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DETAILED ASSESSMENT OF SOLID AND LIQUID WASTE SYSTEMS - HAZARDS EVALUATION, VOL. 4

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ABSTRACT

The ORNL waste disposal facilities and operations, particularly the ground disposal operations, are described and their effectiveness is assessed in terms of fission product movement out of White Oak Creek drainage area into the Clinch River. Where repeated measurements of fission product concentration in biota and in the physical environment have been made, and where these measurements have been substantiated experimentally, an attempt is made to relate these data to the stability of a sizable inventory of radio-nuclides in seepage pits, burial grounds, and the bed of former White Oak Lake.

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DETAILED ASSESSMENT OF SOLID AND LIQUID WASTE SYSTEMS

1.0 MONITORING OF RADIOACTIVE WASTE WATER RELEASED TO WHITE OAK CREEK
AND CLINCH RIVER

Virtually all radioactivity reaching the Clinch River comes through White Oak Creek (Fig. 1). The creek constitutes a direct continuation of the process waste water flow. It also receives contamination released by seepage from the liquid waste pits and the burial grounds. In addition, drainage from the various reactor facilities located in Melton Valley as well as cooling water from the LITR and ORR and the laundry waste are directly discharged to streams leading to White Oak Creek. Erosion of White Oak Lake bed and the Intermediate Pond also contributes activity to the creek.

The original monitoring system was instituted with the primary objective of determining that the concentration of mixed fission products in the Clinch River at downstream points of use be maintained below the MPC values recommended by NCRP and ICRP. This objective is being met.* However, the original monitoring system, even with subsequent improvements, was not designed for a detailed study of the fate of radionuclides released to the creek.

In 1943 the Graphite Reactor went critical, and the separations pilot plant associated with it started operations. The same year the dam was constructed across White Oak Creek, and the following year the settling basin went into operation. At that time most of the activity released to White Oak Creek went through the settling basin. In recent years other sources such as seepage from the waste pits and the fact that the more radioactive process wastes have been diverted from the basin to the Process Waste Treatment Plant have changed the picture, so that, in terms of gross activity, the settling basin is no longer the only large contributor.

1.1 Sampling

Prior to 1950 the settling basin was grab-sampled periodically. From 1948 to 1950 the sampling period was 4 hr,** and in-flow measurements were made by means of v-notch weirs. In November 1950 a v-notch weir, continuous water-stage recorder, and proportional sampler were installed at the basin outlet.

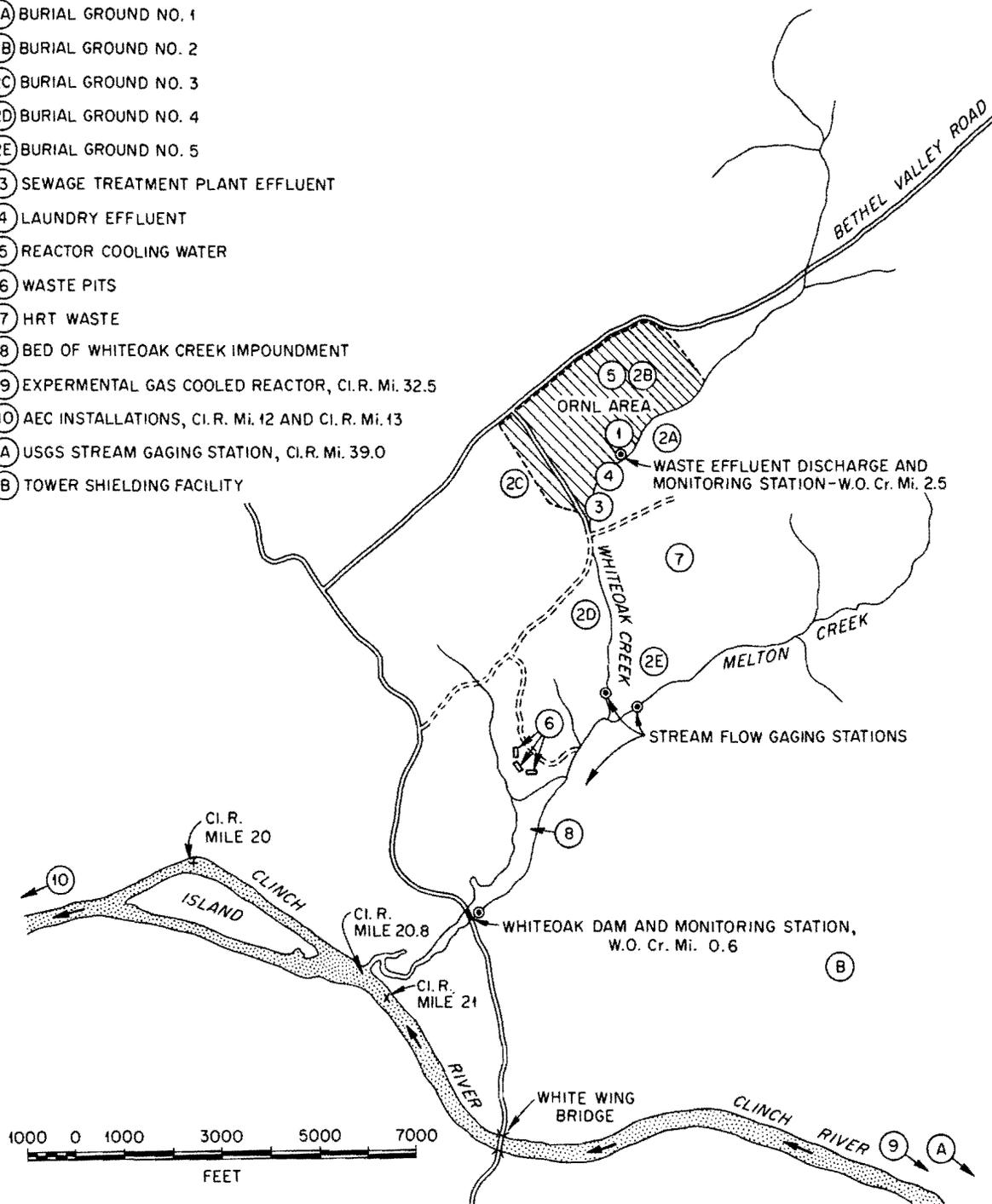
The process waste is continuously monitored for gross activity and the Process Waste Treatment Plant and settling basin effluents are proportionally sampled. Most of the other contributing sources are grab-sampled only, and it is not possible to arrive at accurate figures for the total in-flow of a particular radionuclide to White Oak Creek.

Prior to draining White Oak Lake (September 1955), a gaging station at the dam was used for flow measurement. After the lake was drained, the flow

* F. N. Browder, "Radioactive Waste Management at ORNL," ORNL-2601, 1959.

**H. H. Abee, "Liquid Waste Monitoring Summary Techniques and Data, 1948-1957" (Unpublished), 1958.

- ① PROCESS WASTE TREATMENT PLANT
- ②A BURIAL GROUND NO. 1
- ②B BURIAL GROUND NO. 2
- ②C BURIAL GROUND NO. 3
- ②D BURIAL GROUND NO. 4
- ②E BURIAL GROUND NO. 5
- ③ SEWAGE TREATMENT PLANT EFFLUENT
- ④ LAUNDRY EFFLUENT
- ⑤ REACTOR COOLING WATER
- ⑥ WASTE PITS
- ⑦ HRT WASTE
- ⑧ BED OF WHITEOAK CREEK IMPOUNDMENT
- ⑨ EXPERIMENTAL GAS COOLED REACTOR, Cl. R. Mi. 32.5
- ⑩ AEC INSTALLATIONS, Cl. R. Mi. 12 AND Cl. R. Mi. 13
- Ⓐ USGS STREAM GAGING STATION, Cl. R. Mi. 39.0
- Ⓑ TOWER SHIELDING FACILITY



Sources of Radioactive Contamination in the ORNL Environment

Fig. 1.

across the dam was based on the summation of separate measurements of flow in White Oak Creek and Melton Branch, the principal contributors of waste water and surface drainage. A correction factor of 1.16 was applied during precipitation periods to compensate for the runoff associated with the area between the gaging points and the dam. Prior to June 1958, White Oak Creek was sampled by taking an 8-gal grab sample at White Oak Dam once a day. During June 1958, a continuous sampler (nonproportional) and a scintillation probe and remote recorder were installed at the dam. Construction work is now underway to raise the elevation of the spillway, and a proportional sampler will be installed upon completion of the new spillway.

1.2 Analyses

Analyses of water samples include gross beta determinations and chemical separation and counting of radioactive strontium, cerium, ruthenium, trivalent rare earths, cesium, zirconium, niobium, barium, and cobalt. The gross beta activity in microcuries per milliliter is calculated from the net counts per minute. The total beta curies released is then calculated from volume measurements. A Pa^{234} standard is used for calibrating the counting equipment. The concentration of specific radionuclides may be approximated as a ratio of the counting rate of the particular radionuclide to the gross beta activity multiplied by the total beta curies. Some error is inherent in this procedure, since the beta energies of the various radionuclides differ and counting efficiencies must also differ.

1.3 Conclusions

Daily gross beta determinations of the settling basin effluent fluctuate from 30 to 3000 c/m/ml; the corresponding values for White Oak Dam lie between 2 and 200 c/m/ml. These fluctuations introduce a high degree of uncertainty in results obtained by the grab-sampling method.

A constant amount of each daily grab sample (140 ml) is included in the monthly composite sample. This monthly sample represents the average monthly concentration of activity in the 8-gal grab samples, not the average monthly activity released. Since neither the grab-sampling prior to June 1958 nor the continuous sampling thereafter takes into account the daily flows, sampling at the dam discriminates against days of higher flow. Comparatively large amounts of activity may be discharged to the river during floods, although the average concentration during such periods might well be below the average concentration during periods of normal flow.

The curies of activity released per year for the purpose of this report is derived by multiplying the yearly amounts of gross beta activity discharged by the percentage of activity for the particular isotope found in the monthly composite. This calculation is inaccurate because the daily samples are not aliquoted in volumes proportional to the daily flow. Based on the 1956-58 data from White Oak Dam, the probable error at the 95% confidence level for strontium released per year is 28%, neglecting sampling error.

2.0 LIQUID WASTE PITS*

Pits dug in the earth to receive liquid radioactive waste for disposal may function either as storage basins or as ion exchange columns, as explained by Struxness, Morton, and Straub when they reported on the early experience with waste disposal pits at Oak Ridge.** The concept of the pit as a tank has been largely abandoned, for few if any earth materials are sufficiently impermeable. The three waste disposal pits now in use at Oak Ridge are of value not for their storage capacity, but for the ability of the soil around them to trap and hold the radioactive constituents in the waste as it seeps out. The total amount of waste that such pits can handle and the safety of their operation depend on how good an ion exchange column is provided by the earth around the pits. Determination of the efficiency is difficult because detailed knowledge is required of the hydraulic and chemical properties of an underground mass of rock which, in most cases, is far from homogeneous.

The waste pits at Oak Ridge have advantages that they provide secure storage, for the waste cannot be released suddenly from the clay as it could be from a ruptured tank and the cost is much less than for tank storage. The disadvantage is the inability to determine just what is happening to the waste or how its component elements may disperse. For this reason there is some question in the minds of most of the members of the Committee on Waste Disposal of the National Academy of Sciences as to the long-term safety of this disposal method, and they specifically recommend that continuing disposal of large volumes of low-level waste in the vadose zone, above the water table, be of limited application because it probably involves unacceptable long-term risks.***

2.1 Present Operating Procedures

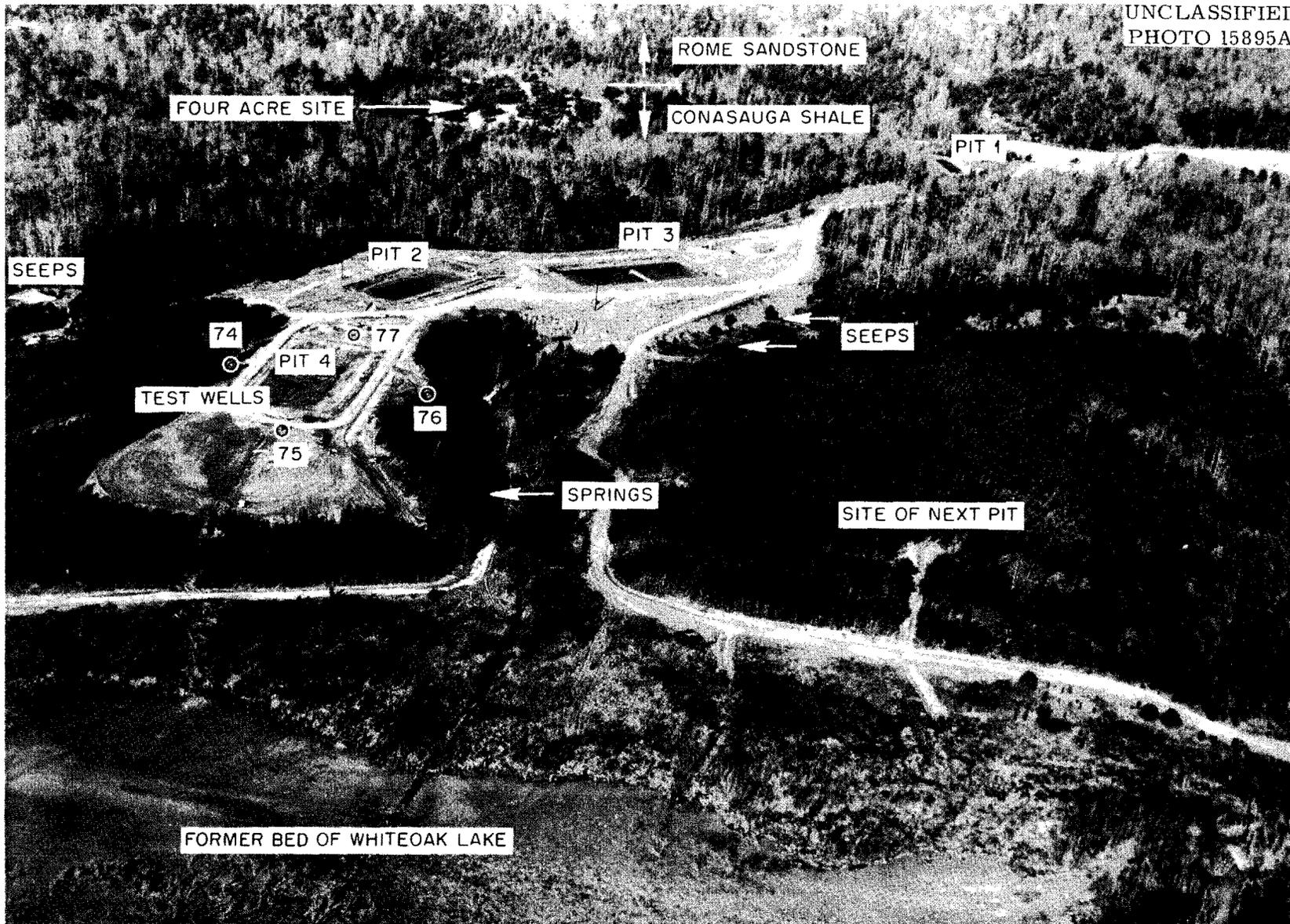
At present there are three pits in service. They are arranged end to end on the crest of a low ridge and are operated as a unit (Fig. 2). Each pit is roughly 200 ft long, 100 ft wide, and 15 ft deep and has a nominal capacity at the 12-ft stage of 1,000,000 gal. Table 1 shows the volumes of waste that have been pumped to each of the pits, and Table 2 shows the total amounts of the more important radionuclides. At present, every 10 days or so a batch of roughly 80,000 gal of waste is pumped to pit 3. Enough is retained in pit 3, which leaks very slowly, to maintain its stage at about 12 ft; the balance is overflowed into pit 2. Pit 2 also is maintained at about the 12-ft stage, and the

* W. de Laguna, K. E. Cowser, and F. L. Parker, "Disposal of High-Level Radioactive Liquid Wastes in Terrestrial Pits; a Sequel," Peaceful Uses of Atomic Energy, Paper 2351, Vol. 18, p. 101, New York (1956).

** E. G. Struxness, R. J. Morton, and C. P. Straub, "Disposal of Radioactive Wastes into Terrestrial Pits," *ibid.*, Paper 140, Vol. 9, p. 684.

*** "The Disposal of Radioactive Waste on Land," report of the Committee on Waste Disposal of the Division of Earth Sciences, Natl. Acad. Sci., National Research Council, Publ. 519 (1957).

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Aerial View of Waste Pits.

Fig. 2.

Table 1. Volumes of Waste Discharged to Pits

Year	Volume, gal			
	Pit 2	Pit 3	Pit 4	
1952	42,900	-	-	
1953	235,100	-	-	
1954	909,600	-	-	
1955	639,000	1,035,000	-	
1956	1,270,300	201,400	1,307,500	
1957	<u>797,500</u>	<u>-577,900</u>	<u>2,682,000</u>	
Total	3,894,400	658,500	3,989,500	8,542,400

Table 2. Amounts of Radioactive Materials Discharged to Pits

Radioactive Materials	Amount, curies
Ru ¹⁰⁶ and Rh ¹⁰⁶	20,000
Cs ¹³⁷ and Ba ¹³⁷	75,000
Sr ⁹⁰ and Y ⁹⁰	2,000
Others	<u>3,000</u>
Total	100,000

balance, if any, is overflowed into pit 4. In summer, when evaporation is high, pits 3 and 2 can dispose of all the waste. In winter some must be run into pit 4 (Fig. 3). Pits 3 and 2 are kept filled because their shale sides have adsorbed much cesium, and when they are exposed the radiation field in the area increases markedly. Once, when the stage of pit 3 was lowered 3 ft, the radiation field at the edge increased to 5 r/hr. Generally it is much less. The radiation field 100 ft back from the pit is commonly of the order of 25 to 50 mr/hr.

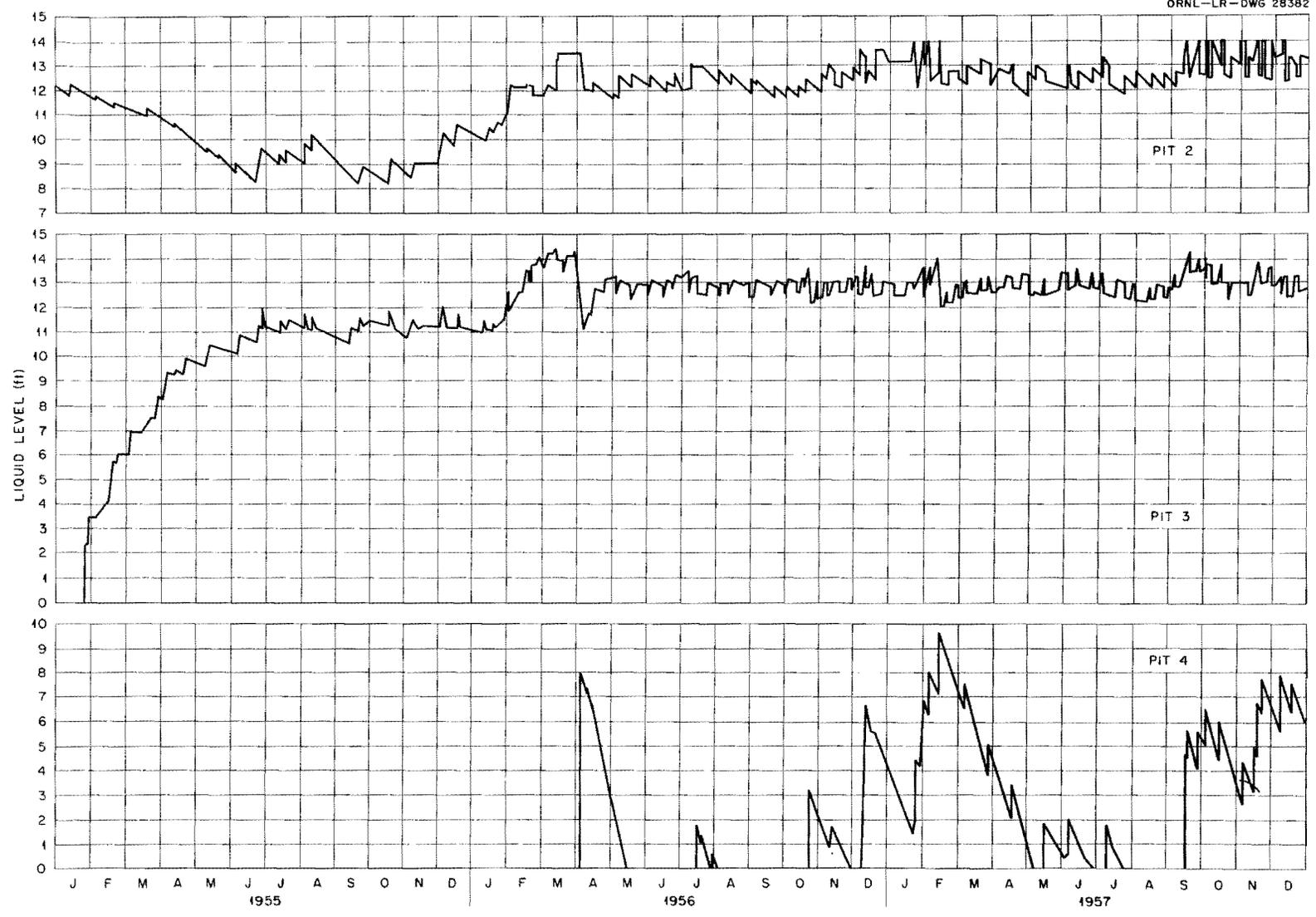
Pit 4 is generally dry in summer and is only partly filled in winter. The waste which has so far reached pit 4 has been relatively low in cesium-137, partly because of dilution by rain water in pits 3 and 2, and partly because of adsorption onto the walls of the first pits. With time, however, the radiation field around pit 4 has also increased to the point that a serious problem has developed. At present working time in the pit area must be limited to a few hours per week, but this depends greatly on location.

To understand how the pits are working and to predict possible future developments, a budget must be maintained to determine the distribution and destination of the various components in the waste. Three budgets are desirable: liquid, chemical, and radiochemical. The liquid budgets for pits 2 and 3 must consider: (a) waste pumped or transferred to the pit, (b) waste transferred to the next pit, (c) changes in volume in storage, (d) gains from rainfall, (e) losses from evaporation, and (f) losses from seepage. The first four items can be measured directly, although the volumes transferred between pits are not so accurately known as we would wish. The evaporation loss from a small lake is seldom easy to determine,* but the problem is harder still in a pit filled with radioactive liquid because of the difficulty of installing and operating the required instrumentation. Therefore it has been necessary to estimate evaporation from the observed losses from several evaporation pans. Evaporation was also calculated from a semiempirical formula developed by Kohler.** Despite the active co-operation of the local United States Weather Bureau, determinations of evaporation have never been entirely satisfactory. Seepage, which is the most important factor in the liquid budget, cannot be measured directly for pits 2 and 3, and must be calculated as the difference between the other items. Pit 4 leaks so rapidly that other factors may be neglected.

During the first few months of operation, the bottom of a new pit becomes nearly sealed with mud and sludge which settle out of the waste. After this, the permeability of the shale around the pits does not appear to change, and one must infer that there is little entrainment of silt, swelling of the clay, or leaching of carbonate from the shale by the moving waste. The seepage rates

* W. B. Langbein, C. H. Hains, and R. C. Culler, "Hydrology of Stock-Water Reservoirs in Arizona - Progress Report," U. S. Geol. Survey Circ. 110, Washington, D. C. (1951).

** M. A. Kohler, T. J. Nordenson, and W. E. Fox, "Evaporation from Pans and Lakes," U. S. Weather Bureau Research Paper No. 38 (1955).



Liquid Levels in Waste Pits.

Fig. 3.

are 15 to 20% higher in summer than in winter, probably due to changes in viscosity of the waste with temperature.

Some progress has been made in maintaining a budget or materials balance for the stable chemical and radioactive nuclides in the waste, but a complete accounting is not yet available. Data from several cores drilled near the pits are now being analyzed and should assist in defining the extent of movement of strontium and cesium. However, collection of numerous cores is not practical because of the radiation exposure hazard. The total amounts of the various waste components going into the pits are fairly well known, and sampling at White Oak Dam, past which flows the drainage from the pits, gives a measure of the amounts leaving the controlled area; but it does not now appear possible to give in detail the distribution of the materials retained in and around the pit system.

Table 3 shows the liquid budgets for pits 2 and 3 for 1956, a typical year.

Pit 2 lost 1,450,000 gal by seepage, or an average of 3900 gal/day, but because rainfall exceeded evaporation by about 450,000 gal the net liquid loss was only 950,000 gal. Pit 3 lost about 375,000 gal by seepage, or roughly 1000 gal/day. However, for pit 3, the rainfall increment exceeded evaporation losses by about 375,000 gal, so there was no net liquid loss or gain for pit 3. This pit serves a useful purpose, for the liquid that seeps away carries some radioactive materials out into the shale, and more are adsorbed on the shale sides of the pit; rain water dilutes the remainder.

Table 3. Liquid Budgets, Pits 2 and 3, 1956

	Volume, gal	
	Pit 2	Pit 3
Losses		
Pumped out	1,300,000	2,600,000
Evaporated	250,000	350,000
Change in storage	<u>300,000</u>	<u>250,000</u>
Gross loss	1,850,000	3,200,000
Gains		
Pumped in	2,600,000	2,850,000
Rainfall	<u>700,000</u>	<u>725,000</u>
Gross gain	3,300,000	3,575,000
Less gross loss	<u>-1,850,000</u>	<u>-3,200,000</u>
Seepage	1,450,000	375,000

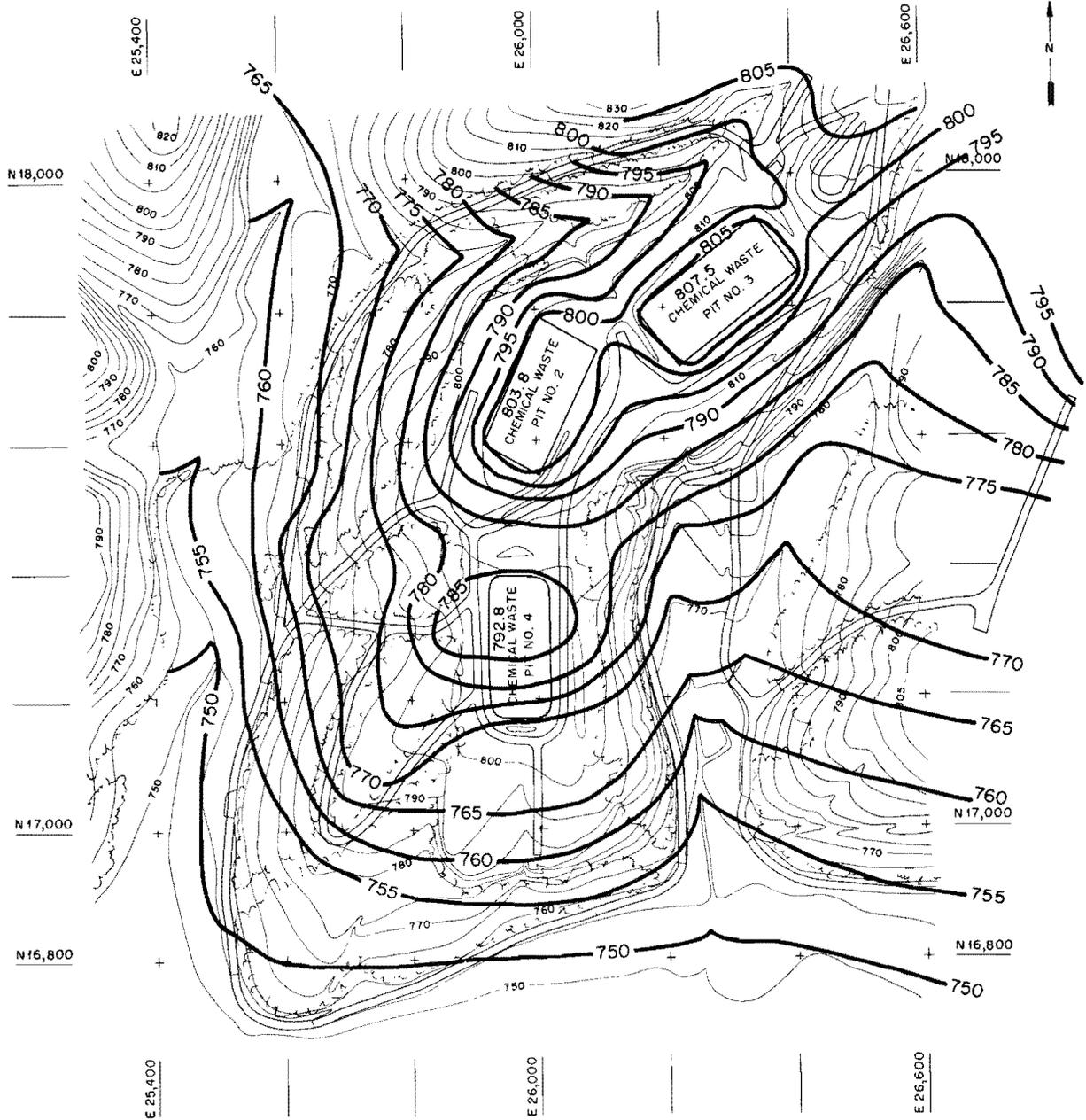
2.2 Hydrologic Characteristics of the Operating Pits

Figure 4 shows the water table on January 10, 1958. For more than a year prior to this time the waste levels in pits 2 and 3 had been maintained at close to the 13-ft stage, and the water table had stabilized around the pits (Fig. 3). Pit 4 was put into service in April 1956, and by the end of 1957 it had received more than 4,000,000 gal of waste. The water table around pit 4 is never stable. Waste is run into the pit in batches, usually during the winter, and it is impossible to maintain a fixed liquid level in the pit. When the level is fairly high the waste leaks out rapidly; 20,000 gal/day at the 8-ft stage. When the stage falls the rate decreases. At the same time the water table rises under and around the pit, water-table gradients are steepened, and the rate of movement of liquid out from the pit area increases (Fig. 5). When the pit stage falls and the leakage rate from the pit decreases, the water table rises less rapidly until eventually an equilibrium is reached. When the pit is empty the water table falls until more liquid is added to the pit. The water-table map of Jan. 10, 1958, was made at a time when there had been liquid in pit 4 for some time, and the water table mound under the pit was approximately at a maximum. The stage of pit 4 at this time was 4.6 ft, and the pit was leaking at a rate of about 16,500 gal/day.

Assuming that all three pits were roughly in equilibrium with the water table around them, it is possible to estimate the permeability of the weathered shale through which the bulk of the waste is moving. The waste is moving out from pit 4 almost entirely along the strike east and west; there is certainly no movement to the north, if only because of the water-table gradients outward from pit 2. The very small amounts of activity found in the observation wells to the south show that no important volume of liquid is moving in this direction. Because the pit was not full, the effective length of the pit was roughly 150 ft. The thickness of saturated weathered shale is roughly 30 ft, and the water-table gradient in the direction of movement about 10 ft in 100. This gives a permeability (Meinzer's units) of 18 gal/day/ft² at unit gradient (300 ft x 30 ft = 9000 ft²; 16,500 gal/day at gradient 0.1 = 165,000 gal/day at unit gradient; 165,000 gal/day/9000 ft² = 18 gal/day at unit gradient).

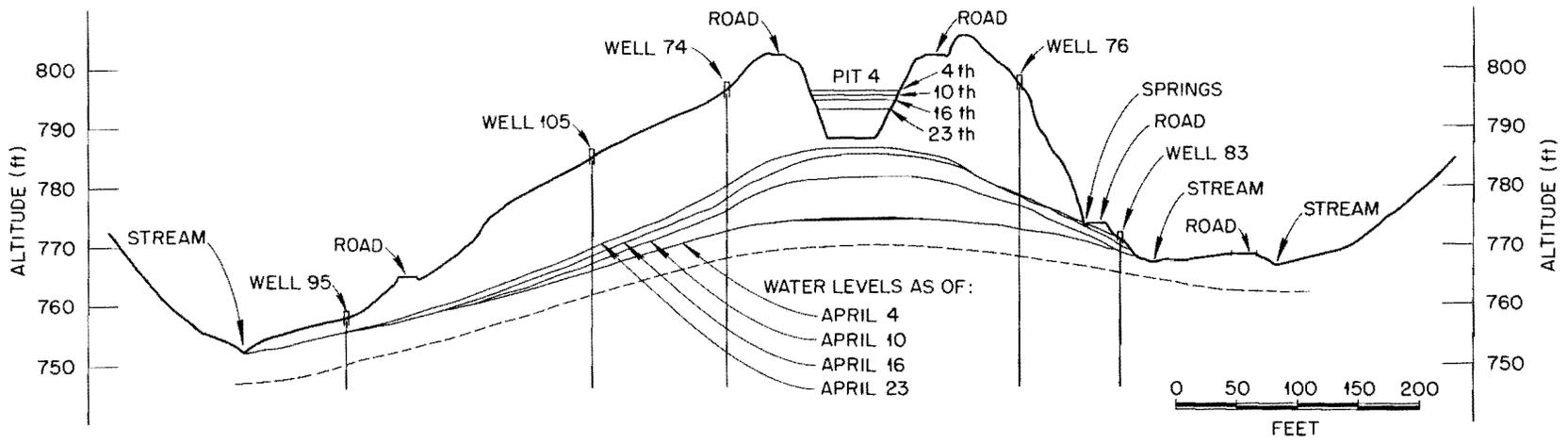
Pit 2 is leaking to the west along a zone about 200 ft wide; to the east it is partly blocked at pit 3, so that the zone of leakage is roughly 100 ft wide. Assuming the same depth to bedrock, a water-table gradient of 15 ft in 100, and leakage rate of 3900 gal/day, the permeability of the weathered shale is 2.9 gal/day in Meinzer's units. The shape of the ridge forced pit 3 to be oriented more nearly parallel to the bedding than is desirable; consequently only 150 ft of bedding is intersected, and much of the potential zone of leakage to the west is blocked by pit 2. Assuming that the leakage zone has a total length of 200 ft, a depth to bedrock of 30 ft, a gradient of 15 ft in 100, and a rate of leakage of 100 gal/day, the permeability of the weathered shale around pit 3 is 1.1 gal/day in Meinzer's units.

Calculation of these values should not suggest that the hydrologic characteristics of the pits or of the shale around them are known. We do not know the thickness of weathered shale through which the waste is moving, and 30 ft may be too large a value, particularly for points a little distance



Water Table Contour Map - January 10, 1958.

Fig. 4.



Cross Section, Pit 4.

Fig. 5.

from the pits. We do not know that movement through the fresh shale below is quantitatively unimportant. Until these questions can be answered it will be impossible to determine the volume or the geometry of the ion exchange column represented by the shale around the pits.

Two small streams flow past the low ridge on which the three pits are located, one on the east, one on the west. The liquid that seeps out of the pits travels underground for 150 to 600 ft; then it seeps up through the beds of the streams and joins their flow. Some of the waste appears at the surface in the form of small surface seeps near the edges of the streams, and some, when pit 4 has waste in it, appears in a line of active springs in the gutter of the road at the foot of the bank to the east of the pit.

None of the waste can move to the north from the pits, and none can pass the streams to the east and west. To the south the two streams drain into the former basin, at White Oak Lake, now dry, out of which the water flows through a weir and sampling point. The liquid escaping from the pits will not therefore inadvertently become a hazard even if details of its movement are unknown.

2.3 Dispersion of Stable Chemical Waste Constituents

Waste that has been pumped out to the pits has come from several different sources and, during different periods, has been pretreated in a variety of ways. Before going to the pits most of the waste is neutralized and stored in tanks. Sludges, which are largely hydroxides and carbonates, form in the tanks and carry down with them most of the radioactive strontium and barium and minor amounts of some of the other fission products. Some of the sludge is carried out to the pits with the liquid waste, and still more sludge appears to have formed in the pits. Because of the several origins and complex history, there have been many variations in the composition of the waste going to the pits; Table 4 gives the average concentration of the chemical ions in waste that has gone into each of the three pits since July 1954.

Table 4. Chemical Composition of Waste Sent to Pits

Pit	Amount, ppm							Total Solids
	Na	NH ₄	Al	NO ₃	SO ₄	Cl	OH	
3	15,600	400	600	23,400	3,400	200	3,500	55,800
2	13,600	100	400	20,300	2,700	200	2,500	47,300
4	11,200	100	300	18,600	2,900	100	1,800	40,700

By July 1, 1957, a total of about 1,800,000 lb of nitrate and 1,400,000 lb of sodium had gone into the three pits. There is about a 10% decrease in nitrate concentration from pit 3 to pit 2, and again from pit 2 to pit 4; this represents loss of nitrate by seepage into the shale and dilution by rainfall. The nitrate is not held in the soil, and virtually all of it appears in the two small streams that flow into the Clinch River. The surface seep 600 ft west of pit 2 contains about 1800 ppm of nitrate, roughly a 10-fold decrease in concentration of nitrate from that of the liquid in the pit. Springs 150 ft to the east of pit 4 show an approximate 2- to 3-fold decrease in nitrate concentration. The dilution factors vary with season of the year and with the weather; this makes an exact inventory very difficult. If all the nitrate discharged to the pits reaches the Clinch River, the concentration in that stream, assuming complete mixing, would be 0.036 ppm (nitrate, 360,000 lb/year; Clinch River flow, 5000 ft³/sec, or 10¹³ lb/year). This is about 0.1% of the maximum permissible concentration. The movement and dilution of the nitrate that is not adsorbed by the soil may prove a valuable guide, when more data are available, to what is happening to the radioactive constituents in the waste.

The sodium, like the nitrate, is decreased about 10% in concentration by dilution as the waste moves from pit 3 to pit 2, and again from pit 2 to pit 4. However, in the 600-ft underground journey from pit 2 to the surface seep to the west, the sodium concentration is decreased roughly 50-fold, as against a 10-fold decrease of the nitrate. The water in the seep, however, has much more calcium in it than normal ground water; sodium is apparently being taken up by the shale in exchange for calcium. The possibility has long been recognized that the steep bank that forms the east side of pit 4 (Fig. 5) might collapse if the pit should be nearly full at the time of a heavy rain, even though the bedding in the shale is at right angles to the direction of potential movement. The substitution of sodium for calcium may well weaken the shale mechanically and the replacement reaction is therefore of some interest.

2.4 Well Logging and Sampling

Movement of active material out of the pits in the escaping liquid has been followed by well logging and sampling in some fifty observation wells in the area and by the analysis of samples taken from surface seeps and streams. The well-logging units in use are of two general types. The first is a trailer-mounted count rate meter to which is attached a long cable and a water-tight aluminum casing containing a halogen-type GM tube and a preamplifier. The second is similar but uses scintillation detectors. The GM equipped units are more satisfactory for field use than the scintillation type, particularly in wells in which the activity is fairly high. Although both types of instruments have been calibrated in air and in simulated "wells" made of short sections of pipe inserted in barrels filled with measured amounts of radioactive solution, it has not been possible to convert instrument response at all points in a well into specific activity.* The parameters that determine instrument response at

* K. E. Cowser and F. L. Parker, "Soil Disposal of Radioactive Wastes at ORNL. Criteria and Techniques of Site Selection and Monitoring," Health Physics, Vol. 1, No. 1 (1958).

any point include concentration of activity in solution, well diameter, active material adsorbed on the walls of the well, and active material in fractures extending back into the rock. It is seldom possible to separate these several factors. Nevertheless, well logging is of great value in detecting the arrival of activity in a well and in locating the first points of entry; later the whole well tends to become radioactive, and the record is blurred. Well logging is also of value in indicating the places in a well at which samples should be taken. The map shown in Fig. 6 is based on the results of our sampling.

Despite the care taken with the well sampling and with the analysis of the samples, the locations of the wells themselves had to be chosen arbitrarily and they may not intersect the channels of highest waste concentration. The wells are cased down to firm shale; therefore the samples do not necessarily represent the liquid in the weathered shale which is the important zone of flow. Pumping the monitoring wells, although perhaps desirable in theory, is hardly practical, and in any case would introduce still other complexities that would be difficult to evaluate.

The level of activity in the wells around pits 2 and 3 has been approximately constant for some time, although it is anticipated that eventually the concentrations of the radionuclides will increase. The level of activity in the observation wells around pit 4 fluctuates in response to the amount of waste in the pit and to the rate at which the pit is leaking. The lag in this response, as well as the time required for activity to reach the well after the pit was first filled, gives a measure of the rate of movement of waste out from the pit.

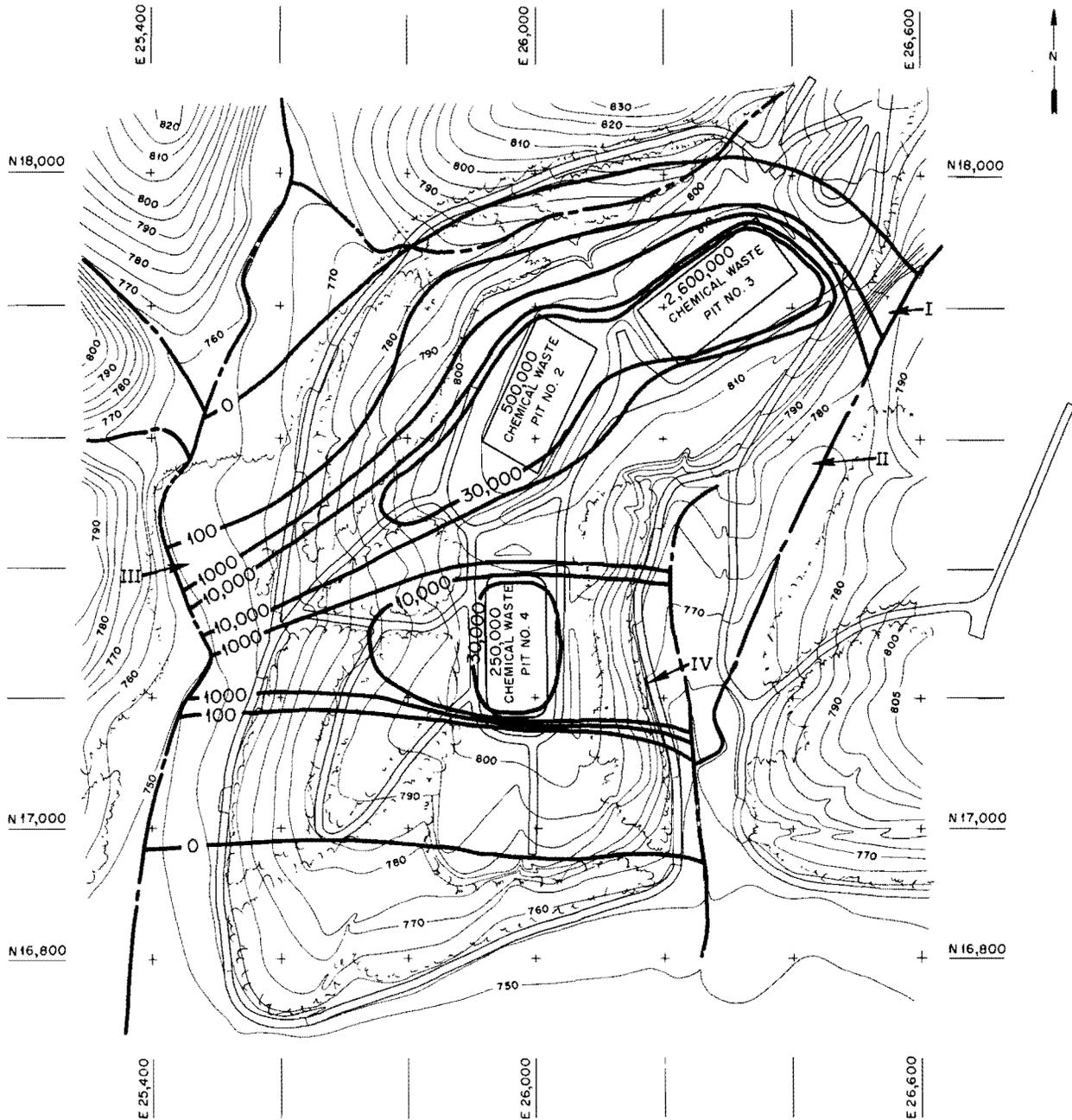
Logging in a sealed hole below pit 4, made by grouting a string of tubing into the central well used in the pumping test, shows that the waste had thoroughly penetrated the shale to a depth of 6 ft below the bottom of the pit within approximately 4 weeks after the first waste was added, and that some 8 weeks later it had reached down as far as 20 ft, to the bottom of the weathered zone. Locally, in some of the observation wells, there is evidence of waste moving in fractures in fresh shale down to a depth of 60 ft and in one well perhaps even to 200 ft, but this is exceptional and quantitatively unimportant. The bulk of the waste, as anticipated, is moving through the weathered shale, and in particular through a zone a few feet thick lying directly on top of the unweathered rock. Not infrequently the first appearance of activity in a well is at the water table.

2.5 Dispersion of Radioactive Waste Constituents

The concentration of radionuclides in the waste, like the concentration of stable chemical constituents, varies from time to time with changes in operation. Table 5 shows the radioactive nuclides in a sample of the waste transferred from pit 3 to pit 2 on Oct. 19, 1956.* The waste pumped into pit 3 has a somewhat higher activity, but the values shown serve as an illustration.

* R. L. Blanchard, B. Kahn, G. G. Robeck, "Laboratory Studies on the Ground Disposal of ORNL Intermediate-Level Liquid Radioactive Waste," ORNL-2475, March (1958).

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I, II, III, IV RADIOACTIVE SEEPS AND SPRINGS
UNITS OF CONCENTRATION = $\mu\mu\text{C}/\text{ml}$

Distribution of Activity; February 1958.

Fig. 6.

Table 5. Radionuclides in Waste Transferred from Pit 3 to Pit 2, Oct. 19, 1956

Radionuclide	Activity μuc/ml
Cesium-137	660,000
Ruthenium-106	94,000
Niobium-95	13,000
Strontium 89 and 90	7,000
Cobalt-60	7,000
Rare earths and yttrium-90	7,000
Antimony-125	4,000
Zirconium-95	1,000

As the waste moves from pit 3 to pit 2, there is a 3-fold decrease in total activity and approximately the same decrease between pits 2 and 4, far too much in each case to be accounted for by dilution. During storage in the pits active material is lost by decay, precipitation, and adsorption on the shale walls.

Waste moves out laterally from pits 2 and 3 along the strike at a rate of about 2 to 6 ft/day. Movement from pit 3 has been largely to the east from the north end of the pit toward the seep marked I on Fig. 6. In all probability a similar tongue of waste is moving to the east from pit 2, or 3, to form the surface seep marked II, but if so it has not been picked up by the monitoring wells. The principal movement out of pit 2 is to the southwest, in a direction somewhat across the strike, suggesting that there are unknown structural or other geologic factors in this area which determine the direction of movement of the waste, or that our present network of observation wells and methods of well logging and sampling do not give a complete and accurate picture of the movement of the waste underground.

Waste moves east and west from pit 4 at a rate of 10 to 30 ft/day and appears in springs along the road at the foot of the ridge in 10 days to two weeks. It was anticipated that pit 4 would leak rapidly to the east and that springs would form. The site was accepted because it would permit the new pit to take overflow from pits 2 and 3, and because the amount of activity involved does not represent a hazard sufficiently great to require the higher cost of the new wells and pipeline necessary to develop a new area.

Very little activity, however, has moved south from pit 4. The south observation well (No. 75), which showed a large change in water level during the pumping test in the area, is only some 30 ft south of the end of the pit. The records are incomplete, but apparently about a year elapsed before activity reached this well, and the concentration is still very low. The activity has appeared at a depth of 50 ft, just below the water table, but not at 110 ft where pressure testing had found a fracture. This corroborates some of the conclusions from the pumping test, namely, that movement from or to this well would be impeded by a hydraulic barrier immediately to the south of it, and that when there was movement it would be at 50-ft, not at the 110-ft level. Other questions need to be answered. For example, well No. 93, some 150 ft south of pit 4, had activity in it only a little over a month after waste was first run into the pit. How it got there, without being found also in well No. 75, is not known.

Ruthenium is not retained completely by the waste-pit system, and consequently some of this material finds its way into the Clinch River after passing through White Oak Creek. Figure 7 shows the fluctuations in concentration and the total transport of radioactivity for 1957 and 1958 in the streams east and west of the pits. Integration of the area under the total transport curve gives an estimated release of Ru^{106} from the pit area to White Oak Creek during 1957 and 1958 of 200 curies and 160 curies, respectively. The volume of liquid lost by seepage from the pits was 4,920,000 gal in 1957 and 4,840,000 gal in 1958. Therefore the average concentration of Ru^{106} in liquid seeping to the streams was 0.011 $\mu\text{c}/\text{ml}$ in 1957 and 0.0087 $\mu\text{c}/\text{ml}$ in 1958. The cumulative average concentration of Ru^{106} in waste released to the pits was 0.47 $\mu\text{c}/\text{ml}$ in 1957 and 0.35 $\mu\text{c}/\text{ml}$ in 1958. Considering the concentration of Ru^{106} in the waste and in the liquid seeping to the streams, the decrease in Ru^{106} due to dilution, decay, and sorption is calculated to have been 97.7% in 1957 and 97.5% in 1958.

With the waste-pit system acting as the primary source of Ru^{106} discharged to the river, it was estimated by Cowser* that the upper limit of Ru^{106} concentration in waste released to the pit system should not exceed 25 $\mu\text{c}/\text{ml}$; this amounts to 350,000 curies of Ru^{106} in 3,700,000 gal of waste each year. Special treatment of the waste to enhance the removal of Ru^{106} was not considered in the calculations. The concentration of ruthenium in waste released to the pits in September 1959 was 3.6 times greater than this estimated upper limit, and a continued discharge at this level could create a potential hazard in the river.

The establishment of the Low-Level Analytical Laboratory in the Analytical Chemistry Division (ORNL) in May 1959 made possible precision analyses of small concentrations of fission products in samples grossly contaminated with Ru^{106} . As a result of more recent work, about 5×10^{-6} $\mu\text{c}/\text{ml}$ of Cs^{137} was detected in one well (well 52 located 50 ft east of pit 2). Underground movement of Sr^{90} has been somewhat more extensive. Wells within 50 ft of pits 2 and 3 (wells 52, 54, 55, and 56) have shown concentrations of Sr^{90} up to 2×10^{-5} $\mu\text{c}/\text{ml}$. Beyond these points and adjacent to pit 4, Sr^{90} has been detected in some wells but generally in concentrations less than 10^{-6} $\mu\text{c}/\text{ml}$. Also, when Sr^{90} was detected in the streams east and west of the pits, it was in concentrations less than

* K. E. Cowser, "Potential Hazard of Ruthenium in the ORNL Waste-Pit System," ORNL-CF-60-3-93 (March 1960).

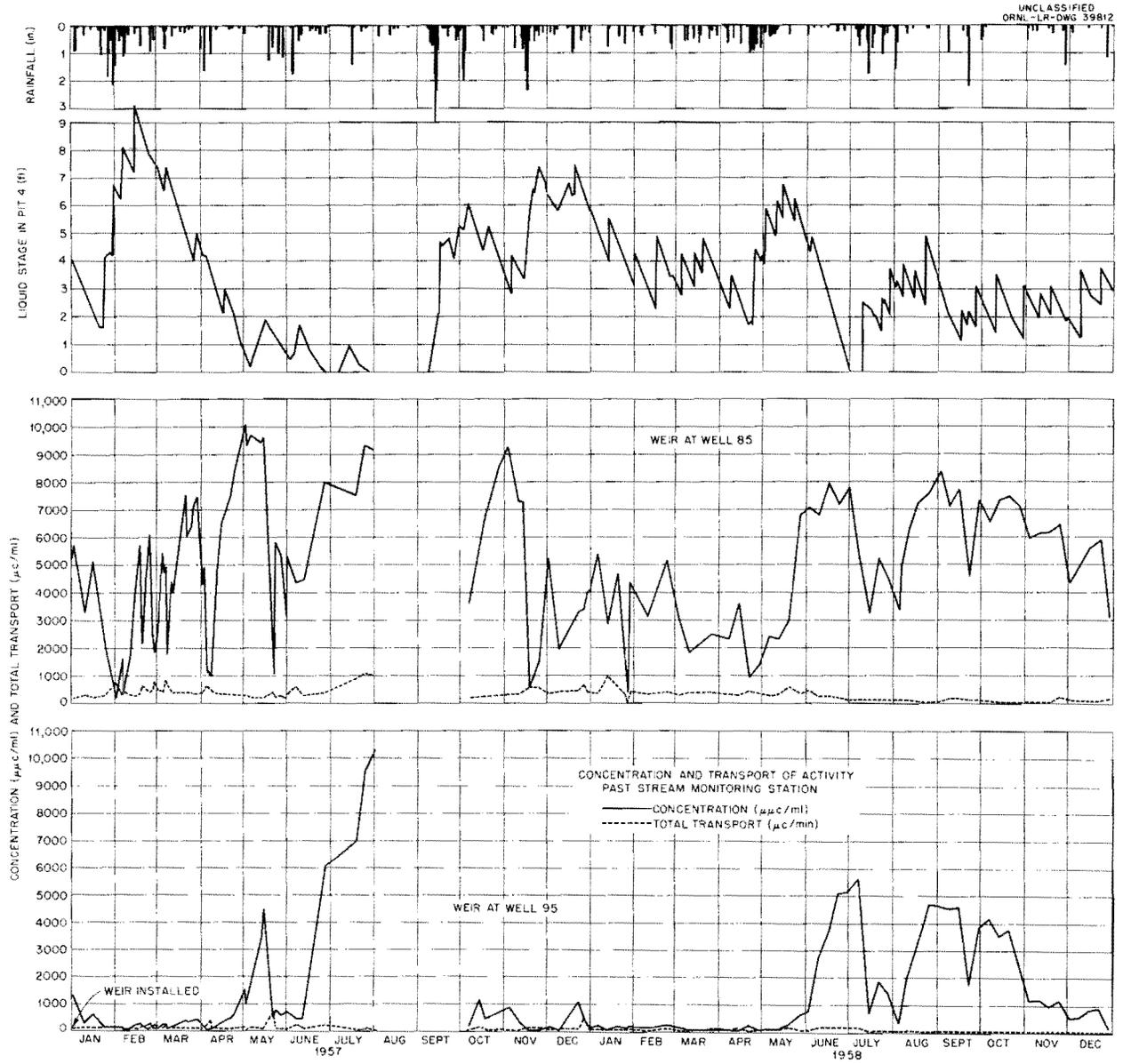


Fig. 7.

10^{-6} $\mu\text{c}/\text{ml}$. These two small streams receive surface drainage from about 150 acres. By assuming that all the rain falling on the drainage area will pass the monitoring weirs (about 200,000,000 gal/year at 4 ft of rain per year) and that 10^{-6} $\mu\text{c}/\text{ml}$ of Sr^{90} was present in the liquid passing the weirs, less than 1 curie per year of Sr^{90} would have been contributed to White Oak Creek.

In a general way the waste appears to be moving out of the pit as anticipated, namely, through the weathered shale in a direction parallel to the bedding. Although the expense and time required for the present monitoring program is a major part of the cost of the disposal operation, we have been unable to explore in detail the pattern of movement of the liquid waste and of its several chemical and radioactive components.

2.6 Future Pit Development

As a result of these studies it is believed possible to build pits which will be better than those now in use. The site for the next pit has been chosen on a ridge just to the east of pit 4. The depth to ground water here is about 40 ft, nearly what it was at the pit 4 site, so that the leakage rates should be rapid enough for economical operation. This ridge is wider than the pit 4 ridge, and there will probably be no active springs, although surface seeps may develop near the streams. The pit will probably be 300 ft long, 25 ft wide at the top, 15 ft deep, and with sides that slope in so that the width of the bottom will be negligible. The present pits leak almost entirely through their sides. The pit will be oriented at right angles to the strike to intercept the maximum number of bedding planes. In this way the escaping liquid will move through, and come in contact with, the largest volume possible of shale. Because a decrease in the storage capacity of the pit is not a disadvantage, the pit will be filled with coarse crushed rock, and a mound of dirt compacted over it. This should not affect the seepage rate very much, but the effects of both rainfall and evaporation will be eliminated, a net gain in itself and a great aid in establishing the pit inventory. The potential danger of air-borne contamination will be eliminated and the radiation hazard for those making studies of the pit operation will be very greatly decreased. If a critical situation developed near the present pits, it would be nearly impossible to conduct a thorough investigation or to take remedial measures. Although the first cost of a rock-filled covered pit would be somewhat higher, the final cost might well be less. When the present pits are abandoned they will have to be filled in, if only to cut down on the radiation field and the potential dust hazard. This will be expensive, because the work will have to be done under very restrictive working conditions. The proposed new pit could be abandoned simply by turning a valve. The design of the first pit built at Oak Ridge placed much emphasis on waste storage; the design and location of the next three pits appear to give joint recognition to the need for both storage and ion exchange. The fifth pit will be designed solely as an ion exchange device.

2.7 Conclusions

The waste disposal pits at Oak Ridge have been operating for about seven years, and have handled with apparent safety many millions of gallons of radioactive waste. There is some concern, however, about their future. Although rather simple changes in location and design can probably correct some of the more obvious defects of the present system, it is unlikely that it will ever be possible to determine in as much detail as desirable the movement of the radioactive materials away from pits of any type. In an area of more homogeneous rocks and simpler structure, it would be possible to learn more, but it does not appear that such a device can ever be operated safely near important centers of population if it is required to handle substantial amounts of radioactive materials. Disposal pits can serve a useful purpose, as they have at Oak Ridge, as a temporary measure to handle limited amounts of waste while better methods are being developed. The better methods may also involve the fixation of radioactive strontium and cesium on clay, but the operation would be conducted under conditions that would permit a more thorough understanding and provide better control.

3.0 PROCESS WASTE TREATMENT PLANT

ORNL process wastes originate from equipment cooling water, decontamination-pad drains, floor drains, storage canals, evaporator condensate, and laboratory sink drains. They are characterized by volumes ranging from 2×10^8 to 3×10^8 gal/year and usually contain less than 2×10^{-6} curie/gal.

3.1 Removal Efficiencies

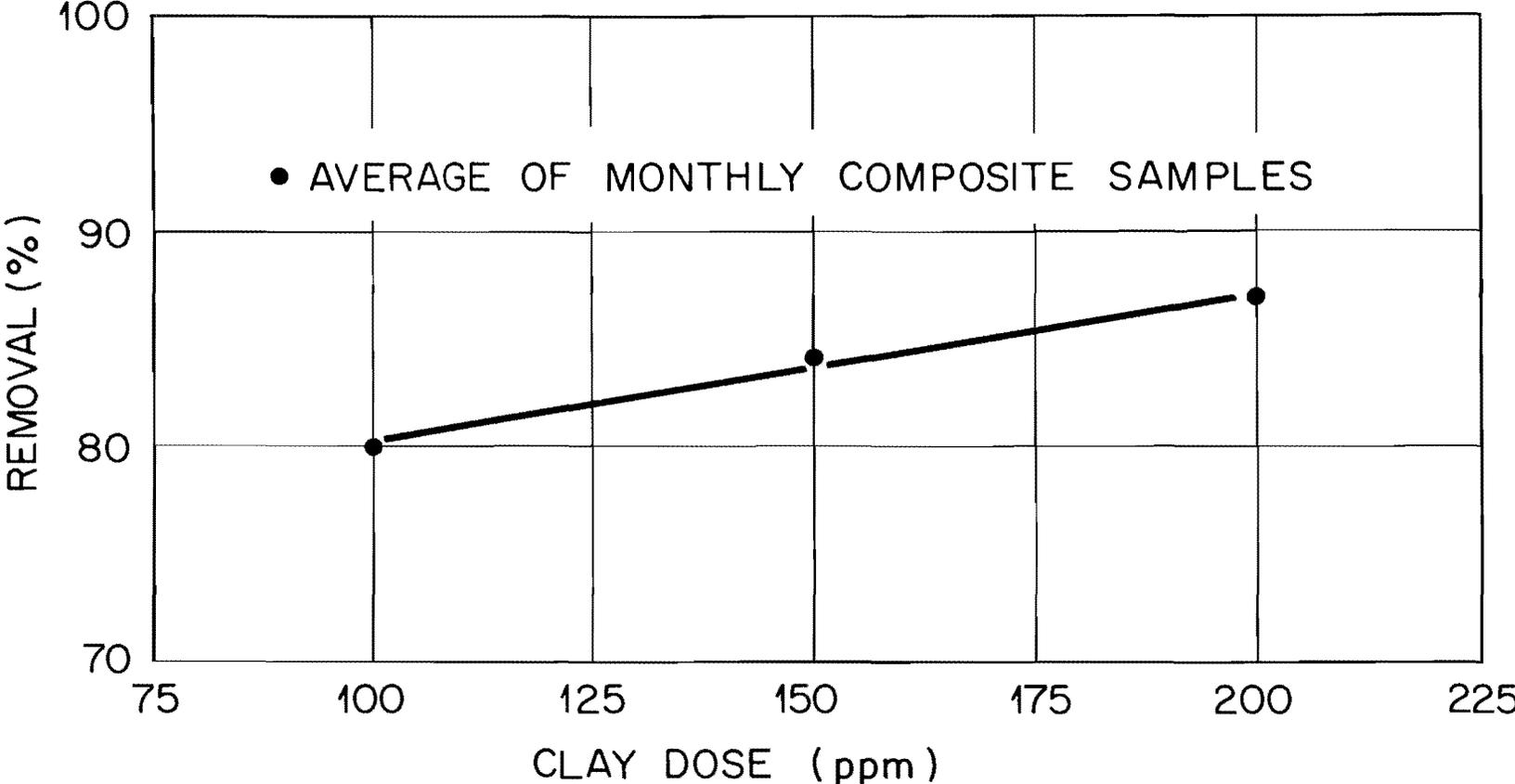
The lime-soda softening process is used for removing strontium and the rare earths; clay is added to remove cesium.* The percentage removal of the important radionuclides is shown in Table 6. During the first 6 months, plant trials were made with various lime and soda ash mixtures, thus varying over-all plant efficiency. Since March 1958, the stoichiometric amount of lime and 200 ppm of excess soda ash have been added to the waste. Strontium removals during this interval averaged 84% and have been as high as 94% for individual periods of a week; removal of total rare earths has averaged 86%. Recently Pm^{147} was released to the process system in quantities exceeding other fission products; the treatment plant removed an average of 94%.

Since the continuous addition of clay was started in October 1958, cobalt removal has averaged 78% and ruthenium removal 76%. The decrease in gross radioactivity depends on the radionuclide composition of the waste and the type and amount of chemicals added. Through September 1958 removal of gross radioactivity averaged 66%; since continuous addition of clay was started, removal has averaged 88%.

Lime-soda treatment does not remove cesium. Prior to the continuous addition of clay, cesium removals were erratic, ranging from zero to 42% removal, and were probably affected by the amount of silt in the equalization basin. Cesium removals, based on analyses of monthly composite samples, were 80, 84, and 86% with additional suspensions of clay of 100, 150, and 200 ppm (see Fig. 8). A dramatic increase in cesium removal by increasing the amount of clay would not be expected, since the removal of cesium approaches 100% asymptotically as the clay dose is increased. Excess soda ash, helpful in removing strontium, adversely affects the removal of cesium.

Not all the solids that are precipitated are removed from solution. For short periods of time the amount of suspended material leaving the plant is as high as 10% of the total solids. Approximately 50% of the radionuclides present in the plant effluent are associated with the suspended solids. During the first

* K. E. Cowser and R. J. Morton, "Radioactive Contaminant Removal from Waste Water: Evaluation of Performance," Proc. Amer. Soc. Civil Engineers, J. San. Eng'g Div., SA3, May 1959, pp. 55-76; M. C. Culbreath, "Radioactive Contaminant Removal from Waste Water: Engineering Design Features," *ibid.*, pp. 41-53.



Process Waste Treatment Plant Removal of Cesium by Grundite.

Fig. 8.

Table 6. Removal of Radionuclides by Treatment Plant
(Monthly Composite Samples)

Date	Percentage Removal						
	Gross β Activity	Sr ⁸⁹⁺⁹⁰	TRE ^a	Cs ¹³⁷	Co ⁶⁰	Ru ¹⁰⁶	Pm ¹⁴⁷
<u>1957</u>							
Sept.	67	87	89	9.5	51	7.6	
Oct.	68	73	77	19	23	0	
Nov.	87	80	80	21	72	83	
Dec.	79	84	87	40	83	67	
<u>1958</u>							
Jan.	53	74	74		76	65	
Feb.	57	66	67	42	56	42	
Mar.	73	85	88	32	69	66	
Apr.	70	84	86	40	74	74	
May	59	78	80	0	61	69	
June	57	84	86	0	67	24	
July	55	85	86	0	70	30	
Aug.	54	85	89	2.1	64	73	
Sept.	77	79	81	48	73	38	
Oct.	84	89	71	80	82	84	
Nov.		82	85	84			
Dec.	84	90	89	84	65	59	
<u>1959</u>							
Jan.	86	87	89	86	78	47	
Feb.	83	79	86	84	78	73	
Mar.	90	88	95	84	65	72	
Apr.	92	80	90	84	89	70	94
May	84	81	90	80	75	76	87
June	89	77	87	81	86	88	96
July	90	83	91	87	76	91	
Aug.	98	84		90	88	96	99

^aTotal Rare Earths.

Table 7. Process Waste Treatment and Discharge to White Oak Creek

Volume of waste treated this quarter: 41,275,000 gal
 Total volume of waste discharged this quarter: 62,099,000 gal

Contaminants	Activity, curies							
	Process Waste Treatment Plant Influent			Process Waste Treatment ^a Plant Effluent			Total Discharge to White Oak Creek	
	Oct.	Nov.	Dec.	Oct.	Nov.	Dec.	Nov.	Dec.
Sr ⁸⁹	0.2	0.2	0.6	0.0	0.1	0.2	1.2	0.2
Sr ⁹⁰	3.4	2.3	4.7	0.7	0.1	1.3	22.6	1.7
Ru ¹⁰⁶	0.7	117.9	27.4	0.1	56.0	2.5	28.1	2.2
Co ⁶⁰	0.2	1.0	3.3	0.1	0.8	1.5	1.0	2.0
Cs ¹³⁷	3.4	65.9	82.7	0.4	14.1	11.7	16.2	20.5
Total rare earths	7.3	349.2	75.7	1.3	67.7	2.4	49.4	4.8

^aPrior to November 1959 the activity discharged to White Oak Creek was estimated from gross beta values. Analyses for individual radionuclides were begun in November to improve reporting accuracy. The total discharge of activity in the untreated waste during October, estimated from gross beta values, was 4.26 curies.

22 months of plant operation 125 curies of Sr^{89 + 90}, 205 curies of the total rare earths, 91 curies of Cs¹³⁷, 13 curies of Co⁶⁰, 8 curies of Ru¹⁰⁶, and 793 curies of Pm¹⁴⁷ were removed from the waste stream. These radionuclides, associated with the sludge resulting from the treatment process, were transferred to the waste pits.

3.2 Conclusions

Under present operating conditions, the total activity discharged to the creek is the sum of that discharged from the Process Waste Treatment Plant and the activity contained in untreated waste. Table 7 shows the curies of Sr⁸⁹, Sr⁹⁰, Ru¹⁰⁶, Co⁶⁰, Cs¹³⁷, and TRE discharged from the plant in the last quarter of 1959 and the total discharge to White Oak Creek in November and December of the same year. These data suggest, at least for the period considered, that the settling basin effluent, i.e., the large volume of untreated waste, is the principal source of radioactive contaminants in the creek and in the Clinch River.

4.0 BURIAL GROUNDS

The burial grounds are special areas which serve as repositories for packaged and unpackaged solid radioactive wastes from the Oak Ridge plants and off-site installations. The burial grounds contribute minor amounts of radioactivity to White Oak Creek. In the following account only burial grounds 3, 4, and 5 are considered; Nos. 1 and 2, the first grounds, are comparatively small and were operated during the early years when the quantities of material handled were low.

4.1 Burial Ground 3

Burial Ground 3, comprising about 7 acres, lies in Bethel Valley about 3000 ft southwest of the present boundary of the ORNL plant site. It was opened in May 1946 and closed during the early part of 1951. The rate of use during this time was about 1.5 acres/year. The burial procedure consisted in removing the soil mantle, generally less than 15 ft deep, down to bedrock, dumping the waste into the excavation, and covering with the original soil. Alpha-contaminated material was covered with concrete before backfilling. Indications are that this area contributes very little, if any, activity to White Oak Creek, and it is believed unlikely that the area will contribute any significant amount of activity in the near future. This opinion is not based on continuous monitoring of the tributary flowing from the site, which would be necessary to determine accurately the amount of activity leaving the area. Also, information on underground leaching and movement of the material buried is lacking, and predictions about future activity release from this area to White Oak Creek are speculative.

Owing to the surface-and-ground-water divide at the westernmost portion of the site, a small part of the area drains southwestward into Raccoon Creek. However, the area involved is small. In 1950, water samples from two of the ten exploratory wells surrounding the site showed activities of 20.7 and 9.1×10^{-9} $\mu\text{c}/\text{cc}$. This was higher than background in the area. Since this time radiologging of the wells has indicated no significant amounts of activity. No beta or alpha activity above background was found in water samples taken in April 1960 or in grab samples taken along the White Oak Creek tributary flowing from the burial ground.

Core drilling indicated that the rock underlying the burial ground is devoid of sizable underground solution channels or caverns, and rapid transfer of ground water is therefore not likely.

4.2 Burial Ground 4

Burial Ground 4 is situated in Melton Valley approximately 0.5 mile southwest of the plant site. The area was opened in February 1951 and closed to routine burial operations in July 1959. It is currently used for special burials and as a standby if the new burial ground should be temporarily closed.

The burial procedure here was similar to that in Burial Ground 3. Trenches, generally 12 to 14 ft deep, were excavated in the weathered shale and the contaminated solids dumped into the hole and then covered with soil. Trenches containing alpha-contaminated materials were covered with concrete. Auger holes, 1 to 2 ft in diameter and approximately 12 ft deep, were used to dispose of extremely high-level waste.

A series of auger holes, 5 to 20 ft deep, was recently completed in and around the burial ground to aid in determining more specifically the geology, hydrology, and movement of radionuclides in the area. Water-level measurements, taken about once each week since August 1959, indicate that the water table fluctuates between 2 and 3 ft below the surface in low areas of the burial ground and 10 to 15 ft beneath higher elevations. Thus solid waste is in contact with ground water during most periods of the year. Beta-gamma contamination has been observed in eight of the sixteen wells in the burial ground; two of these eight, Nos. 185 and 186, have shown significant amounts of alpha activity. With the exception of well 182, all wells showing activity are in areas where the water table is relatively near the surface. This indicates that the effect of ground water has been to move some radionuclides through the soil.

Activity has been detected in numerous seeps throughout the area, many of which seem to be emerging from the downslope end of covered trenches, and also in the intermittent stream that flows from the area into White Oak Creek. Temporary monitoring stations have been installed along the creek immediately above and below the site. Water is pumped continuously from the creek and once each day a 1-liter sample is taken from the collecting container. These daily samples are composited, usually at intervals of a week, and radiochemically analyzed. Five such samples have been analyzed (Table 8). Stream-flow measurements taken at the two stations indicate that the flow is about 5% greater below the burial ground. Results from the five composite samples show that, in general, there is less activity in the stream below the burial ground than above. This may be accounted for by the settling of suspended contaminated solids in the water from the Process Waste Treatment Plant. It is also possible that some activity is taken out of the water as it comes in contact with the banks and the bottom of the stream. In the case of strontium and gross alpha there is a slight increase in concentration below the burial ground, but with the limited amount of data available, it cannot be attributed to the burial ground. The activity in the wells, seeps, and stream at the site is summarized in Table 9.

4.3 Burial Ground 5

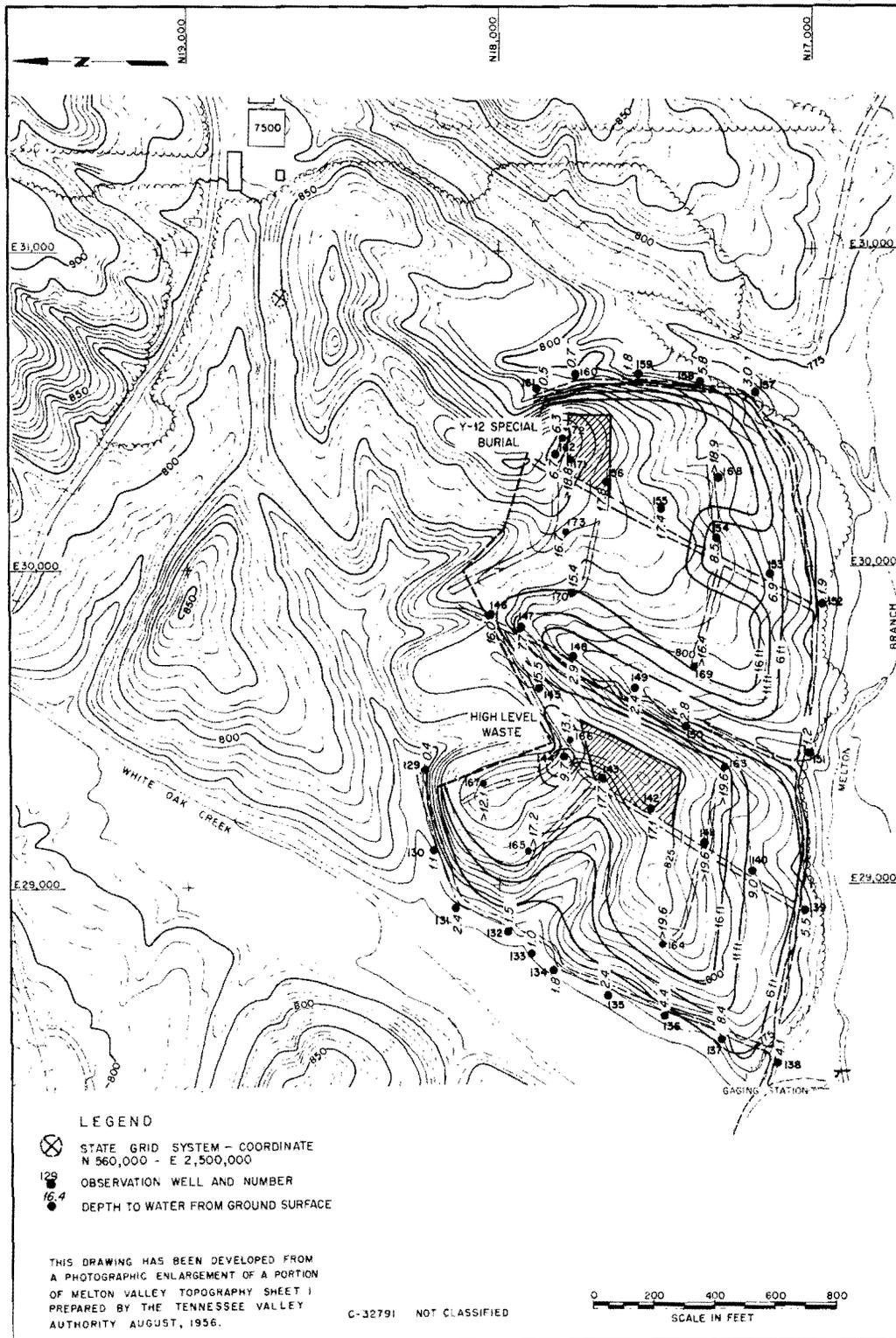
Burial Ground 5 is expected to meet the needs of the Laboratory for at least the next 4 years. Unlike previous sites, a geologic map was made of the area and water table conditions were defined before the burial of any waste. The site of the burial ground is in Melton Valley, about 0.6 mile southeast of ORNL, in the Conasauga shale. The site is situated on the line of knobs underlain by more silty layers along the northwest side of Melton Valley. The topography is that typically developed on shale, with numerous steep-sided gullies, some of which are more than 10 ft deep. Within the site elevations range from 765 to 875 ft, a maximum relief of 110 ft. Stream and well samples taken recently in the vicinity of the buried material showed no activity.

For most efficient use of the burial ground a plan for future disposal operations covering some reasonable period is necessary. To estimate the requirements of land usage, records of solid waste burial dating back to 1957 were analyzed. By linear extrapolation of these data estimates of the volumes of waste expected through 1964 were made (Table 10). Since the policy of segregating contaminated solids is followed, the values for alpha and beta-gamma wastes are estimated separately.

The depth-to-water map (see Fig. 9) can be used to determine the volume available for burial of solid waste. With the restriction that solid waste should be buried 1 ft above the highest water level, the depth of burial in the area between the 6- and 11-ft depth-to-water contours is limited to 5 ft. Similarly, the depth of burial in the area between the 11-ft and 16-ft contours and above the 16-ft contours is limited to 10 and 15 ft, respectively. A 15-ft-deep trench is the maximum depth of burial due to the limitations of existing equipment. The volume available for disposal of solid waste in the outlined area is approximately $21 \times 10^6 \text{ ft}^3$. The areas reserved for high-level waste and other special wastes are not included in the calculations. Where the depth to water is less than 6 ft, these areas can be used for disposal of noncontaminated solids.

The total volume of a trench is not occupied by solid waste, the occupied volume being conservatively estimated at 50%. For convenience of operation a trench is normally 10 ft wide, and a 5-ft spacing between trenches ensures the integrity of each trench and a reasonable working area. Therefore two-thirds of the available area would actually be used. With these limitations, about one-third of the total volume in the burial ground will be occupied by solid waste. Considering all restrictions, the area east of the road along which wells 152 and 156 are located provides about $1.4 \times 10^6 \text{ ft}^3$ of burial space; this should be ample for the alpha-contaminated waste expected over the next 5.5 years. Similarly, east of the road along which wells 146 through 151 are located and west of the site for alpha waste burial about $2.2 \times 10^6 \text{ ft}^3$ is available for burial of beta-gamma-contaminated waste.

To simplify and improve monitoring, a new trench design was adopted for the disposal of solid waste (Fig. 10). The bottom of the trench, covered with 6 in. of gravel, is sloped to an asphalt-lined sump at one end in which a 6-in. perforated casing is installed. Any liquid entering the trench will flow through the gravel underdrain to the collecting sump, from which samples can be withdrawn



Depth to Water Contours of New ORNL Burial Ground.
Fig. 9.



New Trench for Disposal of Solid Wastes Showing Gravel Underdrains and Collecting Sump.
Fig. 10.

Table 8. Radioactivity Measured at Monitoring Stations on White Oak Creek

Composite Date	Volume Discharge, 10 ³ /gal	Activity, mc								
		Gross α	Sr ⁹⁰	Ru ¹⁰⁶	Cs ¹³⁷	Zr Nb ⁹⁵	Co ⁶⁰	Ce ¹⁴⁴	TRE	
Upstream and Downstream from Burial Ground 4										
2/16-19/60	58,330	7.0	298.5	*	*	*	*	*	*	944.9
2/25-3/2/60	44,085	30.0	1,090.0	342.2	225.6	63.9	45.9	580.9	1,495.9	
3/8-14/60	44,822	*	351.5	359.1	298.1	168.1	160.5	5,251.7	1,039.4	
3/22-28/60	38,498	36.1	1,498.9	4,070.1	787.8	722.2	197.0	*	10,208.0	
3/29-4/3/60	64,470	16.6	549.8	*	*	*	*	*	1,813.8	
Totals	250,205	89.7	3,438.7	4,782.4	1,311.5	953.2	403.4	5,832.6	15,502.0	
Monitoring Station on White Oak Creek Downstream from Burial Ground 4										
2/16-19/60	61,400	12.5	418.8	*	*	*	*	*	1,361.2	
2/25-3/2/60	46,405	31.6	1,107.8	363.9	189.9	27.8	50.5	495.4	1,194.8	
3/8-14/60	47,181	*	418.4	378.1	321.8	144.8	160.9	3,178.5	876.8	
3/22-28/60	40,524	41.4	1,036.5	3,040.3	552.8	552.8	207.4	*	6,633.4	
3/29-4/3/60	67,863	10.5	578.7	*	*	*	*	*	1,793.7	
Totals	263,373	96.0	3,560.2	3,782.3	1,064.5	725.4	418.8	3,773.9	11,859.9	

*Results not yet available.

Table 9. Radioactivity in Wells, Seeps and Streams Near Burial Ground 4

	Max. Activity, c/m/ml		Radionuclides	Concentration, d/m/ml
	Gross β (~8% geometry)	Gross α (~47% geometry)		
Wells	25	5	Sr ⁹⁰ (Well 196)	50
Seeps	375	250	TRE (Well 196)	60
			Co ⁶⁰	200
			Cs ¹³⁷	2 x 10 ³
			Ru ¹⁰³⁻¹⁰⁶	Trace
			Po ²¹⁰	Trace
Streams	20	92	Ru ¹⁰³⁻¹⁰⁶	Trace
			Co ⁶⁰	Trace
			Po ²¹⁰	Trace

Table 10. Estimated Volumes of Solid Radioactive Waste
For 1959 Through 1964

Waste	Volumes, 10 ³ ft ³						Total
	1959 (last half)	1960	1961	1962	1963	1964	
Alpha	63	197	194	237	224	262	1177
Beta-gamma	77	163	173	186	193	201	993
Total	140	360	367	423	417	463	2170

and analyzed. After the trench is filled with waste, the void space around the contaminated material is backfilled with shale. A layer of shale near the top of the trench is compacted by tamping, providing a base for an asphalt cap. About 1 in. of asphalt is sprayed on the tamped shale, and after it hardens the remainder of the opening is backfilled with shale. The composition and amount of liquid collected in the sump will be used to evaluate the extent of leaching of radioactive materials from the waste and will serve as an indicator of the effectiveness of the asphalt cap in diverting rainfall. To date, 35 trenches, each approximately 100 ft long, 8 ft wide, and 12 to 14 ft deep, have been opened and filled.

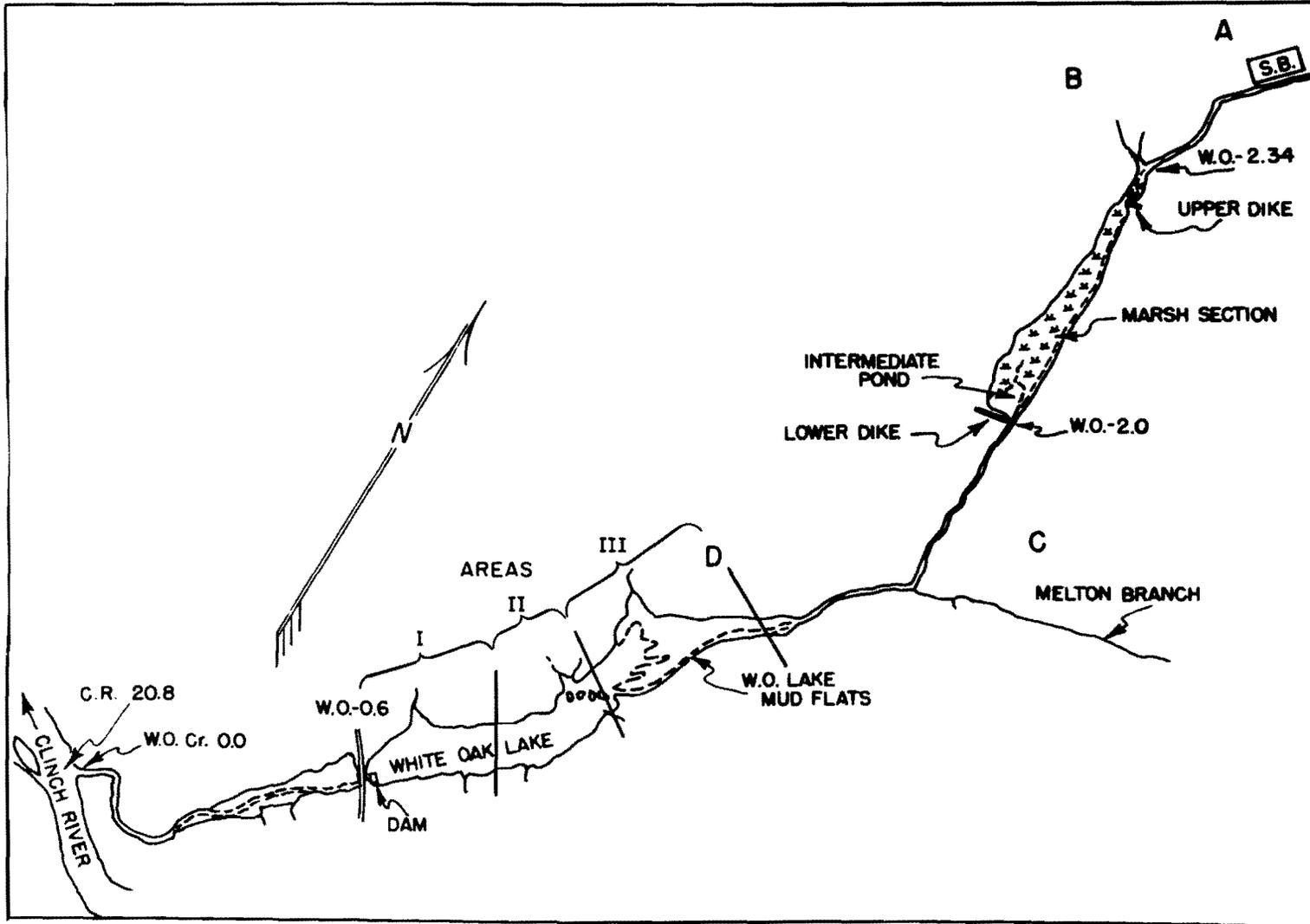
5.0 WHITE OAK LAKE BED AND INTERMEDIATE-POND SEDIMENTS

The quantity of radionuclides in the bed of former White Oak Lake and in the bed of the former intermediate pond located above the lake has been reported earlier.* A dike (see Fig. 11) was constructed across the White Oak Creek in the spring of 1944 at mile 2.0, which created the intermediate pond. In a flood on September 29, 1944, this dike failed. The dam at WOC mile 0.6, constructed in the fall of 1943, is still in existence. The formation of White Oak Lake in 1943 resulted in the deposition and accumulation of sediments on the bottom of the lake. In 1955 White Oak Lake was drained, and the area now comprises an ecological study preserve.

5.1 Distribution of Fission Products in Sediments

The bed of White Oak Lake comprises a total of 44 acres. For survey purposes the bed was divided into three areas or zones (see Fig. 11). Zones II and III were investigated in the 1956, 1958, 1959, and 1960 surveys; zone I was investigated only in the later years. The area of zone I is 11.9 acres; of zone II, 10.8 acres; and of zone III, 21.7 acres.

* Overstreet and Jacobson, CN-2039, CH-3330, CH-2346, 1944; L. H. Weeks, "Assay of Fission Product Contamination in the Mud of White Oak Creek Drainage System," CL-CF-45-5-121, 1945; J. S. Cheka and K. Z. Morgan, "Radioactive Fission Product Contamination in the Mud of White Oak Drainage System," MON-H-258, 1947; K. Z. Morgan and F. Western, "Contamination of Water Discharge from Clinton Laboratories," MON-H-259, 1947; H. H. Abee, "Radioactivity in the Mud of White Oak Lake," ORNL-1580, 1953.



White Oak Creek and Lake Bed.
Fig. 11.

The volume of sediment in each zone was determined from data obtained by the Tennessee Valley Authority in June 1953.* They reported 1,030,000 ft³ of sediment in the lake at that time, and an annual buildup 100,000 ft³. Assuming that further deposition in 1954 and 1955 was offset by equal losses when the lake was drained in October 1955, zone I now contains 570,000 ft³; zone II, 260,000 ft³; and zone III, 200,000 ft³.

Cores of sediment recovered from the lake bed and the intermediate pond were divided into two sections: the upper 6-in. section and the remaining lower section, which varies in depth. The bulk density of the upper 6 in. averaged 1.7 g/cc; that of the lower section averaged 1.9 g/cc. From these values the weights of sediment in each section of the three zones were calculated:

Zone	Volume, ft ³	Weight, g
Zone I, Upper 6 in.	2.58 x 10 ⁵	12.4 x 10 ⁹
	Lower section 3.12 x 10 ⁵	16.8 x 10 ⁹
Zone II, Upper 6 in.	2.34 x 10 ⁵	11.3 x 10 ⁹
	Lower section 0.26 x 10 ⁵	1.4 x 10 ⁹
Zone III, Upper section	2.0 x 10 ⁵	9.6 x 10 ⁹

From the 1958, 1959, and 1960 surveys, it is estimated that 11.2 curies of Sr⁹⁰, 477 curies of Cs¹³⁷, and 58.2 curies of Co⁶⁰ are present in the total sediment (Table 11). The apparent discrepancy between the 1956 and 1958-60 data for Cs¹³⁷ in zone III is probably due to nonrepresentative sampling. An intensive survey in 1958 suggests that the values for that year are more representative of the Cs¹³⁷ and Co⁶⁰ concentrations.

The bed of the Intermediate Pond comprises an area of about 3.62 x 10⁵ ft² or 8.3 acres. During the sampling distinct differences in the sediment were noticed at the 5-in. depth. The upper section of the core showed characteristics of deposited sediment; below 6 in. the character of the material was difficult to evaluate. The cores were readily recoverable and were obtained to a depth of 3.5 ft. Since Burial Ground 4 is contiguous with the west side of the Intermediate Pond, it is not unlikely that radionuclides are reaching the lower depths of pond sediment in ground water moving from the burial ground.

Based on the bulk densities reported for lake bed sediment, the upper 6-in. section of pond sediment is estimated to contain 8.70 x 10⁹ g of soil and the lower section, 58.4 x 10⁹ g. It is estimated that 3.7 curies of Sr⁹⁰, 17.0 curies of total rare earths, 113.6 curies of Cs¹³⁷, and 12.6 curies of Co⁶⁰ are present in the pond sediments (Table 11).

* A. S. Fry, ORNL, memorandum to M. A. Churchill, "Sediment Investigations, White Oak Lake," June 30, 1953.

Table 11. Concentrations of Radionuclides in White Oak Lake Bed and Intermediate Pond
 Dry weight corrected to wet weight by factor 1.3

	$\mu\text{c} \times 10^{-4} / \text{g wet wt}$				Curies			
	Sr ⁹⁰	TRE ^a	Cs ¹³⁷	Co ⁶⁰	Sr ⁹⁰	TRE ^a	Cs ¹³⁷	Co ⁶⁰
White Oak Lake Bed, Summer 1956								
Zone II, upper 6 in	12	-	140	68	13.9	-	160.7	77.1
Zone III, sediment section	8.5	-	150	92	8.2	-	140.7	88.4
White Oak Lake Bed, 1958-1960								
Zone I ^b								
Upper 6 in.	2.1	0.46	78	5.1	2.6	5.7	74.0	4.9
Lower section	0.37	0.10	7.1	0.69	0.6	1.7	9.1	0.9
Zone II ^c								
Upper 6 in.	4.3	-	120	22	4.9	-	129.9	24.3
Lower section	0.62	-	130	0.69	0.9	-	1.8	0.01
Zone III, sediment section	2.3 ^c	-	270 ^d	29 ^d	<u>2.2^c</u> 11.2	-	<u>262.0^d</u> 476.8	<u>28.1^d</u> 58.2
Intermediate Pond, 1960								
Upper 6 in.	3.3	11	155	11	2.8	9.8	86.5	6.3
Next 3 ft	0.15	1.2	72	1.7	<u>0.9</u> 3.7	<u>7.2</u> 17.0	<u>27.1</u> 113.6	<u>6.3</u> 12.6

^aTotal rare earths including Y⁹⁰

^bResults of survey April 1960

^cResults of survey July 1959

^dResults of survey August 1958

Since the Sr^{90} concentration is the limiting factor, it is interesting to compare the Sr^{90} value for 1956 with that of 1958-60. Data in Table 11 show that 14.9 curies of Sr^{90} is distributed in the sediments of the lake bed and intermediate pond. The available data for 1956 do not include Sr^{90} concentrations in zone I, the lower section of zone II, and the Intermediate Pond sediment. To get an estimate of the Sr^{90} concentration in these three sections, the missing values were calculated assuming a factor of 3.3 greater concentration in 1956 than in 1958-60 (this factor determined from the values of 1956 and 1958-60). The total Sr^{90} budget estimate for 1956 in the lake bed and Intermediate Pond was 47.8 curies.

5.2 Losses by Solution Transport

In order to estimate the losses of Sr^{90} from the White Oak Lake bed by solution transport, surface samples (6 in. depth) of contaminated soil were obtained from the upper lake bed area (zone III) and subjected to leaching in the laboratory. Sixty-two-gram portions of oven-dried soil (equivalent to a cylinder 1 in. dia and 6 in. deep) were leached with several leaching solutions. No appreciable strontium was removed until the equilibrium pH was appreciably less than neutral (Table 12). The pH data of the remaining solutions indicate extensive hydrolysis of the free carbonates in the sediment and leaching of calcium. Results were similar with different techniques.* Substantial strontium removal by the 0.1 M HNO_3 solution indicates that there is no silicate fixation of the strontium since this concentration of acid and the short contact time would not be expected to seriously disrupt such lattices. The average ratio of the amount of strontium per gram of sediment to the amount in 1 ml of solution, excluding values for 0.1 M and 1.0 M HNO_3 , was 1650, a value which agrees very closely with that obtained for Clinch River sediments.

Though the simple laboratory leaching experiments do not accurately describe the leaching behavior of Sr^{90} from the lake bed, they serve as a guide. One of the errors in extrapolating these results to field conditions is the short leaching time, which decreases the effectiveness of the leaching agent. On the other hand, in the field the water table fluctuates, and there is not a continuous downward percolation of the incident rainfall. Also, evaporation and surface runoff would tend to decrease the leaching effectiveness in the field as compared to the laboratory.

In order to calculate the percentage removal of Sr^{90} from the lake bed, the following conditions were assumed:

Effective $K_d = 1650$

An acre half-foot of soil weighs 10^9 g

Average annual rainfall = 1.3 meters

*S. I. Auerbach et al., "Health Physics Ann. Prog. Rept. for Period Ending July 31, 1958," p. 35, ORNL-2590.

Table 12. Leaching of Contaminated White Oak Lake Bed Soil
 62-g samples of oven-dried soil leached with volume of solution indicated

Solution	Volume ml	Strontium, μc			% in Sol'n	Equilibrium pH	K_d^a
		Leached	On Soil	Total			
Dist. H_2O	500	0.000247	0.0433	0.0435	0.57	8.08	1409
	1000	0.000405	0.0427	0.0431	0.94	8.20	1700
	1500	0.000468	0.0617	0.0622	0.76	6.42	3189
1 <u>M</u> HNO_3	500	0.0441	0.00240	0.0465	94.8	0.02	0.44
0.1 <u>M</u> HNO_3	500	0.0432	0.0069	0.0502	86.2	3.87	1.24
10^{-3} <u>M</u> HNO_3	500	0.000682	0.0450	0.0456	1.22	7.42	531
10^{-5} <u>M</u> HNO_3	500	0.000216	0.0391	<u>0.0393</u>	0.55	8.18	1458
Avg.				0.0450 $\mu\text{c}/62 \text{ g}$ or 0.00073 $\mu\text{c}/\text{g}$			

$$^a K_d = \frac{\text{Sr/g of soil}}{\text{Sr/ml of solution}}$$

Surface soil of lake bed is uniformly leached by all incident rainfall. Let $X = \% \text{ Sr}^{90}$ leached and $100 - X = \% \text{ Sr}^{90}$ remaining on the soil. With an annual rainfall per acre of 5.2×10^{10} cc,

$$\frac{100-X}{X} = \frac{(1650)(10^9)}{(5.2 \times 10^{10})} = 32; \quad X = 3.3\% \text{ Sr}^{90} \text{ leached per year}$$

Another estimate can be made by assuming that the total flow of White Oak Creek at White Oak Dam has uniformly leached the surface soil of the lake bed. Assuming an average flow of $10 \text{ ft}^3/\text{sec}$ (9×10^{12} cc/yr) for White Oak Creek and an area of 40 acres for the lake bed (4×10^{10} g of soil) a similar calculation can be made:

$$\frac{100-X}{X} = \frac{(1650)(4 \times 10^{10})}{(9 \times 10^{12})} = 7.3; \quad X = 12\% \text{ Sr}^{90} \text{ leached per year}$$

If White Oak Lake Bed is being leached in a manner similar to the leaching of an ion exchange column, that is, if water starts percolating at the higher elevations of the lake bed and moves laterally through the bed, the upper areas would be depleted in Sr^{90} first, and the apparent loss would be greater than that calculated from the K_d values. The percolating solution would contain equilibrium amounts of Sr^{90} before it reached the lower elevations of the lake bed, and in this lower region the loss of Sr^{90} would be much less. Nearly constant annual losses of Sr^{90} at the dam would be expected. The amount contributed by this source would begin to decline when the Sr^{90} content of the sediment in the region of the dam was depleted. It seems unrealistic to assume that more than 10% of the Sr^{90} would leave the lake bed in any year. Using the Sr^{90} budget for the year 1956, the loss of this element by leaching would amount to approximately 5 curies/year.

5.3 Losses by Sediment Transport

Radionuclides can leave the lake bed area as ions dissolved in water or sorbed on particles. From information on the mineralogical character of the sediment, the specific radionuclide affinity of minerals, and the total radionuclide activity discharged to the river, it is possible to infer the form in which radionuclides are leaving the area. Discharge of Cs^{137} and Sr^{90} to the Clinch River is evaluated in terms of the form in which these elements leave the drainage area.

Mineral Character of Sediment. Several materials, including lake bed sediment, river mud, and source material from areas surrounding the lake bed, were analyzed mineralogically. Though the relative distribution of minerals varies, 50-80% of the material in all samples tested is illite; the lower value would be characteristic of silts and the higher value of clayey material. Quartz, the principal non-clay material, makes up approximately 25% of the total weight. The vermiculite concentration is proportional to the degree of weathering of the material; as much as 25% has been allocated to this mineral, though 15% would represent a more normal concentration. Approximately 10% kaolinite has been identified in several samples. Calcite (or dolomite) is

not readily identified by x-ray methods, but has been found in low concentrations. It exerts a strong influence on any chemical reactions involved. Its presence can be inferred from pH measurements; calcareous sediments have pH values from 7.0 to 8.2. In the leaching tests of lake bed sediment the pH in the distilled water system is about 7.5.

Radionuclide Specificity of Minerals. An outstanding example of specificity is the illite-cesium reaction. An expression of this affinity is the ratio of the activity per unit weight of clay to the activity per unit volume of solution (K_d). For cesium sorption by illite, the K_d is about 150,000. Other clays show K_d values of the order of 2000 for cesium (Table 13a). Especially noteworthy is the relatively low affinity of illite for strontium, a value of 350. In comparison, kaolinite has a K_d of about 4000 for strontium (Table 13b).

The high cesium K_d values for a sample of a river sediment suggest that the sediment is illitic; x-ray diffraction diagrams confirm this (approximately 50% illite). The strontium K_d suggests about 25% kaolinite; x-ray tests show about 10% of this mineral. The sensitivity of K_d to pH change also suggests that calcite may be involved in the sediment K_d for strontium. Another interesting relation in the sediment is the very high affinity for cobalt (Table 13c).

Activity Discharged to Clinch River. Discharge data from White Oak Creek during 1954 through 1958* show that approximately 25 curies of Cs-Ba¹³⁷ was discharged in 1954 and 1955; in 1956, 160 curies; in 1957 105 curies; and in 1958, 55 curies.

The Sr⁹⁰ discharges show a gradually decreasing trend, from approximately 120 curies in 1954 to about 70 curies in 1958.

These discharge data suggest several interesting relations. The extremely high affinity of the sediment for cesium would produce a very low concentration of cesium in the ionic phase. The lake condition in 1954 and 1955 would allow the sediments to settle to the lake bottom; the low discharges recorded for these years tend to corroborate the existence of a relatively quiescent lake.

The sudden increase in cesium discharge in 1956 suggests that the conditions for sedimentation were disrupted by more turbulent conditions. In late 1955 the lake was drained, and the increase in cesium discharge was due to the transport of suspended particles containing the sorbed cesium.

While cesium discharge may be due primarily to suspended particle transport, strontium movement as a dissolved ion is suggested. The strontium K_d is 1/50 that of the cesium K_d for lake bed sediment; this means then that the sediment will remove much smaller concentrations of strontium. The recorded discharges suggest that by 1954 most of the strontium was in an ionic state when it left White Oak Lake. If sediment transport played a dominant role in removing strontium from the lake bed, an increase in strontium discharge in 1956 would have been expected, similar to the discharge pattern observed for

* W. D. Cottrell, "Radioactivity in Silt of the Clinch and Tennessee Rivers," ORNL-2847, November 1959.

Table 13. Fission Product Sorption by Clays and Sediment

Standard Clay	Contact Time	Activity Sorbed, %		K_d		Amount Clay Used
		pH 6	pH 9	pH 6	pH 9	
a. Cesium						
Illite	1 hr	89.99	90.48	27,000	28,500	0.1 g/300 ml
	3 days	97.92	98.60	141,000	217,800	
	7 days	98.39	98.56	183,000	205,000	
Kaolinite	1 hr	74.23	93.19	2,900	13,700	0.1 g/100 ml
	3 days	73.96	62.35	2,800	1,600	
	7 days	68.47	51.20	2,200	1,100	
Montmorillonite	1 hr	61.20	58.26	1,600	1,400	0.1 g/100 ml
	3 days	55.14	56.15	1,200	1,300	
	7 days	49.95	50.54	1,000	1,000	
Vermiculite	1 hr	96.63	95.77	1,430	1,300	1.0 g/50 ml
	2 days	99.41	99.64	8,430	14,100	
River Sediment	1 hr	53.76	61.31	2,300	3,200	0.1 g/200 ml
	3 days	96.17	96.16	50,200	50,200	
	7 days	97.78	97.64	88,000	82,800	
b. Strontium						
Illite	1 hr	23.42	31.67	310	320	0.1 g/100 ml
	3 days	26.69	41.05	360	700	
	7 days	26.88	43.17	370	760	
Kaolinite	1 hr	62.77	71.24	3,400	4,900	0.1 g/200 ml
	3 days	67.49	68.55	4,200	4,400	
	7 days	66.435	66.28	4,000	3,900	
Montmorillonite	1 hr	70.85	71.88	2,400	2,600	0.1 g/100 ml
	3 days	66.88	68.65	2,000	2,200	
	7 days	67.31	68.67	2,100	2,200	
Vermiculite	1 hr	77.45	67.14	170	100	1 g/50 ml
	2 days	96.95	96.46	1,600	1,400	
River Sediment	1 hr	21.42	24.79	540	660	0.1 g/200 ml
	3 days	45.79	63.87	1,700	3,500	
	7 days	41.83	66.80	1,400	4,000	
c. Cobalt						
Illite	1 hr	28.98	78.04	410	3,600	0.1 g/100 ml
	3 days	85.49	94.65	5,900	17,700	
	7 days	86.43	95.94	6,400	23,600	
Kaolinite	1 hr	63.91	69.17	3,500	4,500	0.1 g/300 ml
	3 days	71.54	51.51	4,000	2,100	
	7 days	60.92	46.58	3,100	1,700	
Montmorillonite	1 hr	69.70	56.38	2,300	1,300	0.1 g/100 ml
	3 days	63.91	45.68	1,800	840	
	7 days	62.36	45.37	1,700	830	
Vermiculite	1 hr	70.63	72.87	120	130	1.0 g/50 ml
	2 days	78.63	84.62	3,600	275	
River Sediment	1 hr	46.44	71.91	1,700	5,100	0.1 g/200 ml
	3 days	93.34	82.38	28,000	9,400	
	7 days	97.28	85.12	71,600	11,400	

cesium. However, a decrease in strontium discharge is recorded for 1956, which indicates that very little strontium was sorbed on the sediment relative to the amount normally discharged.

Assuming that the 160 curies of cesium discharged in 1956 was associated with suspended solids and using 2.7×10^{-6} curie of cesium per 100 g, approximately 6.0 acre half-ft of sediment was discharged (assuming 10^9 g of soil per acre half-ft). Since there is approximately 0.8 curie/acre half-ft, the strontium discharge associated with suspended solids would approximate 4.8 curies. The volume of sediment discharged under these conditions would be 130,000 ft³.

Since the recorded discharges in the year after 1956 show a decrease in cesium discharge, strontium discharge by sediment transport would be expected to decrease proportionately. From these data it may be concluded that cesium movement from White Oak Lake bed is primarily by sediment transport and strontium movement is primarily by solution transport. The strontium contribution from lake bed sediments to the total strontium discharged at White Oak Dam was less than 10 curies/year in 1956 and more nearly 5 curies/year in 1959.

5.4 Biological Uptake

Lake Bed Vegetation. The pattern of vegetation on the lake bed has increased in complexity with each succeeding growing season. The successional trend has been toward an increase in numbers of the trees (particularly willow) at the expense of herbaceous vegetation. The sedge-rush community maintained about the same amount of coverage in 1952 as in 1957 (see Fig. 12). The predominant plant communities now in the lake bed are black willow, sedge-rush, smartweed-grass, Spanish needle, and lespedeza. In addition to willow the most abundant new trees are sycamore, smooth sumac, and green ash. Tree growth has been rapid, averaging 3 ft/year.

Two aspects of the soil-to-plant movement of radionuclides have been explored. Many of the species sampled* in 1957 were again sampled and analyzed for radionuclide content to evaluate the extent of radioisotope accumulation over a period of two growing seasons. Duplicate 0.5-m² harvests of some of the plant communities were then taken to provide information on radionuclide accumulation per unit area of vegetation and soil.

The plant taxa sampled for the first phase were Solidago spp., Eupatorium serotinum Michx., Impatiens spp., Polygonum spp., Bidens frondosa (L.), Rhus spp., Salix nigra Marsh., and Fraxinus pennsylvanica Fern. In addition samples of Platanus occidentalis (L.) and Phytolacca americana (L.) were taken to provide additional data for future comparisons. All samples were analyzed for Sr⁹⁰, Cs¹³⁷, Co⁶⁰, and the more abundant cations.

Comparison of the Sr⁹⁰ and Cs¹³⁷ content of many of these species shows more similarities between 1957 and 1958 samples than differences (Table 14). Mean values for each plant part for the two years were tested for significance of difference by a t test. Variability of the lake bed samples was considerable

* S. I. Auerbach et al., op. cit., p. 32 (July 31, 1958).

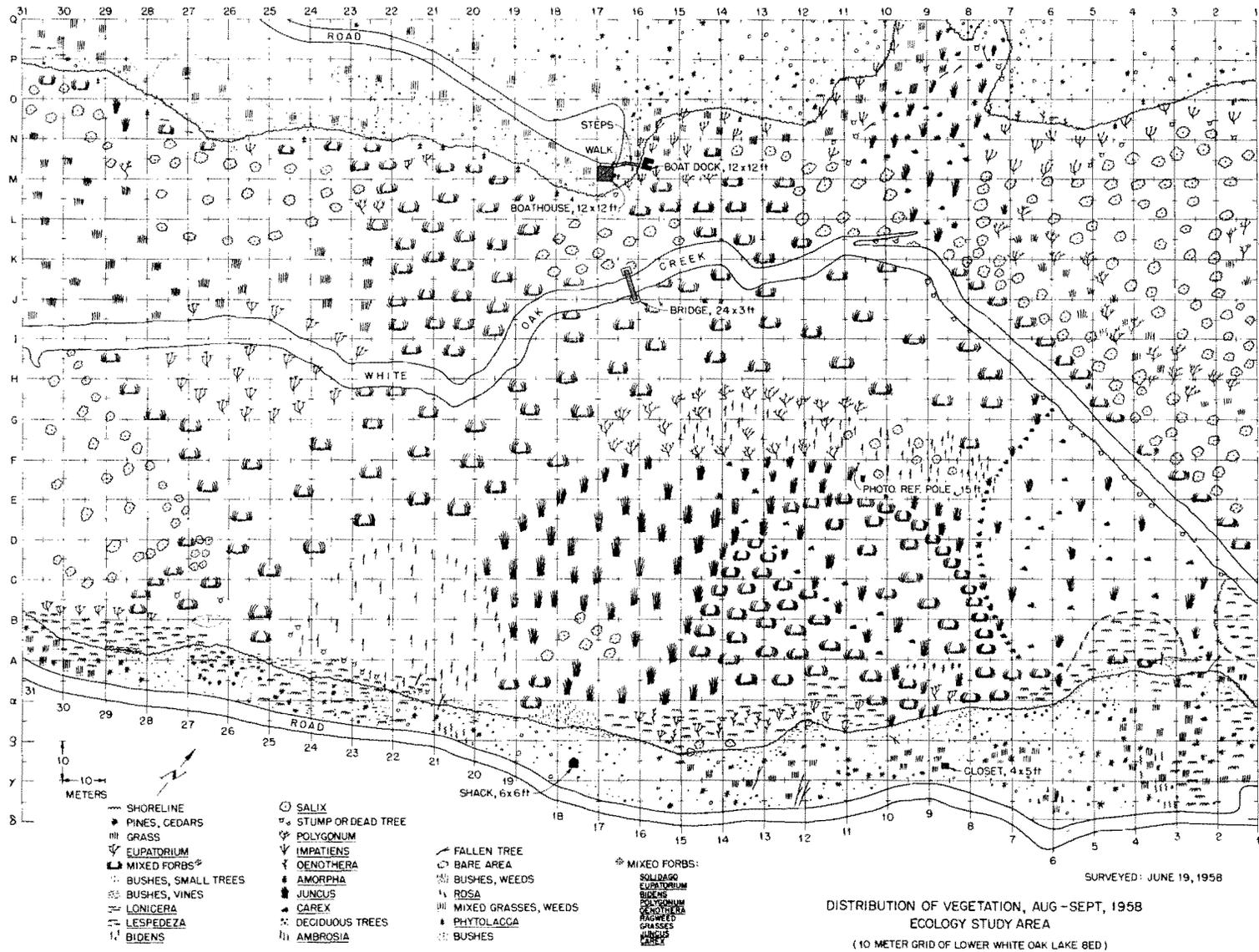


Fig. 12.

and the number of replicates was not great. Consequently the degree of difference and the significance placed on this difference should be interpreted on the basis of the considerable variance in the samples. Nevertheless, the general trend over the past three years indicates no cumulative concentration. The four tree genera analyzed (Rhus, Salix, Platanus, Fraxinus) show no evidence of increasing accumulation in leaves or stems. Within this group, Rhus (sumacs) seem to be the most effective concentrators of Sr^{90} . The Cs^{137} and Co^{60} concentrations are much the same for the four genera. Even though sumacs have shallow, spreading roots, this physical characteristic in itself would not account for the differences.

Table 14. Comparisons of Radionuclide Content in Native Plant Species 1957 and 1958

Differences are not significant except as indicated by asterisks

Plants	Plant Part	Amount, $\mu\text{c}/100\text{ g}$				
		Sr^{90}		Cs^{137}		
		1957	1958	1957	1958	
<u>Polygonum</u>	Leaves	0.151	0.107			
	Stems	0.057	0.051			
<u>Eupatorium</u>	Upper lake bed	Leaves	0.153	0.0745	0.012	0.042
		Stems	0.031	0.021	0.006	0.026
		Flowers	0.050	0.034	0.023	0.040
	Lower lake bed	Leaves	0.062	0.121*	0.005	0.039
		Stems	0.021	0.030**	0.004	0.012
		Flowers	0.026	0.045**	0.008	0.025
<u>Rhus</u>	Upper lake bed	Leaves	0.122	0.137	0.028	0.022
		Stems	0.095	0.139	0.009	0.020*
		Petioles	0.200	0.199	0.028	0.035
	Lower lake bed	Leaves	0.111	0.151	0.005	0.003*
		Stems	0.094	0.232	0.003	0.004
		Petioles	0.159	0.410		
<u>Solidago</u>	Leaves	0.098	0.074	0.028	0.065**	
	Stems	0.044	0.036	0.018	0.014	
	Flowers	0.028	0.0318	0.040	0.052	
<u>Bidens</u>	Leaves	0.042	0.099			
	Stems	0.033	0.028			
	Flowers	0.031	0.024			
<u>Impatiens</u>	Leaves	0.241	0.177			
	Stems	0.385	0.158**			
<u>Fraxinus</u>	Leaves			0.023	0.018	
	Stems			0.007	0.013	
	Petioles			0.023	0.027	

*Significant at $P = 0.05$.

**Significant at $P = 0.01$.

From a synoptic viewpoint, the most important general fact emerging from these studies is that the plants are maintaining the same concentrations of radionuclides, especially Sr⁹⁰, over a period of three years. This fact becomes particularly significant when the total accumulation of radionuclides per unit area is considered. The total Sr⁹⁰, Cs¹³⁷, and Co⁶⁰ per square meter of vegetation is given in Table 15.

Table 15. Accumulation of Radionuclides in 1 m² of Vegetation

Quadrat No.	Plant Type	% of Total Soil Burden of Radionuclides		
		Sr ⁹⁰	Cs ¹³⁷	Co ⁶⁰
1	Barnyard grass	0.12	0.017	0.003
2	(<u>Echinochloa</u>)	0.004	0.0004	0.041
3		0.09	0.007	0.018
4		0.007	0.001	0.006
5		0.06	0.002	0.001
6		0.05	0.001	0.001
7	Smartweed	0.69	0.006	0.019
8	(<u>Polygonum</u>)	0.93	0.015	0.048
9	Sericea	2.3	0.011	0.007
10	(<u>Lespedeza</u>)	1.3	0.009	0.004
11	Begger-tick	0.62	0.006	0.006
12	(<u>Bidens</u>)	0.54	0.041	0.006

Only one plant community - a legume (Lespedeza cuneta) - removed more than 1% of the soil burden of radionuclides. The percentage Cs¹³⁷ accumulation was small even though in the first nine quadrats the concentration in the soil was high, ranging from 3 to 11 mc/m². Removal of Sr⁹⁰ by the plants was far more effective because Sr⁹⁰ concentrations in the soil of the same quadrats ranged from 18 to 235 µc/m².

The possibility that increasing amounts of Sr⁹⁰ are being tied up in the vegetation and undecayed litter can be ruled out at present because accumulation of strontium by the vegetation has been approximately the same since studies were started. The total Sr⁹⁰ burden in vegetation on 40 acres of the lake bed, using the highest values for accumulation per unit area, would be 150 mc. If it is assumed that equal quantities are contained in the two-year (1956 and 1957) accumulation of undecayed litter, a maximum of 500 mc is bound in these compartments of the biological system. Most of the vegetation has a lower concentration, and this would be reflected in the litter. Since much of the 1956 litter has been decomposed and returned to the soil, the actual quantity in the vegetation is probably even less. Movement of Sr⁹⁰ out of the litter

may not be in equilibrium with the rate of income from vegetation for several more years. Yet decay rates on the lake-bed site are probably fast enough that high storage will not occur here.

As more Sr^{90} (and calcium) is leached out of the surface soil, the possibility remains that the vegetation might translocate a relatively greater proportion of this fission product than it has the past three years. Under those circumstances a continuous replenishment of the surface soil in hazardous quantities might occur for several decades. In spite of the considerable loss of Sr^{90} in the past three years, there has not been any reduction in the hazard potential of the vegetation when considered from the food-chain aspect. In terms of strontium units (micromicrocuries of Sr^{90} per gram of calcium) the 1958 samples ranged from 8.0 to 10^4 to 7.0×10^2 , which strongly suggests a long-term hazard for areas heavily contaminated by this fission product unless effective means of depressing plant uptake or of removal from the soil are developed.

Insects. The three major objectives of the sampling regimen for insects on vegetation of the lake bed, initiated in 1956,* are (a) to discover how quickly insects invade the lake-bed vegetation, (b) to estimate the weight of insects per unit area (biomass), and (c) to obtain data on the accumulation of fission products by these insects. This program provides an estimation of the role of the insects in the environmental cycling of fission products.

Samples have been taken in three vegetation types: sedge-rush, smartweed, and willow. Generally, the insect communities in each of these areas were composed of a few dominant species during the first year the vegetation became established on the lake bed (1956 for sedge-rush and smartweed, 1957 for willow). In the herbaceous communities during subsequent seasons the trend has been for these dominant insect species to become less numerous and for additional insect species to appear. However, the willow insect community is still dominated by one group.

In the sedge-rush vegetation type, 1956 samples showed a community dominated by two leafhopper species, two grasshopper species, a predaceous lygaeid bug, and a number of fly species. These accounted for about 80% (by number) of the insects collected. In 1957 these groups of insects (leafhoppers, grasshoppers, true bugs, and flies) were still the most abundant, but no species was of such abundance as to greatly exceed the other species in numbers of individuals. About 50-60% of the insects collected belonged to these groups. In 1958 the insect community was similar to that of 1957.

The smartweed vegetation type supported in 1956 a community dominated by one species of a plant bug, bees, and flies. About 70% of the insects collected belonged to these groups. In 1957 other plant bugs, leafhoppers, bees, and flies were the most numerous, but many additional insect species appeared. All these groups were less numerous in 1957 than in 1956, and still less numerous in 1958.

* R. M. Anderson et al., "H. P. Ann. Prog. Rept. July 31, 1957," ORNL-2384, p. 21

The willow vegetation type was dominated by a single species of flea beetle during the first year the willow insects were sampled (1957). A variety of other insect species was present but none was abundant. The 1958 samples showed a very similar community, consisting of many insect species but still dominated by the flea beetle. The numbers of the flea beetle were greater in 1958 than in 1957.

Evidently, then, the insects characteristic of the sedge-rush and smartweed vegetation types required two or more seasons to develop, with the greatest change in the composition of the insect fauna occurring between the first and second seasons. Little change occurred in the willow insect community. Changes in the abundance of the vegetation types themselves may be reflected in these changes in their insect communities. The sedge-rush association maintained about the same amount of coverage on the lake bed during the period of study, but smartweed decreased in abundance and willow increased markedly. The expanded willow habitat available to the insects in each season may be responsible for the continued high populations of the dominant flea beetle in that community.

A sampling method has been developed for the estimation of the numbers and weights of insects per square meter of vegetation. Briefly, this method involves a series of replicated sweeps with an insect net which was checked against the results from a box trap. The box trap was a portable plywood box open at the bottom, standing 5 ft high, and covering 1 m². On one side was a plastic funnel, to which a killing jar could be attached. When the box was dropped over the vegetation, many insects were attracted to the light visible through the plastic funnel, and were collected in the jar. The two methods yielded about the same number of insects and were used in estimation of biomass of insects.

A summary of the biomass estimates of insects is given in Table 16 for the three vegetation types. The correlation coefficient, r , was 0.26 for numbers of insects per sample vs. weight of samples. This correlation, although highly significant statistically, is too low to permit prediction of weights of samples from numbers of insects in samples. While the means for 1958 are higher than those for 1957, the differences between the years are not significant statistically. For the smartweed and sedge-rush areas the average was 239 mg/m² for these years. Estimates for old-field areas in Georgia were similar.* Estimates of biomass of insects on willow could not be checked against the box trap. By 1958 the willows had reached such size that the sweep method for estimating biomass of insects was no longer applicable.

Previous estimates of the radionuclide content of insects on White Oak Lake bed based on biomass samples,** were refined by taking larger samples in 1958. Samples were analyzed by gamma spectrometry for Cs¹³⁷, Co⁶⁰, and Ru¹⁰⁶, and then by radiochemical methods for Sr⁹⁰.

* W. H. Cross, "The Arthropod Component of Old Field Ecosystems. Herb Populations with Special Emphasis on the Orthoptera," unpublished doctoral dissertation, Univ. Ga., 1956.

** S. I. Auerbach et al., op. cit., p. 38 (July 31, 1958).

Table 16. Biomass of Herbivorous Insects on Vegetation Growing on White Oak Lake Bed

Year	Amount, mg/m ² (dry weight)					
	Sedge-Rush		Smartweed		Willow	
	Mean	Standard Error	Mean	Standard Error	Mean	Standard Error
1956			192	38		
1957	197	34	202	26	88	9
1958	273	78	330	92		

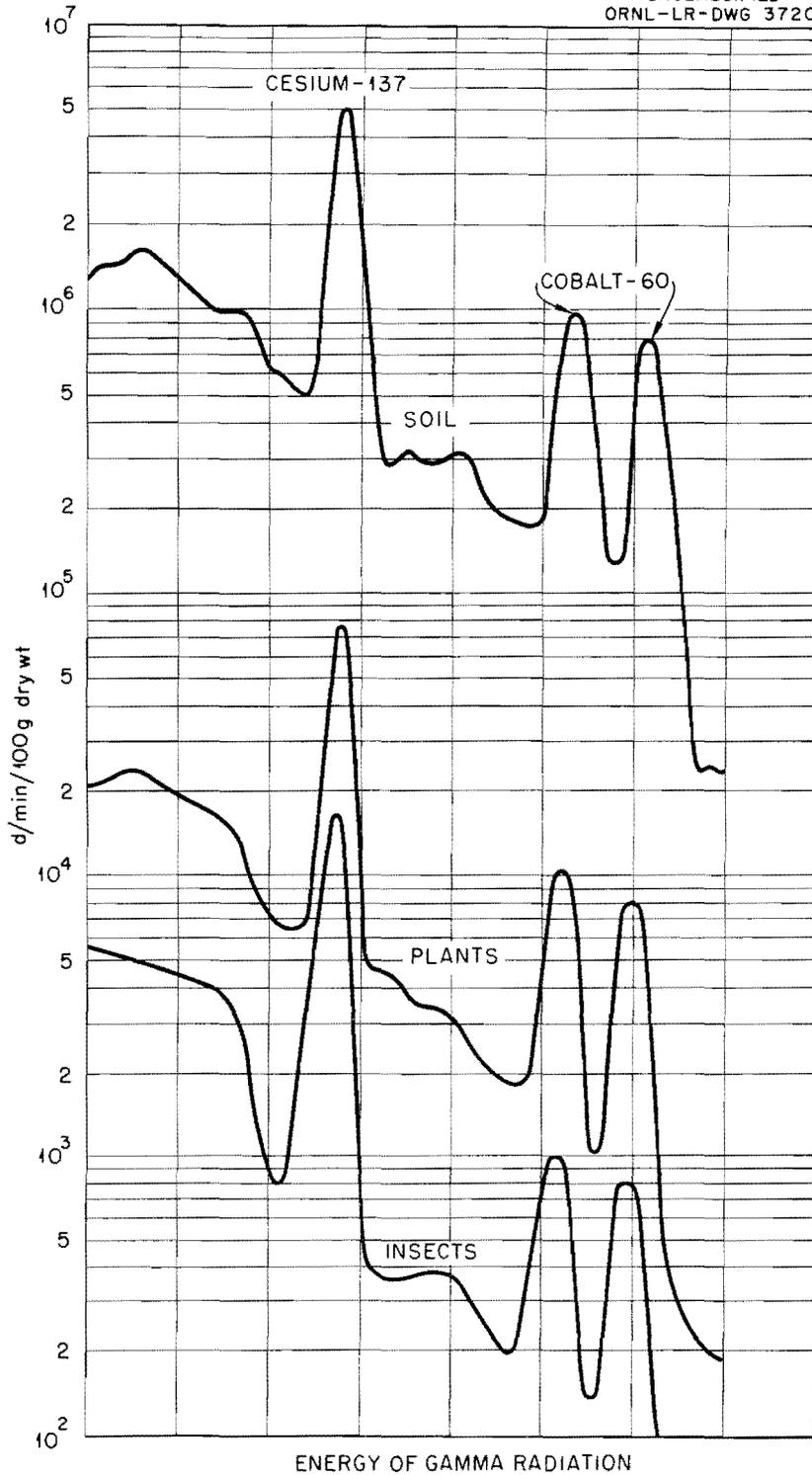
Figure 13 shows a gamma scan for insects, plants, and soil of White Oak Lake bed. In the samples of insects, Cs¹³⁷, Co⁶⁰, and Ru¹⁰⁶ were present in detectable amounts, with Cs¹³⁷ concentrations the highest; Sr⁹⁰ also was detected by radiochemical methods.

Table 17 gives the Cs¹³⁷ and Sr⁹⁰ concentrations in six groups of insects. Of these, Conocephalus and Melanoplus are grasshoppers, Systema is a flea beetle, "bees" includes a number of species, Chauligognathus is a soldier beetle, and Hemiptera-Homoptera includes mostly aphids and leafhoppers. Transfer of nuclides from plant hosts to these insect species is illustrated in Table 18. The Cs¹³⁷ concentrations in the insects are very close to the plant concentrations. Only in the case of insects feeding on Juncus is there a marked discrepancy between insect and plant concentrations of Cs¹³⁷. Since Juncus, a rush, has a low growth form, values for this species may include some soil contamination. In any case, it is evident that insects accumulate Cs¹³⁷ to such an extent that concentrations in plants and insects are similar.

Table 17. Concentrations of Sr⁹⁰ and Cs¹³⁷ in Insects Feeding on Vegetation of White Oak Lake Bed

Insect Species	Concentration, μg/g dry wt	
	Cs ¹³⁷	Sr ⁹⁰
<u>conocephalus</u>	1.50 x 10 ⁻⁴	1.40 x 10 ⁻⁴
<u>Systema</u>	1.40 x 10 ⁻⁴	1.78 x 10 ⁻⁴
Bees	0.65 x 10 ⁻⁴	0.71 x 10 ⁻⁴
<u>Chauligognathus</u>	0.62 x 10 ⁻⁴	1.40 x 10 ⁻⁴
<u>Melanoplus</u>	0.59 x 10 ⁻⁴	0.10 x 10 ⁻⁴
Hemiptera-Homoptera	0.43 x 10 ⁻⁴	0.06 x 10 ⁻⁴

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ENERGY OF GAMMA RADIATION
White Oak Lake Bed Studies.
Relative Concentration of Radioisotopes

Fig. 13.

Table 18. Transfer of Cs¹³⁷ and Sr⁹⁰ from Plants to Insects

Insect Species	Host Plants ^a (leaves)
Cs ¹³⁷	
<u>Conocephalus</u> (1.50) ←	<u>Polygonum</u> (1.75)
<u>Systema</u> (1.40) ←	<u>Polygonum</u> (1.75)
Bees (0.65) ←	<u>Bidens</u> (0.83)
<u>Chauligognathus</u> (0.62) ←	<u>Eupatorium</u> (0.78)
<u>Melanoplus</u> (0.59) ←	<u>Juncus</u> (1.05)
Hemiptera-Homopters (0.43) ←	<u>Juncus</u> (1.05)
Sr ⁹⁰	
<u>Conocephalus</u> (1.40) ←	<u>Polygonum</u> (10.4)
<u>Systema</u> (1.78) ←	<u>Polygonum</u> (10.4)
Bees (0.71) ←	<u>Bidens</u> (5.1)
<u>Chauligognathus</u> (1.40) ←	<u>Eupatorium</u> (9.8)
<u>Melanoplus</u> (0.10) ←	<u>Juncus</u> (2.6)
Hemiptera-Homopters (0.06) ←	<u>Juncus</u> (2.6)

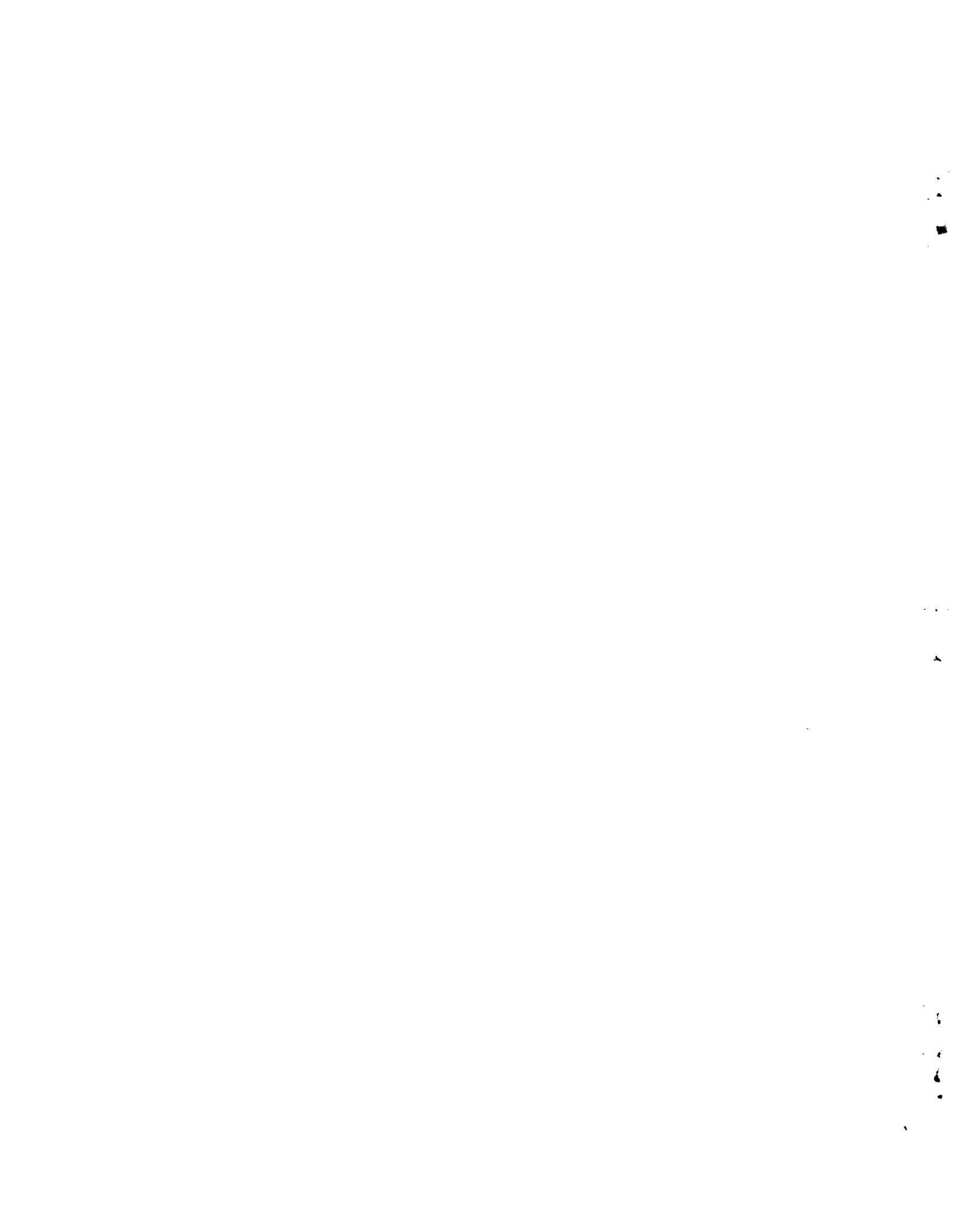
^aValues are in units of 10⁻⁴ µc/g dry weight.

In contrast, Sr^{90} concentrations in insects are nearly an order of magnitude lower than plant concentrations, but the Sr^{90} concentrations in the insects are clearly related to those in the host plants. The insects accumulate a larger amount of Cs^{137} than Sr^{90} in relation to the Cs^{137} and Sr^{90} in the host plants, and concentrations of these nuclides in insects are therefore quite similar, although Sr^{90} concentrations are higher than Cs^{137} concentrations in the plants.

These data afford an interesting example of the interplay of biological materials in the environmental cycling of fission products. White Oak Lake bed soils contain more Cs^{137} than Sr^{90} , but native vegetation accumulates higher concentrations of Sr^{90} than Cs^{137} (by a ratio of approximately 7 to 1). In the herbivorous insects, however, the process is reversed, and Cs^{137} and Sr^{90} concentrations become approximately equal.

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