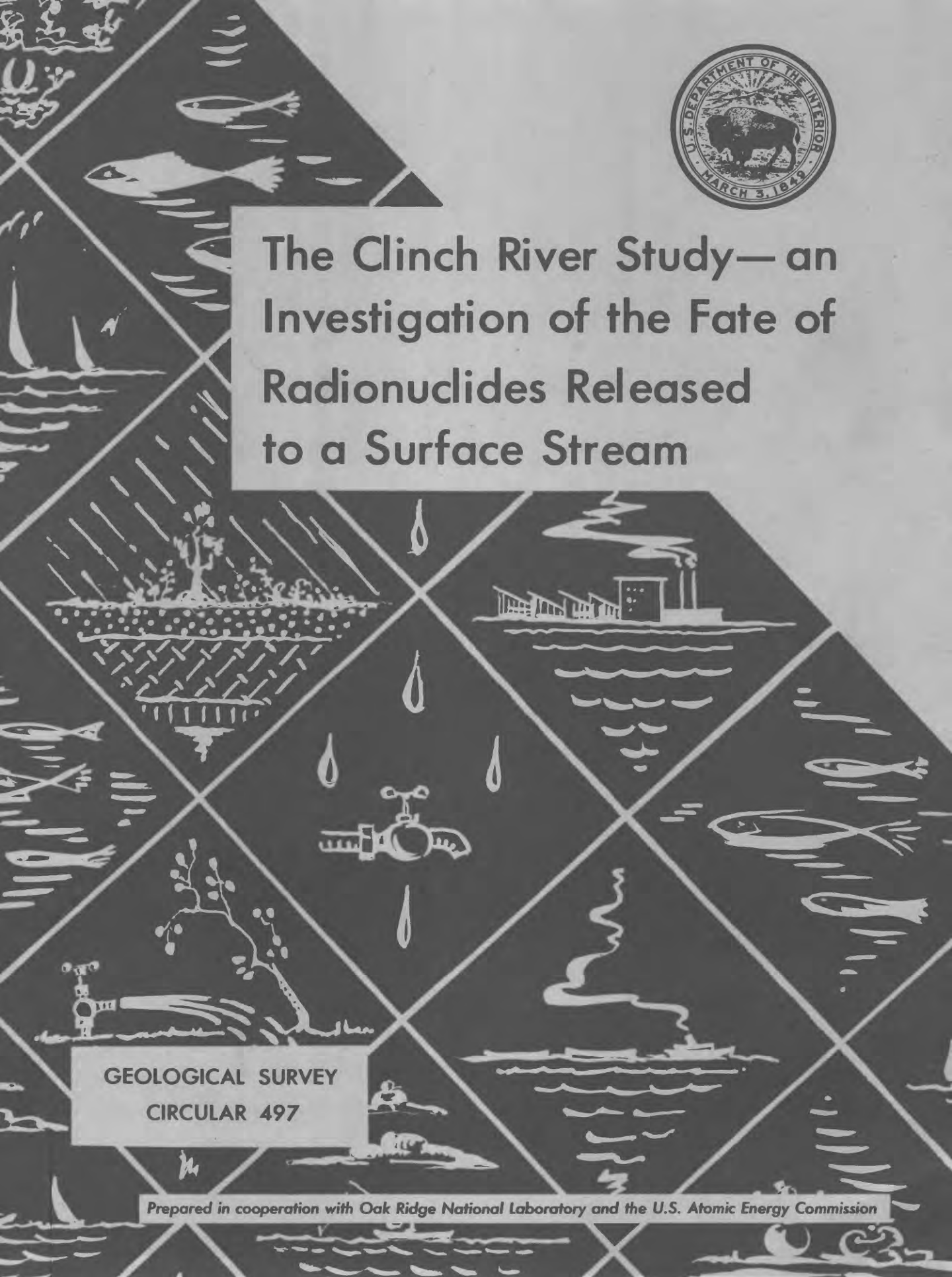

The Clinch River Study—an Investigation of the Fate of Radionuclides Released to a Surface Stream

GEOLOGICAL SURVEY
CIRCULAR 497

Prepared in cooperation with Oak Ridge National Laboratory and the U.S. Atomic Energy Commission

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By R. J. Pickering, P. H. Carrigan, Jr., and F. L. Parker

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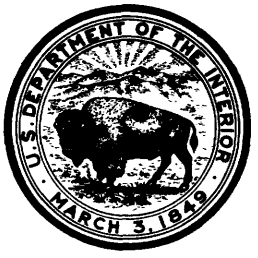
STEWART L. UDALL, SECRETARY



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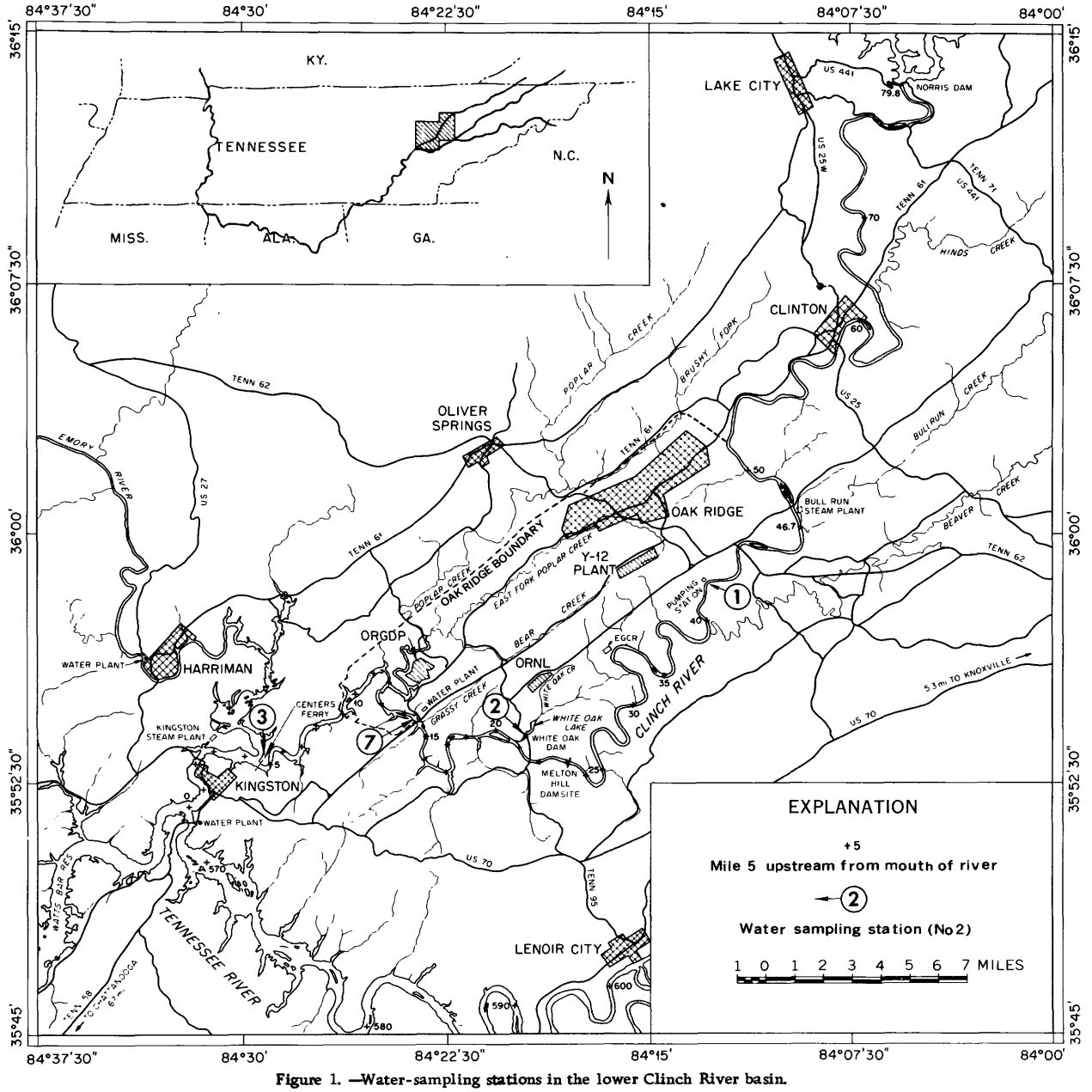

Figure 1. —Water-sampling stations in the lower Clinch River basin.

The Clinch River Study—an Investigation of the Fate of Radionuclides Released to a Surface Stream

By R. J. Pickering, P. H. Carrigan, Jr., and F. L. Parker [1]

ABSTRACT

The Clinch River Study is a multiagency effort to evaluate the physical, chemical, and biological effects of the release to the Clinch River of low-level radioactive wastes from the Oak Ridge National Laboratory. The major radionuclides released are ruthenium–106, cesium-137, cobalt-60, and strontium-90. Hydrologic and biologic studies have indicated that the radiation dosages in the river are well below maximum acceptable levels. Radionuclide concentrations in river water have been measured at seven sampling stations on the Clinch and Tennessee Rivers. Mass-balance calculations for 44 weeks of sampling indicate that losses of radionuclides from the water phase to the river-bottom sediments represent only a very small part of the total radioactivity released to the river.

A study of the Clinch River bottom-sediment cores collected in 1962 has disclosed a recurring pattern of variation in radioactivity with depth which may reflect past events in waste-disposal operations at the laboratory. Current investigations are expected to provide information about the chemical forms in which the major radionuclides exist and the mechanisms by which they were incorporated in the sediments.

INTRODUCTION

The Clinch River Study is a multiagency effort to evaluate the physical, chemical, and biological effects caused by the disposal in the Clinch River of low-level radioactive wastes from the Oak Ridge National Laboratory[2] at Oak Ridge, Tenn. The study, begun in 1959, has the following objectives:

1. To determine the fate of the radioactive materials currently being discharged to the river.
2. To determine and understand the mechanisms of dispersion of radionuclides released to the river.
3. To evaluate the direct and indirect hazards of current disposal practices in the river.
4. To evaluate the overall usefulness of this river for radioactive-waste-disposal purposes.
5. To recommend long-term monitoring procedures.

The study is under the general supervision of a steering committee composed of representatives of the Oak Ridge National Laboratory (ORNL), the Tennessee State Department of Public Health (TDPH), the Tennessee State Game and Fish Commission (TGFC), the Tennessee Valley Authority (TVA), the U.S. Atomic Energy Commission (AEC), the U.S. Geological Survey (USGS), and the U.S. Public Health Service (USPHS) (Morton, 1961). Investigations are carried out by personnel of the various agencies under the direction of four subcommittees of the steering committee, namely: Aquatic Biology, Safety Evaluation, Water Sampling and Analysis, and Bottom Sediment Sampling and Analysis.

The Clinch River heads in Virginia and flows southwestward to join the Tennessee River at Kingston, Tenn. (fig. 1). At its mouth the drainage area of the Clinch River is 4,413 square miles. At the farthest downstream gaging station, 39.0 miles upstream from its mouth, the average flow of the Clinch River is 4,612 cfs (cubic feet per second), based on a 26-year period of measurement.

During the first 3 years of the Clinch River Study, Norris Dam, at mile 79.8 on the Clinch River (CRM [3] 79.8) was the only major control structure on the river. Releases from

[1] R. J. Pickering and P. H. Carrigan, Jr., U.S. Geological Survey, and F. L. Parker, Oak Ridge National Laboratory.

[2] Operated by Union Carbide Corp. for the U.S. Atomic Energy Commission.

[3] The abbreviation "CRM" (Clinch River Mile) followed by a number has been used in this report to designate distance upstream, in miles, from the mouth of the river. This terminology is consistent with prior usage in the Clinch River Study (Morton, 1961).

Norris Reservoir are largely made on the basis of demand for electric power. In May 1963 control of flow began at Melton Hill Dam which is at CRM 23.1 on the Clinch River, 2.3 miles upstream from the mouth of White-oak Creek, where radioactive waste from ORNL enters the river. The Melton Hill system is to be a "peaking" powerplant, which is one that is operated mainly during periods of peak demand for power and shut down at other times.

The Clinch River below Whiteoak Creek is affected by backwater from Watts Bar Dam, which is on the Tennessee River 37.8 miles downstream from the mouth of the Clinch River and a total of 58.6 miles downstream from the mouth of Whiteoak Creek. Before construction of Melton Hill Dam, thermally stratified flow occurred in the lower Clinch River from spring to fall because the cold water released from Norris Reservoir flowed under the relatively still, warmed backwater of Watts Bar Reservoir. The effect of the operation of Melton Hill Dam on temperature conditions in the river is not yet known.

Since 1943, low-level radioactive wastes derived from the Laboratory's Process Waste Water Treatment Plant, solid-waste burial grounds, liquid-waste seepage pits, and other minor sources of radioactivity have entered the Clinch River through Whiteoak Creek (fig. 1). The radionuclides in the wastes are largely fission products—cesium-137, strontium-89 and 90, ruthenium-103 and 106, and others—but also include some activation products, such as cobalt-60 and zinc-65. Continous monitoring of radionuclide concentrations at the point of release to the river and downstream at CRM 4.5 has shown that concentrations are well within the limits recommended by the National Committee on Radiation Protection.

Whiteoak Creek drains an area of 6 square miles, which includes the Laboratory area. Its average discharge is about 10 cfs, most of which is derived from surface runoff. At the present time, its waters are impounded in Whiteoak Lake by Whiteoak Dam, which is downstream from all sources of radioactive wastes and 0.6 mile upstream from the Clinch River. The area covered by Whiteoak Lake at the present elevation of the gates, 744 feet above mean sea level, is 19.1 acres. The use of Whiteoak Lake for impoundment of radioactive-waste water was described in more detail by Morton (1961).

AQUATIC BIOLOGY AND SAFETY EVALUATION

The radionuclide content of plant and animal life in the Clinch River has been studied by the Ecology Section, Health Physics Division, ORNL, and the Radiological Health Research Activities Group, Radiological Health Division, USPHS. All preliminary estimates of public consumption of fish, ingestion of water, and exposure due to use of the river for recreational purposes indicated radiation dosages that were below maximum permissible levels. Calculation of estimated future dose rates is continuing. Results of the biological studies were reported by Morton (1961, 1962a, 1962b, 1963).

HYDRAULIC STUDIES

The capacity of the Clinch River to disperse radioactive materials has been assessed through a series of hydraulic tests. Radioactive gold-198 was used as a tracer in the early tests, whereas rhodamine-B and pontacyl brilliant pink-B fluorescent dyes have been used in more recent tests. In these tests, the longitudinal distance downstream to the cross section of complete lateral diffusion, the variation of peak concentration with distance, and the times of travel have been determined. Times of water travel observed in the tests compare closely with times computed on the basis of the continuity equation. Nearly uniform lateral mixing occurs within 4–6 miles downstream from the mouth of Whiteoak Creek. Variation of concentration with distance is shown in figure 2 for two of the tracer tests.

WATER SAMPLING AND ANALYSIS

A network of seven water-sampling stations on the Clinch and Tennessee Rivers was established under the direction of the Subcommittee on Water Sampling and Analysis. Samples were collected regularly at the stations for material-balance studies of the radionuclides released to the river system and for information on the stable-chemical composition of the river water. Station locations are listed in table 1. Water samples were composited weekly on the basis of water discharge and were analyzed for major and minor radioactive- and stable-chemical constituents. Analyses of stable-chemical constituents were made by personnel of the Tennessee Stream Pollution Control Board at Nashville, Tenn., and radiochemical analyses, by personnel of the Division of Radiological

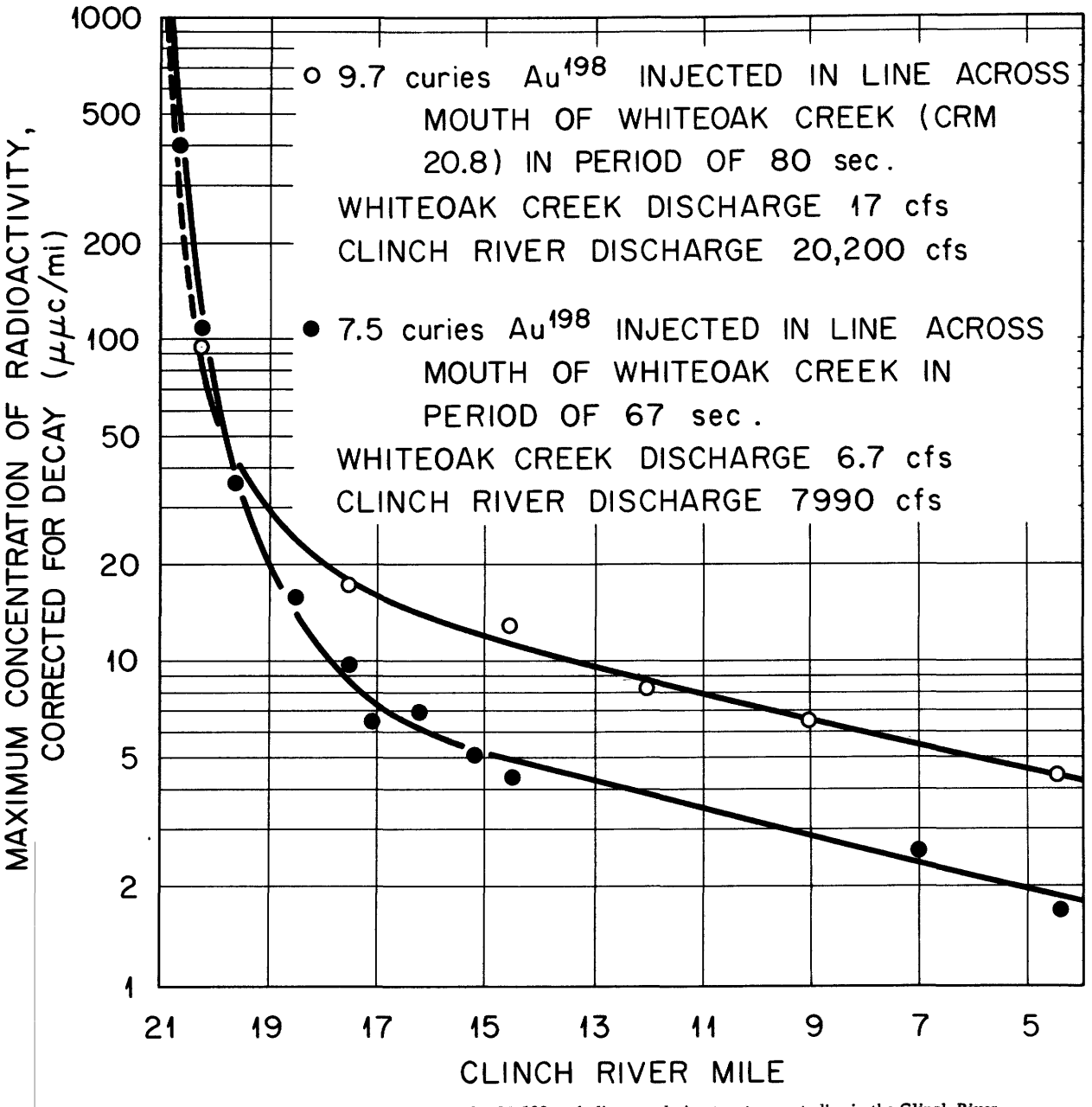

Figure 2.—Reduction of maximum concentration of gold-198 with distance during two tracer studies in the Clinch River.

Table 1.—*Water-sampling stations, Clinch River Study*

Station no.	Location	Stream	Stream mile (above mouth)
1	Oak Ridge water-treatment-plant intake	Clinch River	41.5
2	Whiteoak Dam	Whiteoak Creek	.6
3	Centers Ferry	Clinch River	5.5
4	Loudon, Tenn	Tennessee River	591.8
5	Watts Bar Dam	do	529.9
6	Chickamauga Dam	do	471.0
7	Oak Ridge Gaseous Diffusion Plant, water-treatment-plant intake.	Clinch River	14.6

Health, USPHS, at the Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio. The major radionuclides in the Clinch River water are ruthenium-106, cesium-137, cobalt-60, and strontium-90. Most of the cesium-137 is associated with suspended sediment having a diameter greater than 0.7 micron, whereas the other three radionuclides are associated with finer suspended sediment or are dissolved in the water.

The amount, or load, of a given radionuclide which passes a sampling point during a short period of time can be assumed to be equal to the product of the average radionuclide concentration at the sampling point during that period of time and the flow of water that passes the point during the same period. If there were no loss of the radionuclide from the Clinch River water as it moved downstream, the load passing the "background" station at the Oak Ridge water-treatment plant at CRM 41.5 plus the load released through the Whiteoak Creek sampling station at Whiteoak Dam should equal the load passing the sampling station at CRM 5.5 (see fig. 1).

The results of mass-balance (load) calculations (table 2; figs. 3, 4, 5, 6) indicate that during the study period virtually all the ruthenium-106 in the Clinch River water was transported downstream into the Tennessee River. A small part of the strontium-90 in the river water was apparently lost in 15 miles of downstream movement. The calculated loads of cesium-137 and cobalt-60, which show apparent gains between the point of release to the river and the downstream sampling station, cannot be considered reliable because most of the reported concentrations of the two nuclides in samples from

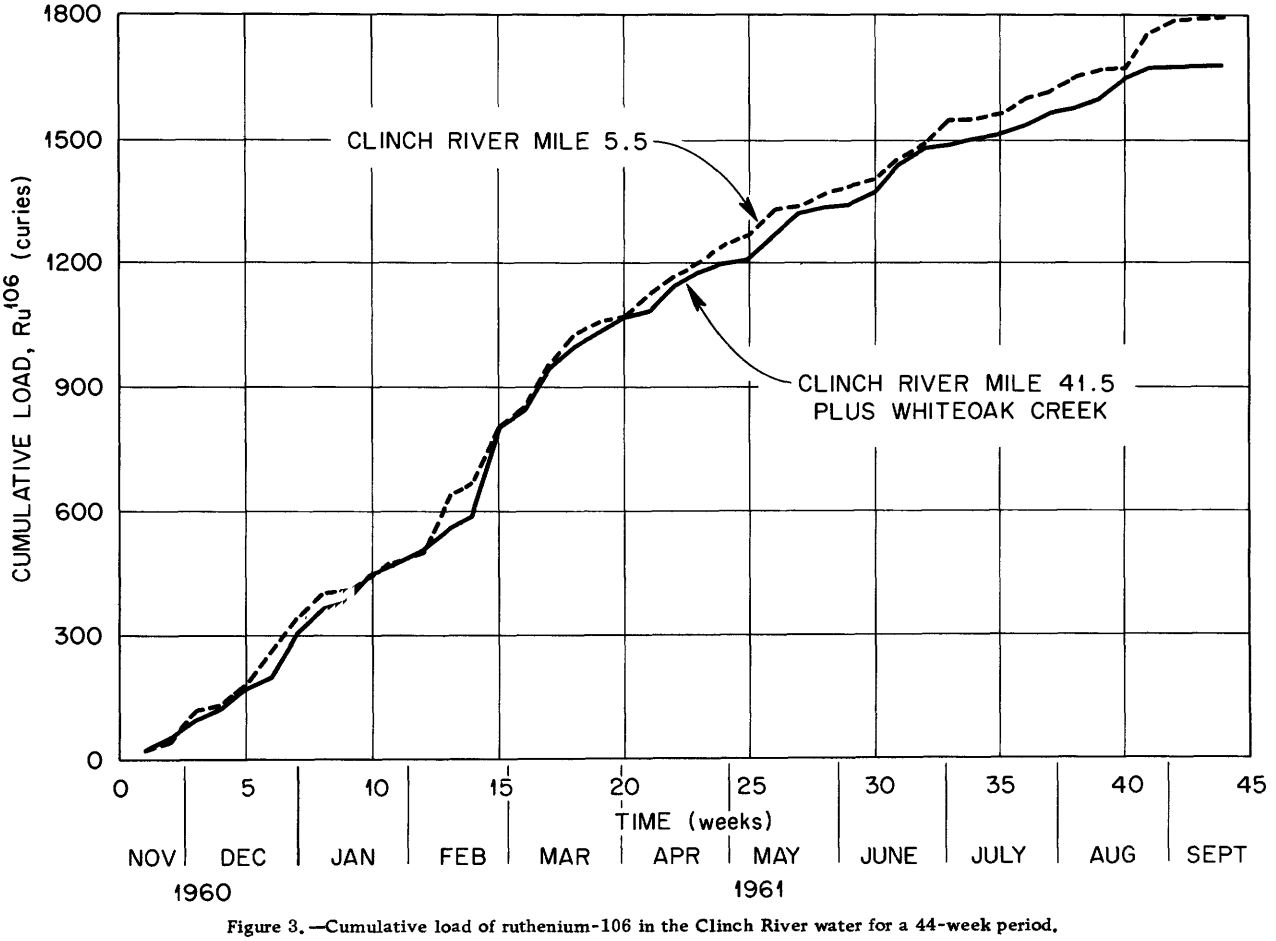

Figure 3.—Cumulative load of ruthenium-106 in the Clinch River water for a 44-week period.

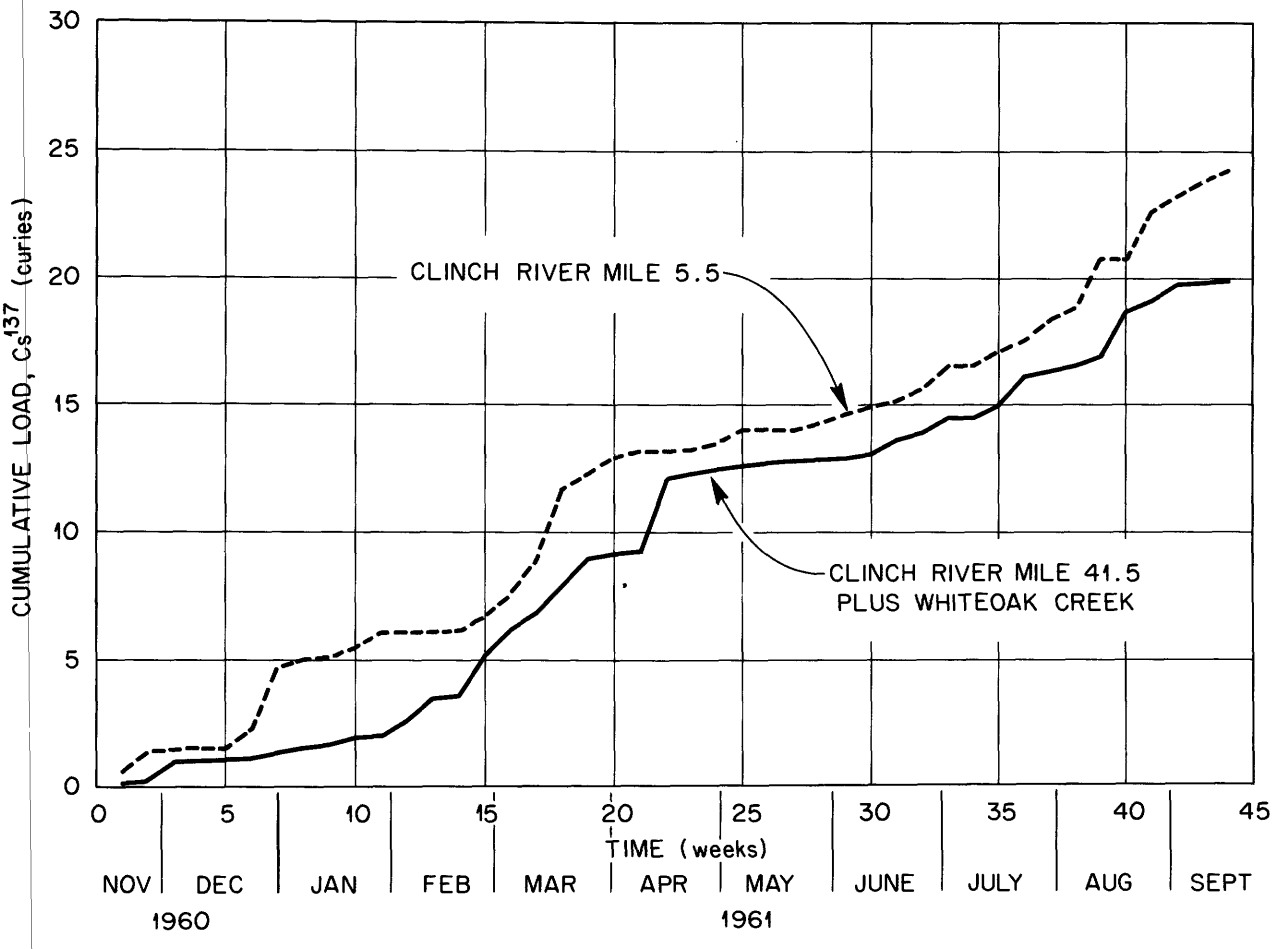

Figure 4.—Cumulative load of cesium-137 in the Clinch River water for a 44-week period.

CRM 5.5 (32 of 44 determined cesium-137 concentrations and 26 of 44 determined cobalt-60 concentrations) were at or below the lower limit of detectability for the analytical procedure used.[4] True gains may have occurred as a result of local movement of bottom sediment or desorption reactions, but this cannot be proved or disproved with the available data.

Stable-chemical analyses of the Clinch River water from the sampling station at the Oak Ridge water-treatment plant show the water to have an average hardness (as calcium carbonate) of 106 ppm (parts per million), a dissolved-solids content of 125 ppm, and a pH of 7.7. The Clinch River water may be classed as moderately hard. On the basis of its chemical composition, which does not vary greatly, it may be called a calcium bicarbonate type of water.

[4] Concentrations of ruthenium-106, cesium-137, and cobalt-60 in water samples were determined by gamma-spectrum analysis programmed through a digital computer. Strontium-90 concentrations were determined by wet-chemical separation and beta counting.

Table 2.—*Cumulative radionuclide loads in the Clinch River water for the period November 13, 1960, to September 16, 1961*

Radionuclide	Cumulative load (curies)		Gain or loss (percent)
	Mile 41.5 plus Whiteoak Dam	Mile 5.5	
Ruthenium-106	1680	1800	+ 7
Cesium-137	19.9	24.3	+22
Cobalt-60	24.8	26.3	+ 6
Strontium-90	27.7	24.1	-13

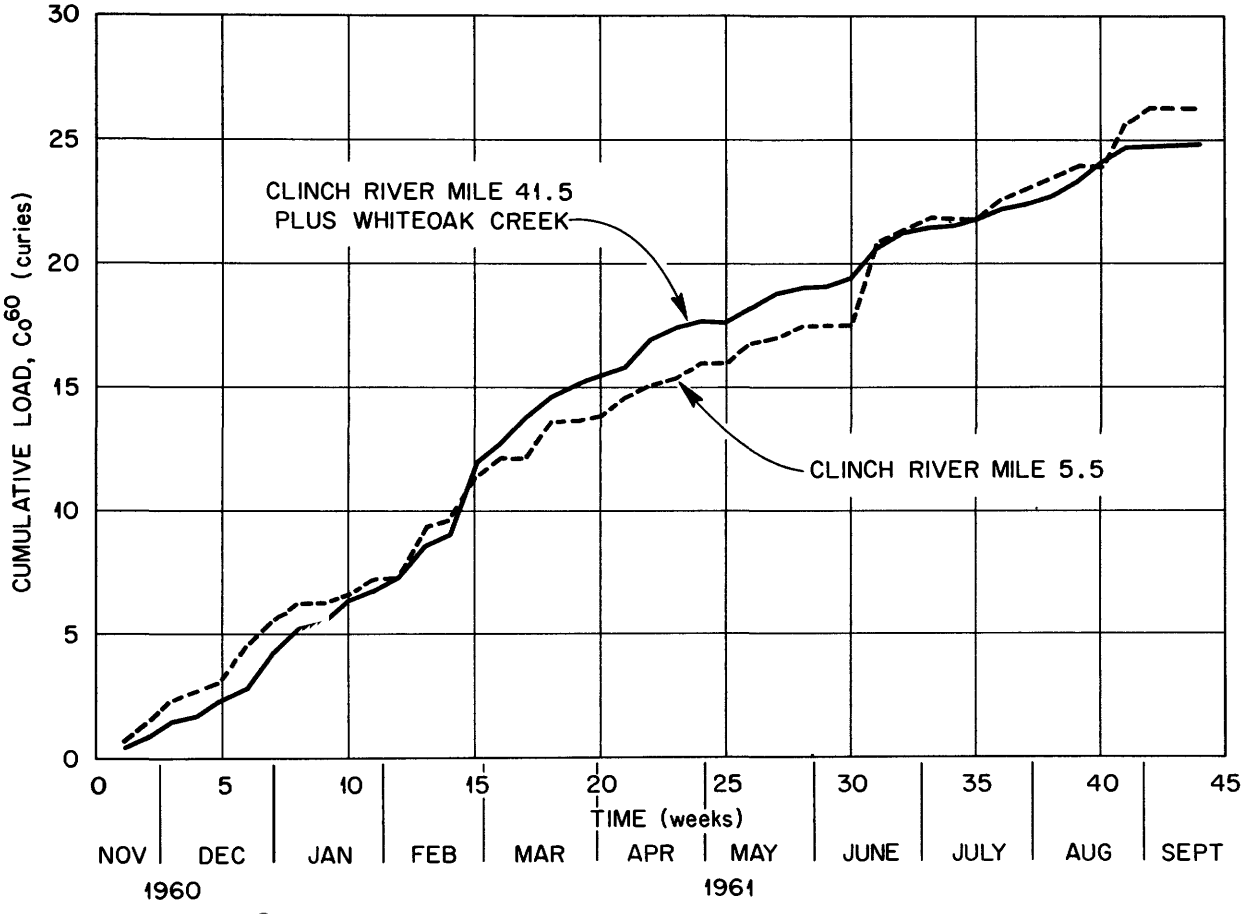

Figure 5.—Cumulative load of cobalt-60 in the Clinch River water for a 44-week period.

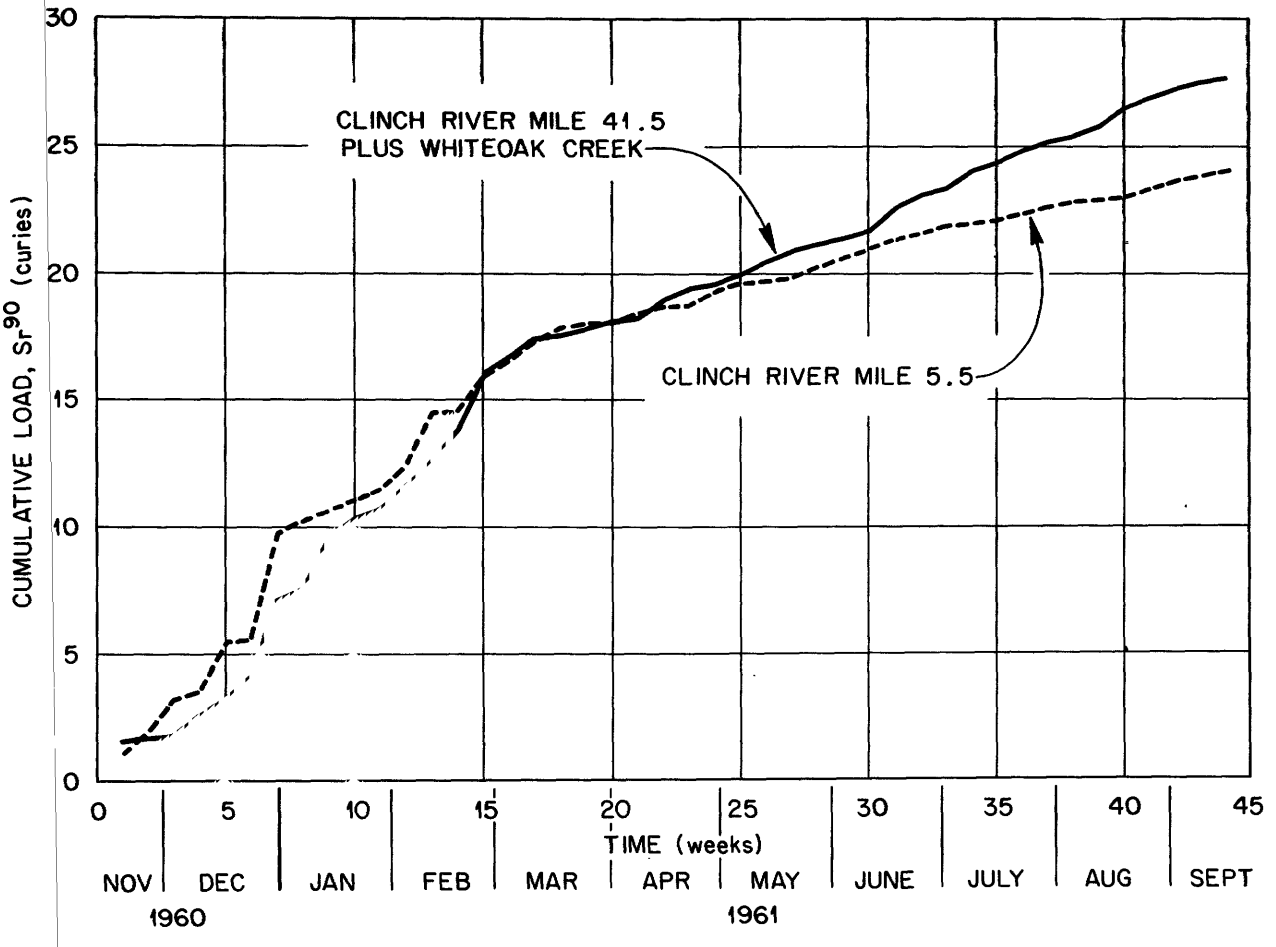

Figure 6.—Cumulative load of strontium-90 in the Clinch River water for a 44-week period.

BOTTOM-SEDIMENT SAMPLING AND ANALYSIS

Cross-section composite samples, obtained from five cross sections in the Clinch River downstream from Whiteoak Creek, have shown the particle-size distribution in the bottom sediments to be 15–20 percent clay (<2 microns), 50–60 percent silt (2–62 microns), and 25–30 percent sand (>62 microns) (Morton, 1962b). The clay-size fraction of the sediment is composed largely of vermiculite, mica,[5] kaolinite, randomly interstratified vermiculite-mica, and fine-grained quartz. The sand and silt fractions of the sediment are composed almost entirely of quartz. An appreciable content of calcium carbonate in the sediment has been indicated by indirect evidence, but relative concentrations have not yet been determined.

The uptake, or sorption (McBain, 1950), of chemical constituents from liquid systems by natural sediment-forming minerals has been investigated extensively during the past several years. Sorption of radionuclides by river sediments was studied by Carritt and Goodgal (1953), Barker (1958), Jacobs and Tamura (1960), and others. The importance of radiocesium uptake by the clay minerals has been well established, and Tamura and Jacobs (1960) showed that the 2:1-layer clay minerals having a c spacing of 10 A selectively sorb cesium. Tamura (1963) further showed that hydrous iron oxides and aluminum oxides, which are common minor constituents in many sediments, selectively sorb strontium. The well-known fact that certain radionuclide-sorption reactions are reversible has been emphasized by Reynolds' (1963) description of desorption reactions which occur when fresh-water sediments enter a radically different chemical environment, such as the ocean. Work on radionuclide uptake by river sediments has been summarized by Sayre and others (1963) and by Kornegay and others (1963).

[5]No distinction is made here between mica and illite.

Table 3.—*Concentrations of specific radionuclides in the upper part of the Clinch River bottom sediments*

Clinch River mile	Radionuclide concentration (10^{-2} μc/kg)				
	Ruthenium-106[1]	Cesium-137[1]	Cobalt-60[1]	Strontium-90[2]	TRE[2 3]
4.7	1.05	6.22	0.788	0.086	0.99
5.8	1.91	10.6	1.42	.194	2.28
6.9	1.43	7.21	.995	.158	1.30
8.0	4.13	11.2	1.55	.189	4.86
9.0	3.34	9.95	1.03	.171	4.42
10.0	1.81	7.97	.752	.113	2.14
11.0	4.50	19.7	1.89	.423	7.03
12.0	2.81	11.9	1.24	.212	5.40
13.0	3.12	17.6	1.84	.252	7.66
14.0	1.33	10.6	1.19	.194	2.90
14.6	4.41	18.1	1.85	.347	5.63
15.3	2.16	9.77	.977	.149	3.11
16.0	.815	3.54	.347	.077	1.50
16.9	5.99	13.2	1.22	.140	5.86
18.1	2.12	16.1	1.29	.230	3.59
19.5	2.79	7.12	.923	.108	2.28
20.8	3.90	106	8.32	1.68	20.4
21.6	2.54	7.03	.653	.144	3.07
22.5	1.36	2.48	.334	.009	1.40

[1]Gamma-spectrometer analysis.
[2]Chemically separated before counting.
[3]Trivalent rare earths plus yttrium-90.

Surveys of radioactivity in the Clinch River bottom sediments have been conducted annually since 1951 by the Applied Health Physics Section of the Health Physics Division, ORNL, and have indicated that a part of the radionuclides released to the river has been retained in the bottom sediments (Garner and Kochtitzky, 1956; Cottrell, 1959). Samples collected by the Applied Health Physics Section and the USPHS and a bottom-sediment-coring program conducted in 1960 by personnel of the Radioactive Waste Disposal Research Section of the Health Physics Division, ORNL (table 3), have provided a measure of the content of cesium-137, ruthenium-106, cerium-144, zinc-65, cobalt-60, zirconium-niobium-95, strontium-90, and the trivalent rare earths in the upper part of the sediments (Morton, 1962b).

Although material balances computed from analyses of water samples indicate that the mass of radionuclides in bottom sediments represents only a small part of the total mass released to the river, physical and chemical processes in the river have concentrated the radionuclides in the bottom sediments enough to make them easily measureable. In areas of the river where continuous sedimentation has taken place, the sediments provide a record of net radionuclide removal from river water as it passed downstream throughout the period of release from the Laboratory.

In the summer of 1962 a comprehensive sampling of the Clinch River bottom sediments through the entire thickness of the radioactive zone was conducted by personnel of the USGS and ORNL. The samples were obtained through the use of the Swedish Foil Sampler (Pickering, 1965), a device which takes undisturbed sediment cores 2-1/2 inches in diameter. The sampler utilizes a piston and thin steel strips (foils) to decrease compaction due to friction between the sample tube and the sediment while the core is being taken and to prevent loss of core while the core tube is being raised to the surface. More than 150 cores were obtained at 14 cross sections on the Clinch River and

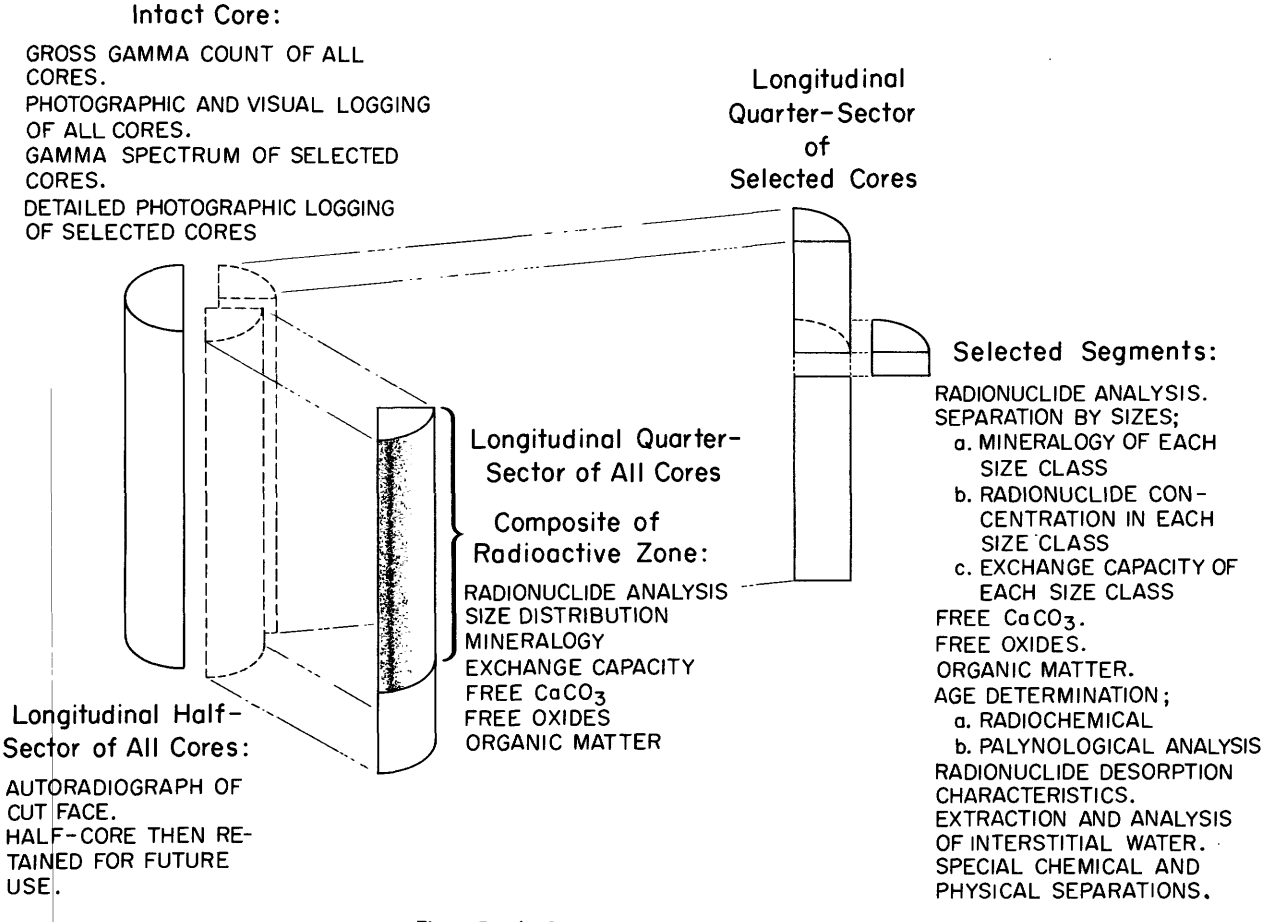

Figure 7.—Analysis of bottom-sediment cores.

4 cross sections on two tributary streams. Core recovery was excellent at most sites. The longest core obtained was 14 feet long.

Plans for processing the cores are indicated diagrammatically in figure 7. It is expected that the analyses listed will provide sufficient information to determine the following:

1. The total load of each of the major radionuclides in the Clinch River bottom sediments.
2. The distribution of the major radionuclides in the sediments.
3. The distribution of the various size, mineralogical, and organic fractions of the sediments.
4. The relationships between the distribution of the major radionuclides and the composition of the sediments.
5. The chemical form of each of the major radionuclides in the sediments.

Determination of the total load of radionuclides in the Clinch River bottom sediments, when compared with total releases to the river from Whiteoak Creek since 1943 (14,000 curies), will permit a quantitative estimate of the effectiveness of the sediments in removing radionuclides from the river water. It will also provide a check on material-balance calculations based on analyses of water samples. The other determinations listed in figure 7 are directed primarily toward an understanding of the mechanisms by which the radionuclides were incorporated in the sediments. Sorption, particle sedimentation, and chemical precipitation were probably the principal mechanisms involved.

The lower limit of the radioactive zone and the vertical distribution of gross gamma radioactivity in the cores collected in 1962 have been determined by scanning each intact core with a device known as a core scanner (fig. 8). The core scanner consists of a 3 x 3-

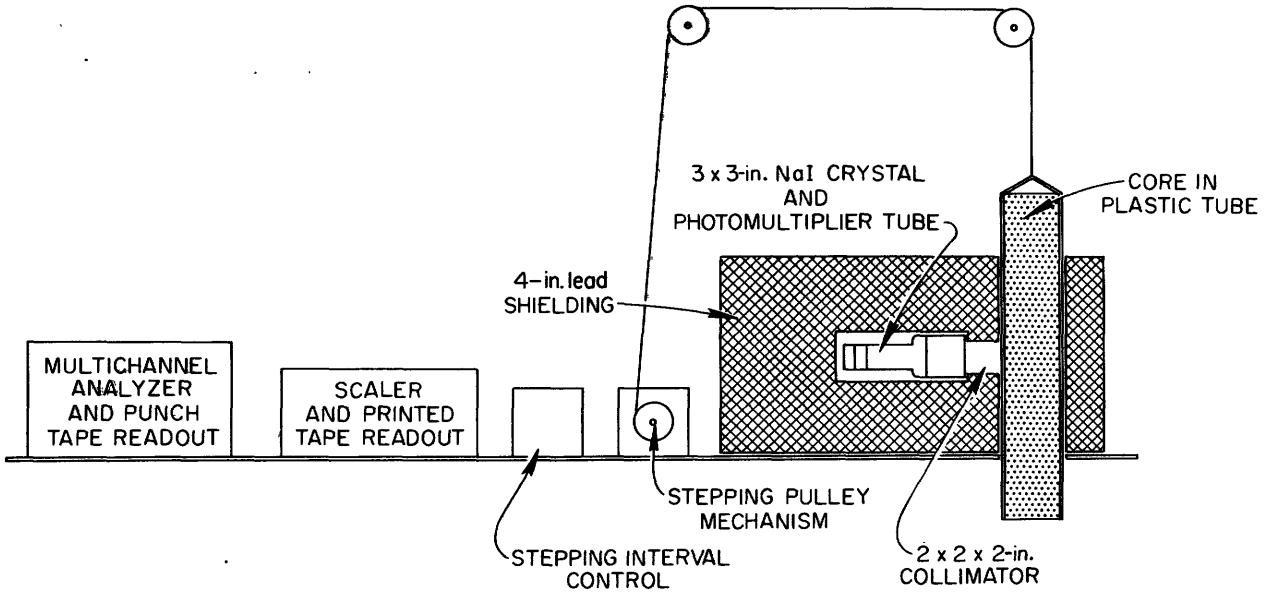

Figure 8.—Core scanner.

inch sodium iodide scintillation crystal and matched phototube enclosed in a 4-inch-thick cylindrical lead shield. A collimating opening, 2 x 2 x 2 inches, leads from the core "well" to the scintillation crystal. A calibrated hoist automatically moves the core vertically past the collimator in 2-inch increments. The phototube output can be routed either through a scaler for determination of gross gamma radioactivity or to a multichannel analyzer for determination of the concentration of individual gamma-emitting radionuclides in each increment.

Results of gross gamma scanning of cores from CRM 7.5 are shown in figure 9. In the cross section note the distribution of the radioactive sediments and the recurring pattern of gross gamma activity with depth in several of the cores. The results of visual logging and the determination of the vertical distribution of individual radionuclides in selected cores are hoped to provide information which can be used to correlate specific sediment horizons in some of the cores with specific past events in waste disposal operations at the Laboratory. The existence of other "dateable" physical or chemical properties of the sediments will also be investigated.

STATUS OF THE CLINCH RIVER STUDY

Since the Clinch River Study was initiated in 1959, considerable progress has been made toward meeting the objectives listed in the introduction of this paper. Material-balance studies have shown that most of the radioactivity discharged to the Clinch River is transported downstream in the Clinch and Tennessee Rivers in the water phase. Very low radionuclide concentrations in the lower Tennessee River make detection farther downstream impractical. A small percentage of the radioactivity has been incorporated in bottom sediments of the two rivers. A part of the radioactivity in both the water and bottom sediments is associated with organic matter.

Time-of-travel and dispersion studies have outlined some of the hydrologic characteristics of the Clinch River which affect the concentration of radioactive waste passing downstream points. A few additional tests after power releases begin at Melton Hill Dam will satisfactorily complete this phase of the study.

Laboratory and field studies have indicated that cesium-137 is introduced to the river and transported largely as a sorbed ion on clay-mineral particles. Because this particular sorption reaction is not readily reversible under fresh-water conditons, the incorporation of cesium in river-bottom sediments is probably controlled largely by sedimentation processes. Results of desorption, exchange capacity, and other tests being performed on Clinch River bottom-sediment samples will provide further information on the chemical form of the other radionuclides in the sediments and on the overall usefulness of the river for disposal of radioactive wastes.

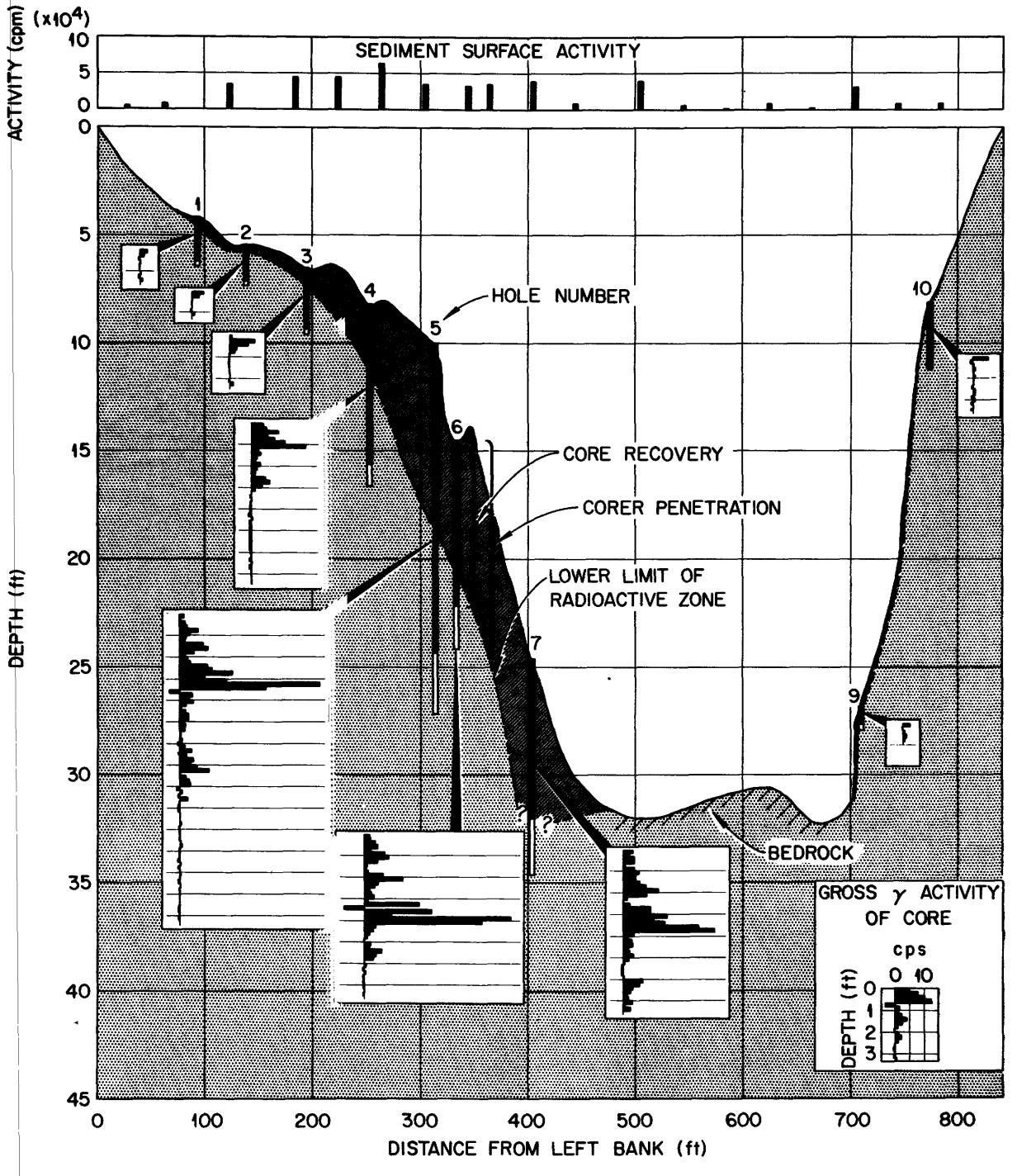

Figure 9. —Cross section at CRM 7.5 showing penetration, recovery, and gross-gamma-radioactivity variations with depth for 1962 bottom-sediment core samples (vertical exaggeration, 10:1). Cps, counts per second; cpm, counts per minute.

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