

THE OCCURRENCE AND RETENTION OF RADIONUCLIDES IN THE SEDIMENTS OF WHITE OAK LAKE*

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Abstract—As a result of the discharges of large volumes of low-level radioactive liquid wastes to surface streams at the Oak Ridge National Laboratory, large quantities of radionuclides have accumulated in the bottom sediments of White Oak Lake. Ruthenium-106 (1038 c) and cesium-137 (704 c) account for more than 90 per cent of the total activity now present at the site, while ^{60}Co (152 c), the rare earths (17 c, exclusive of ^{90}Y), and ^{90}Sr (15 c) make up the remainder. More than half of the activity is associated with the upper 6-in. sediment layer, while progressively smaller quantities of activity are found with depth. The ruthenium, which is restricted to a small area in the now dry upper lake bed, is partially water soluble; however, its rate of movement through the soil is slow enough so that radioactive decay reduces the concentration of that reaching surface streams to insignificant levels. Most of the ^{137}Cs occupies highly selective exchange sites on the illitic fraction of the clay in the sediment and can be desorbed only by disruption of the lattice structure. Only a small fraction of the ^{60}Co in the soil was found to be exchangeable. It is, therefore, unlikely that any large fraction of the ^{137}Cs or ^{60}Co would move from the area except through erosion of the sediment. About one-half of the ^{90}Sr and the rare earths in the sediment appears to be exchangeable, while the other half is in the form of slightly soluble salts. Through leaching by ground water, a slow depletion of strontium from the dry part of the lake bed occurs.

The accumulation of radionuclides in the sediments of White Oak Lake illustrates the effectiveness of relatively quiescent bodies of water in concentrating activity in stream beds and in retarding the downstream movement of these materials.

INTRODUCTION

AT MOST nuclear energy installations radioactive liquid wastes may be classified as "high-level" or "low-level". The high-level wastes, which are relatively small in volume but contain most of the radioactive materials, are stored in tanks. The low-level wastes, which occur in large volumes but contain comparatively little activity, are treated to reduce the concentrations of the radionuclides and then are diluted and dispersed to the environment. For those wastes released at permissible levels that remain diluted and dispersed in the environment, there is no harmful effect on the public. However, because of natural or artificial processes, radionuclides may accumulate and be reconcentrated in selected areas of the environment.

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Through natural geologic processes all radionuclides that enter surface bodies of water will sooner or later find their way downstream. Those which move in solution with the water itself will move downstream at rates approximating the average rate of movement of the water, while those which enter into the biologic cycle or are sorbed on suspended or fixed surfaces will move at a much slower rate. Relatively quiescent waters, whether natural or artificial, greatly reduce the rate of downstream transport of the radioactivity associated with the sediment phase, and, as a consequence, may allow for the accumulation and concentration of activity within selected areas of the stream bed. White Oak Lake Bed, located within the controlled area of the Oak Ridge Reservation, exemplifies the accumulation and concentration of activity downstream from the point of continuous discharges of low-level liquid wastes to surface streams.

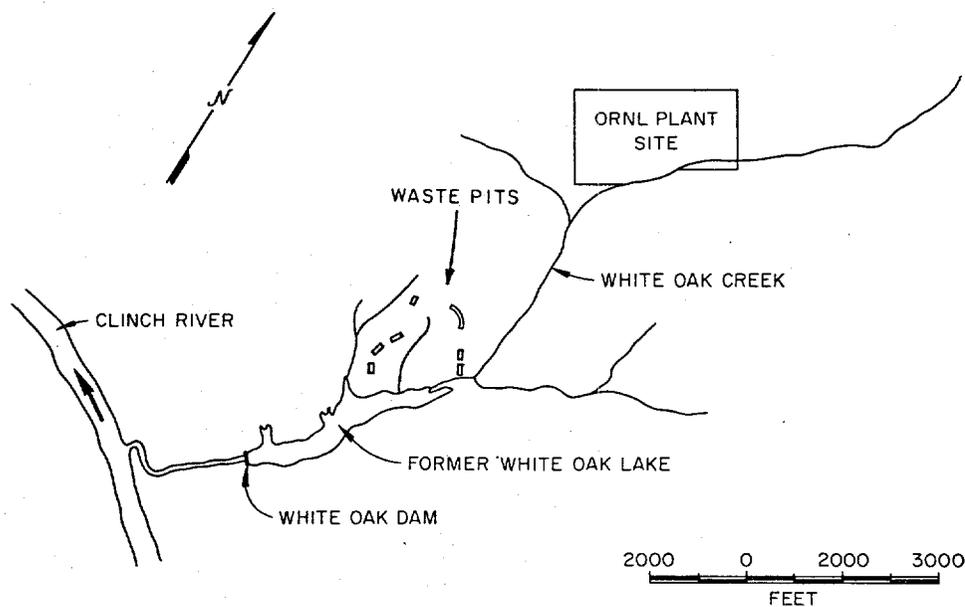


Fig. 1. Location map.

For the past 20 years large quantities of low-level radioactive liquid wastes have been routinely discharged into surface streams at the Oak Ridge National Laboratory. From the Laboratory area the waste water is transported by White Oak Creek through White Oak Lake and into the Clinch River. These releases have caused only small increases in the concentration of activity in the waters and bottom sediments of the Clinch River; however, large quantities of radionuclides have accumulated within the controlled area of the Laboratory in the sediments of White Oak Lake. The lake was formed in 1943 to serve as a final control in the water stream between the Laboratory and the Clinch River (see Fig. 1). The temporary holdup of the contaminated creek water afforded some dilution and permitted the decay of short-lived radioisotopes, but the standing pool also allowed for the deposition and accumulation of large quantities of contaminated sediments. At full pool the inundated area covered some 44 acres and extended about 1 mile upstream. For the first 12 years of its operation, the lake was maintained at or near full capacity, but since 1955, when it was completely drained, the lake stage has been kept low. At present, the inundated area covers an area that is about one-third that of full capacity.

The lake bed now contains about 1 million ft^3 of contaminated sediments. The sediment layer is as much as 2 ft thick in the lower part of the area near the dam, with a gradual thinning of the material upstream and toward the shore line of the lake. The sediment, which has been classified as a silty clay loam⁽¹⁾ is underlain by a 6- to 12-ft-thick section of alluvium and interbedded shales and thinly bedded limestones of the Conasauga formation of Cambrian age. In addition to the accumulation of radioactive contaminants in the lake bed concurrent with the deposition of the sediments, waste water presently flows onto the upper part of the now dry lake bed from the nearby intermediate-level chemical waste pits (see Fig. 1). As the waste water traverses the lake bed, some of the radioactive contaminants are removed from solution. The water table generally lies from 2 to 5 ft below the surface, but, where waste water from the seepage pits flows over the area, the ground water is closer to the land surface. The rate of ground-water movement is considerably greater in the upper layers of soil than in the underlying material, ranging from 1 to 5 ft a day in the upper 2 ft of soil and from 0.05 to 0.25 ft a day in the material 2-5 ft below the surface.⁽¹⁾

SOIL SAMPLING AND ANALYSES

A statistical sampling procedure based on the area of zones of similar activity concentrations and the variation within the zones was employed to determine sample locations in the 44-acre tract of former White Oak Lake. Data taken from annual surveys made in the lake bed during the period, 1950-1954, and showing the gross beta-gamma activity for the first 3 in. of bottom material^(2,3) were used to divide the lake bed into zones of four different radioactivity intensities (see Fig. 2) defined as follows:

Class of zone	Gross beta-gamma activity (10^{-2} $\mu\text{c/g}$)	Standard deviation (10^{-2} $\mu\text{c/g}$)
A (low)	<1.5	0.764
B (medium)	1.5 to 2.5	1.163
C (high)	2.5 to 4.5	1.451
D (very high)	>4.5	1.896

The area of the zones and the amount of variability within them were taken into account in the method of optimum allocation⁽⁴⁾ of sample points which can be expressed as

$$N_i = Na_i s_i / \sum_{i=1}^x a_i s_i$$

where N_i = number of samples allocated to the i th (any designated) zone,
 N = total number of samples in all zones,
 a_i = area of the i th zone,
 s_i = standard deviation of the i th zone,
 x = total number of zones.

By using the previously collected data for determining the standard deviation of activity concentrations in the zones and by designating 250 as the total number of samples, the expression defined above was used to determine the number of sample points allocated to each zone of the lake bed. The calculated numbers of samples were spotted in the zones in a random manner.

Core samples were obtained with a 2-in.-diameter thin wall tube, known as the Shelby Tube Sampler, which was driven into the earth and recovered manually. Because previous work indicated that practically all of the contamination was associated with the upper

few feet of soil, all cores were taken to a depth of 2 ft. The samples were sliced and segmented into four equal segments of 6 in. in length. Approximately 10 g from each segment was oven-dried weighed and scanned, using a Packard automatic gamma-counting system which consists of a spectrometer with well-type-scintillation detector, automatic sample changer and control unit, digital printer and tape punch. Concentrations of ^{106}Ru , ^{137}Cs and ^{60}Co were determined by use of an IBM 7090 computer program for stripping the gamma spectrum. In this manner a large quantity of samples (about 1000) were analyzed for gamma-emitting isotopes at a low cost and in a short period of time. Standard radiochemical procedures were used for strontium and the rare earth analyses. A random selection of 93 of the cores were analyzed for ^{90}Sr and the rare earths, while all 250 of the cores were analyzed for ^{137}Cs , ^{106}Ru and ^{60}Co .

QUANTITY AND DISTRIBUTION OF ACTIVITY

General

Ruthenium-106 (1038 c) and ^{137}Cs (704 c) account for about 90 per cent of the total activity present in the lake bed, while ^{60}Co (152 c), the rare earths exclusive of ^{90}Y and ^{90}Sr (15 c) make up the remainder (see Table 1). For all of the radionuclides it is observed that the greatest quantities of activity occur in the surface layer of soil, and progressively smaller quantities of activity are associated with successively deeper soil layers. The 0- to 6-in. soil layer contains almost 80 per cent of the ^{60}Co and the rare earths, a little more than 65 per cent of the ^{137}Cs and ^{90}Sr and about 55 per cent of the ^{106}Ru . Most of the remaining fractions of the various radionuclides occur in the 6- to 12-in. layer, while the 12- to 18-in. and 18- to 24-in. layers contain relatively little activity.

Ruthenium-106

Ruthenium-106 is concentrated in only a small area of the upper lake bed (see Fig. 3). The ruthenium as well as ^{60}Co is transported onto the lake bed by two surface streams that drain the Laboratory's intermediate-level radioactive liquid waste seepage pit area.⁽⁵⁾ In the upper 6 in. of lake bed soil the areas of ruthenium contamination coincide roughly with the

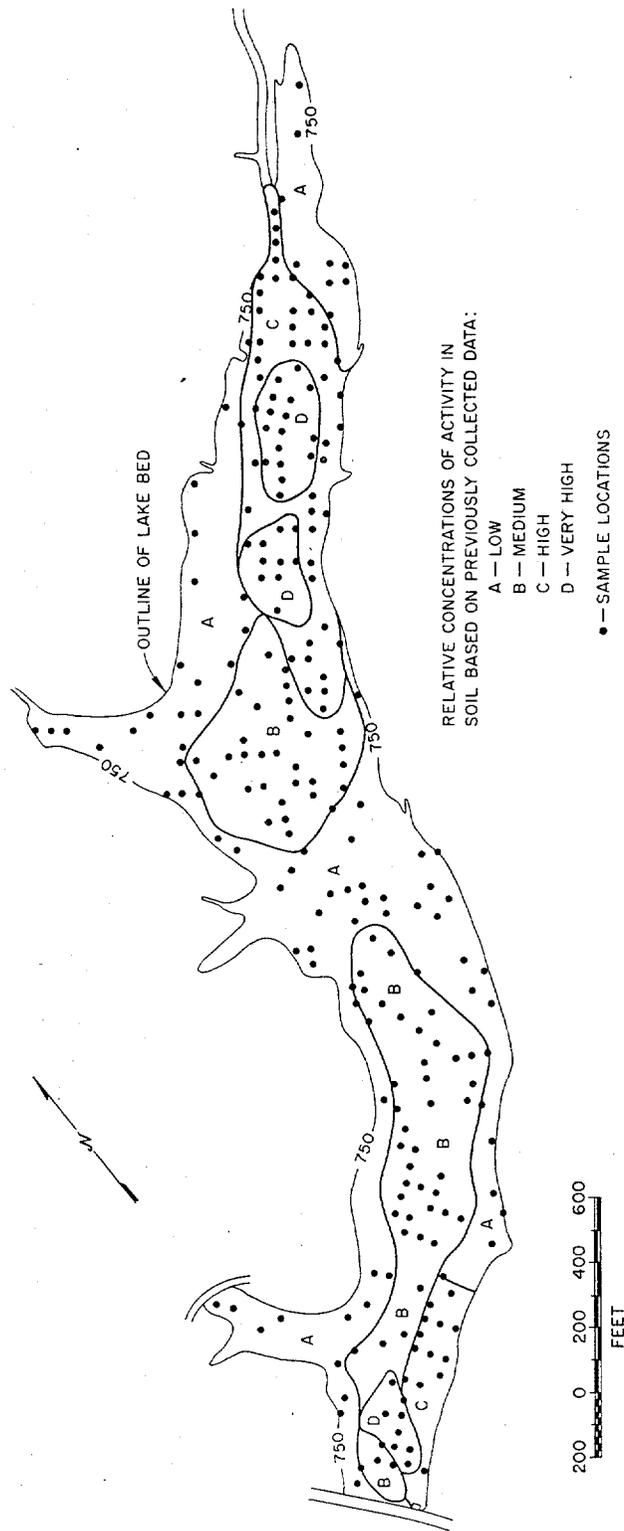


Fig. 2. Locations of core samples taken in the bed of White Oak Lake.

Table 1. Quantity and distribution of radionuclides in White Oak Lake bed—December 1962

Radionuclides	Depth from surface (in.)										
	0 to 6		6 to 12		12 to 18		18 to 24		Totals		
	Curies	%	Curies	%	Curies	%	Curies	%	Curies	%	
¹⁰⁶ Ru	594 ± 76*	57	276 ± 38	27	112 ± 19	11	56 ± 10				
¹³⁷ Cs	468 ± 20	66	204 ± 25	29	29 ± 12	5	3 ± 1	<1	1038 ± 88	100	
⁶⁰ Co	119 ± 9	78	22 ± 2	15	8 ± 1	5	3 ± 0.5	2	704 ± 35	100	
TRE†	13 ± 3	78	2.5 ± 1.0	15	1.0 ± 0.7	6	0.1 ± 0.1	1	152 ± 9	100	
⁹⁰ Sr	10 ± 0.9	68	3.5 ± 0.5	24	1.0 ± 0.3	7	0.1 ± 0.1	1	16.6 ± 3.3	100	
									14.6 ± 1.1	100	

* Numbers following ± are estimated standard errors.

† Trivalent rare earths exclusive of ⁹⁰Y.

areas of the surface flow of waste water from the two streams (see Fig. 3). The areas of highest ruthenium concentration are also the regions of most frequent flow. The zones of ruthenium contamination do not extend to the creek, except where surface drainage from the contaminated area enters White Oak Creek. The same pattern of ruthenium distribution exists at the 6- to 12-in. and deeper soil depths as that shown for the 0- to 6-in. soil section, but the ruthenium on the soil is much less at the greater depth than in the surface layer. This suggests that upon entering the lake bed soil most of the ruthenium is sorbed on the upper few inches of the soil, while the unsorbed ruthenium migrates slowly underground. Since there is no appreciable lateral spread in the ruthenium-contaminated zone at depth, it is concluded that the lateral movement of ruthenium through the soil is so slow that radioactive decay reduces the concentration of that which reaches White Oak Creek by subsurface water flow to insignificant levels. These findings confirm the results of an earlier study of the movement of ruthenium in the lake bed.⁽¹⁾

Cesium-137

Nearly all of the ¹³⁷Cs in the lake bed is associated with the lacustrine sediment. The sediment layer is usually thinnest near the shore line and thickest toward the center part of the lake bed, and ¹³⁷Cs follows the same distribution. The close association of ¹³⁷Cs with the sediment layer is illustrated in Fig. 4 which shows the concentration of cesium in various core samples. It is observed that only very small quantities of cesium have migrated from the sediment layer into the underlying alluvium. Also, there is apparently no relationship between sediment

thickness and cesium concentration. Finally, it is observed that the concentration of cesium in the sediments varies greatly in some cores with depth, while in others it is relatively uniform. This is probably the result of variations in the cesium content of the sediment deposited.

In order to define further the distribution of cesium in the sediment, the size fraction of several samples of the material from various parts of the lake bed was determined, using JACKSON'S method.⁽⁶⁾ Each fraction was analyzed to determine the quantity of cesium associated with it. Only trace quantities of cesium were removed from the sediment during the separations. The quantities of cesium associated with the various particle-size groups of a typical sediment sample are shown in Table 2. The clay fraction of the sediment comprised only about 35 per cent of the weight of the material, but more than 80 per cent of the cesium was associated with it. In contrast the silt fraction, which constitutes more than 60 per cent of the weight of the sediment, was found to sorb only about 15 per cent of the cesium. Only a trace of cesium was associated with the sand fraction which made up less than 5 per cent of the total weight of the sediment. It is obvious that the association of cesium with the sediment is due to the high affinity of the clay minerals for cesium sorption. Through X-ray diffraction techniques, it has been found that the clay fraction of the sediment is predominantly illite.⁽⁷⁾

The accumulation of ¹³⁷Cs in the sediments of White Oak Lake illustrates clearly the effectiveness of a quiescent body of water in concentrating radionuclides in bottom sediments when the radionuclides are associated with

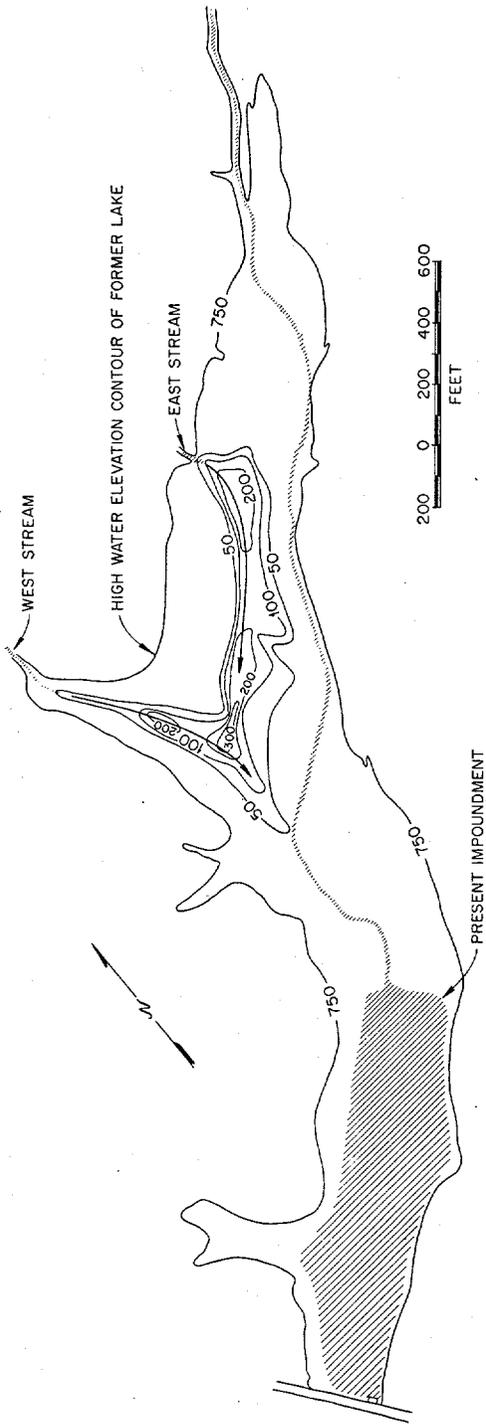
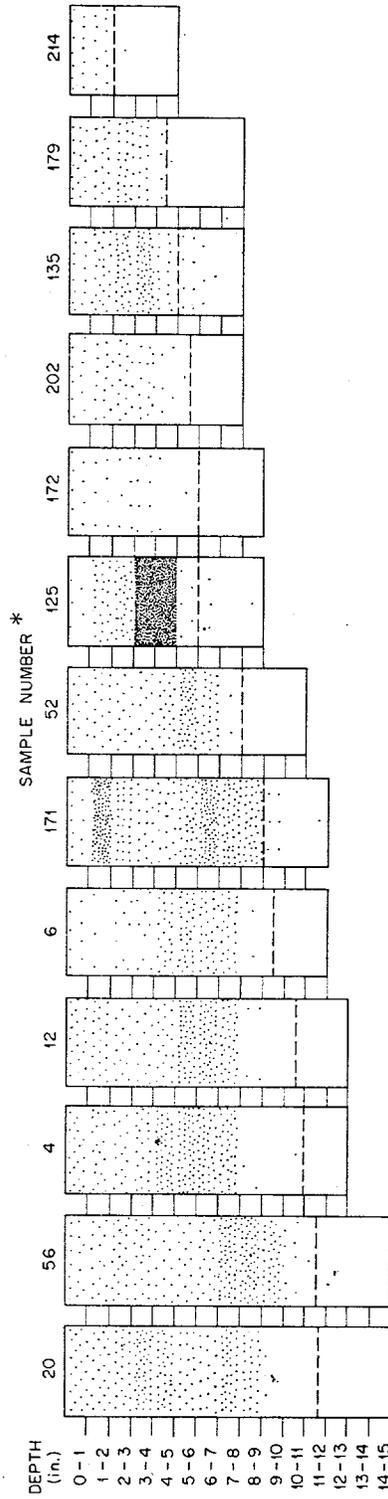


Fig. 3. White Oak Lake bed, showing ^{106}Ru concentrations in $10^{-3} \mu\text{c/g}$ dry weight for 0- to 6-in. layer.



1 dot per 1 in. depth = $1000 \mu\text{c/g}$.
 --- Contact between recent lacustrine sediment and alluvium.
 Fig. 4. Concentrations of ^{137}Cs in selected White Oak Lake bed core samples.

* Low numbers refer to sample locations in lower portion of lake bed with progressively higher numbers identifying upstream samples.

suspended sediments. During the period, 1944–1955, which represents the principal sedimentation period and the time of deposition of the 700 c of ^{137}Cs presently in the lake bed, it is estimated that only 600 c of cesium (or less than 50 per cent of that discharged to surface streams at ORNL during this time) passed White Oak Dam into the Clinch River.⁽⁸⁾

Cobalt-60

Cobalt-60, the third most abundant radionuclide in the lake bed, occurs throughout the lake bed but is most concentrated in the relatively small part of the upper bed that is contaminated by seepage from the ORNL intermediate-level radioactive liquid waste pits.

Table 2. Particle-size analyses and distribution of cesium-137 in lacustrine sediment

	Size (μ)	Weight (%)	Cesium sorbed (%)
Sand	>50	2	Trace
Silt	50–2	63	16
Clay	<2	35	84

Only a small portion of the cobalt presently in the lake bed was accumulated during the deposition of the lacustrine sediments. This activity is for the most part, still associated with these sediments and is widely distributed over the lake bed. Like ^{137}Cs , the greatest quantities of the cobalt associated with lacustrine sediments occur in those areas where the sediment layer is thickest. However 75–80 per cent of the cobalt in the lake bed lies along the paths of flow of the two streams that drain the waste pits. Most of the cobalt, like the ruthenium in this contaminated zone, is confined to the upper layers of soil, and only small quantities have migrated vertically or laterally through the soil.

Strontium-90 and the rare earths

Strontium-90, the least plentiful of the radionuclides detected in the lake bed, contributes less than 1 per cent of the total activity. Small quantities of strontium are found throughout the lake bed; however, the greatest concentrations occur in the presently inundated lower part of the site (see Fig. 5). More than 50 per cent of the total amount of strontium within the lake bed is restricted to this area. Most of

the strontium in the area behind the dam is restricted to the uppermost few inches of the sediment layer, while the quantity of strontium in the deeper sediments is comparatively low and roughly comparable to concentrations in other parts of the lake bed. These conditions provide strong evidence that strontium is presently being removed from lake water and is accumulating in the bottom sediments. The strontium associated with the deeper sediments in the inundated area and that found in other parts of the lake bed represent the remaining portion of the strontium accumulated in the sediments during the time of their deposition.

The quantity of rare earths in the lake bed is only slightly greater than the strontium, and their distribution is, with few exceptions, similar to that of strontium.

RETENTION CHARACTERISTICS OF THE RADIONUCLIDES IN LAKE BED SEDIMENT

To determine the degree of fixation of the several radionuclides by the lacustrine sediments samples of the sediment from several locations in the lake bed were leached with various solutions in the laboratory. The leaching procedure consisted of mixing 5-g portions of the air-dried sediment with 30-ml aliquots of various concentrations of KMnO_4 , NaOH , NaCl , CaCl_2 , HNO_3 , as well as tap water and distilled water. After a contact time of 24 hr the mix was centrifuged and the leachate, as well as the sediment, were radiochemically analyzed to determine the amount of leachable activity. A summary of the average quantities of the particular radionuclides removed from the sediment samples by the leaching agents is presented in Table 3. It is observed that ^{106}Ru is partially soluble in water, but is much more soluble in concentrated acid or base solutions (Table 3). It also appears that there is little or no exchangeable ruthenium in the system, since water removes about the same amount as 1 N salt solutions. The activity leached from the sediment is increased markedly only when the pH of the leaching solution is raised or lowered beyond the limits of percolating ground water in the lake bed, which ranges from pH 6.5 to pH 8.5, or surface water, which varies between pH 7 and pH 8. Thus only a relatively small fraction of the ruthenium in the sediment would

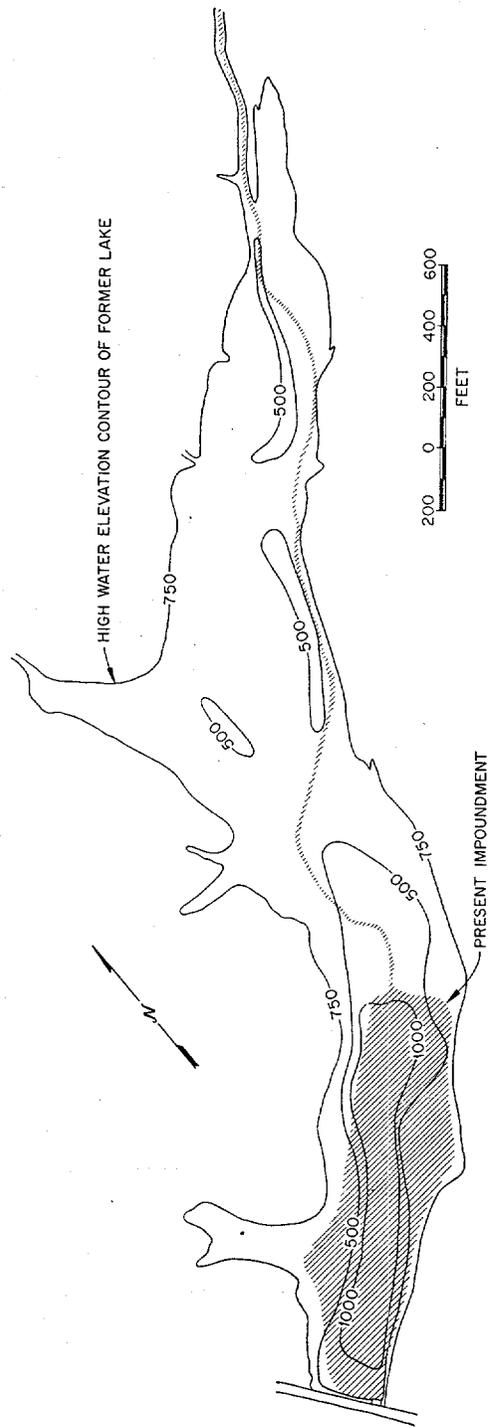


FIG. 5. White Oak Lake bed, showing ^{90}Sr concentrations in $\mu\mu\text{c/g}$ dry weight for 0- to 6-in. layer.

Table 3. Leaching of radionuclides from lake bed sediments*

Solution	Equilibrium (pH)	% Leached				
		¹⁰⁶ Ru	¹³⁷ Cs	⁶⁰ Co	⁹⁰ Sr	TRE†
HNO ₃						
6 N	<0.1	65	19	76	92	70
1 N	<0.1	50	<1	70	86	73
10 ⁻³ N	7.4	10	<1	2	4	7
NaOH						
6 N	11.2	53	<1	23	4	3
1 N	12.1	37	<1	5	1	1
10 ⁻³ N	8.8	16	<1	2	2	1
CaCl ₂						
1 N	6.2	14	<1	1	49	31
10 ⁻³ N	7.5	18	<1	2	4	3
NaCl						
1 N	6.4	7	<1	2	34	48
10 ⁻³	7.3	5	<1	2	7	5
KMnO ₄						
1 N	7.7	24	<1	5	—	—
10 ⁻³ N	7.2	16	<1	2	—	—
Tap H ₂ O‡	7.6	14	<1	2	4	1
Distilled H ₂ O	7.7	14	<1	1	3	1

* Five-gram samples of oven-dried sediment leached with 30 ml of solution indicated.

† Trivalent rare earths exclusive of ⁹⁰Y.

‡ Commonly contains 25–50 ppm calcium bicarbonate.

be expected to move through the soil by downward percolating rainfall or be carried off in solution by surface water, which on occasion floods the area, though some of the sediments might be removed in flood water.

It is apparent that there is little or no easily exchangeable cesium in the sediment, since the only leaching solution to remove more than trace amounts of cesium from the sediment was 6 N HNO₃ which apparently broke down the clay structure. As shown above, most of the cesium is associated with the illitic clay fraction of the sediment. TAMURA and JACOBS⁽⁹⁾ have shown that the mineral *illite* has a high affinity for cesium at concentrations of the same order of magnitude as those sorbed on lake bed soil. Their work also suggests that the marked sorption of cesium by illite is due to the *c*-axis spacing of 10 Å which allows the cesium to be held with a high affinity at exchange sites on the edges of the interlayer spacings. The cesium sorbed on the illitic samples of TAMURA and JACOBS was somewhat easier to desorb

than that observed to be leached by comparable solution on the lake bed sediment. This is believed to be due to the much longer contact time that cesium has had with the lake sediment (as much as 20 years), permitting the cesium to diffuse deeper into the lattice than was the case in laboratory studies of TAMURA and JACOBS in which the contact time was only a few days. This belief is supported by the recent work of JACOBS⁽¹⁰⁾ which shows that the process of cesium sorption by collapsed-lattice clay minerals is controlled by particle diffusion, with cesium loading increasing with time due to slow penetration of the cesium to the interior exchange sites. It is concluded that the cesium in the lake bed would, therefore, be extremely difficult to remove from the sediment by percolating ground water, and only through erosion or scouring of the sediment from the lake bed would any significant quantities of cesium be transported from the area.

The relatively small quantities of ⁶⁰Co leached from the sediment samples by the salt and water

solutions show that ^{60}Co does not exist in an exchangeable or soluble form in any appreciable quantities in the lake bed. High base concentrations are effective in solubilizing about 25 per cent of the cobalt, but high concentrations of acid are much more effective, removing more than 70 per cent of the activity. Since the pH of the leaching solutions that effectively remove cobalt from the lake bed sediment are either much higher or much lower than the pH of the ground and surface waters in the area, it is expected that only a small fraction of the cobalt will be carried in solution on or through the soil. The absence of any appreciable vertical or lateral movement of the cobalt from its points of entry in the soil in the upper part of the lake bed where most of the cobalt occurs confirms this belief.

The leaching of 30–50 per cent of the ^{90}Sr and TRE from the lake bed sediments with salt solutions indicates that as much as one-half of the total quantity of these nuclides in the area probably exists in a rather easily exchangeable form. The exchange is repressed, however, by high concentrations of NaOH. The removal of ^{90}Sr and TRE with the acid leach is much more than that observed by the salt solutions, indicating that another 30–50 per cent of these nuclides are present in the soil in the form of slightly soluble salts of weak acids.

The removal of 3–4 per cent of the ^{90}Sr from the sediment samples by the water solutions indicates that small quantities of strontium are presently being leached by rainfall and ground water from the dry upper lake bed. However, these losses are probably offset by the accumulation of strontium in the presently inundated area.

CONCLUSIONS

Analyses of a series of 250 strategically located core samples has revealed that the bed of former White Oak Lake contains 1038 c of ^{106}Ru , 704 c of ^{137}Cs , 152 c of ^{60}Co , 17 c of the rare earths (exclusive of ^{90}Y) and 15 c of ^{90}Sr . More than half of the activity was found in the 0- to 6-in. layer of soil, while progressively smaller quantities were detected at the 6- to 12-in., 12- to 18-in. and 18- to 24-in. layers of soil.

Practically all of the ^{106}Ru and a large part

of the ^{60}Co are confined to a small part of the upper lake bed that receives surface flow from the nearby radioactive waste seepage pits. The ^{106}Ru in the lake bed is partially water soluble; however, its rate of movement through the soil is slow enough so that radioactive decay reduces the concentration of that reaching surface streams to insignificant levels. Most of the cesium occupies highly selective exchange sites on the illitic fraction of the clay in the sediment and can be desorbed only by disruption of the lattice structure. Only a small fraction of the ^{60}Co in the soil was found to be exchangeable. It is unlikely that any large fraction of the ^{137}Cs or ^{60}Co would move from the area except through erosion of the sediment. About one-half of the ^{90}Sr and the rare earths in the sediment appears to be exchangeable, while the other half is in the form of slightly soluble salts. Through leaching by ground water, a slow depletion of strontium from the dry part of the lake bed occurs; however, it appears that strontium is presently accumulating in the now inundated area.

Because of the short half life of ^{106}Ru (1 year), the relatively small quantities of ^{60}Co , ^{90}Sr and the rare earths, and the tightly bound nature of ^{137}Cs , the presence of these radionuclides in the sediments of White Oak Lake does not appear to constitute any long-term hazard. However, ^{137}Cs , even though it is firmly fixed in the clay fraction of the sediment, will for many years remain as a possible source of contamination to the Clinch River that could, through large-scale erosion of the area, add significantly to the concentration of cesium in the bottom sediments of the Clinch River.

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