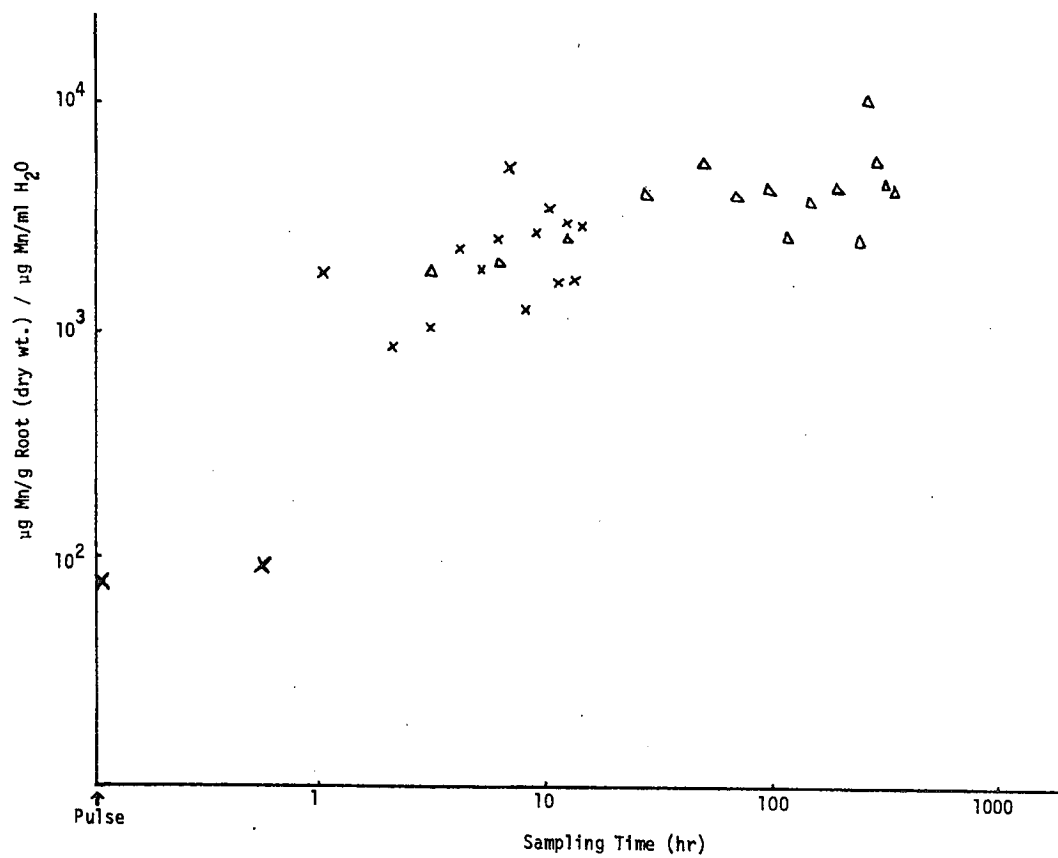


FIGURE 3-3

Mn run #1. Plot of the log of bioaccumulation of Mn versus the log of sampling time. x = Radioactive introduction.  $\Delta$  = INAA. Arrow indicates pulse introduction.

following the initial introduction of the tracer. By INAA it was determined that the Mn remained concentrated at the roots and did not show a significant decrease in concentration in samples taken after the time when water and roots showed no radioactivity. The data of Figure 3-3 demonstrate the feasibility of using short lived radiotracers in combination with stable tracers for long term tracer experiments in the environment. This is evidenced in Figure 3-3 by the overlap of three points calculated by both radioactive tracer introduction and INAA.

The oscillations observed with time in both the  $\mu\text{g Mn/g}$  of root and the coefficient of accumulation might be accounted for by the recirculation of the radioactivity which was introduced. This seems reasonable because it was the only major difference in the four experimental runs. In the remaining three runs, the activity was introduced over a longer period of time and was allowed to go past the roots only one time. This was believed to be a closer approximation of the situation which would exist in nature.

#### Manganese Run #2

For the second Mn run, 1.38 mCi in 100 ml of water was introduced over a 84 minute period, allowing for a good comparison with the first Mn run. Immediately after introduction, the water had less than 1  $\mu\text{g Mn/ml}$  (Fig.3-4). A maximum of 13.6  $\mu\text{g Mn/ml}$  was attained after 1.5 hours which was shortly after the introduction of Mn was completed. Four hours after introduction began, the activity of  $^{56}\text{Mn}$  showed less than 0.5  $\mu\text{g Mn/ml}$  where it remained for the rest of the experiment. It took longer for the maximum value to be reached because the Mn was introduced

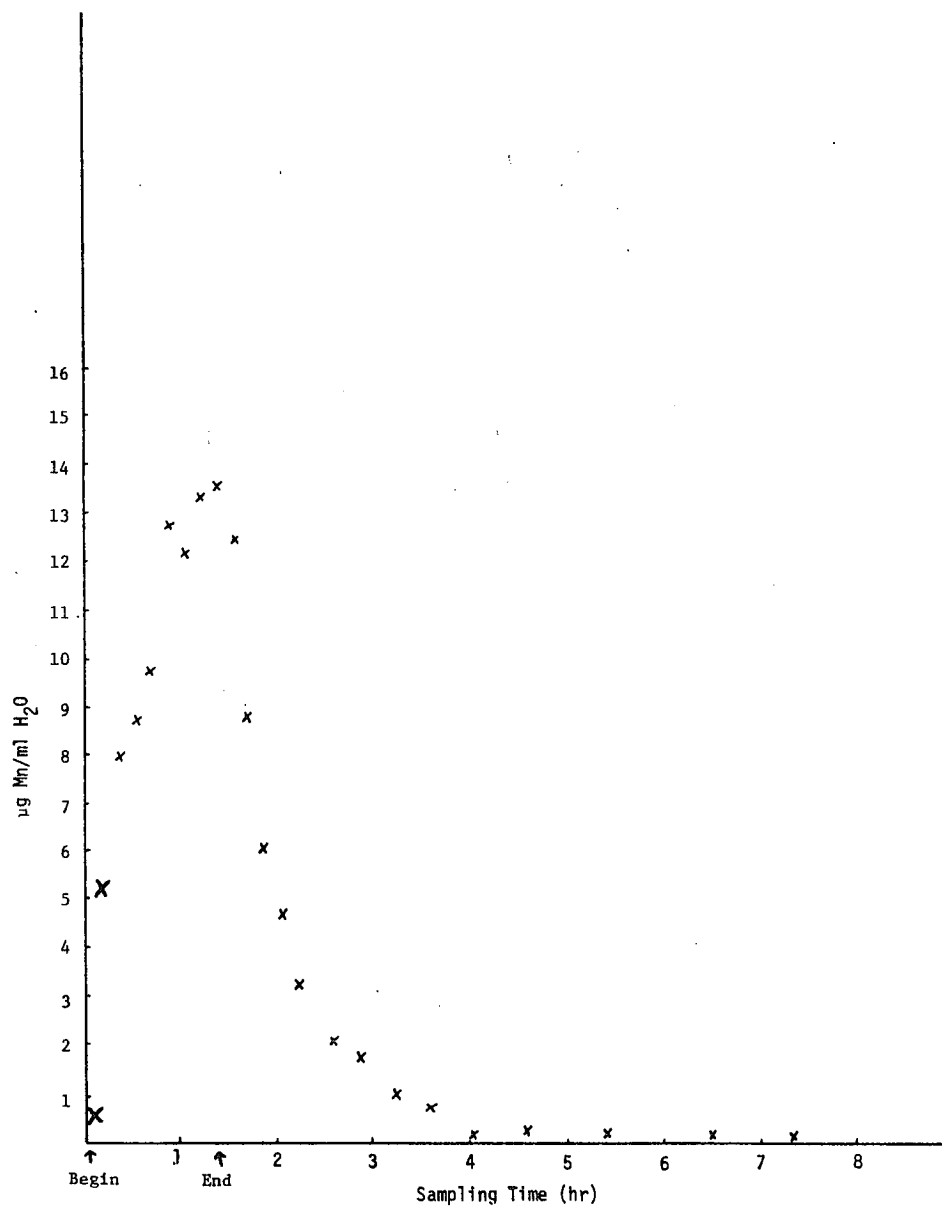


FIGURE 3-4

Mn run #2. Plot of  $\mu\text{g Mn/ml}$  water versus sampling time. x = Radioactive introduction. Arrows indicate beginning and end of introduction.

over an extended period of time, not as a pulse as in the first Mn run. The data from Mn run #2 show a much smoother rise and fall of Mn since the activity in this experiment was not allowed to recirculate. The minimum concentration of Mn was reached in less than half as much time as it took in the first Mn run.

Shortly after introduction began, the roots showed a concentration of 500  $\mu\text{g Mn/g}$  of root dry weight (Fig. 3-5). This value climbed to over 6000  $\mu\text{g Mn/g}$  after 1.5 hours when the introduction was completed. A peak was reached after 4.5 hours with a value of over 10,000  $\mu\text{g Mn/g}$ . The first Mn run did not reach a maximum half as large. The reduced flow rate and slow infusion of radioactivity allowed the activity to remain in the immediate vicinity of the roots for longer periods of time than in Mn run #1. It was assumed this increased the chances of the Mn being either adsorbed or absorbed by the willow roots.

The coefficient of accumulation steadily increased with time from a minimum of 85 at a fraction of an hour to over 5000 after 4 hours (Fig. 3-6). The roots continued to accumulate the Mn long after the majority of the activity passed the roots, despite the fact that the roots were only given one chance to collect the Mn (Fig. 3-5). In the first Mn run, the activity was passed through the roots a number of times, but they did not concentrate even half as much Mn as in the second Mn run (Fig. 3-2). This may be evidence that the fast flow rate (52 l/min versus 10 l/min) could have kept the Mn suspended in solution and not have allowed it to settle out or adsorb on the surface of the roots.

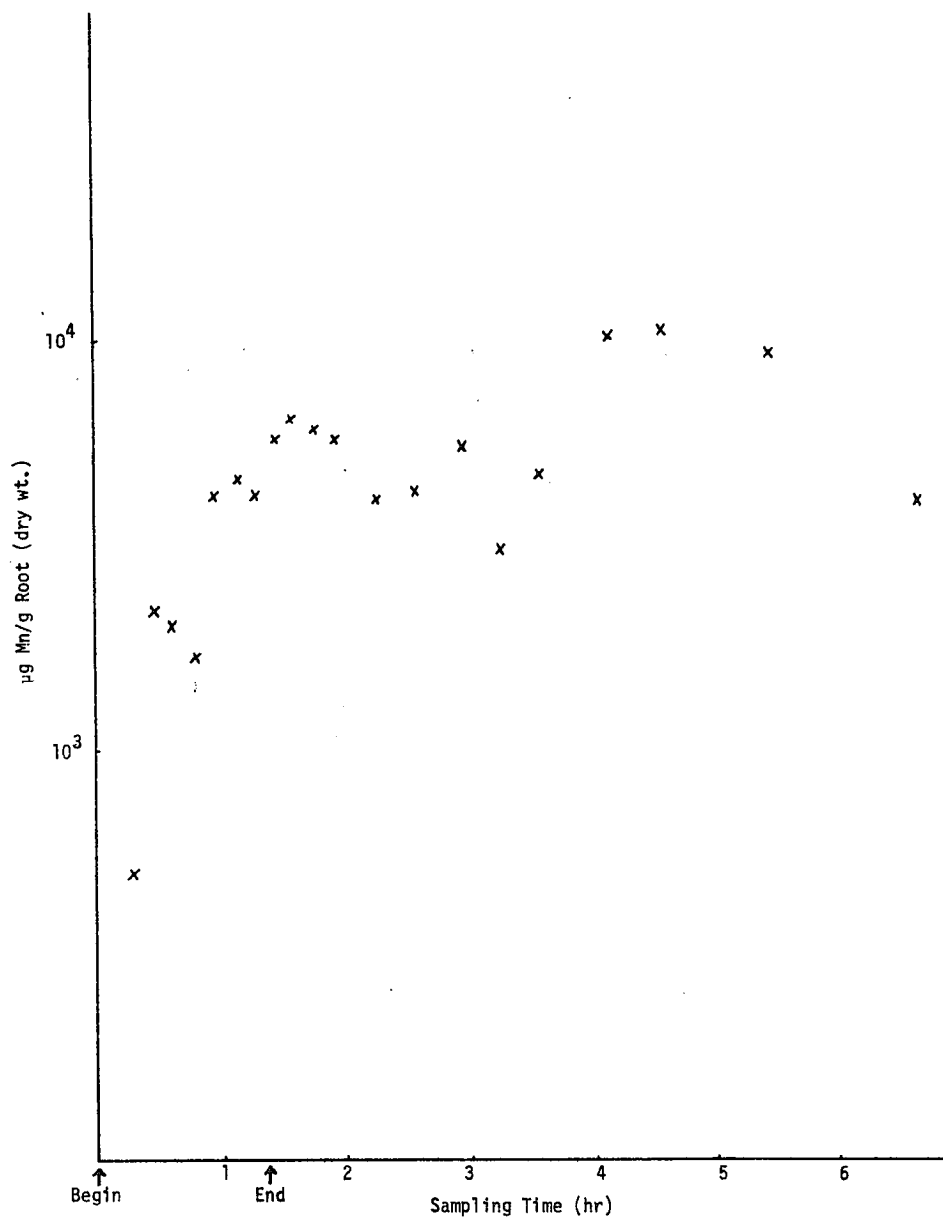


FIGURE 3-5

Mn run #2. Semilog plot of  $\mu\text{g Mn/g}$  root versus sampling time. x = Radioactive introduction. Arrows indicate beginning and end of introduction.

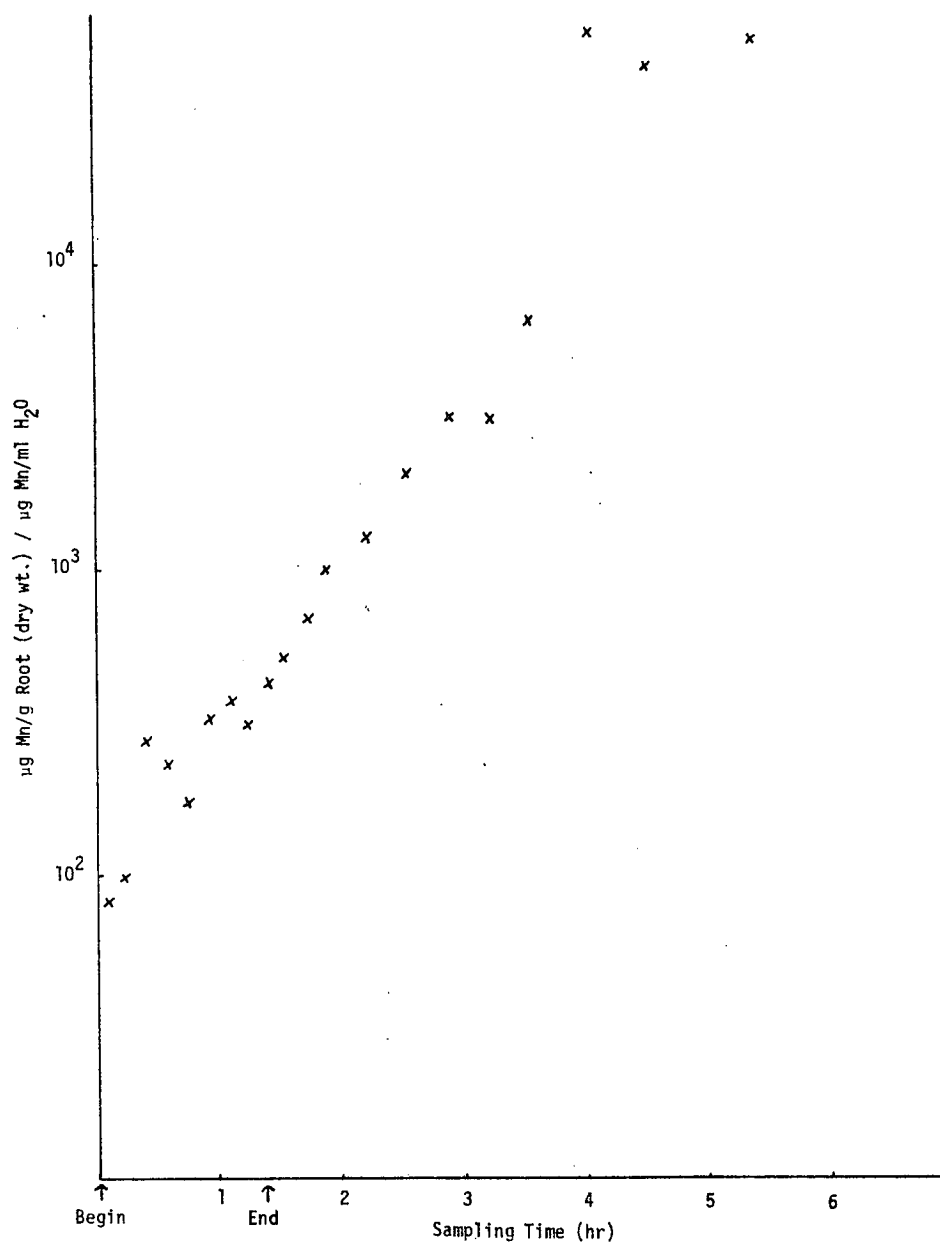


FIGURE 3-6

Mn run #2. Semilog plot of bioaccumulation of Mn versus sampling time. x = Radioactive introduction. Arrows indicate beginning and end of introduction.

Lanthanum Run #1

Only 22  $\mu\text{Ci}$  of  $^{140}\text{La}$  was introduced into the stream in the first La run. The neutron flux produced by the  $^{252}\text{Cf}$  sources was not sufficient to produce over 100  $\mu\text{Ci}$  of  $^{140}\text{La}$  calculated to be necessary for quantitative results. Also, the physical form which was necessary for use as an environmental tracer made activation less efficient. Because of the great dilution factor and high flow rate of the stream (25 l/min), this small amount of activity was not ample to provide water and root samples with statistically valid counts. However, the data did show a relative decrease in activity in the water samples after the La was introduced. In contrast, the roots showed an increase.

By INAA of the samples from the first La run, statistically valid information was obtained by calculating and comparing the photopeak of interest in each activated sample. The water samples produced a sharp, smooth peak shortly after the introduction was completed (Fig. 3-7). In trough 8 the samples went from 0.11  $\mu\text{g La/ml}$  of water at 0.9 hours to a maximum of 0.36  $\mu\text{g La/ml}$  after 1.3 hours. This value then dropped to less than 0.05  $\mu\text{g La/ml}$  after 2 hours.

The activated root samples did show an increase in concentration of La with time, but the increase was not consistent (Fig. 3-8). The maximum concentration of only 21  $\mu\text{g La/g}$  of root dry weight was obtained after 1.8 hours. Between the point of introduction and the sampling area was a number of materials with large surface areas for adsorption such as sand, dirt, rocks, and the stream walls. Therefore, not enough of the La reached the roots in the sampling area.

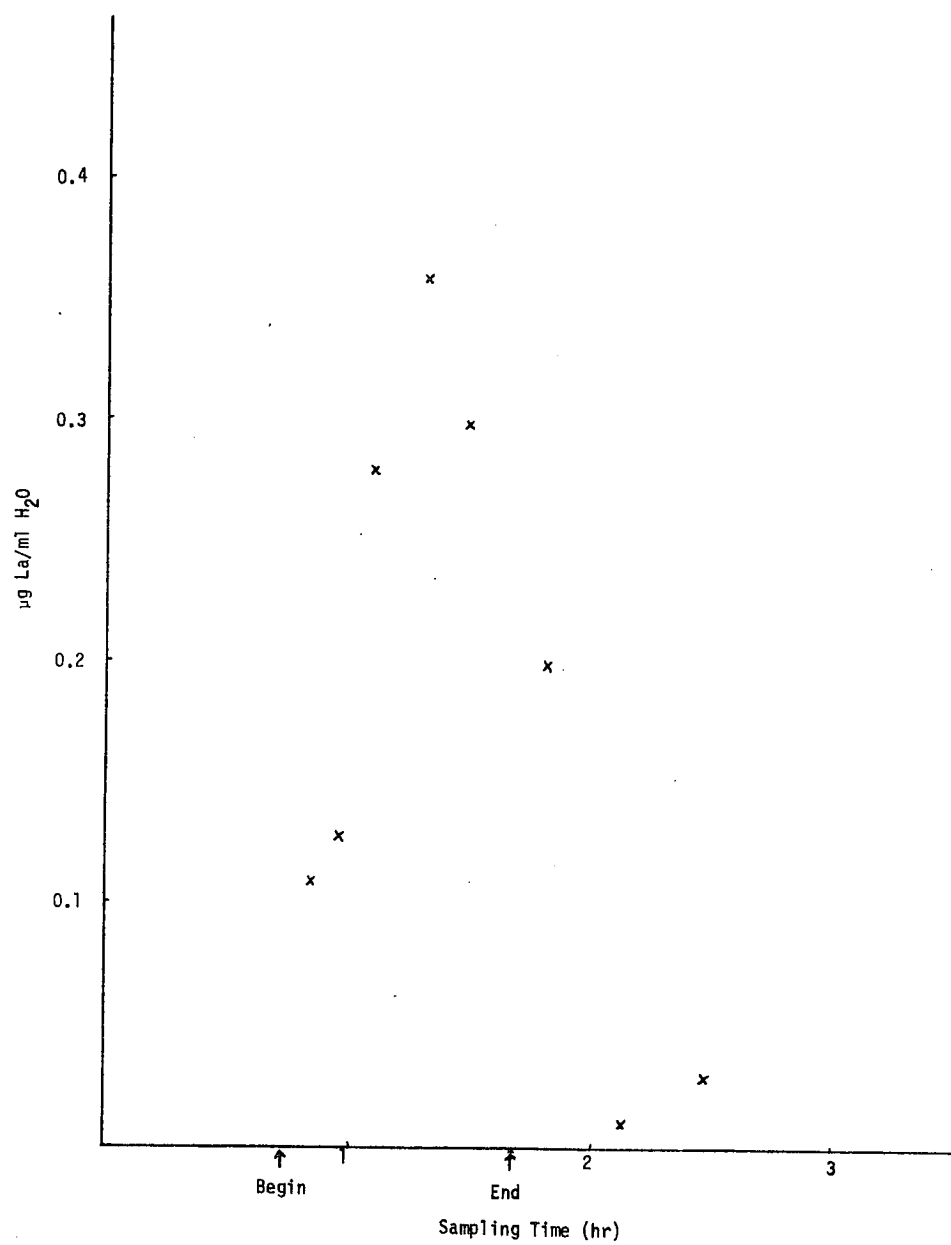


FIGURE 3-7

La run #1. Plot of  $\mu\text{g La/ml water}$  versus sampling time. x = INAA. Arrows indicate beginning and end of introduction.

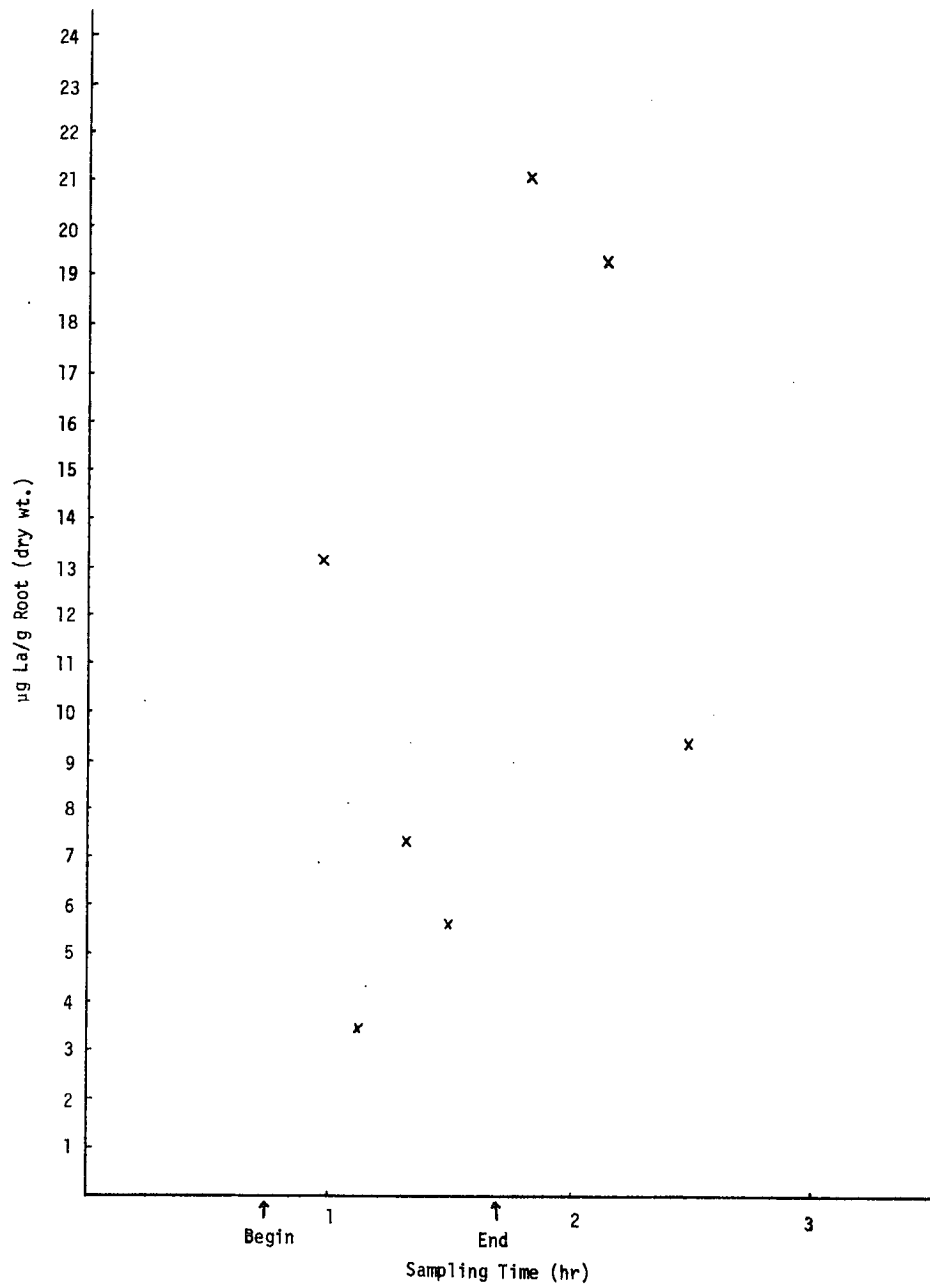


FIGURE 3-8

La run #1. Plot of  $\mu\text{g La/g}$  root versus sampling time. x = INAA. Arrows indicate beginning and end of introduction.

The coefficients of accumulation determined by INAA for the first La run were extremely low, most of them being below 100 (Fig. 3-9). Although the data showed no regular pattern, the first La run did provide useful information in that it showed that the roots did indeed tend to concentrate the radioisotope over water even though the  $^{140}\text{La}$  was present in only  $\mu\text{Ci}$  quantity.

#### Lanthanum Run #2

In the second La run, 0.45 mCi was introduced into the stream. The activity was introduced into trough 11, reducing the chances of losing the La before it reached the roots. The flux of neutrons provided by the Georgia Tech Research Reactor increased the amount of  $^{140}\text{La}$  produced even though the amount of La activated was 30 times less than in the first La run (Table 3-1).

The amount of La in the water samples went from 0.0008  $\mu\text{g La/ml}$  in the sample taken immediately after introduction to a maximum of 0.092  $\mu\text{g La/ml}$  in the sample taken 19 minutes after the completion of introduction (Fig. 3-10). This value dropped very quickly to a minimum of 0.0001  $\mu\text{g La/ml}$  1.45 hours later.

Lanthanum uptake by the roots was very rapid (Fig. 3-11). The first sample taken 6 minutes after introduction began had 0.088  $\mu\text{g La/g}$  of root dry weight. A maximum value was reached of 24.98  $\mu\text{g La/g}$  35 minutes after introduction was completed. The concentration in all later samples remained above 5  $\mu\text{g La/g}$  of root dry weight.

The coefficient of accumulation of La in the roots went from 20 shortly after introduction to a maximum of over 130,000 which was reached

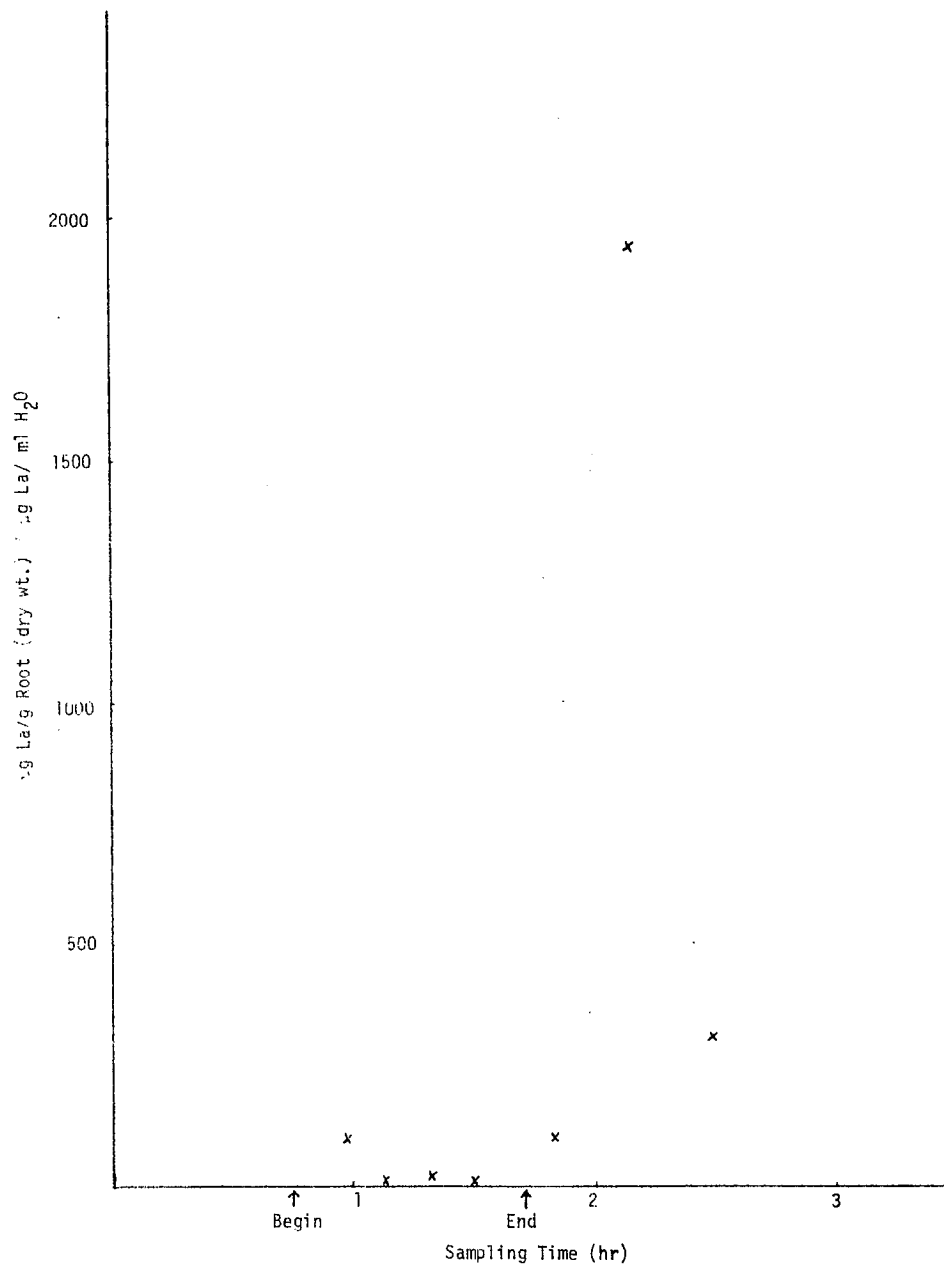


FIGURE 3-9

La run #1. Plot of bioaccumulation of La versus sampling time. x = INAA. Arrows indicate beginning and end of introduction.

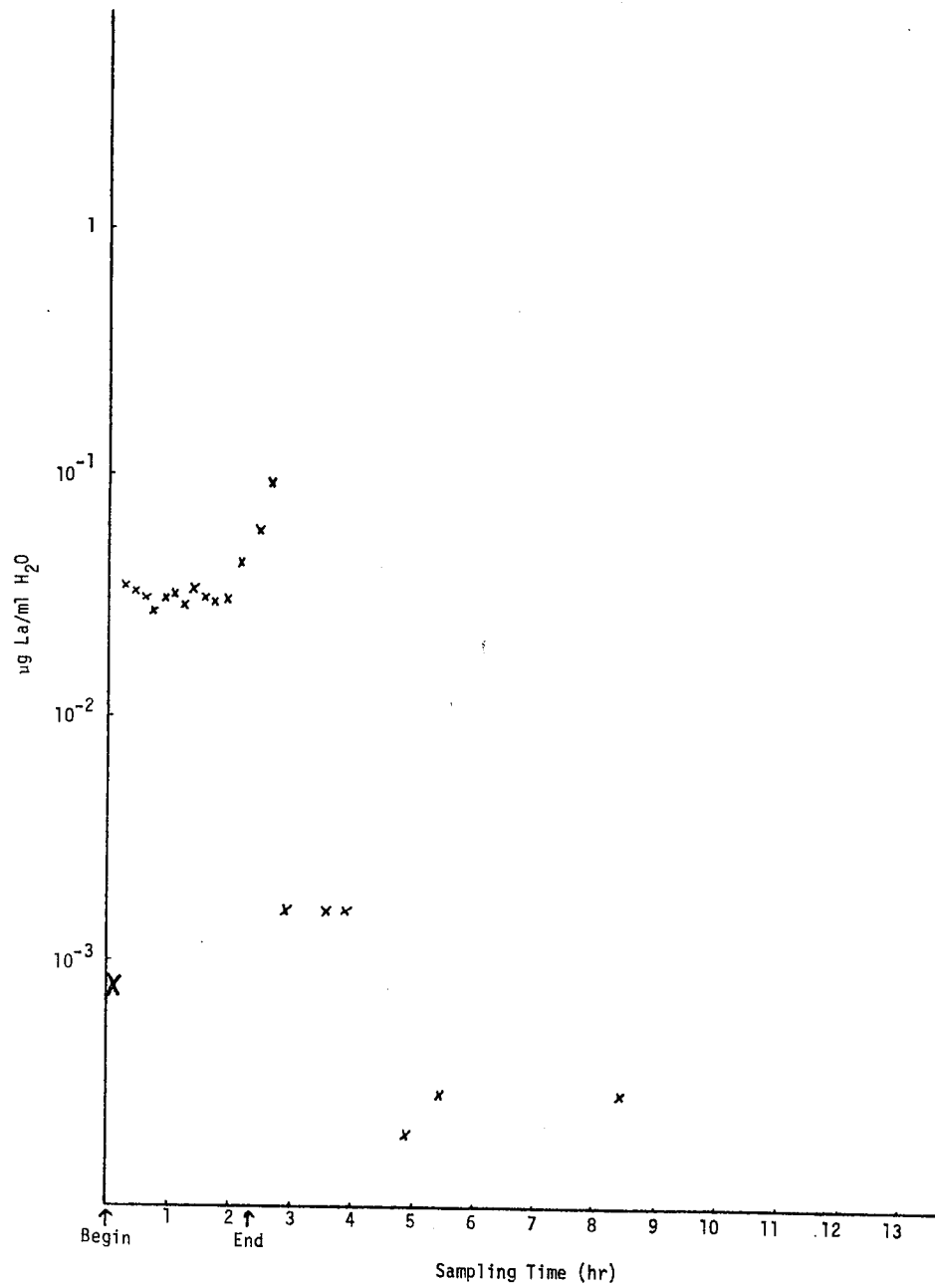


FIGURE 3-10

La run #2. Semilog plot of  $\mu\text{g La/ml}$  water versus sampling time. x = Radioactive introduction. Arrows indicate beginning and end of introduction.

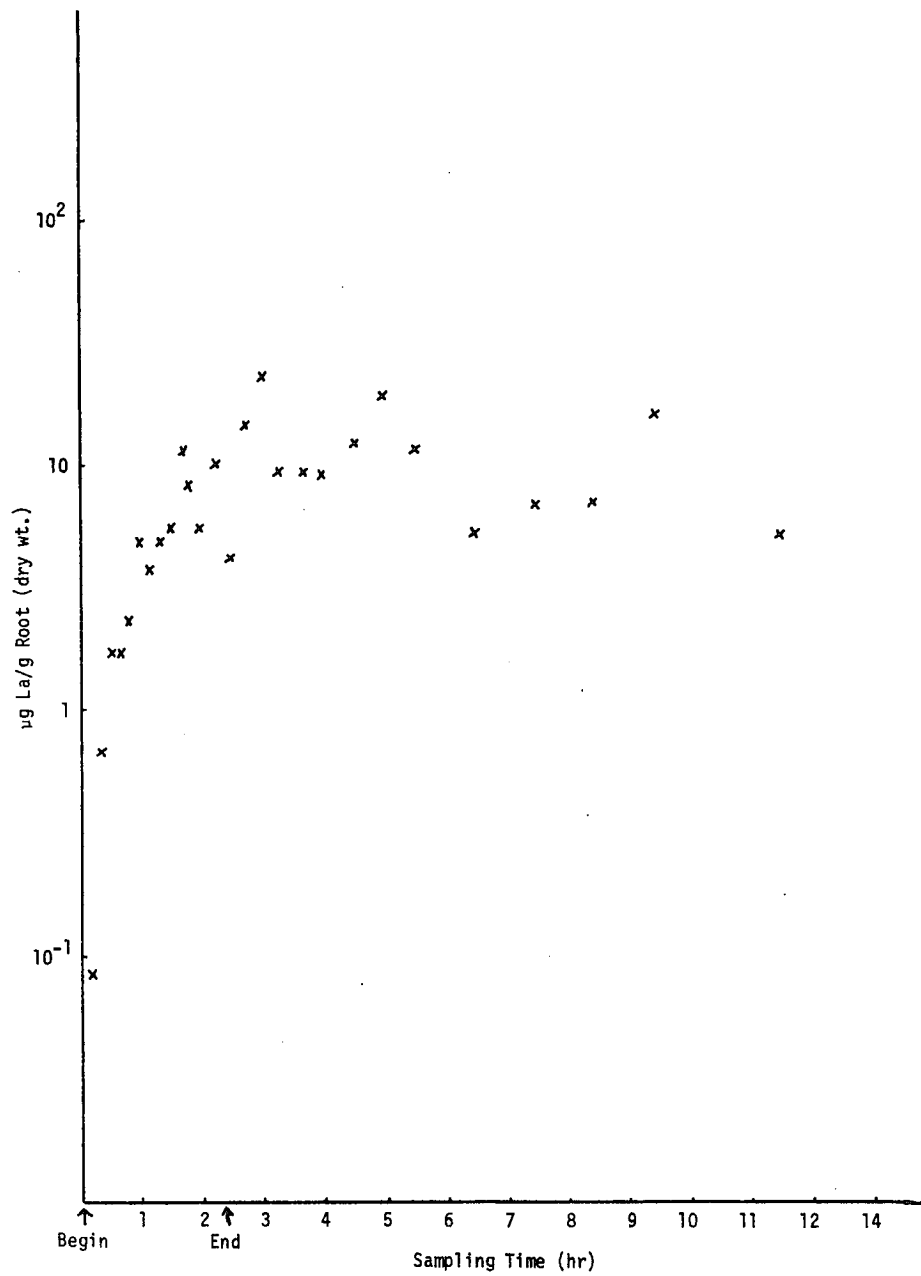


FIGURE 3-11

La run #2. Semilog plot of  $\mu\text{g La/g root}$  versus sampling time. x = Radioactive introduction. Arrows indicate beginning and end of introduction.

one hour after the introduction was completed (Fig. 3-12). During this experiment the slow infusion caused the maximum coefficient of accumulation to be reached after an extended period of time. The coefficient was reduced by a factor of 5 to about 25,000 after 4 additional hours, showing that the La was retained in the roots to a large extent.

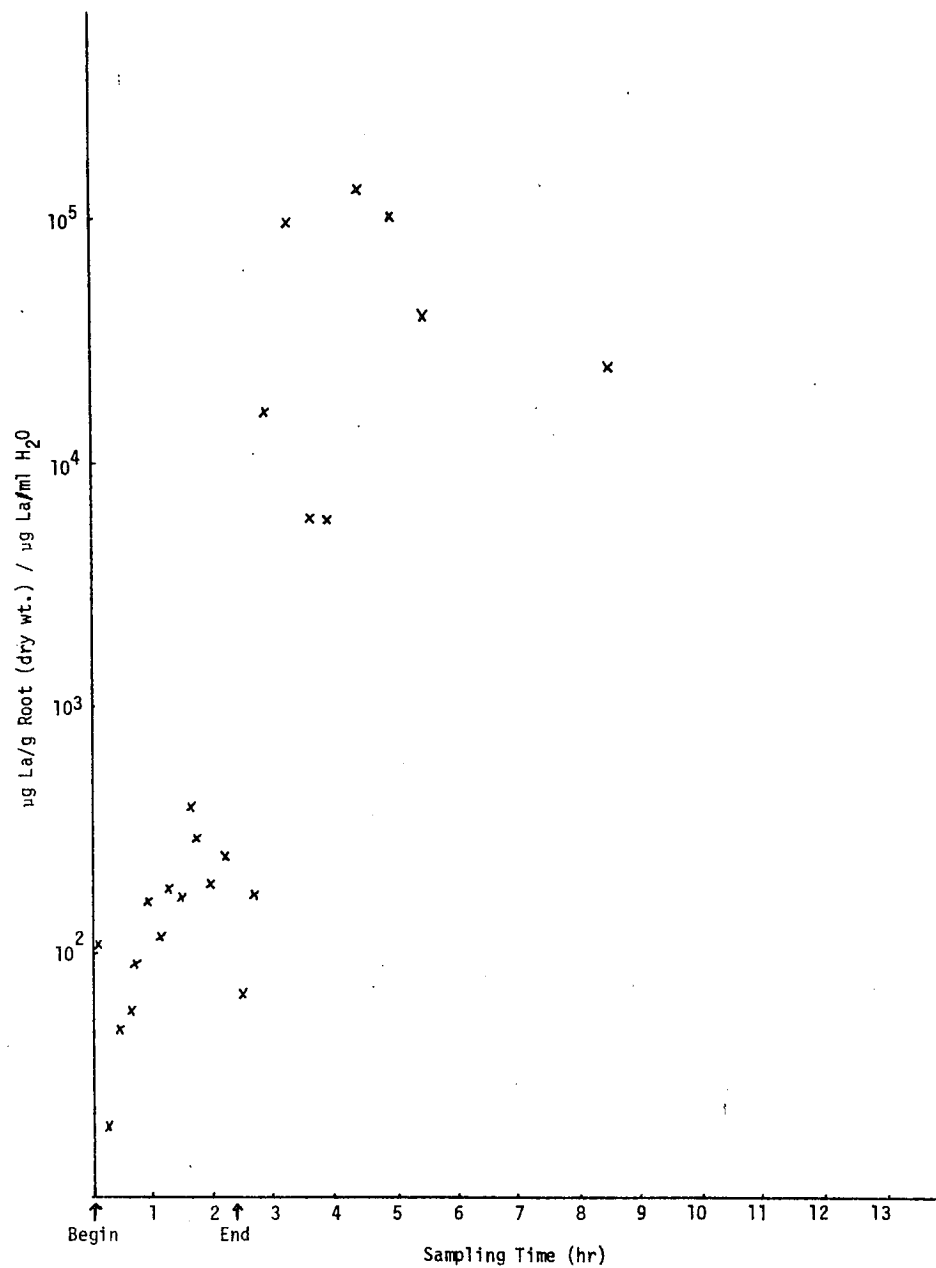


FIGURE 3-12

La run #2. Semilog plot of bioaccumulation of La versus sampling time. x = Radioactive introduction. Arrows indicate beginning and end of introduction.

## CHAPTER IV

## Conclusions

Both Mn and La, after being introduced into the simulated stream ecosystem, were concentrated at the roots of the Black Willow (Salix nigra Marsh.). The data obtained by short lived radiotracers combined with data obtained from instrumental neutron activation analysis give evidence that these two techniques can be used for long term tracer experiments in the environment.

Since the gamma energies are essentially identical for  $^{54}\text{Mn}$  and  $^{56}\text{Mn}$  and gammas follow every disintegration, the counting yield for the two isotopes (based on 0.84 MeV  $\gamma$ ) will be almost the same. Therefore, data on detection of  $^{56}\text{Mn}$  is directly applicable to  $^{54}\text{Mn}$ .

For the long lived  $^{54}\text{Mn}$ , over a short time period the specific activity will be almost constant, and the coefficient of accumulation C can be written in terms of activities:

$$C = \frac{A_{\text{Mn}}/\text{g root}}{A_{\text{Mn}}/\text{ml H}_2\text{O}}$$

Assuming a conservative value of  $5 \times 10^3$  for the coefficient of accumulation,

$$A_{\text{Mn}}/\text{ml H}_2\text{O} = (2 \times 10^{-4})(A_{\text{Mn}}/\text{g root})$$

In practice, counting  $^{54}\text{Mn}$  with a multichannel analyzer to allow peak selection, coupled to a large NaI(Tl) crystal should provide a conservative estimate of counting yield of 10%:

$$R = (1 \times 10^{-1})A$$

If the background in the peak region can be identified by valley-to-valley

extrapolation to allow for a peak-area estimation, and long counts can be made, a "one-half background" criteria for detection limit is not unreasonable. Assuming a background in the peak region of 10 cpm, then the lower limit of detection for  $^{54}\text{Mn}$  will be on the order of 5 cpm.

This corresponds to an activity expressed in  $\mu\text{Ci}$  of

$$A \approx (5 \text{ cpm}) / [(2.2 \times 10^6 \text{ dpm}/\mu\text{Ci})(1 \times 10^{-1})]$$

$$A \approx 2.3 \times 10^{-5} \mu\text{Ci}$$

Assuming that this is in a 5 g sample, then

$$\begin{aligned} (A_{\text{Mn}}/\text{g root}) &\approx (2.3 \times 10^{-5} \mu\text{Ci})/5\text{g} \\ &\approx 4.6 \times 10^{-6} \mu\text{Ci/g root} \end{aligned}$$

Therefore, the lower limit of detection of  $^{54}\text{Mn}$  in the water by bio-accumulation in the Black Willow is

$$\begin{aligned} A_{\text{Mn}}/\text{ml H}_2\text{O} &= (2 \times 10^{-4})(4.5 \times 10^{-6}) \\ &= 9 \times 10^{-10} \mu\text{Ci/ml H}_2\text{O} \end{aligned}$$

If the flow rate in the Mississippi River is on the order of  $3 \times 10^{12}$  ml/min, and a four hour release of  $^{54}\text{Mn}$  occurred at a constant rate, then the average concentration of  $^{54}\text{Mn}$  in the river water which could be detected would be  $9 \times 10^{-10} \mu\text{Ci/ml}$ , and the total  $^{54}\text{Mn}$  in the release would be

$$\begin{aligned} &(3 \times 10^{12} \text{ ml/min})(9 \times 10^{-10} \mu\text{Ci/ml})(2.4 \times 10^2 \text{ min}) \\ &\approx 6.5 \times 10^5 \mu\text{Ci} \\ &\approx 0.65 \text{ Ci,} \end{aligned}$$

or approximately 0.16 Ci/hr.

This series of approximations suggest that a release of 0.2 Ci/hr of  $^{54}\text{Mn}$  from a nuclear reactor into the Mississippi River could be

detected by the bioaccumulation technique using Black Willow roots. Additional elements should be tested for bioaccumulation, and future studies should be carried out under varying conditions of pH, dissolved oxygen, and temperature.

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## VITA

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