Distribution of Radionuclides in Bottom Sediment of the Clinch River Eastern Tennessee

---

GEOLOGICAL SURVEY PROFESSIONAL PAPER 433-H

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

U.S. DEPARTMENT OF THE INTERIOR

Distribution of Radionuclides in Bottom Sediment of the Clinch River Eastern Tennessee

By R. J. PICKERING

TRANSPORT OF RADIONUCLIDES BY STREAMS

GEOLOGICAL SURVEY PROFESSIONAL PAPER 433-H

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

U.S. DEPARTMENT OF THE INTERIOR

UNITED STATES GOVERNMENT PRINTING OFFICE, WASHINGTON : 1969

UNITED STATES DEPARTMENT OF THE INTERIOR

WALTER J. HICKEL, *Secretary*

GEOLOGICAL SURVEY

William T. Pecora, *Director*

For sale by the Superintendent of Documents, U.S. Government Printing Office
Washington, D.C. 20402 - Price 40 cents (paper cover)

CONTENTS

ILLUSTRATIONS

TABLES

TRANSPORT OF RADIONUCLIDES BY STREAMS

DISTRIBUTION OF RADIONUCLIDES IN BOTTOM SEDIMENT OF THE CLINCH RIVER, EASTERN TENNESSEE

By R. J. Pickering

ABSTRACT

Radioactive bottom sediment in the Clinch River in eastern Tennessee was investigated through a study of the distribution of radioactivity in sediment cores. The cores were taken from a 21-mile-long reach of river downstream from Oak Ridge National Laboratory. Low-level-radioactive liquid wastes are released from the Laboratory to the Clinch River via Whiteoak Creek.

In the upper 8 miles of the reach, radioactive sediment is found only along the sides of the stream channel, but in the lower 13 miles of the river, it extends out into the main part of the channel as well and attains its greatest thicknesses. Several sediment cores from the lower part of the river showed the same general pattern of variation of gross gamma radioactivity with depth. This pattern resembled the pattern of annual releases of cesium-137 from the Oak Ridge National Laboratory. Statistical comparison of the patterns confirmed the similarity.

Measurements of the vertical distribution of the major gamma-ray-emitting radionuclides in nine cores showed that approximately 81 percent of the gamma radioactivity was due to cesium-137, 12 percent to cobalt-60, and 7 percent to ruthenium-106. The vertical distribution of cobalt-60 correlated strongly with that of cesium-137.

It is concluded that cesium-137 and cobalt-60 were incorporated in Clinch River bottom sediment by deposition of suspended radioactive solids which entered the river from Whiteoak Creek. The two radionuclides are not necessarily associated with the same solids, and cobalt-60 in the bottom sediment probably represents only a small fraction of that released to the river.

A study of the physical and chemical compositions of sediment samples from two cores showed that cation-exchange properties of the sediment are controlled largely by its content of mica and clay minerals. These minerals are found primarily in the finest sediment fraction, but can occur also in the coarser fractions in the form of mineral aggregates and shale particles. Calcium and magnesium are the major leachable cations in the sediment; potassium and sodium are not present in measurable quantities. The effects of chemical and physical properties of the sediment on its radionuclide content are obscured as a result of variations in annual releases of the radionuclides and as a result of dilution of radioactive sediment by nonradioactive sediment in the river. Compositional effects were discernible only after corrections were made for variations in annual releases of radionuclides.

INTRODUCTION

This report describes one of several contributions of the U.S. Geological Survey to the Clinch River Study. The Clinch River Study was a multiagency effort to evaluate the past, present, and future use of the Clinch River for disposal of low-level-radioactive liquid waste from the Oak Ridge National Laboratory, which is in eastern Tennessee (Morton, 1961, 1962b, 1963; Pickering and others, 1965; Struxness and others, 1967) and is operated by the Union Carbide Corp. for the U.S. Atomic Energy Commission. The agencies that participated in the study are: Oak Ridge National Laboratory (ORNL); Tennessee Game and Fish Commission; Tennessee State Department of Public Health, Stream Pollution Control Board; Tennessee Valley Authority (TVA); U.S. Atomic Energy Commission (AEC); U.S. Geological Survey (USGS); and U.S. Public Health Service (PHS).

When the study was begun in 1960, the following objectives were established by the Clinch River Study Steering Committee, an advisory group composed of representatives of each of the participating agencies (Morton, 1963, p. 1):

1. To determine the fate of radioactive materials currently being discharged to the Clinch 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 the river for radioactive waste disposal purposes.
5. To provide appropriate conclusions regarding long-term monitoring procedures.

Work described in this report was part of a cooperative program with the Health Physics Division, ORNL;

the Oak Ridge Operations Office, AEC; and the Division of Reactor Development, AEC.

The release of low-level-radioactive liquid waste to the Clinch River via Whiteoak Creek (fig. 1), which drains the Oak Ridge National Laboratory area, was begun soon after establishment of the Laboratory in 1943 for the processing of radioactive materials. Radioactive liquids have entered Whiteoak Creek as a result of direct releases of processed waste water from the Laboratory, seepage from liquid-waste holdup pits, and drainage from solid-waste-disposal trenches (Browder, 1959). This waste-disposal practice has resulted in the incorporation of some of the introduced radionuclides (Glasstone, 1958, p. 213) in bottom sediment in the reach of river downstream from the mouth of Whiteoak Creek (Cottrell, 1959; Pickering and others, 1966).

Throughout most of the Laboratory's history, the waters of Whiteoak Creek have been impounded in Whiteoak Lake by Whiteoak Dam, which is located 0.6 mile upstream from the mouth of the creek. The lake was created as a holdup facility for the radioactive waste carried in the creek water. Radioactive waste waters in Whiteoak Creek flow into the Clinch River at a point 3.3 miles downstream from the Laboratory area. The diluted wastes in the Clinch River flow into the Tennessee River 20.8 miles downstream from the entry of Whiteoak Creek.

The continuous release of radioactive wastes to the Clinch River during nearly 20 years of Laboratory operations has provided a unique opportunity for studying the effects of such releases on the river and the effects of physical, chemical, hydrologic, and biological characteristics of the river on the fate of the radioactive materials.

A knowledge of the amount, distribution, and nature of the radioactive sediment was needed for determining the fraction of the released radionuclides that is being retained in the river system, and the locus and duration of retention. With such information, an estimate can be made of present and potential radiation hazards resulting from the use of the river for disposal of radioactive wastes. An evaluation of the advisability of continued use of the river for such disposal can be made also. Knowledge of the physical, mineralogical, and chemical composition of the radioactive sediment was required for an understanding of the mechanisms whereby the various radionuclides were incorporated in the sediment.

A series of core samples of radioactive bottom sediment was taken at 14 sampling sections in the Clinch River and at two sampling sections in each of two tributary streams—Poplar Creek and the Emory River (fig. 1), by means of a Swedish Foil Sampler. Sampling sections were selected for adequate coverage of the study reach and, wherever possible, were made to coincide with sediment ranges used by TVA for periodic measurements of bottom-sediment accumulation or erosion. Two sampling sections were located just upstream and just downstream from the mouth of Poplar Creek to assess the effect of that tributary on bottom-sediment radioac-

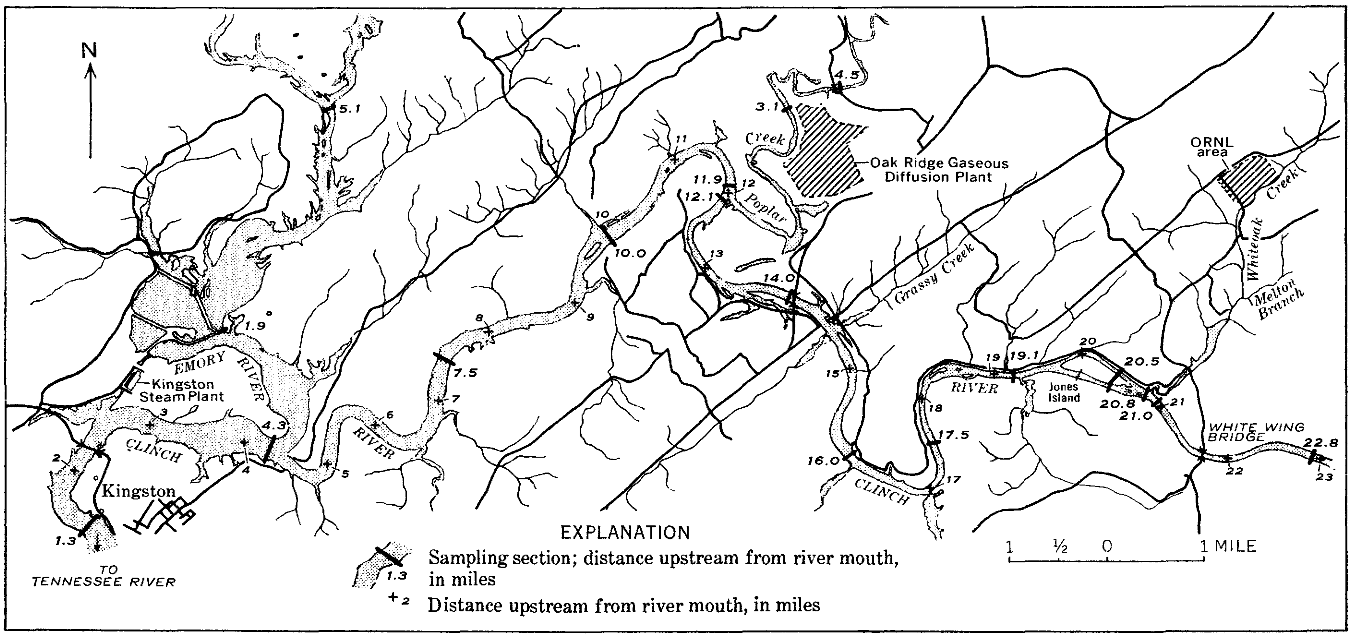


FIGURE 1.—Study reach and locations of bottom-sediment core-sampling sections.

tivity. The most downstream sampling section was at CRM (Clinch River mile) 1.3 and the most upstream section was at CRM 22.8. The abbreviation CRM followed by a number has been used in this report to designate distance, in miles, upstream from the mouth of the river. This terminology is consistent with prior usage in the Clinch River Study (Morton, 1961).

Radioactive waste enters the Clinch River via Whiteoak Creek at CRM 20.8. The entire study reach of the Clinch River is part of Watts Bar Lake, which is formed by Watts Bar Dam on the Tennessee River.

Initially, 10 coring sites were selected in each of the six sampling sections in the Clinch River downstream from CRM 13. Five sites were selected in each of the eight sections upstream from CRM 13 and in the four sections in the two tributary streams. Coring sites were selected on the basis of known sediment thicknesses calculated by TVA from sediment-range surveys, and the results of sediment-probe measurements and a survey of sediment-surface radioactivity which were made at each sampling section just prior to core sampling.

The vertical distribution of gross gamma radioactivity and of the major gamma-ray-emitting radionuclides in the cores was measured by means of a device, known as a core scanner, which was developed especially for that purpose.

The chemical forms and associations of radionuclides in the bottom sediment were investigated through a program of detailed analyses of 23 sediment samples from two cores. The two cores were obtained from areas in which the accumulated radioactive sediment was several feet in thickness. The samples consisted of alternate 2-inch-thick slices of sediment taken from the two cores. A series of physical and chemical tests, as well as radionuclide analyses, were performed on the 23 samples in order to characterize their compositional variability. Properties measured were the mineralogy, particle-size distribution, cation-exchange capacity, leachable-cation content, free aluminum oxide content, free iron oxide content, inorganic and organic carbon contents, and the amounts of potassium, rubidium, cesium, and strontium in each of the samples. Correlations of variations in concentration with depth for each core were determined for selected constituent pairs by means of a digital computer program in an effort to determine what properties of the sediment were responsible for the presence of each of the radionuclides.

Composite samples of 45 additional cores were analyzed for particle-size distribution. The radionuclide content of these cores had been determined previously (Carrigan, 1969; Carrigan and Pickering, 1967).

ACKNOWLEDGMENTS

The help and support of E. G. Struxness, Assistant Division Director, Health Physics Division, ORNL; F. L. Parker, Section Chief, Radioactive Waste Disposal Section, Health Physics Division, ORNL, and coordinator of the Clinch River Study; and P. H. Carrigan, Jr., chief of the USGS project for the Clinch River Study, are gratefully acknowledged. Tsuneo Tamura and D. G. Jacobs, ORNL, have discussed the results of their research and the general concepts in this report at length with the author and have thereby contributed greatly to the author's knowledge of sorption mechanisms in aluminosilicate minerals. Similar thanks are due to V. C. Kennedy and E. A. Jenne, USGS, and R. W. Andrew, Jr., PHS.

E. R. Eastwood, ORNL, prepared the samples used in the study and assisted in tabulation and inspection of data. He also performed most of the gross gamma and gamma spectrum scanning of the sediment cores and assisted in the field while coring was in progress. He was assisted in the core scanning by J. E. Jackson, ORNL, and W. E. Parker, ORNL and USGS; B. P. Glass, ORNL, assisted with coring operations.

Construction of the core scanner was under the direction of P. N. Hensley, ORNL. W. P. Bonner, ORNL, acted as consultant on the use of the 512-channel pulse-height analyzer in conjunction with the core scanner. Advice on gamma spectrum analysis was furnished also by C. L. Burros, Analytical Chemistry Division, ORNL.

The digital computer program used for the analysis of gamma spectra of the sediment samples was developed by Ernest Schonfeld, Chemical Technology Division, ORNL, who also contributed his personal services and advice in the handling of the raw data and in the evaluation of analytical results.

Analysis of sediment samples for content of Strontium-90 was performed at the Low Level Radiochemistry Laboratory, Analytical Chemistry Division, ORNL. Particle-size analyses were made at the Water Quality Laboratory, USGS, Raleigh, N.C. Cation-exchange capacity, leachable-cation content, and mineralogical analyses were performed at the Denver, Colo., laboratories of the Branch of Analytical Laboratories, USGS. Determinations of free iron oxide content, adsorbed water content, and contents of mineral, organic, and total carbon were performed at the same Denver laboratories. Analyses of minor and trace elements in the sediment samples were performed at the Laboratory for X-ray and Spectrochemical Analysis, Analytical Chemistry Division, ORNL.

The digital computer program used for correlation of gross gamma core scans was conceived by P. H. Carrigan, Jr., USGS, and written by M. T. Harkrider, ORNL Mathematics Division. Other computer programs used in interpreting the data reported here were written by M. T. Harkrider and by R. P. Leinius and A. M. Craig, Jr., ORNL Mathematics Division.

Data on sediment deposition in the Clinch River were provided by J. W. Beverage and James Smallshaw, TVA. Information concerning the distribution of radioactivity in the upper part of Clinch River bottom sediment was obtained through H. H. Abee and W. D. Cottrell, Applied Health Physics Section, Health Physics Division, ORNL.

Russell Lenker and Leon Dowdy, Jr., employees of Sprague and Henwood, Inc., drilling contractor for coring of the bottom sediment, provided much helpful information and instruction on the use of the Swedish Foil Sampler. Sprague and Henwood, Inc., constructed the special cutting shoe with basket core retainer which was designed during the coring operations and used to obtain increased recovery of cores consisting of rather fluid sediment. Contractual arrangements for drilling were under the supervision of F. L. Parker, Section Chief, Radioactive Waste Disposal Section, ORNL.

Figures 1, 2, and 3 were prepared by the Graphic Arts Services, ORNL.

PREVIOUS WORK

Uptake of chemical constituents from liquid systems by sediment-forming minerals has been investigated extensively during the past several years, but the specific processes involved in the uptake of radioactive materials by stream sediment are for the most part not precisely known. For this reason, the general term "sorption" has been used in this paper; for an explanation of the term see Sayre, Guy, and Chamberlain (1963, p. 11) and McBain (1950, p. 38). Sorption of radionuclides by river sediment has been studied by Carritt and Goodgal (1953), Barker (1958), Jacobs (1960), Jacobs and Tamura (1960), and others. The importance of radiocesium uptake by the clay minerals has been well established, and Tamura and Jacobs (1960) have shown that cesium is selectively sorbed by the micas and the 2:1-layer clay minerals (minerals that have an aluminum-oxygen layer between two silicon-oxygen layers; Grim, 1953, p. 65) which have a layer-to-layer spacing of 10 A (angstroms). Tamura (1963) has further shown that hydrous iron oxides and aluminum oxides, common minor constituents in many sediments, selectively sorb strontium in neutral and basic solutions. Work on radionuclide uptake by river sediments has been summarized by Sayre, Guy, and Chamberlain (1963), Kornegay, Vaughan, Jamison, and Morgan (1963), and Robinson (1962).

Sorathesn, Bruscia, Tamura, and Struxness (1960) measured the capacity of a sample of Clinch River bottom sediment to sorb cesium and found it to be quite high. This they attributed to the high illite content of the sediment. The higher-than-expected sorption capacity for cobalt-60 and strontium-90 in one sample of Clinch River bottom sediment was attributed to organic matter in the sediment.

Jenne and Wahlberg (1965) have noted that during the progressive extraction of iron and manganese oxides from bottom sediment from Whiteoak Creek, cobalt-60 was also extracted. A study of extraction rates has indicated that more of the cobalt-60 occurs in association with manganese oxides than with iron oxide.

R. W. Andrew, Jr. (PHS, written commun., 1964) noted a correlation between particle size and content of cesium-137 and cobalt-60 in sediment samples from the Clinch River and the Tennessee River when the clay-size fraction was divided into fine (less than 0.2 micron) and coarse (0.2–2.0 microns) subfractions. The content of ruthenium-106 correlated best with the content of fine clay in samples from the Clinch River, but the content of organic matter provided the best correlation in the Tennessee River. Organic matter correlated also with the contents of cesium-137, cobalt-60, and strontium-90 in the Tennessee River. Through the use of multiple correlation techniques, Andrew determined that 50 to 90 percent of the observed variation in the radionuclide content of bottom-sediment samples from the Tennessee River could be explained on the basis of particle size and content of organic matter in the samples. Less than 50 percent of the variation in samples from the Clinch River was explained by the same variables.

Annual monitoring surveys by the Applied Health Physics Section, ORNL, have shown that most of the radioactivity in Clinch River bottom sediment is due to cesium-137. The remainder of the radioactivity is primarily due to cobalt-60, ruthenium-106, certain rare earths of the lanthanide series, and strontium-90. The annual surveys have also shown that concentrations of the major radionuclides in the surface layer of sediment vary with changes in radionuclide loads released by the Laboratory (Morton, 1965, p. 30). However, the longitudinal pattern of distribution of gross gamma radioactivity at the surface of the sediment in the Clinch River has remained much the same since the first survey was made in 1951. Strong similarities in the longitudinal distributions of cesium-137, cobalt-

60, ruthenium-106, and certain rare earths in the surface layer of the sediment, observed in 1961, suggested that similar mechanisms controlled the distribution of those radionuclides. The observed rates of decrease in concentration of the radionuclides with distance downstream were much greater than rates calculated on the basis of flow dilution alone. This evidence has been interpreted to mean that the mechanism controlling the distribution pattern may be a sedimentation process. The longitudinal distribution pattern of strontium-90, which was different from the distribution patterns of the three other radionuclides, suggests that a different mechanism may be controlling the distribution of strontium-90 (Morton, 1965, p. 65).

A series of bottom sediment cores was collected from the Clinch River in 1960 by ORNL personnel, but the coring device that was used failed to penetrate the full thickness of radioactive sediment (Morton, 1962a, p. 45–57). Analysis of the cores showed that cesium-137 was the predominant radionuclide at all sampling sections. The relative abundances of cesium-137, cobalt-60, and strontium-90, which were much the same throughout the study reach, suggest that the same, or at least concurrent, processes governed the deposition of the three radionuclides in the riverbed (Morton, 1962a, p. 53).

METHODS AND EQUIPMENT

CORE SAMPLING

Relatively undisturbed cores of bottom sediment were obtained by means of a Swedish Foil Sampler. The Swedish Foil Sampler is a 2½-inch-diameter, piston-type sampler in which thin axial metal strips (foils) are used to decrease friction between the sample tube and the sediment while coring is proceeding (Pickering, 1966). As the sampler is pushed into the sediment, the foils unroll and form a sheath that encloses the core and prevents its contact with the interior of the sample tube. The core does not move relative to the foil strips, and the only friction in the sample tube is the metal-to-metal friction between the foils and the interior of the tube. Compaction and blocking of the core due to friction between the sediment and the walls of the tube is thus prevented.

Core recovery, at coring sites where radionuclides were believed to be present throughout the full thickness of the sediment penetrated, generally ranged from 80 to 100 percent. When a satisfactory core recovery was not obtained, a second coring attempt was made. At a few locations, satisfactory cores could not be obtained with the unmodified sampler. Use of a cutting shoe containing a basket-type core retainer generally produced adequate samples.

Sediment cores were transferred in the field from the sample tube to plastic storage tubes and were then frozen to facilitate handling and to suppress biological and chemical action prior to processing of the samples.

CORE COUNTING

Variations of radionuclide content with depth in the intact sediment cores were measured with a core scanner which was designed and constructed at ORNL. The scanner consists of a 3- by 3-inch, thallium-activated sodium iodide scintillation crystal and a matched photomultiplier tube housed in one end of a 4-inch-thick cylindrical lead shield. In the other end of the shield is a cylindrical hole, or core well, whose axis is perpendicular to that of the lead cylinder and through which the cores, still encased in their plastic tubes and foils, can be moved. A 2-inch-long, 2- by 2-inch collimator channel extends from the face of the scintillation crystal to the core well. A diagrammatic representation of the core scanner is shown in figure 2.

The use of a 2-inch-long collimator channel in the core scanner represented a compromise between the desire for high collimation of the gamma-ray emission from the 2-inch-thick increment of core opposite the channel opening and the desire for minimum dropoff of total gamma-ray radioactivity due to the distance between the scintillation crystal and the sediment core. With the chosen channel length, gamma-ray radioactivity was high enough for adequate measurement, but a certain amount of this radioactivity represented emission from parts of the core above and below the 2-inch-thick increment being measured. Each incremental gamma-ray radioactivity measurement was corrected for the effect of this extraneous radioactivity by means of a calibration curve based on measurements of plane sources of radioactivity. The correction was applied by means of a digital computer program, which also was used for plotting the corrected incremental radioactivity against depth in the core for gross gamma scans.

For counting of gross gamma radioactivity, the photomultiplier-tube output was routed through a scaler and automatic timer-printout system. The number of seconds required for the accumulation of a preset number of counts (usually 2,048 counts) from the 2-inch-thick increment of core opposite the collimator channel was recorded, the core was automatically raised 2 inches by means of a calibrated hoist, and counting of the next increment of core was begun.

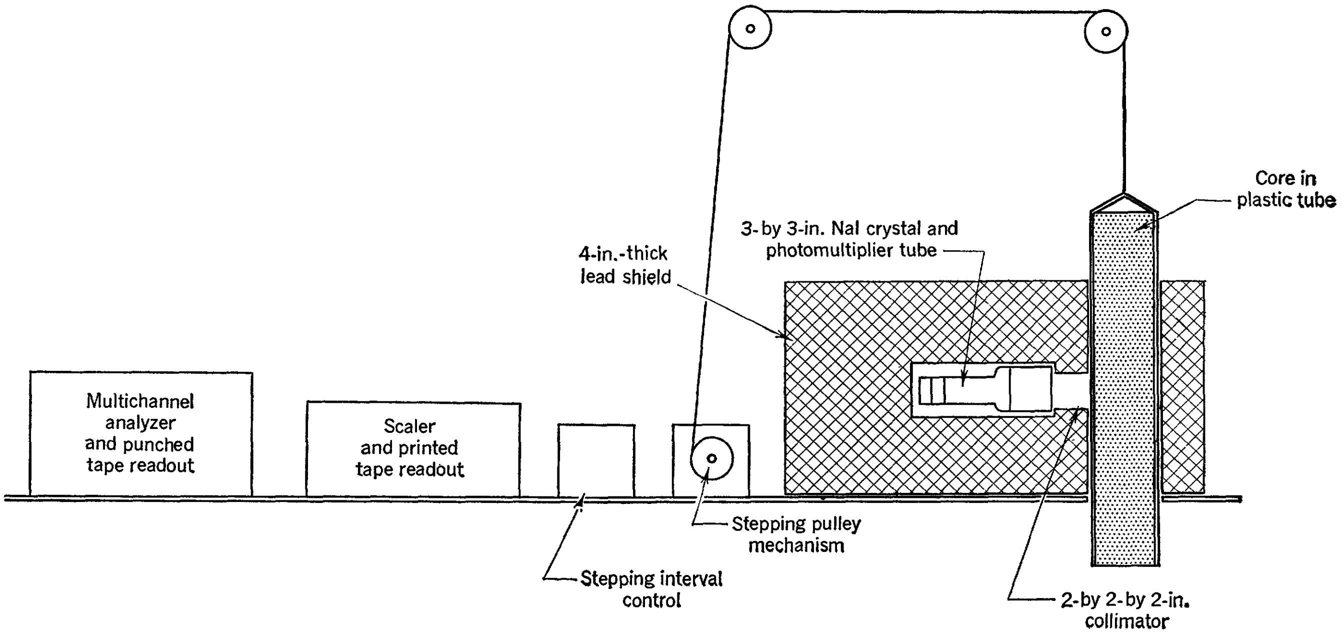


FIGURE 2.—Diagrammatic representation of core scanner.

For gamma-ray-spectrum scanning, the photomultiplier-tube output of the core scanner was routed through a 512-channel pulse-height analyzer, and the analyzer output (gamma-ray spectrum) for each 2-inch-thick increment of core was recorded. The output data were then examined for analyzer malfunctions by means of a computer program. The gamma-ray spectrum produced by each 2-inch-thick increment of the cores was analyzed through the use of a computer program developed at ORNL (Schonfeld and others, 1965). The results of each analysis, which included specific activities with computed standard deviations for eight radionuclides, were corrected for imperfect collimation and plotted for visual inspection by means of a third computer program. The eight radionuclides used in the gamma-ray-spectrum analysis included the nuclear-fission products cesium-137, ruthenium-106, cesium-134, europium-152, europium-154, cerium-144, zirconium-95, niobium-95, the neutron-activation product cobalt-60, and the naturally occurring radionuclide potassium-40.

PHYSICOCHEMICAL ANALYSIS OF SEDIMENT

Comprehensive particle-size analyses of aliquots of 23 sediment samples were made by wet sieving and by bottom-withdrawal tube (U.S. Inter-agency River Basin Committee, 1953). Size ranges, determined in microns, were: <2, 2–4, 4–8, 8–16, 16–31, 31–62, 62–125, 125–250, 250–1000, >1000. The method of size analysis used involves dispersal of the sediment particles by chemical as well as mechanical means and can be expected to break up aggregates of clay minerals which existed in the bottom sediment as silt- or sand-size masses. The method thus results in a determined particle-size distribution which is not necessarily fully representative of the actual distribution of particle sizes in the natural environment. However, it gives a measure of the primary size distribution, which is useful in some hydraulic studies of sediment movement and in mineralogical studies of sediment composition.

Particle-size separations of aliquots of the same samples were performed by wet sieving and by centrifugation after the samples had been initially dried. Dispersal was by hand stirring or, for some samples, by ultrasonic agitation. Size ranges, determined in microns, were: <2, 2–62, >62. This method of separation tended to preserve aggregates of smaller particles, but additional aggregates may have formed as a result of the attachment of clay-size particles to sand- and silt-size particles when the samples were dried. Leaf fragments and twigs tended to be concentrated in the >62-micron-size fraction, and finer, lightweight organic material was decanted with the <2-micron-size fraction following centrifugation. Determinations of cation-exchange capacities and mineralogical composition were made of the three size fractions.

The significance of the two methods of sizing in respect to the analysis of the physico-chemical characteristics of the sediment is discussed in another section of this report.

Cation-exchange capacities were determined by means of an ammonium chloride leach followed by measurement of the sorbed ammonium after extraction by distillation. The content of leachable cations in the raw sediment samples was determined through a chemical analysis of the leachates obtained as a result of the cation-exchange-capacity determinations. Mineralogical determinations were made by X-ray diffractometer.

The content of free alumina in the samples was determined by extraction in a 0.5 *N* solution of sodium hydroxide (Hashimoto and Jackson, 1960) and colorimetric analysis by the "aluminon" method (Jackson, 1958, p. 296–300). Free iron oxides were extracted in a buffered dithionate-citrate solution (Mehra and Jackson, 1960) and analyzed by the potassium thiocyanate method (Jackson, 1958, p. 168–169). The extraction methods used are believed to have caused very little dissolution of silicate minerals present in the sediment.

The content of organic carbon in each of the sediment samples was determined as the difference between the total amount of carbon, determined through combustion of the sample at 1350°C (Tourtelot and others, 1964, p. D74), and the carbonate-derived (mineral) carbon, determined by the gas-evolution method of Rader and Grimaldi (1961, p. 33). Analysis of the samples for minor and trace elements was performed by flame spectrophotometric methods in which recently developed precision methods for the determination of strontium (Rains and others, 1962) and cesium (Feldman and Rains, 1964) were used.

The content of the gamma-ray-emitting radionuclides in each sample was determined by analysis of the gamma-ray spectrum obtained by means of a 512-channel pulse-height analyzer. The detection system consisted of a 4- by 4-inch thallium-activated sodium iodide scintillation crystal housed in a cadmium-lined lead shield which was 4 inches thick. Gamma-ray spectra were analyzed by means of a digital computer program developed at ORNL (Schonfeld and others, 1965). The content of strontium-90 in each sample was determined by "wet" chemical separation methods and by counting of the beta-ray radioactivity of the purified sample.

CORE SAMPLING RESULTS

A total of 163 bottom-sediment cores (including cores resulting from repeated attempts at a single coring site) were obtained at 135 coring sites. Sediment thicknesses measured with a sediment probe were generally confirmed by core sampling. At sampling sections which coincided with TVA sediment ranges, coring results generally agreed with TVA measurements of sediment thicknesses.

In the Clinch River upstream from CRM 13.0, periodic high discharges have swept the central part of the stream channel relatively free of all sediment except pebbles and boulders, and accumulation of finer sediment is confined largely to the sides of the channel. Downstream from CRM 13.0, fine sediment extends over a considerable part of the channel bottom, but is generally thickest on the less steep side of the stream channel. At the sampling sections at CRM 10.0, 11.9, and 12.1, fine sediment from a few inches to several feet thick extends across almost the entire width of the channel. The depositional pattern of the sediment in the study reach probably reflects the distribution of velocity in the river channel and, at four cross sections in the Clinch River, is consistent with the measured velocity distribution (Morton, 1962a, p. 81–85).

VARIATION OF RADIONUCLIDE CONTENT WITH DEPTH IN CORES

GROSS GAMMA CORE SCANNING

Scanning the bottom-sediment cores for variations in gross gamma radioactivity with depth revealed the general pattern of variation in each core and provided a basis for determining the thickness of radioactive sediment at each coring site. As used in this report, gross gamma radioactivity refers to the measured quantity of radioactivity given off by all gamma-ray-emitting nuclides in the sample. The fraction of the total radioactivity detected depends on the design and sensitivity of the analytical equipment used.

Variations in gross gamma radioactivity in the cores were great enough to permit the base of the radioactive zone to be picked from plots of the gross gamma scans and to encourage attempts at gamma spectrum core scanning. In addition, the results of gross gamma core scanning indicated that the entire thickness of radioactive sediment had been sampled in most cores.

The greatest thicknesses of radioactive sediment occur in the lower part of the river, downstream from CRM 13.0. The core showing the greatest thickness (8.7 feet) of radioactive sediment was obtained at CRM 7.5.

In accord with the general pattern of sediment accumulation in the river, the layer of radioactive sediment is thickest along the sides of the stream channel, but in sections downstream from CRM 13.0 it extends out into the main part of the channel as well. Near the mouth of Poplar Creek at CRM 12.0, radioactive sediment extends across the entire width of the river channel. The effect of this wider distribution of radioactive sediment in the cross section on the average cross-sectional radioactivity level at the sediment surface can be seen in the results of annual measurements made by the Applied Health Physics Section, ORNL. This average, which represents the mean of all sediment-surface radioactivity readings at a cross section, has been highest for cross sections in the vicinity of CRM 12.0 in several annual surveys (Morton, 1962b, p. 62).

The results of gross gamma scanning of core samples from CRM 7.5 are shown in figure 3. Sediment deposition, and therefore accumulation of radioactive sediment, has been largely confined to the more gently sloping left half of the stream channel which constituted the side of the former stream channel and a part of the flood plain of the stream prior to inundation by Watts Bar Lake. (Note that the vertical exaggeration of the cross section plot is 10:1). Parts of the cross section showing a water depth of less than 6 feet are not submerged during low winter lake levels. The resulting nondeposition of sediments and erosion probably account for the very thin layer of radioactive sediment found in cores from such sites (cores 1 and 2).

Cores 4, 5, 6, and 7–1 in figure 3 show a persistent general pattern of variation of gross gamma radioactivity with depth. Similar distribution patterns of gross gamma radioactivity were observed in several cores from other sampling sections in the part of the study reach downstream from CRM 18. The general pattern which they exhibit is strikingly similar to the pattern of annual releases of cesium-137 to the Clinch River as measured at Whiteoak Dam (table 1). This similarity is illustrated in figure 4 for several of the cores. The zone of highest radioactivity in each of the four cores is assumed to correspond to the period during which releases of cesium-137 were highest—that is, 1956 (Cottrell, 1959, p. 39).

The patterns of variation of gross gamma radioactivity with depth in the sediment cores were compared by means of a digital computer program. With this program, the variation of radioactivity in a core of any length could be compared to that in any other core, provided two reference points at different depths could

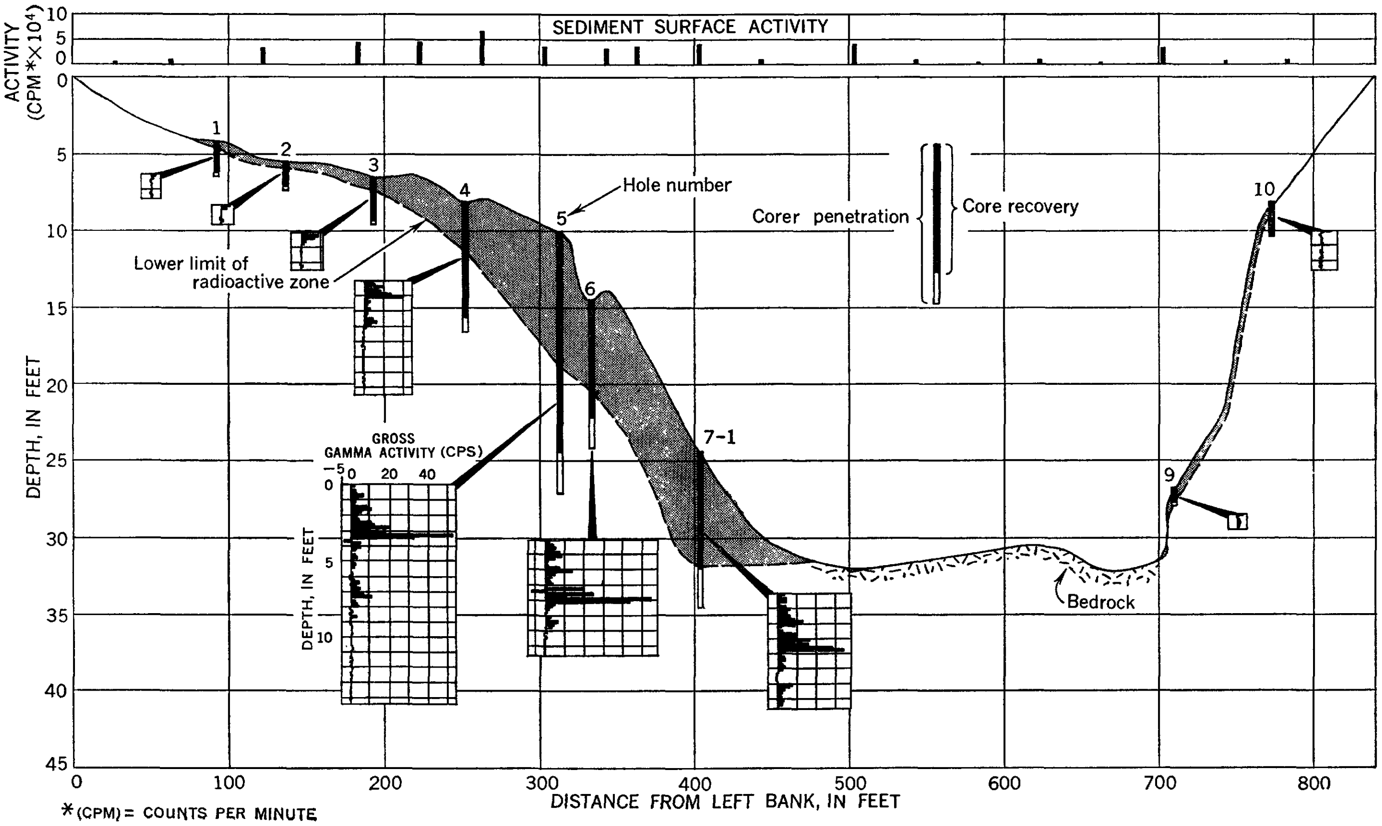


FIGURE 3.—Cross section at CRM 7.5 showing penetration, recovery, and gross gamma radioactivity variations with depth for 1962 bottom-sediment core samples (vertical exaggeration 10:1).

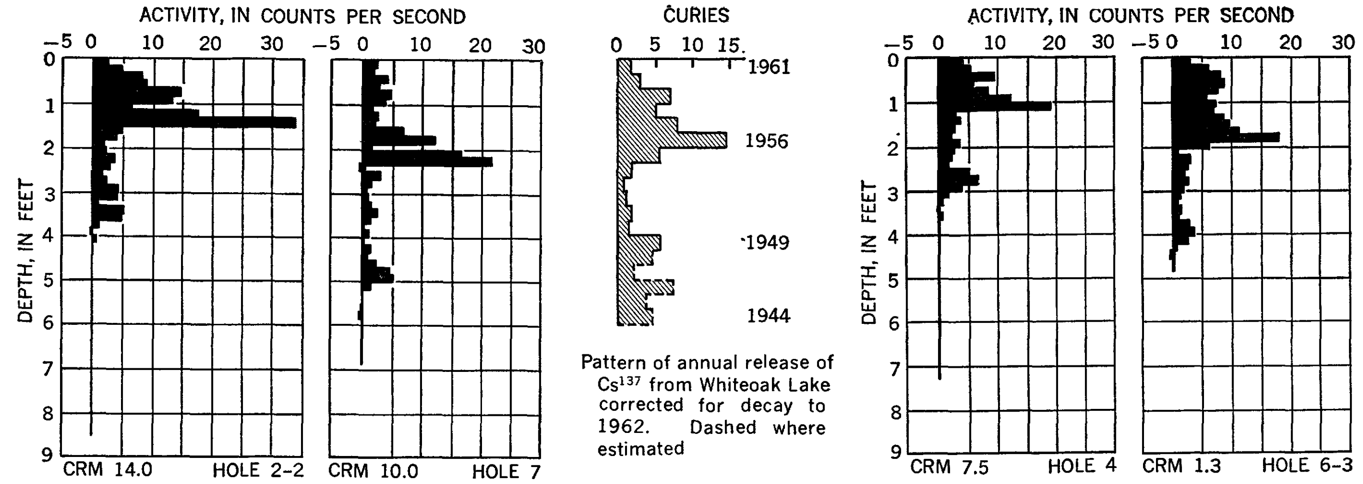


Figure 4.—Graphical comparison of patterns of variation of gross gamma radioactivity with depth in four bottom-sediment cores to variations in annual releases of cesium-137 to the Clinch River.

Table 1.—*Yearly discharges of radionuclides to the Clinch River*

[Results in curies. From Cowser and Snyder (1966). TRE (—Ce), trivalent rare earths excluding cerium; Nd, not determined]

Year	Gross beta	Cs^{137}	Ru^{106}	Sr^{90}	TRE (—Ce)	Ce^{144}	Zr^{95}	Nb^{95}	I^{131}	Co^{60}
1949	718	77	110	150	77	18	180	22	77	Nd
1950	191	19	23	38	30	Nd	15	42	19	Nd
1951	101	20	18	29	11	Nd	4.5	2.2	18	Nd
1952	214	9.9	15	72	26	23	19	18	20	Nd
1953	304	6.4	26	130	110	6.7	7.6	3.6	2.1	Nd
1954	384	22	11	140	160	24	14	9.2	3.5	Nd
1955	437	63	31	93	150	85	5.2	5.7	7.0	6.6
1956	582	170	29	100	140	59	12	15	3.5	46
1957	397	89	60	83	110	13	23	7.1	1.2	4.8
1958	544	55	42	150	240	30	6.0	6.0	8.2	8.7
1959	937	76	520	60	94	48	27	30	.5	77
1960	2,190	31	1,900	28	48	27	38	45	5.3	72
1961	2,230	15	2,000	22	24	4.2	20	70	3.7	31
1962	1,440	5.6	1,400	9.4	11	1.2	2.2	7.7	.36	14
1963	470	3.5	430	7.8	9.4	1.5	.34	.71	.44	14

be assigned as corresponding to the same parts of the patterns in each of the cores compared. Core lengths were adjusted for the comparison on the basis of the distance between the two reference points picked for each core. A series of correlations were attempted for each pair of cores, a different adjustment factor being used in each correlation attempt. The output of the program included the coefficients of correlation and regression for the best correlation attempted with each pair of cores.

By means of the computer program, gross gamma scans of 27 cores which exhibited a similar pattern of variation of gross gamma radioactivity with depth were compared with the pattern of annual releases of cesium-137 to the Clinch River, the releases being corrected for decay to the date of sampling (table 2). Coefficients of correlation ranged from 0.60 to 0.90 and had a mean of 0.76. When annual releases of cobalt-60 were added to those of cesium-137 and correction was made for the relative gamma-ray emission of the two nuclides, the comparison showed improved correlation with the cores and resulted in a mean coefficient of correlation of 0.80 and a range of 0.64 to 0.98. When the annual releases of ruthenium-106 were added to those of cesium-137 and cobalt-60, after making corrections for relative gamma ray emission, the correlation was not as good, giving a mean coefficient of correlation of 0.68 and a range of 0.44 to 0.97.

Coefficients of correlation resulting from a comparison of the distribution of radioactivity in four cores shown in figure 4 with annual releases of radionuclides to the Clinch River are included in table 2. In all four cores, 18 increments were used for comparison.

TABLE 2.—*Coefficients of correlation for comparison of vertical distribution of gross gamma radioactivity in sediment cores with annual releases of radionuclides*

Annual releases were corrected for decay to date of sampling. Where releases are combined, correction has been made for the relative gamma emission of the radionuclides. Dash leaders indicate correlation was not obtained]

Sampling section	Core	Cs^{137}	$Cs^{137}+Co^{60}$	$Cs^{137}+Co^{60}+Ru^{106}$
CRM 1.3	3	0.64	0.67	0.68
1.3	5	.78	.84	.72
1.3	6–2	.63	.72	.64
1.3	6–3	.73	.78	.78
1.3	7	.60	.64	.65
1.3	7–2	.88	.93	.80
4.3	2–2	.67	.83	.69
4.3	11–2	.73	.79	.63
7.5	4	.81	.82	.62
7.5	5	.80	.72	.51
7.5	6–1	.81	.75	.49
7.5	7–1	.75	.79	.57
7.5	7–2	.72	.64	.50
10.0	2	.83	.82	.61
10.0	3–3	.77	.90	.81
10.0	7	.83	.79	.44
11.9	9	.76	.76	--------------
11.9	10	.60	.79	.71
12.1	2	.88	.89	.80
12.1	6–1	.66	.82	.80
12.1	7	.79	.78	.77
12.1	9	.89	.98	.87
14.0	2–1	.82	.85	.60
14.0	2–2	.80	.84	.62
14.0	3	.76	.87	.97
17.5	3–1	.90	.79	.83
17.5	3–2	.76	.66	.63
Mean		0.76	0.80	0.68
Standard deviation		.084	.083	.126

The correlation results suggest that, in most of the 27 cores tested, the pattern of variation in gross gamma radioactivity exhibited by the cores reflects not only the pattern of annual releases of cesium-137 to the Clinch River but to a minor extent that of annual releases of cobalt-60 as well.

GAMMA-RAY SPECTRUM CORE SCANNING

Nine cores were selected for gamma-ray spectrum scanning on the basis of gross gamma scan results. Cores selected were those with a zone of radioactive sediment which was several feet thick and which showed the characteristic pattern of variation in gross gamma radioactivity with depth illustrated in figures 3 and 4.

Four gamma-ray-emitting radionuclides were present in the bottom sediment in concentrations high enough to provide acceptable data for the full thickness of the radioactive zone in each core scanned; they were cesium-137, cobalt-60, ruthenium-106, and potassium-40. Because of its short half life (1 year), ruthenium-106 was detected in only the upper parts of most of the cores. The presence of the naturally occurring radionuclide potassium-40 was not due to releases from ORNL.

Variations in contents with depth of the three most abundant gamma-ray-emitting radionuclides in a core from hole 6, CRM 7.5 are illustrated in figure 5; values whose standard deviations represented greater than 75 percent of the analytical values computed prior to collimation correction have been plotted as zero values.

An examination of the first four plots in figure 5 demonstrates the relative contributions of the three radionuclides to the total gamma-ray radioactivity of the core. Over 80 percent of the radioactivity is contributed by cesium-137. The contribution of radioactivity from ruthenium-106 is confined to the upper part of the core. (The isolated positive values for ruthenium-106 below a depth of 20 inches on the plot are questionable.) Similar results obtained for the eight other cores that were scanned are shown in table 3.

TABLE 3.—*Fractional contributions of cesium-137, cobalt-60, and ruthenium-106 to total gamma-ray radioactivity of selected Clinch River sediment cores*

Sampling section	Core	Cs^{137}	Co^{60}	Ru^{106}
CRM 1.3	3	0.82	0.11	0.07
1.3	6–3	.77	.13	.10
7.5	5	.81	.13	.16
7.5	6	.82	.12	.06
7.5	7–2	.77	.13	.10
10.0	2	.79	.10	.11
14.0	2	.80	.13	.07
14.0	2–2	.84	.11	.05
17.5	3–1	.86	.10	.04
Mean value		0.81	0.12	0.07

The similarity between the distributions of cesium-137 and cobalt-60 with depth in the core can be observed with the aid of the fifth plot in figure 5, which shows variations in cobalt-60 concentration with depth at an expanded scale. The same relationship was exhibited by the eight other cores that were gamma spectrum scanned. Statistical treatment of the relationship between cesium-137 and cobalt-60 in all nine cores further emphasizes the similarity in distribution of the two radionuclides (see table 4).

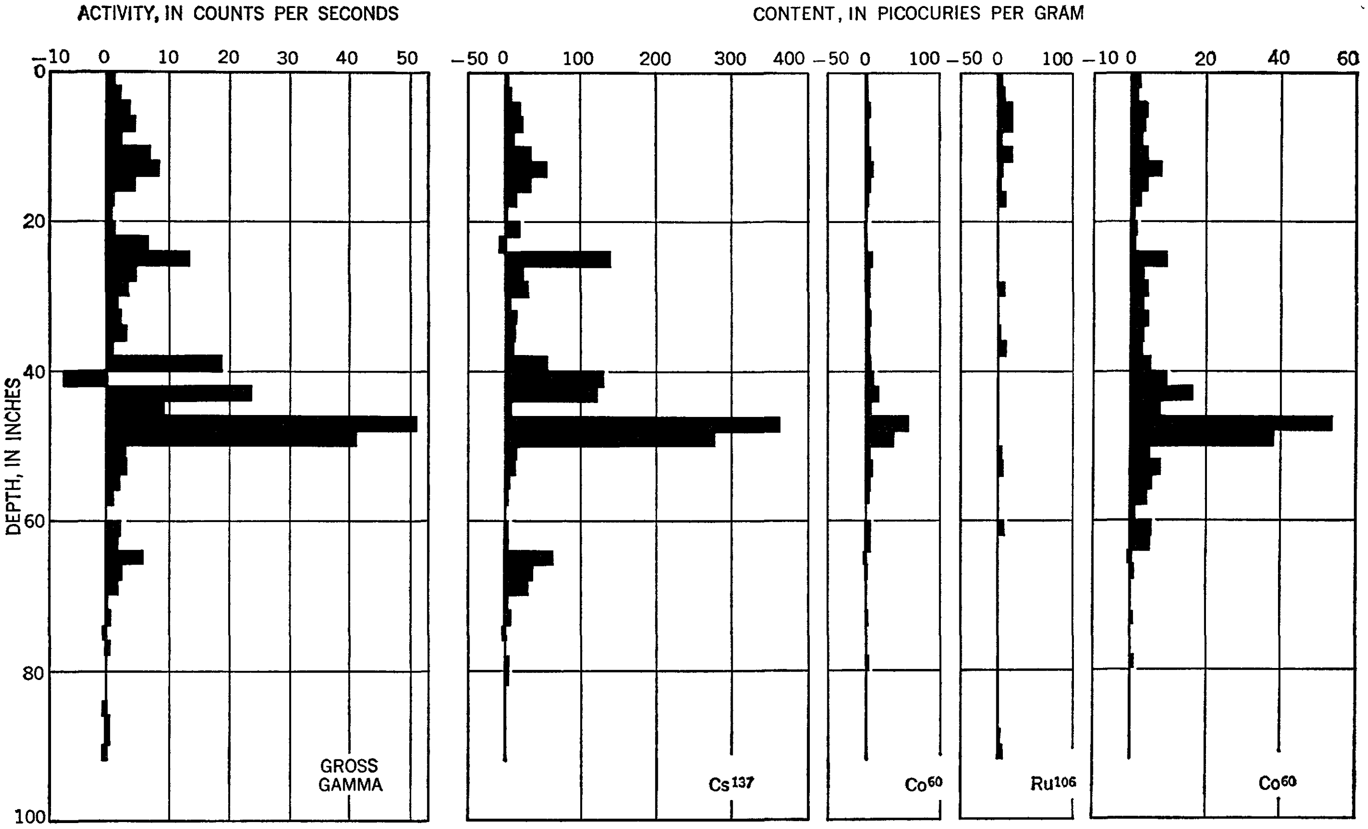


FIGURE 5.—Variations in contents of cesium-137, colbalt-60, and ruthenium-106 with depth in hole 6, CRM 7.5.

TABLE 4.—*Relationship of cobalt-60 to cesium-137 in selected bottom-sediment cores from the Clinch River*

Sampling section	Core	Correlation coefficient	Regression coefficient	Ratio of mean concentrations (Cs:Co)
CRM 1.3	3	0.62	0.09	7.39
1.3	6–3	.89	.12	5.83
7.5	5	.90	.13	5.77
7.5	6	.93	.13	7.34
7.5	7–2	.91	.13	5.86
10.0	2	.87	.11	7.83
14.0	2	.90	.13	6.37
14.0	2–2	.87	.13	7.25
17.5	3–1	.99	.09	9.11
Mean value		0.88	0.12	6.97

RELATIONSHIP OF VERTICAL DISTRIBUTION TO ANNUAL RELEASES

Radiochemical analyses of water samples collected weekly at Whiteoak Dam for a period of 2 years indicate that on the average nearly 70 percent of the cesium-137 present in the water is associated with suspended solids having a diameter greater than 0.7 micron Churchill and others, 1965, p. 37). Cesium is known to be preferentially sorbed by layered aluminosilicate minerals in which the *c*-axis spacing is 10 A (Jacobs and Tamura, 1960; Tamura, 1963). Such minerals are abundant in Whiteoak Creek basin (Jacobs, 1960; McMaster and Waller, 1965). Furthermore, this sorption reaction is known to be not readily reversible. It is safe to assume, then, that incorporation of cesium-137 in Clinch River bottom sediment is primarily a result of sedimentation of suspended radioactive aluminosilicate minerals which enter the river in water from Whiteoak Creek.

The year of highest release of cesium-137 to the Clinch River was 1956. This high release resulted from the draining of Whiteoak Lake during the latter half of 1955 (Cottrell, 1959, p. 39) and from the exposure to the air of the accumulation of fine sediment on the lake bottom. Rapid erosion of the exposed radioactive lake sediment during the subsequent period of high winter rainfall resulted in an increase in the quantity of radioactive sediment entering the Clinch River and in higher content of cesium-137 in bottom sediment deposited in the river during 1956. Through the operation of processes such as this, variations in annual releases of cesium-137 have been recorded as variations in the vertical distribution of the cesium-137 in the bottom sediment in parts of the river where fairly regular and persistent deposition of sediment has taken place.

The pattern of variation in the content of cobalt-60 in the sediment does not strongly resemble the pattern of annual releases of cobalt-60 to the Clinch River (see table 1). Some effect of annual releases is implied, however, by the improved correlation which resulted when annual cobalt-60 releases were taken into account in the comparison of gross gamma core scan patterns described previously. The most striking feature of the pattern of cobalt-60 variation in the cores gamma spectrum scanned is its great similarity to the pattern of cesium-137 variations (see fig. 5 and table 3). This similarity suggests that cobalt-60, like cesium-137, was incorporated in Clinch River bottom sediment primarily by sedimentation of suspended solids which had obtained their content of cobalt-60 before entering the Clinch River. It also suggests that a rather small fraction of the total cobalt-60 released to the river became incorporated in Clinch River bottom sediment; otherwise, distribution of cobalt-60 in the cores would resemble the pattern of annual releases of cobalt-60 to the river. This second suggestion is supported by the results of water sampling which show that only about 20 percent of the cobalt-60 released from Whiteoak Creek is associated with suspended sediment with a diameter greater than 0.7 micron (Churchill and others, 1965, p. 52). Although the results of water sampling do not preclude association of the remaining 80 percent of the cobalt-60 with smaller diameter suspended solids, some of which might be deposited in the Clinch River, it is possible that the nuclide may be present in Whiteoak Creek water also in a second form, such as a dissolved ion. The presence of two chemical forms of cobalt-60 in Whiteoak Creek water does not conflict with the results of gamma-ray spectrum core scanning if it is assumed that the ratio of dissolved cobalt-60 to solids-associated cobalt-60 in the water did not remain constant during the period in which the sampled sediment was deposited.

The suggestion that some of the cobalt-60, like most of the cesium-137, is associated with suspended solids when it enters the Clinch River from Whiteoak Creek does not necessarily imply that the two radionuclides are associated with the same type of solid. It is assumed that cesium-137 is primarily associated with layered aluminosilicate minerals. Leaching experiments (Morton, 1965, p. 60) have indicated that cobalt-60 may be present in the bottom sediment as a hydrated oxide. The studies further indicate that cobalt-60, like cesium-137 and ruthenium-106, is not readily removed by leaching after it becomes associated with the bottom sediment. Other studies have indicated that cobalt-60 in the sediment is associated with iron and manganese oxides (Jenne and Whalberg, 1965).

The contribution of ruthenium-106 to gamma radioactivity in the upper part of the sediment cores studied is much less than one would expect if its relative retention in the sediment were as great as that of cesium-137. It can be assumed, then, that a relatively small fraction of the total amount of ruthenium-106 that has been released to the Clinch River has become incorporated in the bottom sediment. This assumption has been confirmed by the results of an inventory of radioactivity in Clinch River bottom sediment (Carrigan, 1969).

Too little is known about the chemical form of ruthenium-106 in the Clinch River to permit definite inferences concerning the manner in which it is incorporated in Clinch River bottom sediment. The ruthenium-106 content of annual samples of the surface layer of Clinch River bottom sediment has been shown to reflect the amount of ruthenium-106 released to the river during the preceding year (Morton, 1965, p. 30). Analyses of water samples from Whiteoak Lake indicate that less than 10 percent of the ruthenium-106 released to the Clinch River is associated with suspended sediment greater than 0.7 micron in diameter (Churchill and others, 1965). Even with the benefit of this information, it can be stated only that the mechanism by which ruthenium-106 is incorporated in the sediment may be one or more of the following: (1) Sedimentation of radioactive solids suspended in Whiteoak Creek water, (2) precipitation of a ruthenium-bearing compound from Clinch River water, (3) an ion-exchange reaction between Clinch River water and bottom sediment in the river. It has been suggested (Morton, 1965, p. 60; Lomenick, 1963, p. 844) that some of the ruthenium-106 in bottom sediment may be present in the form of nitrosyl ruthenium hydroxide—$RuNO(OH)_3(H_2O)_2$. This compound, which is presumed to have been formed as a result of the high nitrate content of the original waste solutions, is not easily decomposed.

The general pattern of variation in gross gamma radioactivity with depth that occurs in many of the longer sediment cores taken from the Clinch River implies a similar history of sediment deposition at each of those coring sites. Furthermore, the resemblance of the pattern, which is due primarily to the cesium-137 content of the sediment, to variations in the annual release of cesium-137 from Whiteoak Creek to the Clinch River, implies fairly regular, persistent deposition at those coring sites.

Regular, persistent deposition at certain coring sites in a cross section does not necessarily require regular, persistent deposition over the entire cross section. Periodic short-term net sediment losses for the cross section are possible. However, a long-term net gain for the entire period of deposition of radioactive sediment is implied for all cross sections at which cores showing the characteristic pattern were collected. This sedimentation model is confirmed by TVA's record of sedimentation for the reaches of the Clinch River from which the cores were collected. The record shows net losses for some of the reaches downstream from CRM 13 for some of the 5-year intervals between surveys, but a net increase in sediment was computed for all reaches for the entire period of record. This net accumulation of sediment in the downstream reaches of the Clinch River is no doubt the result of readjustment of channel shape caused by the filling of Watts Bar Lake, which began at about the same time as release of radioactivity to the Clinch River was started by ORNL (U.S. Geological Survey, 1958, p. 228).

It should be emphasized that regular and persistent sediment deposition can occur without continual sediment deposition. Because the radioactive part of the sediment cores represents a sedimentation history of approximately 20 years, conclusions pertaining to sedimentation are at best valid for time intervals of a year or more. Annual sedimentation would be expected to be irregularly distributed throughout the year.

PHYSICOCHEMICAL COMPOSITIONS OF SELECTED CORE SAMPLES

The results of physical and chemical analyses of 23 sediment samples from the two cores selected for detailed study are listed in table 5. In general, radioactive bottom sediment in the Clinch River may be classed, according to particle size and mineralogical composition, as clayey silt (Shepard, 1954) composed of approximately 35 percent mica and clay minerals and 65 percent quartz. The mineralogical compositions and cation-exchange capacities of all 23 samples studied showed surprisingly little variation.

PHYSICAL CHARACTERISTICS

PARTICLE-SIZE DISTRIBUTION

The results of particle-size analyses of the 23 samples studied are shown in tables 5 and 6 and figure 6. A diameter of 4 microns was used as the lower limit of silt-size particles (Lane, 1947, p. 937) in the two figures for the purpose of classification of the sediment according to particle size, which is normally based on hydraulic characteristics. A lower limit of 2 microns was used for the summarized data presented in table 5 (cols. 2–7) and in constituent correlation studies, because it was believed that data on the finer size range would be more useful in relating particle-size distribution to the physical and chemical characteristics of the sediment. Lack of agreement between the two sets of particle-size data (cols. 2–4 and cols. 5–7, table 5) is the result of the use of two different methods of treatment and analysis.

The results of the particle-size analyses by wet sieving and bottom-withdrawal tube, plotted on triangular sand-silt-clay diagrams (Shepard, 1954), are shown in figure 6. Most of the samples fall in the clayey silt part of the diagram.

Particle-size analyses were also made of composite samples of 45 Clinch River bottom-sediment cores. Figure 7 is a plot of the results of these analyses. It is apparent that the particle-size distributions in the composite samples, which represent the radioactive parts of cores that were taken throughout a 21-mile-long reach of river, are more variable than those in the 2-inch-thick incremental samples of the two cores studied in detail. Greater variations occur in the contents of sand- and silt-size particles than in the content of clay-size particles. Nevertheless, more analyses fall in the clayey silt field of the diagram than in any of the other fields. The mean contents of particles in the three size groups for all composite samples are: sand, 23 percent; silt, 54 percent; clay (<4 microns), 23 percent. Because only composite samples with a relatively thick zone of radioactive sediment were chosen for particle-size analysis, the average particle-size distribution listed above, which would fall in the sand-silt-clay field of the diagram, is probably somewhat lower in sand content and higher in silt content than a true average of bottom sediment throughout the study reach would be. The greatest thicknesses of radioactive sediment in the Clinch River occur in the lower part of the river, where stream velocity is low and conditions are favorable for deposition of fine sediment. The two cores whose incremental compositions were studied in detail have average contents of sand-size particles that are still lower than the average of the composite samples (compare figs. 6, 7).

CATION-EXCHANGE CAPACITY

The cation-exchange capacities of the unsized samples from the two cores studied in detail do not vary greatly (see table 5). The cation-exchange capacities of the clay- and silt-size fractions of the same samples do not vary greatly either (table 7), a fact suggesting that most of the variation in cation-exchange capacity of the unsized samples may be due to variations in content of clay-size

TABLE 5.—*Physical and chemical characteristics of selected samples of bottom sediment from the Clinch River*

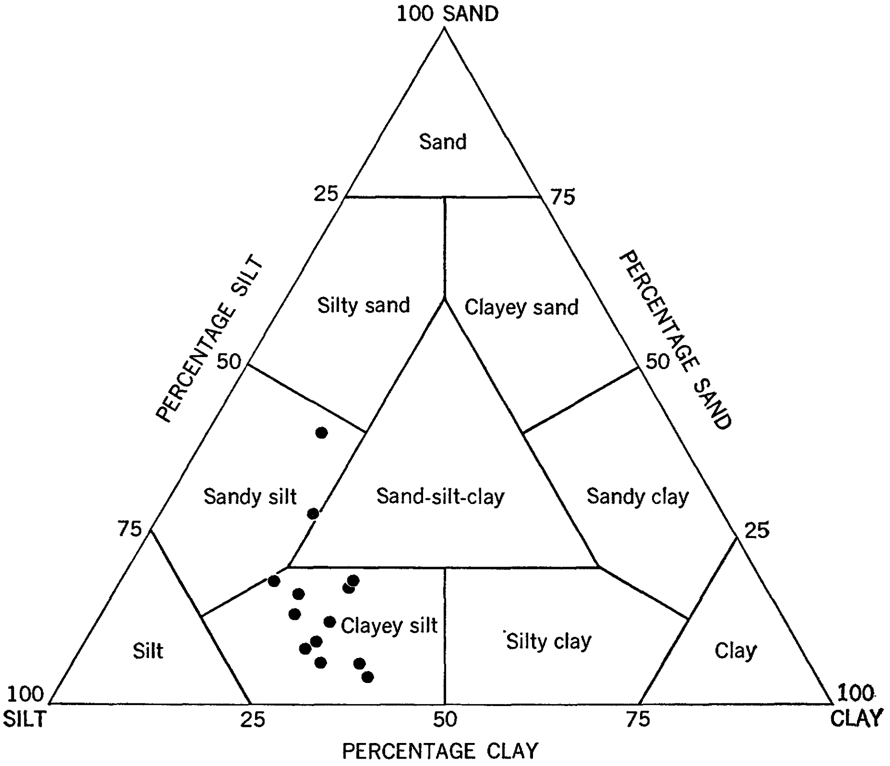
[Results are on a dry-weight basis, except for adsorbed water, and are in percent, except as indicated. Clay-size particles, <2 microns in diameter; silt-size particles, 2–62 microns in diameter; sand-size particles, >62 microns in diameter. Analysts: USGS—P. D. Blackmon, H. C. Starkey, H. E. Reeder, E. J. Fennelly, I. C. Frost; ORNL—T. C. Rains, J. H. Moneyhun]

Depth interval (in.)	Particle-size distribution[1]			Particle-size distribution[2]			Adsorbed water	Cation-exchange capacity (meq per 100 g)	Leachable-cation content (meq per 100 g)		Carbon content			Free oxide content		Minor element content				Radionuclide content[3]			
	Sand	Silt	Clay	Sand	Silt	Clay			Ca	Mg	Mineral	Organic	Total	Fe_2O_3	Al_2O_3	K	Rb	Cs	Sr	Sr^{90}	Cs^{137}	Co^{60}	Ru^{106}
CRM 7.5, hole 7–2																							
10–12	40	48	12	55	34	11	34.2	12.6	21.6	8.2	0.35	2.2	2.55	1.89	0.93	1.67	0.0067	0.00044	0.0093	≈0.45	37.07	3.891	6.738
14–16	28	58	14	59	28	13	34.0	12.3	18.2	5.7	.28	2.5	2.75	1.78	.92	1.47	.0069	.00031	.0097	.81	57.39	5.370	5.628
18–20	18	66	16	33	48	19	32.4	12.2	19.5	5.6	.62	1.6	2.22	2.22	1.28	1.98	.0097	.00047	.0105	2.0	58.28	4.682	5.476
22–24	16	63	21	23	58	19	35.1	15.1	23.7	5.6	.15	2.4	2.50	2.32	1.41	1.96	.0096	.00038	.0118	2.4	153.7	13.55	10.45
30–32	13	68	19	16	58	25	36.2	16.3	15.0	5.4	.14	2.8	2.97	2.51	1.52	2.02	.0107	.00047	.0090	3.0	305.6	21.46	−.2331
34–36	6	64	30	21	53	26	35.1	17.9	15.0	5.8	.21	2.8	3.05	2.48	1.53	1.91	.0101	.00048	.0081	3.0	155.8	18.87	−.04510
40–42	8	68	24	22	47	31	39.7	15.7	15.3	6.7	.20	2.8	2.98	2.42	1.54	1.85	.0096	.00040	.0079	2.4	29.14	.6705	.6520
44–46	12	63	25	16	58	26	39.3	18.8	14.1	6.0	.21	2.8	3.02	2.66	1.63	1.82	.0098	.00047	.0088	3.0	34.18	1.212	.9307
50–52	17	59	24	25	52	23	46.3	23.0	18.6	7.0	.21	4.2	4.39	2.47	1.33	1.75	.0096	.00046	.0086	1.5	17.08	4.538	.5210
52–54	18	56	26	25	51	24	36.0	18.0	16.5	5.8	.22	2.5	2.75	2.28	1.37	1.74	.0095	.00038	.0097	.81	25.51	3.588	.1235
56–58	9	68	23	18	56	26	36.9	18.7	12.9	7.5	.14	2.3	2.40	2.35	1.37	1.85	.0099	.00039	.0118	.50	60.02	.2848	.6987
60–62	4	65	31	5	64	31	35.3	20.0	12.7	5.0	.08	2.2	2.28	3.03	1.87	2.01	.0113	.00065	.0136	≈.38	34.61	.2122	.6145
62–64	6	65	29	11	59	30	36.8	16.9	11.5	7.4	.08	2.1	2.16	2.99	1.86	1.90	.0116	.00046	.0153	≈.41	51.46	.04010	.2155
Mean value	15.0	62.4	22.6	25.3	51.3	23.4	36.7	16.7	16.5	6.3	0.22	2.6	2.77	2.42	1.43	1.84	0.0096	0.00044	0.0103	1.6	78.44	6.028	2.443
CRM 14.0, hole 2																							
4–6	11	69	20	15	61	24	38.6	18.3	19.4	8.2	0.11	2.2	2.35	2.68	1.50	1.89	0.0100	0.00039	0.0059	1.7	127.6	11.30	16.40
8–10	8	66	26	15	60	25	46.2	25.0	17.2	10.2	.15	2.4	2.59	2.85	1.51	2.01	.0112	.00039	.0066	4.91	123.1	15.94	7.211
12–14	6	69	25	18	57	25	40.7	18.4	13.6	8.8	.12	2.5	2.63	2.56	1.41	1.95	.0107	.00038	.0061	3.2	229.4	13.39	.8430
16–18	13	63	24	21	41	38	38.1	17.5	15.7	8.7	.13	2.7	2.80	2.65	1.68	1.87	.0103	.00045	.0063	8.60	605.1	64.63	−2.805
20–22	28	54	18	36	46	18	37.3	14.4	11.9	7.4	.14	3.2	3.29	2.22	1.17	1.64	.0083	.00052	.0059	2.7	44.16	10.20	.6095
24–26	13	66	21	24	51	25	39.7	17.3	12.7	7.0	.25	2.3	2.59	2.31	1.51	1.84	.0096	.00041	.0060	2.2	21.04	8.868	1.289
28–30	13	67	20	31	44	25	37.2	15.0	13.2	4.9	.22	2.3	2.49	2.22	1.40	1.82	.0093	.00040	.0055	1.3	31.64	16.30	1.045
32–34	15	61	24	32	47	21	35.8	16.4	15.1	10.2	.36	2.4	2.73	2.30	1.38	1.87	.0096	.00037	.0070	1.2	33.07	4.123	.6315
36–38	13	64	23	24	53	23	36.1	17.4	11.8	6.7	.32	2.2	2.53	2.20	1.38	1.74	.0092	.00035	.0054	1.4	82.55	.4458	.3147
40–42	13	63	24	23	55	22	33.5	17.9	12.8	8.6	.33	2.1	2.40	2.23	1.28	1.77	.0088	.00038	.0058	≈.47	39.88	.2382	.2847
Mean value	13.3	64.2	22.5	23.9	51.5	24.6	38.3	17.8	14.3	8.1	0.21	2.4	2.64	2.42	1.42	1.84	0.0097	0.00040	0.0061	2.77	143.7	14.54	2.582

[1] Separation of sand-size particles by wet sieving of wet sample. Separation of silt- and clay-size particles by bottom-withdrawal tube.

[2] Separation of sand-size particles by wet sieving of previously dried sample. Separation of silt- and clay-size particles by centrifugation.

[3] Radionuclide contents are in picocuries per gram. Negative values of Ru^{106} are the result of the method of gamma-ray spectrum analysis used; true values are near zero.



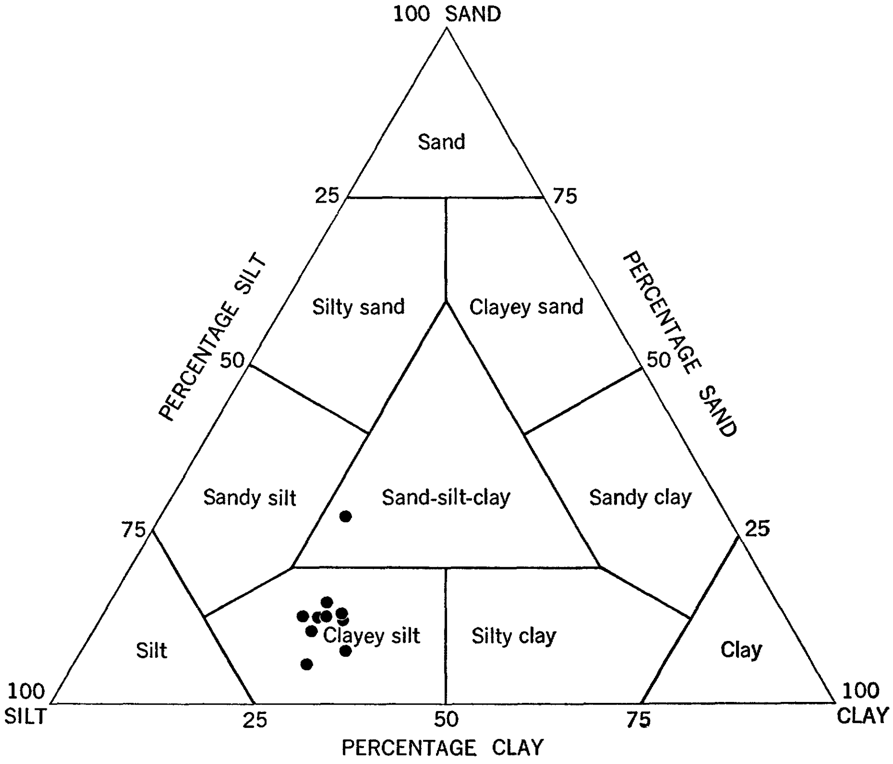


FIGURE 6.—Results of particle-size analyses of selected samples. Upper: From hole 7–2, CRM 7.5. Lower: From hole 2, CRM 14.0. Nomenclature from Shepard (1954).

TABLE 6.—*Particle-size analyses of selected samples of bottom sediment from the Clinch River*

[Analysis by wet sieving and bottom-withdrawal tube; H. E. Reeder, analyst]

Depth interval (in.)	Percent finer than size indicated, in millimeters									
	0.002	0.004	0.008	0.016	0.031	0.062	0.125	0.250	0.500	1.000
CRM 7.5, hole 7-2										
10–12	12	14	18	25	37	60	93	96	98	100
14–16	14	19	23	30	43	72	88	93	95	96
18–20	16	19	26	39	55	82	91	97	100	----
22–24	21	23	34	55	72	84	91	97	98	99
30–32	19	24	36	54	72	87	92	97	100	----
34–36	30	31	45	60	78	94	98	99	100	----
40–42	24	28	38	50	68	91	98	99	100	----
44–46	25	29	43	60	79	88	95	99	100	----
50–52	24	29	39	54	69	83	89	93	96	98
52–54	26	29	40	55	70	82	88	95	98	99
56–58	23	29	42	60	76	91	96	98	99	100
60–62	31	38	52	68	86	96	97	98	99	100
62–64	29	36	51	70	84	94	97	98	99	100
CRM 14.0, hole 2										
4–6	20	27	39	54	74	89	95	98	100	----
8–10	26	33	44	62	79	92	96	97	98	99
12–14	25	29	42	60	78	94	97	98	99	100
16–18	24	30	43	60	75	87	93	97	99	100
20–22	18	23	33	47	60	72	84	95	100	----
24–26	21	28	38	51	69	87	96	98	99	100
28–30	20	25	34	48	66	87	96	99	100	----
32–34	24	27	37	51	66	85	95	97	98	100
36–38	23	30	39	49	69	87	92	98	99	100
40–42	24	27	40	52	64	87	97	98	99	100

material, the size fraction with the highest cation-exchange capacity.

Size separations for cation-exchange capacity determinations were made by wet sieving and centrifugation after the samples had been initially dried (see "Methods and equipment"). This procedure can result in clay-size fragments becoming attached to sand grains and may fail to disperse all aggregates of smaller grains. As noted, it can result also in concentration of organic material in the sand- and clay-size fractions. The data reported in table 7 therefore do not represent the exchange capacities of the ultimate particle-size fractions of the sediment but do represent those of the size fractions approximately as they existed in the natural environment. The data in table 7 are based on the same size separation as the data in columns 5–7, table 5.

The cation-exchange capacity of the clay-size sediment fraction was higher than that of the silt-size fraction in all samples, probably primarily due to the higher content of layered aluminosilicate minerals in the clay-size fraction of the sediment. Aluminosilicate minerals present in the other two size fractions, as well as their content of organic matter, probably account for most of the measured cation-exchange capacities of these two sediment fractions.

MINERALOGICAL CHARACTERISTICS

Semiquantitative determinations of the mineralogical compositions of the clay- and sand-size fractions were made for all 23 sediment samples studied in detail (table 8). The size fractions studied were separated by wet sieving and centrifugation of previously dried samples, which might be expected to preserve some sand- and silt-size aggregates of smaller particles formed by drying. Careful redispersion of the sediment particles has minimized this effect, however, and the mineralogical compositions reported below are considered to be fairly representative of the three size fractions as they existed in the natural environment.

Results of the mineralogical analyses showed that sand-size sediment fractions of all samples were composed of 70–80 percent quartz with lesser amounts of feldspar, dolomite, and mica. Kaolinite was detected in seven samples.

The silt-size fraction of each sample contained 60–70 percent quartz and lesser amounts of mica, mixed-layered mica-vermiculite, vermiculite (trioctahedral), aluminum-interlayered vermiculite (dioctahedral), kaolinite, feldspar, and dolomite (see Grim, 1953, p. 43–83, for discussion of the structures of the clay minerals). Calcite was reported in three samples.

The clay-size fractions (particles <2 microns in diameter) of each sample contained much less quartz—10–20 percent—than did the other two size fractions. Mica and kaolinite occurred in approximately the same abundance as quartz, along with slightly lesser amounts of vermiculite (trioctahedral), aluminum-interlayered

TABLE 7.—*Cation-exchange capacities of clay-, silt-, and sand-size fractions of selected samples of Clinch River bottom sediment*

[Cation-exchange capacities determined by means of ammonium chloride leach and measurement of sorbed ammonium, by distillation; results are in milliequivalents per 100 grams, dry weight; Harry C. Starkey, analyst. Maximum diameter of clay particles was 2 microns. Nd, not determined]

Depth interval (in.)	Sand	Silt	Clay	Depth interval (in.)	Sand	Silt	Clay
CRM 7.5 hole 7-2				**CRM 14.0, hole 2**			
10–12	Nd	11. 7	42. 4	4–6	Nd	12. 0	34. 8
14–16	Nd	13. 8	33. 5	8–10	Nd	13. 5	33. 4
18–20	Nd	9. 6	37. 1	12–14	Nd	13. 2	35. 6
22–24	Nd	10. 2	33. 2	16–18	Nd	15. 1	28. 1
30–32	Nd	10. 9	33. 5	20–22	Nd	14. 2	32. 8
34–36	7. 8	13. 9	33. 8	24–26	10. 5	12. 3	33. 2
40–42	Nd	8. 9	39. 2	28–30	Nd	11. 0	30. 1
44–46	Nd	14. 1	33. 4	32–34	Nd	11. 5	35. 6
50–52	Nd	14. 9	39. 1	36–38	Nd	12. 2	38. 6
52–54	Nd	12. 6	30. 0	40–42	Nd	9. 3	30. 4
56–58	Nd	11. 6	33. 4				
60–62	Nd	10. 1	34. 2				
62–64	Nd	11. 1	32. 2				

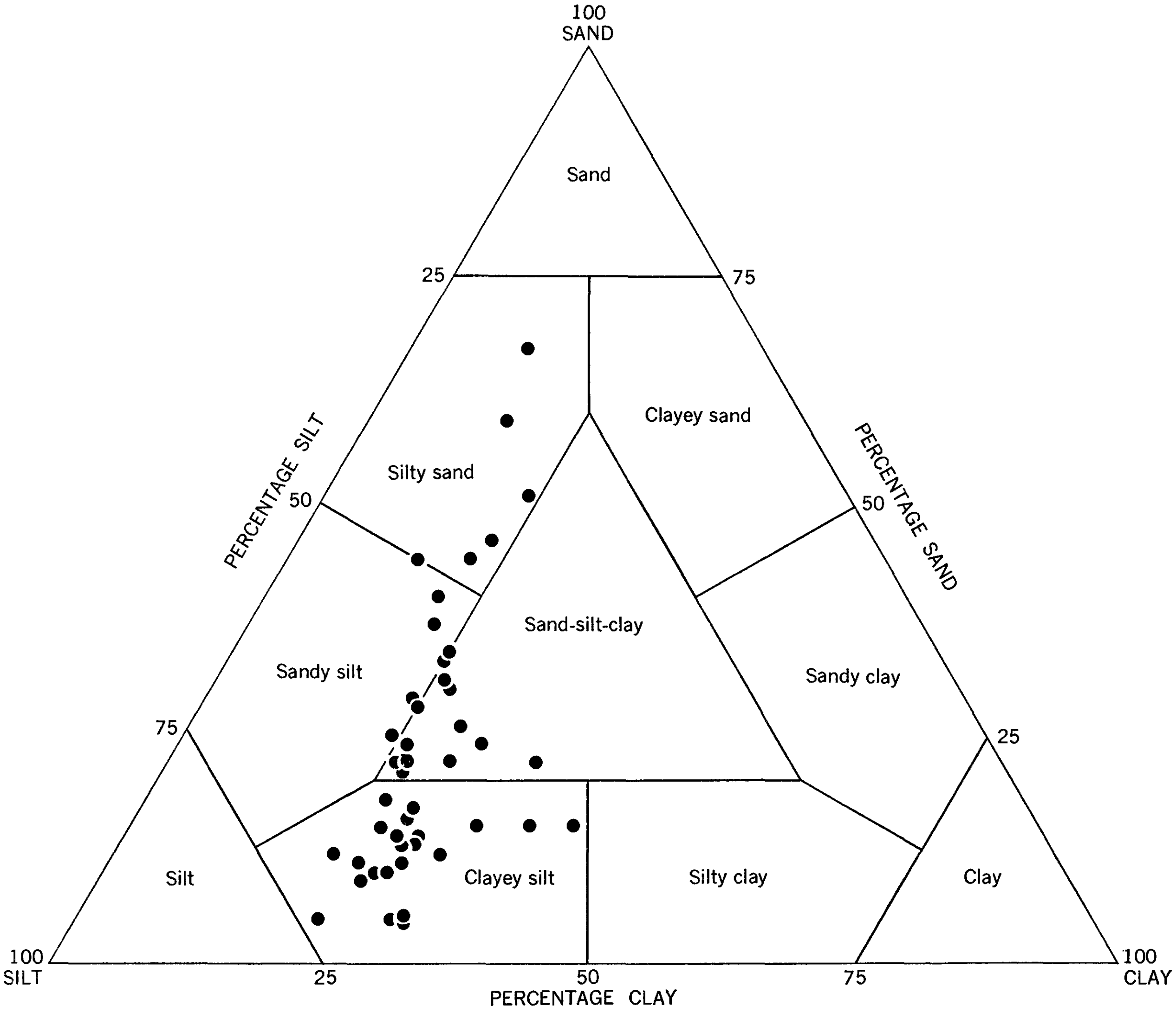


FIGURE 7.—Results of particle-size analyses of composite samples of the radioactive parts of 45 cores of Clinch River bottom sediment. Nomenclature from Shepard (1954).

vermiculite (dioctahedral), and mixed-layered mica-vermiculite. Traces of chlorite, montmorillonite, and feldspar were present in many of the samples.

Dolomite was present in the sand-size fraction of the sediment in some abundance, probably in the form of detrital grains, and was present in the silt-size fractions of all but four of the 23 samples (table 8). Calcite was detected in only a few of the samples; however, calcium carbonate can precipitate in river sediment in a poorly crystallized form which is not susceptible to detection by X-ray methods.

The presence of precipitated calcium carbonate in the bottom sediment was of interest because, in one place where calcite precipitated from Clinch River water on a bimetallic object, the calcite contained an appreciable content of radionuclides. A radiochemical analysis of the calcite showed it to contain 123.9 pc per g (picocuries per gram) ruthenium-103 to ruthenium-106 and 10.1 pc per g strontium-90 (Parker and Blanco, 1963, p. 105). This radionuclide content is within the range commonly found in suspended sediment in Clinch River water downstream from the mouth of Whiteoak Creek. In the place cited, precipitation of the calcite may have been the result of local electrochemical action between the two metals and the river water.

In order to investigate the possible presence of poorly crystallized calcium carbonate in Clinch River bottom sediment, the content of carbonate minerals (mineral carbon) in the raw sediment samples was determined by chemical analysis. In all samples, the content of min-

TABLE 8.—*Mineralogical composition of selected samples of Clinch River bottom sediment*

[Determined by X-ray diffraction method. Because of inherent inaccuracies of method, contents have been rounded to nearest 10 percent, ±5 percent, except where noted. Clay-size fraction, particles <2 microns in diameter; silt-size fraction, particles 2–62 microns in diameter; sand-size fraction, particles >62 microns in diameter. Tr, trace; ----, mineral not detected. P. D. Blackmon, analyst]

Depth interval (in.)	Size fraction	Mica	Vermiculite (trioctahedral)	Mica-vermiculite (mixed-layered)	Mont-morillonite	Chlorite	Aluminum-interlayered vermiculite (dioctahedral)	Kaolinite	Quartz	Feldspar	Dolomite	Calcite
CRM 7.5, hole 7–2												
10–12	Clay	20	10–15	10	Tr	Tr	10–15	20	10–15	----	----	----
	Silt	<10	Tr	Tr	----	----	Tr	Tr	70	<10	<10	----
	Sand	<10	----	----	----	----	----	----	80	<10	10	----
14–16	Clay	20	10–15	<10	Tr	Tr	20	20	10–15	----	----	----
	Silt	10	Tr	Tr	----	Tr	----	Tr	70	Tr	Tr	Tr
	Sand	<10	----	----	----	----	----	----	80	10	<10	----
18–20	Clay	20	10–15	<10	----	Tr	20	20	10–15	Tr	----	----
	Silt	10	Tr	Tr	----	----	Tr	<10	60–65	<10	Tr	Tr
	Sand	<10	----	----	----	----	----	----	70	10	10–15	----
22–24	Clay	10–15	10–15	10	----	Tr	20	20	10–15	----	----	----
	Silt	<10	<10	<10	----	----	<10	<10	60–65	Tr	Tr	----
	Sand	<10	----	----	----	----	----	<10	70	<10	<10	----
30–32	Clay	20	10–15	<10	----	Tr	20	20	10–15	Tr	----	----
	Silt	10	----	<10	----	----	Tr	Tr	60–65	10	----	----
	Sand	<10	----	----	----	----	----	----	70–75	10	10	----
34–36	Clay	20	10–15	<10	----	Tr	10–15	20	20	Tr	----	----
	Silt	10–15	----	Tr	----	----	Tr	<10	70	<10	----	----
	Sand	Tr	----	----	----	----	----	----	70–75	10–15	10	----
40–42	Clay	20	20	10	Tr	----	10	20	20	----	----	----
	Silt	10	----	----	----	----	----	<10	70	<10	Tr	----
	Sand	Tr	----	----	----	----	----	----	70–75	10	10	----
44–46	Clay	20	10–15	10–15	----	Tr	10	20	10–15	----	----	----
	Silt	10	----	<10	----	----	----	Tr	70	<10	Tr	----
	Sand	<10	----	----	----	----	----	Tr	80	<10	Tr	----
50–52	Clay	10–15	10–15	10–15	----	Tr	20	10–15	10–15	----	----	----
	Silt	10	----	----	----	10	----	Tr	70–75	Tr	Tr	----
	Sand	<10	----	----	----	----	----	----	70	10	10–15	----
52–54	Clay	20	10–15	<10	----	Tr	20	20	10–15	----	----	----
	Silt	10	----	----	----	----	10	Tr	60–65	10	Tr	----
	Sand	<10	----	----	----	----	----	----	70–75	10	10	----
56–58	Clay	20	10–15	10	Tr	----	10–15	20	10–15	----	----	----
	Silt	10	<10	----	----	----	<10	----	70	<10	Tr	----
	Sand	Tr	----	----	----	----	----	----	80	<10	<10	----
60–62	Clay	20	10	10	Tr	----	20	20	10–15	----	----	----
	Silt	10	----	Tr	----	----	<10	Tr	70	<10	Tr	Tr
	Sand	Tr	----	----	----	----	----	Tr	70–75	10–15	Tr	----
62–64	Clay	20	10	10	----	----	20	20	20	----	----	----
	Silt	10	----	----	----	----	<10	Tr	70	10	Tr	----
	Sand	10	----	----	----	----	----	Tr	70–75	<10	<10	----
CRM 14.0, hole 2												
4–6	Clay	20	10	10–15	----	Tr	10–15	20	10–15	----	----	----
	Silt	10	----	Tr	----	----	Tr	Tr	70–75	10	----	----
	Sand	<10	----	----	----	----	----	----	80	10	Tr	----
8–10	Clay	20	10–15	10	----	Tr	10–15	20	10–15	----	----	----
	Silt	10–15	----	Tr	----	----	Tr	<10	70	<10	----	----
	Sand	<10	----	----	----	----	----	<10	70	10	<10	----
12–14	Clay	20	10	10–15	----	Tr	10–15	20	10–15	----	----	----
	Silt	10	----	Tr	----	----	Tr	<10	70	<10	Tr	----
	Sand	<10	----	----	----	----	----	----	80	<10	<10	----
16–18	Clay	20–25	10	10	Tr	Tr	10	10–15	20–25	----	----	----
	Silt	10	----	<10	----	----	<10	Tr	70	<10	Tr	----
	Sand	<10	----	----	----	----	----	----	80	10–15	----	----
20–22	Clay	10–15	10–15	10–15	Tr	Tr	10–15	20	10–15	----	----	----
	Silt	<10	----	Tr	----	----	Tr	<10	70	<10	Tr	----
	Sand	Tr	----	----	----	----	----	----	80–85	10	<10	----
24–26	Clay	20	10–15	10–15	----	----	10–15	20	10–15	----	----	----
	Silt	<10	----	Tr	----	----	Tr	Tr	70	10	Tr	----
	Sand	<10	----	----	----	----	----	----	70	10–15	10–15	----
28–30	Clay	10–15	10–15	10–15	----	----	10–15	10–15	20	----	----	----
	Silt	<10	----	Tr	----	----	Tr	<10	70	<10	Tr	----
	Sand	Tr	----	----	----	----	----	Tr	70	10–15	<10	----

TABLE 8.—*Mineralogical composition of selected samples of Clinch River bottom sediment*—Continued

Depth interval (in.)	Size fraction	Mica	Vermiculite (trioctahedral)	Mica-vermiculite (mixed-layered)	Mont-morillonite	Chlorite	Aluminum-interlayered vermiculite (dioctahedral)	Kaolinite	Quartz	Feldspar	Dolomite	Calcite
CRM 14.0 hole 2—Continued												
32–34	Clay	10–15	10	10–15			10–15	20	20	<10		
	Silt	10		Tr			Tr	Tr	70	<10	<10	
	Sand	Tr						Tr	70–75	10	<10	
36–38	Clay	10–15	10–15	10–15	Tr		10–15	20	10–15			
	Silt	10	Tr	Tr			Tr	Tr	70	10	Tr	
	Sand	Tr							70–75	10	10–15	
40–42	Clay	10–15	10–15	10–15			20	20	10–15			
	Silt	10		Tr			Tr		70–75	10	<10	
	Sand	10							70–75	10	<10	

eral carbon was very low compared to the content of organic carbon (table 5). The presence of dolomite as sand- and silt-size particles (table 8) suggests that much of the mineral carbon content was derived from the coarser sediment fractions. Thus, it appears that poorly crystallized calcium carbonate is not abundant in the finer size fractions of Clinch River bottom sediment.

Tamura (1963) has described the strong affinity of strontium-90 for iron and aluminum oxides and hydroxides in basic solutions and has suggested that this affinity is related to the surface area of the sorbent. Results of determinations of free iron oxide and free alumium oxide in the sediment samples are listed in table 5. Constituent correlations indicate that most of the free iron and aluminum oxides occurred in association with clay-size particles.

Organic carbon contents of the samples are listed in table 5. The variation in content of organic carbon between samples is small.

CHEMICAL CHARACTERISTICS

Contents of one minor element and three trace elements in the sediment samples are shown in table 5. Contents of potassium and rubidium were measured as possible indcators of the amount of mica-type silicate minerals in the samples. The contents of cesium and strontium in the samples were of interest because, in chemical sorption and ionic substitution reactions, the amount of the radioactive form of an element that becomes associated with the sediment depends on the relative abundance of the stable form of the element; for example, stable cesium will compete with radiocesium (Cs^{137}) for cation-exchange sites on clay-type minerals. Not only does stable strontium compete with radiostrontium (Sr^{90}) for exchange sites, but both forms compete with the much more abundant calcium. Nelson (1962) measured the ratio of radiostrontium to stable strontium in clamshells in the Clinch River and the Tennessee River and reported that the ratio decreased with distance downstream from the mouth of Whiteoak Creek in the same manner as that predicted on the basis of flow dilution. This relationship is apparently the result of the proportional substitution of radiostronium and stable strontium for some of the calcium ions in the calcium carbonate of the clamshell as the shell is formed.

In 22 of the 23 sediment samples, the contents of leachable calcium and magnesium total more than the cation-exchange capacity of the sediment (table 5). This observation suggests that in addition to sorbed ions displaced by ammonium ions from the leaching solution, minerals dissolved by the leaching solution and interstitial water contained in the sediment contributed to the cation content of the leachate. Dissolution of only a small fraction of the measured content of calcium carbonate (mineral carbon) in the sample could account for the excess calcium and magnesium in the leachates.

The only two major leachable cations found in the sediment were calcium and magnesium. No sodium or potassium was detected. This is to be expected in samples from a humid area in which the predominant rock types are limestone and dolomite and in which the predominant cations in river water are calcium and magnesium (Grim, 1953, p. 144–145).

Results of radiochemical analyses of the 23 sediment samples are shown in table 5.

CORRELATIONS OF PHYSICOCHEMICAL CHARACTERISTICS AND RADIONUCLIDE CONTENT

In general, correlations of patterns of variation in constituent concentration with depth in each of the two sediment cores studied (tables 9 and 10) confirm the interrelationships between cation-exchange capacity, particle size, and mineralogical composition that have been described previously. The correlations also emphasize the close association of potassium and rubidium in geochemical processes. The good correlation of free iron oxide with rubidium in hole 7–2, CRM 7.5, and with both potassium and rubidium in hole 2, CRM 14.0, may be due to a similarity in distribution of particle sizes of free iron oxide and the minerals in which the potassium and rubidium occur. Correlation of cation-exchange capacity with particle-size distribution is better when the particle-size data used are those which were obtained by chemical dispersion and analysis by bottom-withdrawal tube. This method of separation should have resulted in the inclusion of nearly all clay minerals in the clay-size sediment fraction.

Mutual correlations of the contents of strontium-90, cesium-137, and cobalt-60, which occur in both cores, are best in hole 2. For hole 7–2, a correlation of cesium-137 and cobalt-60 contents with those of particles less than 2 microns in diameter was detected after application of a correction for variations in annual releases of cesium-137 to the river (see "Relationship of vertical distribution to annual releases"). The correction was possible for only the upper five samples out of the 13 samples analyzed in hole 7–2. Application of a similar correction to data for all samples from hole 2 did not materially improve the correlations. The corrected contents of cesium-137 and cobalt-60 correlate with cation-exchange capacity and the contents of free iron oxide, free aluminum oxide, adsorbed water, and organic matter in hole 7–2, and with free iron oxide content and free aluminum oxide content in hole 2. The content of ruthenium-106 correlates with leachable calcium in both cores.

In the 45 composite samples of bottom-sediment cores that were analyzed for both particle-size distribution and radionuclide content, the content of sand-size particles shows a strong negative correlation with the content of silt-size particles. This negative correlation emphasizes the previously noted small variation in content of clay-size particles in the composite samples. In the case of a relatively unvarying content of clay-size particles, any major variation in content of silt-size particles will be accompanied by an opposite variation in the content of sand-size particles—hence the reported negative correlation. No correlation of particle-size distribution with the content of individual radionuclides in the samples was noted. A strong correlation of cesium-137 content with cobalt-60 content observed in the composite samples is consistent with the correlation of those two radionuclides observed in other samples of Clinch River bottom sediment.

DISCUSSION

The quantity of a radionuclide sorbed on river sediment might be expected to increase with decreasing particle size of the sediment for two reasons: (1) The relative content of layered aluminosilicate minerals, the mineral group with the highest sorption capacities for many cations (Grim, 1953, p. 131–132), generally increases as the mean particle size of the sediment decreases, and (2) in some reactive materials, including several of the layered aluminosilicate minerals and organic matter, sorption capacity varies directly with surface area, which increases as particle size decreases (Grim, 1953, p. 132; Sayre and others, 1963, p. 15).

The study of the relationship between particle-size distribution and radionuclide content of Clinch River bottom sediment is complicated by the effect of variations in the amounts of each of the radionuclides released annually. Both cores that were studied in detail exhibited patterns of variation in gross gamma radioactivity with depth which were similar to the pattern of annual releases of cesium-137 to the Clinch River from ORNL. Variations in annual releases of cesium-137 are strongly reflected in the contents of cesium-137 and cobalt-60 in the bottom sediment, as has been shown in a preceding section of the report. Because these two radionuclides account for most of the radioactivity in the sediment, corrections must be made for the effect of annual releases in order to investigate the effect of particle-size distribution on the content of the two radionuclides in Clinch River bottom sediment. Attempts at such a correction were made in two ways: (1) In the incremental samples of the two sediment cores, by decreasing the measured radiochemical contents of the individual slices by an amount proportional to the amount of cesium-137 released during the years which the sample is believed to represent, and (2) in the composite samples of the sediment cores, by using cores from parts of the river in which there has been a great preponderance of sediment deposition over sediment erosion, an integrated sample representing sediment deposited during the entire period since 1943 thus being obtained.

Another complicating factor in a study of the relationship of sediment composition to radionuclide content is the effect of dilution of radioactive sediment by nonradioactive sediment. If incorporation of cesium-137 and cobalt-60 in Clinch River bottom sediment is by deposition of suspended radioactive solids, as has been suggested in this report, the concentration of the two radionuclides at a given sample site, and at a given depth, will be greatly affected by the amount of nonradioactive sediment deposited with the radioactive sediment. Because the proportions of the two kinds of

TABLE 9.—*Coefficients of correlation for constituent pairs, hole 7–2, CRM 7.5*

[Clay-size particles, <2 microns in diameter; silt-size particles, 2–62 microns in diameter; sand-size particles, <62 microns in diameter]

	Particle-size distribution[1]			Particle-size distribution[2]			Adsorbed water	Cation-exchange capacity	Leachable-cation content		Carbon content		
	Sand	Silt	Clay	Sand	Silt	Clay			Ca	Mg	Mineral	Organic	Total
Sand[1]		−0.84	−0.85	0.91	−0.81	−0.93	−0.23	−0.57	0.71	0.33	0.52	−0.07	0.05
Silt[1]	−0.84		.43	−.70	.71	.60	.06	.21	−.53	−.36	−.25	−.09	−.16
Clay[1]	−.85	.43		−.83	.87	.90	.32	.74	−.67	−.20	−.63	.20	.07
Sand[2]	.91	−.70	−.83		−.97	−.91	−.29	−.67	.64	.25	.58	−.09	.04
Silt[2]	−.81	.62	.75	−.97		.77	.23	.64	−.48	−.28	−.53	.06	−.07
Clay[2]	−.93	.71	.87	−.91	.77		.36	.63	−.79	−.16	−.58	.14	.01
Adsorbed water	−.23	.06	.32	−.29	.23	.36		.74	−.16	−.29	−.35	.88	.83
Cation-exchange capacity	−.57	.21	.74	−.67	.64	.63	.74		−.47	−.001	−.59	.66	.55
Calcium	.71	−.53	−.67	.64	−.48	−.79	−.16	−.47		.01	.51	.02	.13
Magnesium	.33	−.36	−.20	.25	−.28	−.16	.29	−.001	.01		−.01	.09	.08
Mineral carbon	.52	−.25	−.63	.58	−.53	−.58	−.35	−.59	.51	−.01		−.32	−.09
Organic carbon	−.07	−.09	.20	−.09	.06	.14	.88	.66	.02	−.09	−.32		.97
Total carbon	.05	−.16	.07	.04	−.07	.01	.83	.55	.13	.08	−.09	.97	
Fe_2O_3	−.83	.56	.84	−.92	.87	.87	.29	.64	−.69	−.18	−.58	.06	−.07
Al_2O_3	−.89	.64	.86	−.94	.88	.91	.20	.55	−.69	−.26	−.58	−.01	−.14
Potassium	−.67	.69	.45	−.78	.83	.59	−.09	.23	−.25	−.35	−.17	−.22	−.26
Rubidium	−.90	.74	.79	−.96	.92	.87	.19	.56	−.64	−.29	−.47	−.006	−.11
Cesium	−.44	.23	.50	−.58	.59	.49	.04	.43	−.39	−.26	−.17	−.06	−.08
Strontium	−.31	.21	.31	−.37	.42	.26	−.27	.05	−.30	.04	−.34	−.52	−.65
Sr^{90}	−.15	.28	−.01	−.17	.27	−.04	−.27	−.13	.11	−.53	.12	−.05	−.004
Cs^{137}	−.15	.36	−.10	−.18	.25	.003	−.25	−.14	.05	−.38	−.19	.01	−.02
Co^{60}	−.01	.13	−.10	−.002	.08	−.16	−.20	−.12	.26	−.41	−.04	.18	.19
Ru^{106}	.61	−.37	−.66	.59	−.42	−.76	−.45	−.66	.86	−.02	.42	− .35	−.29
Cs^{137} (corrected)[3]	−.62	.55	.68	−.77	.71	.81	.89	.97	−.49	−.40	−.71	.74	.71
Co^{60} (corrected)[3]	−.60	.49	.74	−.76	.73	.75	.93	.99	−.34	−.38	−.81	.77	.69

	Free oxide content		Minor element content				Radionuclide content					
	Fe_2O_3	Al_2O_3	K	Rb	Cs	Sr	Sr^{90}	Cs^{137}	Co^{60}	Ru^{106}	Cs^{137} (corrected)	Co^{60} (corrected)
Sand[1]	−0.83	−0.89	−0.67	−0.90	−0.44	−0.31	−0.15	−0.15	−0.01	0.61	−0.62	−0.60
Silt[1]	.56	.64	.69	.74	.23	.21	.28	.36	.13	−.37	.55	.49
Clay[1]	.84	.86	.45	.79	.50	.31	−.01	−.10	−.10	−.66	.68	.74
Sand[2]	−.92	−.94	−.78	−.96	−.58	−.37	−.17	−.18	−.002	.59	−.77	−.76
Silt[2]	.87	.88	.83	.92	.59	.42	.27	.25	.08	−.42	.71	.73
Clay[2]	.87	.91	.59	.87	.49	.26	−.04	.003	−.16	−.76	.81	.75
Adsorbed water	.29	.20	−.09	.19	.04	−.27	−.27	−.25	−.20	−.45	.89	.93
Cation-exchange capacity	.64	.55	.23	.56	.43	.05	−.13	−.14	−.42	−.66	.97	.99
Calcium	−.69	−.69	−.25	−.64	−.39	−.30	.11	.05	.26	.86	−.49	−.34
Magnesium	−.18	−.26	−.35	−.29	−.26	.04	−.53	−.38	−.41	−.02	−.40	−.38
Mineral carbon	−.58	−.58	−.17	−.47	−.17	−.34	.12	−.19	−.04	.42	−.71	−.81
Organic carbon	.06	−.01	−.22	−.006	−.06	−.52	−.05	.01	.18	−.35	.74	.77
Total carbon	−.07	−.14	−.26	−.11	−.08	−.65	−.004	−.02	.19	−.29	.71	.69
Fe_2O_3		.97	.68	.92	.72	.51	.04	.03	−.14	−.58	.79	.76
Al_2O_3	.97		.72	.94	.64	.46	.12	.10	−.08	−.57	.77	.75
Potassium	.68	.72		.82	.62	.32	.40	.46	.30	−.19	.56	.53
Rubidium	.92	.94	.82		.59	.47	.17	.24	.06	−.56	.69	.65
Cesium	.72	.64	.62	.59		.30	.06	.02	−.04	−.36	.30	.20
Strontium	.51	.46	.32	.47	.30		−.30	−.13	−.35	.07	−.19	−.07
Sr^{90}	.04	.12	.40	.17	.06	−.30		.69	.72	.11	.78	.76
Cs^{137}	.03	.10	.46	.24	.02	−.13	.69		.90	.03	.99	.96
Co^{60}	−.14	−.08	.30	.06	−.04	−.35	.72	.90		.12	.99	.98
Ru^{106}	−.58	−.57	−.19	−.56	−.36	.07	.11	.03	.12		−.54	−.39
Cs^{137} (corrected)[3]	.79	.77	.56	.69	.30	−.19	.78	.99	.99	−.54		.98
Co^{60} (corrected)[3]	.76	.75	.58	.65	.20	−.07	.76	.96	.98	−.39	.98	

[1] Separation of sand-size particles by wet sieving of wet sample. Separation of silt- and clay-size particles by bottom-withdrawal tube.

[2] Separation of sand-size particles by wet sieving of previously dried sample. Separation of silt- and clay-size particles by centrifugation.

[3] Five values only.

sediment can be expected to vary considerably from place to place, and from time to time at the same place, only a rather imperfect correlation of radionuclide content and sediment composition can be expected at best.

The lack of consistent correlations with particle size of the corrected cesium-137 and cobalt-60 contents of the incremental samples from the two cores is probably the result of the complicating factors just described. Corrections for the effects of annual radionuclide releases were applied to the analyses for the two radionuclides but may not have been adequate. Effects of dilution of radioactive sediment by nonradioactive sediment may have been considerable. Particle-size ranges (sand, silt, clay) used in the comparisons may not have been detailed enough to demonstrate relationships between particle size and radionuclide content. The latter two statements might be used also to explain the poor correlations between particle-size and radionuclide content exhibited by the 45 composite samples analyzed. This conclusion is supported by the results of R. W. Andrew's work, which has been described in a preceding section of this report.

TABLE 10.—*Coefficients of correlation for constituent pairs, hole 2, CRM 14.0*

[Clay-size particles, <2 microns in diameter; silt-size particles, 2-62 microns in diameter; sand-size particles, >62 microns in diameter]

	Particle-size distribution[1]			Particle-size distribution[2]			Adsorbed water	Cation-exchange capacity	Leachable-cation content		Carbon content		
	Sand	Silt	Clay	Sand	Silt	Clay			Ca	Mg	Mineral	Organic	Total
Sand[1]	----	−0.91	−0.69	0.80	−0.52	−0.40	−0.43	−0.63	−0.42	−0.26	0.07	0.69	0.78
Silt[1]	0−.91	----	.33	−.74	.51	.32	.37	.43	.43	−.06	−.20	−.71	−.83
Clay[1]	−.69	.33	----	−.53	.28	.34	.32	.68	.20	.68	.17	.34	−7.32
Sand[2]	.80	−.74	−.53	----	−.72	−.42	−.55	−.77	−.63	−.38	.41	.46	.60
Silt[2]	−.52	.51	.28	−.72	----	−.32	.41	.67	.43	.34	−.14	−.49	−.54
Clay[2]	−.40	.32	.34	−.42	−.32	----	.20	.19	.31	.07	−.36	−.005	−.13
Adsorbed water	−.43	.37	.32	−.55	.41	.20	----	.77	.45	.32	−.60	.12	.009
Cation-exchange capacity	−.63	.43	.68	−.77	.67	.19	.77	----	.54	.59	−.23	−.30	−.35
Calcium	−.42	.43	.20	−.63	.43	.31	.45	.54	----	.50	−.45	−.19	−.34
Magnesium	−.26	−.06	.68	−.38	.34	.07	.32	.59	.50	----	−.04	.05	.07
Mineral carbon	.07	−.20	.17	.41	−.14	−.36	−.60	−.23	−.45	−.04	----	−.41	−.24
Organic carbon	.69	−.71	−.34	.46	−.49	−.005	.12	−.30	−.19	.05	−.48	----	.96
Total carbon	.78	−.83	−.32	.60	−.54	−.13	.009	−.35	−.34	.07	−.24	.96	----
Fe_2O_3	−.55	.46	.45	−.79	.45	.50	.77	.77	.82	.58	−.68	.003	−.17
Al_2O_3	−.60	.56	.38	−.62	−.02	.87	.44	.42	.57	.19	−.33	−.24	−.35
Potassium	−.84	.22	.66	−.73	.44	.41	.71	.74	.67	.54	−.31	−.35	−.46
Rubidium	−.79	.65	.67	−.77	.39	.52	.80	.78	.64	.52	−.46	−.19	−.31
Cesium	.76	−.67	−.56	.44	−.49	.05	.002	−.36	−.16	−.14	−.50	.89	.83
Strontium	−.12	−.13	.50	−.08	−.01	.12	.35	.39	.46	.85	−.004	.18	.22
Sr^{90}	−.12	−.03	.33	−.33	−.29	.82	.47	.33	.31	.31	−.59	.45	.33
Cs^{137}	−.28	.12	.43	−.45	−.22	.90	.30	.29	.38	.33	−.53	.27	.12
Co^{60}	−.08	.02	.15	−.20	−.50	.91	.20	.05	.30	.10	−.52	.37	.23
Ru^{106}	−.27	.44	−.16	−.58	.66	−.05	.37	.41	.84	.18	−.45	−.26	−.40
Cs^{137} (corrected)	−.23	.09	.36	−.41	−.27	.91	.27	.24	.35	.27	−.53	.29	.15
Co^{60} (corrected)	−.08	.02	.14	−.19	−.50	.91	.20	.05	.30	.09	−.52	.37	.23

	Free oxide content		Minor element content				Radionuclide content					
	Fe_2O_3	Al_2O_3	K	Rb	Cs	Sr	Sr^{90}	Cs^{137}	Co^{60}	Ru^{106}	Cs^{137} (corrected)	Co^{60} (corrected)
Sand[1]	−0.55	−0.60	−0.84	−0.79	0.76	−0.12	−0.12	−0.28	−0.08	−0.27	−0.23	−0.08
Silt[1]	.46	.56	.22	.65	−.67	−.13	−.03	.12	.02	.44	.09	.02
Clay[1]	.45	.38	.66	.67	−.56	.50	.33	.43	.15	−.16	.36	.14
Sand[2]	−.79	−.62	−.73	−.77	.44	−.08	−.33	−.45	−.20	−.58	−.41	−.19
Silt[2]	.45	−.02	.44	.39	−.49	−.01	−.29	−.22	−.50	.66	−.27	−.50
Clay[2]	.50	.87	.41	.52	.05	.12	.82	.90	.91	−.05	.91	.91
Adsorbed water	.77	.44	.71	.80	.002	.35	.47	.30	.20	.37	.27	.20
Cation-exchange capacity	.77	.42	.74	.78	−.36	.39	.33	.29	.05	.41	.24	.05
Calcium	.82	.57	.67	.64	−.16	.46	.31	.38	.30	.84	.35	−.30
Magnesium	.58	.19	.54	.52	−.14	.85	.31	.33	.10	.18	.27	.09
Mineral carbon	−.68	−.33	−.31	−.46	−.50	−.004	−.59	−.53	−.52	−.45	−.53	−.52
Organic carbon	.003	−.24	−.35	−.19	.89	.18	.45	.27	.37	−.26	.29	.37
Total carbon	−.17	−.35	−.46	−.31	.83	.22	.33	.12	.23	−.40	.15	.23
Fe_2O_3	----	.67	.81	.88	−.05	.46	.64	.65	.48	.60	.61	.48
Al_2O_3	.67	----	.67	.73	−.19	.28	.68	.72	.70	.27	.72	.70
Potassium	.81	.67	----	.95	−.46	.55	.36	.40	.25	.40	.34	.24
Rubidium	.88	.73	.95	----	−.34	.49	.55	.57	.39	.36	.52	.39
Cesium	−.05	−.19	−.46	−.34	----	−.009	.40	.20	.40	−.14	.25	.41
Strontium	.46	.28	.55	.49	−.009	----	.35	.27	.20	.05	.22	.20
Sr^{90}	.64	.68	.36	.55	.40	.35	----	.94	.92	−.05	.94	.92
Cs^{137}	.65	.72	.40	.57	.20	.27	.94	----	.92	−.009	.99	.92
Co^{60}	.48	.70	.25	.39	.40	.20	.92	.92	----	−.08	.94	≈1.00
Ru^{106}	.60	.27	.40	.36	−.14	.05	−.05	−.009	−.08	----	−.02	−.08
Cs^{137} (corrected)	.61	.72	.34	.52	.25	.22	.94	.99	.94	−.02	----	.94
Co^{60} (corrected)	.48	.70	.24	.39	.41	.20	.92	.92	≈1.00	−.08	.94	----

[1] Separation of sand-size particles by wet sieving of wet sample. Separation of silt- and clay-size particles by bottom-withdrawal tube.

[2] Separation of sand size particles by wet sieving of previously dried sample. Separation of silt and clay-size particles by centrifugation.

Contents of radionuclides which have been incorporated in bottom sediment as a result of simple cation-exchange reactions might be expected to correlate with the total cation-exchange capacity of the sediment. Desorption experiments performed on sediment collected near the mouth of Whiteoak Creek have indicated that, of the four most important radionuclides in Clinch River bottom sediment (strontium-90, cesium-137, cobalt-60, and ruthenium-106), only strontium-90 was held primarily by simple ion exchange (Morton, 1965, p. 53). Even in the case of strontium-90, there is evidence that some of the radionuclide is associated with calcium carbonate as the result of a process other than simple ion exchange.

The content of strontium-90 in the samples from the two sediment cores does not show a good correlation with either the total cation-exchange capacity or the mineral carbon (calcium carbonate) content of the samples. This lack of good correlation may be due to the effect of annual releases of strontium-90. Measured contents of strontium-90 could not be corrected for variations in annual releases as were contents of cesium-137 and cobalt-60. Also, if detrital calcium carbonate predominates over precipitated calcium carbonate in the sediment, the content of strontium-90 cannot be expected to correlate with the mineral carbon content of the samples.

In the two samples for which the cation-exchange capacity of the sand-size sediment fraction was determined, the cation-exchange capacity of the silt-size sediment fraction exceeds that of the sand-size fraction. In all samples the cation-exchange capacity of the clay-size fraction exceeds that of the silt-size fraction by a ratio of nearly 3:1 (see table 7). These relationships between the various size fractions of the sediment can be largely explained by the relative content of layered aluminosilicate minerals in each size fraction. The ratio of the content of these aluminosilicate minerals in the clay-size fracton to that in the silt-size fraction is also almost 3:1. The higher specific surface area of the clay-size fraction no doubt has modified the effect of mineralogy on the cation-exchange capacity of the sediment.

The role which calcium carbonate has played in the incorporation of radionuclides in Clinch River bottom sediment requires further clarification. Evidence for the precipitation of calcium carbonate in the Clinch River has been obtained, yet the carbonate content of the sediment appears to be minor, and much of that which is present appears to be in the form of detrital carbonate grains associated with the larger particles of sediment. It has been observed (Morton, 1965, p. 58) that the content of calcium carbonate is higher in bottom sediment from Whiteoak Lake than in bottom sediment from near the mouth of Whiteoak Creek, 0.6 mile downstream, and that the fraction of the total content of strontium-90 desorbed from Whiteoak Lake sediment through ion-exchange mechanisms is less than in sediment from near the mouth of the creek. The tendency of strontium to substitute for calcium in the lattices of calcium carbonate minerals (Nelson, 1962), the observed inverse relationships between the calcium carbonate content of Whiteoak Creek bottom sediment and the amount of strontium-90 held through ion exchange, the incorporation of both strontium-90 and ruthenium-106 in calcium carbonate that precipitated in the Clinch River, and the correlation of ruthenium-106 with leachable calcium in Clinch River bottom sediment, all illustrate the need for further study of the role which calcium carbonate plays in incorporation of radionuclides in Clinch River bottom sediment.

The significance of the content of free oxides in the samples of bottom sediment requires further investigation. The correlation of free iron oxide with the clay-size sediment fraction suggests that iron oxide coatings on sand grains are not a major source of the iron. Iron and aluminum oxides should be investigated further because of their high specific sorption capacities for strontium. Sorption of cobalt by iron and manganese oxides also should be investigated.

Organic matter (organic carbon) appears to add to the cation-exchange capacity of the sediment in hole 7–2 (see table 9). The reported high cation-exchange capacities of a number of organic materials (Faust, 1963, p. 28) support this observation. Both vegetal material, consisting of individual leaves and leaf mats, twigs, and other woody material, and metabolic products and remains of aquatic animals are present in the river. Coal is an obvious constituent of the coarser sediment fractions in many places throughout the study reach.

The use of minor and trace elements to indicate the relative abundance of certain layered aluminosilicate minerals in samples of Clinch River bottom sediment appears to be complicated by the presence of other minerals, such as feldspar, which contain the same constituents. The content of adsorbed water in the sediment seems to be a better indication of its cation-exchange capacity and possibly, therefore, of its content of layered aluminosilicate minerals.

The mutual correlations between the contents of strontium-90, cesium-137, and cobalt-60 in the two cores (quite strong correlation in hole 2) suggest that the strontium-90 content of Clinch River bottom sediment may be related in some way to the process of sedimentation of suspended matter in the river, which is believed to be the process by which cesium-137 and cobalt-60 are incorporated in the bottom sediment, as has been noted in a preceding section of this report. This relationship could result from sedimentation either of clay minerals on which strontium-90 has been sorbed or of precipitated calcium carbonate containing strontium-90 in its crystal structure.

CONCLUSIONS

On the basis of the information reported in this paper, the following conclusions can be drawn:

1. Variations in gross gamma radioactivity with depth in Clinch River bottom sediment largely reflect variations in the content of cesium-137 in the sediment.
2. A similar pattern of variations in the content of cesium-137 in the sediment at several coring sites in the reach of river downstream from CRM 18 indicates fairly regular, persistent deposition of sediment at those particular sites, and thus long-term net accumulation of sediment at those sampling sections.

3. Incorporation of cesium-137 in Clinch River bottom sediment by sedimentation of cesium-bearing aluminosilicate minerals entering the Clinch River from the Oak Ridge National Laboratory is indicated by: (1) the similarity of the patterns of variations with depth in cesium-137 content of bottom-sediment cores to the pattern of annual releases of cesium-137 from the laboratory to the Clinch River, (2) the knowledge that at least 70 percent of the cesium-137 released from the laboratory to the Clinch River was associated with suspended solids, and (3) the known preferential sorption of cesium by certain layered aluminosilicate minerals.
4. The similarity of the distribution pattern of cobalt-60 to that of cesium-137 in Clinch River bottom sediment suggests that cobalt-60 may be incorporated in the sediment by deposition of suspended solids entering the Clinch River from Whiteoak Creek. The two radionuclides are not necessarily associated with the same solids, however. The fact that the distribution pattern of cobalt-60 does not reflect the pattern of annual releases of cobalt-60 from Whiteoak Creek implies that most of the cobalt-60 released to the Clinch River is not associated with suspended sediment large enough to be deposited in the Clinch River, and that a rather small fraction of the total cobalt-60 released to the river becomes incorporated in Clinch River bottom sediment.
5. The relatively small contribution of ruthenium-106 to the gross gamma radioactivity of the upper part of Clinch River bottom sediment indicates that only a small fraction of the total amount of ruthenium-106 released to the river is incorporated in the bottom sediment. The manner of incorporation of ruthenium-106 in the sediment is not known.
6. Cation-exchange properties of Clinch River bottom sediment are largely controlled by its content of mica and clay minerals. These minerals are found primarily in the finest sediment fraction, but occur also in the coarser fractions in the form of mineral aggregates and shale particles. Calcium and magnesium are the major leachable cations in the sediment; potassium and sodium are not present in measurable quantities.
7. The effects of chemical and physical properties of Clinch River bottom sediment on its radionuclide content are obscured as a result of variations in annual releases of the radionuclides and as a result of dilution of radioactive sediment by nonradioactive sediment in the river. Corrections for these two factors should be applied if an assessment of the effect of sediment composition on radionuclide content is sought.

REFERENCES CITED

Barker, F. B., 1958, Factors affecting the transport of radioactivity by water: Am. Water Works Assoc. Jour., v. 50, no. 5, p. 603–612.

Browder, F. N., 1959, Radioactive waste management at Oak Ridge National Laboratory, *in* Hearings on industrial radioactive waste disposal before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy: U.S. 86th Cong., 1st sess., v. 1, p. 461–514.

Carrigan, P. H., Jr., 1969, Inventory of radionuclides in bottom sediment of the Clinch River, eastern Tennessee: U.S. Geol. Survey Prof. Paper 433–I, in press.

Carrigan, P. H., Jr., and Pickering, R. J., 1967, Radioactive materials in bottom sediment of Clinch River: Part B, Inventory and vertical distribution of radionuclides in undisturbed cores: U.S. Atomic Energy Comm. ORNL–3721, Supp. 2B, 80 p.

Carritt, D. E., and Goodgal, S. H., 1953, The sorptive and zeolitic properties of natural and waterborne silts, with relation to their capacities to remove, transport, concentrate, and regenerate radioactive waste components in natural waters: Department of Sanitary Engineering, Johns Hopkins University NYO–4591, 97 p.

Churchill, M. A., Cragwall, J. S., Jr., Andrew, R. W., Jr., and Jones, S. L., 1965, Concentrations, total stream loads, and mass transport of radionuclides in the Clinch and Tennessee Rivers: U.S. Atomic Energy Comm. ORNL–3721, Supp. 1, 78 p.

Cottrell, W. D., 1959, Radioactivity in silt of the Clinch and Tennessee Rivers: U.S. Atomic Energy Comm. ORNL–2847, 50 p.

Cowser, K. E., and Snyder, W. S., 1966, Safety analysis of radionuclide release to the Clinch River: U.S. Atomic Energy Comm. ORNL–3721, Supp. 3, 115 p.

Faust, G. T., 1963, Physical properties and mineralogy of selected samples of the sediments from the vicinity of the Brookhaven National Laboratory, Long Island, New York: U.S. Geol. Survey Bull. 1156–B, 34 p.

Feldman, Cyrus, and Rains, T. C., 1964, The collection and flame photometric determination of cesium: Anal. Chemistry, v. 36, p. 405–409.

Glasstone, Samuel, 1958, Sourcebook on atomic energy [2d ed.]: New York, D. Van Nostrand Co., Inc., 546 p.

Grim, R. E., 1953, Clay mineralogy: New York, McGraw-Hill Book Co., Inc., 384 p.

Hashimoto, I., and Jackson, M. L., 1960, Rapid dissolution of allophane and kaolinite-halloysite after dehydration, *in* Clays and clay minerals. Monograph 5: New York, Pergamon Press, p. 102–104.

Jackson, M. L., 1958, Soil chemical analysis: Englewood, N.J., Prentice-Hall, Inc., 498 p.

Jacobs, D. G., 1960, Sorption of cesium by Conasauga shale: Health Physics, v. 4, p. 157–163.

Jacobs, D. G., and Tamura, Tsuneo, 1960, The mechanism of ion fixation using radio-isotope techniques: Internat. Cong. Soil Science, 7th, Madison, Wis., Trans., v. 2, p. 206–214.

Jenne, E. A., and Wahlberg, J. S., 1965, Manganese and iron oxide scavenging of cobalt 60 in White Oak Creek sediment (Oak Ridge, Tenn.) [abs]: Am. Geophys. Union Trans., v. 46, no. 1, p. 170.

Kornegay, B. H., Vaughan, W. A., Jamison, D. K., and Morgan, J. M., Jr., eds., 1963, Transport of radionuclides in fresh water systems: U.S. Atomic Energy Comm. TID–7664, 406 p.

Lane, E. W., chm., 1947, Report of the Subcommittee on Sediment Terminology: Am. Geophys. Union Trans., v. 28, no. 6, p. 936–938.

Lomenick, T. F., 1963, Movement of ruthenium in the bed of Whiteoak Lake: Health Physics, v. 9, p. 838–845.

McBain, J. W., 1950, Colloid Science: New York, D. C. Heath and Co., 450 p.

McMaster, W. M., and Waller, H.D., 1965, Geology and soils of Whiteoak Creek basin, Tennessee: U.S. Atomic Energy Comm. ORNL–TM–1108, 37 p.

Mehra, O. P., and Jackson, M. L., 1960, Iron oxide removal from soils and clays by a dithionite-citrate system buffered with sodium bicarbonate, *in* Clays and clay minerals, Monograph 5: New York, Pegamon Press, p. 317–323.

Morton, R. J., ed., 1961, Status report No. 1 on Clinch River Study: U.S. Atomic Energy Comm. ORNL–3319, 81 p.

——— 1962a, Status report No. 2 on Clinch River Study: U.S. Atomic Energy Comm. ORNL–3202, 97 p.

——— 1962b, Status report No. 3 on Clinch River Study: U.S. Atomic Energy Comm. ORNL–3370, 123 p.

——— 1963, Status report No. 4 on Clinch River Study: U.S. Atomic Energy Comm. ORNL–3409, 115 p.

——— 1965, Status report No. 5 on Clinch River Study: U.S. Atomic Energy Comm. ORNL–3721, 149 p.

Nelson, D. J., 1962, Clams as indicators of strontium-90: Science v. 137, p. 38–39.

Parker, F. L., and Blanco, R. E., 1963, Waste treatment and disposal progress report for November–December 1962 and January 1963: U.S. Atomic Energy Comm. ORNL–TM–516, p. 105.

Pickering, R. J., 1966, River-bottom sediment sampling with a Swedish Foil Sampler, *in* Selected techniques in water resources investigations: U.S. Geol. Survey Water-Supply Paper 1822, p. 94–99.

Pickering, R. J., Carrigan, P. H., Jr., and Parker, F. L., 1965, The Clinch River Study—An investigation of the fate of radionuclides released to a surface stream: U.S. Geol. Survey Circular 497, 12 p.

Pickering, R. J., Carrigan, P. H., Jr., Tamura, Tsuneo, Abee, H. H., Beverage, J. W., and Andrew, R W, Jr, 1966, Radioactivity in bottom sediment of the Clinch and Tennessee Rivers, *in* Disposal of radioactive wastes into seas, oceans and surface waters: Vienna, Austria, Internat. Atomic Energy Agency, p. 57–86.

Rader, L. F., and Grimaldi, F. S., 1961, Chemical analyses for selected minor elements in Pierre shale: U.S. Geol. Survey Prof. Paper 391–A, 45 p.

Rains, T. C., Zittel, H. E., and Ferguson, Marion, 1962, Flame spectrophotometric determination of micro concentrations of strontium in calcareous material: Anal. Chemistry, v. 34, p. 778–781.

Robinson, B. P., 1962, Ion-exchange minerals and disposal of radioactive wastes—a survey of literature: U.S. Geol. Survey Water-Supply Paper 1616, 132 p.

Sayre, W. W., Guy, H. P., and Chamberlain, A. R., 1963, Uptake and transport of radionuclides by stream sediments: U.S. Geol. Survey Prof. Paper 433–A, 35 p.

Schonfeld, Ernest, Kibbey, Arlene H., and Davis, Wallace. Jr., 1965, Determination of nuclide concentrations in solution containing low levels of radioactivity by least-squares resolution of the gamma-ray spectra : U.S. Atomic Energy Comm. ORNL–3744, 58 p.

Shepard, F. P., 1954, Nomenclature based on sand-silt-clay ratios: Jour. Sed. Petrology, v. 24, p. 151–158.

Sorathesn, Aroon, Bruscia, Giuseppe, Tamura, Tsuneo, and Struxness, E. G., 1960, Mineral and sediment affinity for radionuclides: U.S. Atomic Energy Comm. ORNL–60–6–93, 28 p.

Struxness, E. G., Carrigan, P. H., Jr., Churchill, M. A., Cowser, K. E., Morton, R. J., Nelson, D. J. and Parker, F. L., 1967, Comprehensive report of the Clinch River Study: U.S. Atomic Energy Comm. ORNL–4035, 121 p.

Tamura, Tsuneo, 1963, Selective ion exchange reactions of cesium and strontium by soil minerals, *in* Colloque International sur la Rétention et la Migration des Ions Radioactifs dans les Sols: Paris, Presses Universitaires de France, p. 95–104.

Tamura, Tsuneo, and Jacobs, D. G., 1960, Structural implications in cesium sorption: Health Physics, v. 2, p. 391–398.

Tourtelot, H. A., Huffman, Claude, Jr., and Rader, L. F., 1964, Cadmium in samples of the Pierre Shale and some equivalent stratigraphic units, Great Plains Region, *in* Short papers in geology and hydrology: U.S. Geol. Survey Prof. Paper 475–D, p. D73–D78.

U.S. Inter-Agency River Basin Committee, 1953, A study of new methods for size analysis of suspended sediment samples, *in* A study of methods used in measurement and analysis of sediment loads in streams: Minneapolis, Minn., St. Anthony Falls Hydraulic Lab. Rept. 7, 102 p.

U.S. Geological Survey 1958, Surface-water supply of the United States 1956, Part 3–B: U.S. Geol. Survey Water-Supply Paper 1436, 262 p.

U.S. GOVERNMENT PRINTING OFFICE: 1969