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RADIOACTIVE MATERIALS IN BOTTOM SEDIMENT OF CLINCH RIVER: PART B, INVENTORY AND VERTICAL DISTRIBUTION OF RADIONUCLIDES IN UNDISTURBED CORES

P. H. Carrigan, Jr.
R. J. Pickering

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RADIOACTIVE MATERIALS IN BOTTOM SEDIMENT OF CLINCH RIVER: PART B, INVENTORY AND VERTICAL DISTRIBUTION OF RADIONUCLIDES IN UNDISTURBED CORES

Supplement No. 2B to Status Report No. 5 on Clinch River Study

By

P. H. Carrigan, Jr. (USGS)

R. J. Pickering (USGS)

Final Progress Report, Part B, of Subcommittee on Bottom Sediment Sampling and Analysis

MARCH 1967

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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.

ABSTRACT

Inventory, retention, and distribution of radionuclides in bottom sediment of the Clinch River, and factors influencing association of radionuclides with this sediment have been experimentally investigated as part of the Clinch River Study. Experimental work included a field sampling program and a variety of physicochemical analyses in the laboratory.

Core samples of bottom sediment were collected by means of a Swedish Foil Sampler from a 21-mile reach of the Clinch River. The study reach extended from the mouth of the river to the mouth of White Oak Creek and included fourteen sampling sections. Four to thirteen cores were obtained from each section, the number depending on the transverse distribution of the bottom sediment.

Physicochemical analyses performed on sediment samples included gross gamma counting, gamma spectrometry, radiochemical extractions and beta counting, particle-size analyses, mineralogical analyses and determinations of cation exchange capacities and leachable cation content.

The total quantities of the principal radionuclides contained in bottom sediment in the 21-mile study reach at the time of sampling (July 1962) were: 150 curies of ^{137}Cs, 18 curies of ^{60}Co, 16 curies of ^{106}Ru, 2.9 curies of ^{90}Sr, and at least 10 curies of rare earths. Through comparison of the inventory to the total release of each radionuclide to the Clinch River for the period of record, 1943-62, (adjusting for radioactive decay) the percentage of retention is computed to be: 21 percent for ^{137}Cs, 9 percent for ^{60}Co, 0.4 percent for ^{106}Ru, and 0.2 percent for ^{90}Sr. Retention of rare earths may approach, and possibly exceeds, 25 percent.

Ninety-five percent of the total amount of radionuclides in the sediment is in the section of channel between CRM 0 and CRM 15. The longitudinal distribution in the river, based on mean sectional concentrations, is similar throughout the study reach for all radionuclides. Highest radionuclide concentrations occur at, or near, the

mouth of White Oak Creek, where the smallest sectional volume of radioactive sediment in the reach is found.

Similar patterns in the distribution of gamma radioactivity with depth in the sediment, largely controlled by the distribution of ^{137}Cs, were observed in cores collected throughout the 21-mile reach. This persistent pattern indicates that more or less regular sediment deposition, as well as net accumulation of sediment, has taken place at the sites sampled. However, there were parts of each sampling section in which either no sediment deposits were found or the sediment present was not radioactive.

The vertical distribution of ^{137}Cs in the sediment reflects annual variations in the release of this radionuclide to the Clinch River. Strong similarities in vertical distribution patterns of ^{137}Cs and ^{60}Co in the sediment suggest that incorporation of these radionuclides in Clinch River bottom sediment was due to deposition of radioactive suspended sediment originating almost exclusively from White Oak Creek.

The radioactive sediment may be classed as a clayey silt, with surprisingly little vertical variation in mineralogical composition and in cation exchange capacity. The sand- and silt-size fractions of the sediment are composed largely of quartz grains. The clay-size fraction is made up of varying mixtures of mica, clay minerals, and quartz.

INTRODUCTION

Oak Ridge National Laboratory (ORNL) began processing radioactive materials in 1943, and has released low-level liquid radioactive waste to the Clinch River via White Oak Creek since that year (see Fig. 1). This waste disposal practice has resulted in incorporation of some of the released radionuclides in bottom sediment in the river[1].

The quantity, distribution, and nature of the radioactive bottom sediment must be known in order to determine the proportion of the released radionuclides being retained in the river system, and the locus and duration of retention. With this information, the possibility of future radiation hazards resulting from current disposal practices can be assessed. An evaluation of continued use of the river for disposal of radioactive wastes can be made also. Through knowledge of the physical, mineralogical, and chemical composition of the radioactive sediment, an understanding of the mechanisms whereby the various radionuclides were incorporated in the sediment may be gained. Sampling techniques and methods of estimating and predicting radionuclide content

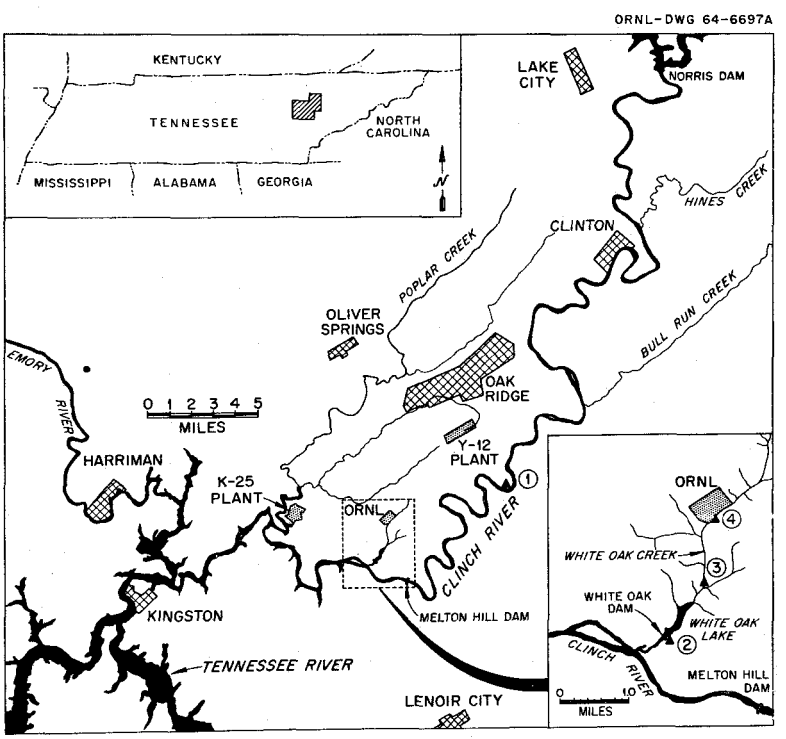

Fig. 1. Map of White Oak Creek, Clinch River, and Tennessee River in Vicinity of Oak Ridge, Tennessee. Numbers in circles indicate streamgaging station.

of the bottom sediment, developed through such a study, may suggest alternate or additional procedures that can be applied to long-term monitoring of the sediment. Bottom sediment studies thus can contribute to the realization of all five stated objectives of the Clinch River Study[2, 3]: (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 this river for radioactive waste disposal purposes, and (5) to provide appropriate conclusions regarding long-term monitoring procedures.

A series of core samples of radioactive bottom sediment was taken during the summer of 1962 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 (figure 2), by means of the Swedish Foil Sampler and other samplers. Sampling sections were selected for adequate coverage of the study reach and, wherever possible, were made to coincide with sediment ranges used by the Tennessee Valley Authority (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 distribution of radioactive sediment. The most downstream sampling section was at CRM 1.3 and the most upstream section was at CRM 22.8.

The vertical distribution of gross gamma radioactivity and the major gamma-emitting radionuclides in the cores was measured by means of a device, known as a core scanner, which was developed especially for that purpose[4]. The radioactive portions of the cores, as determined from the core scans, were mixed, sampled, and analyzed for radionuclide content in order to compute an inventory of radionuclides in bottom sediment in the Clinch River. Composite samples of 45 of the cores were analyzed for particle-size distribution.

The chemical form and associations of radionuclides in the

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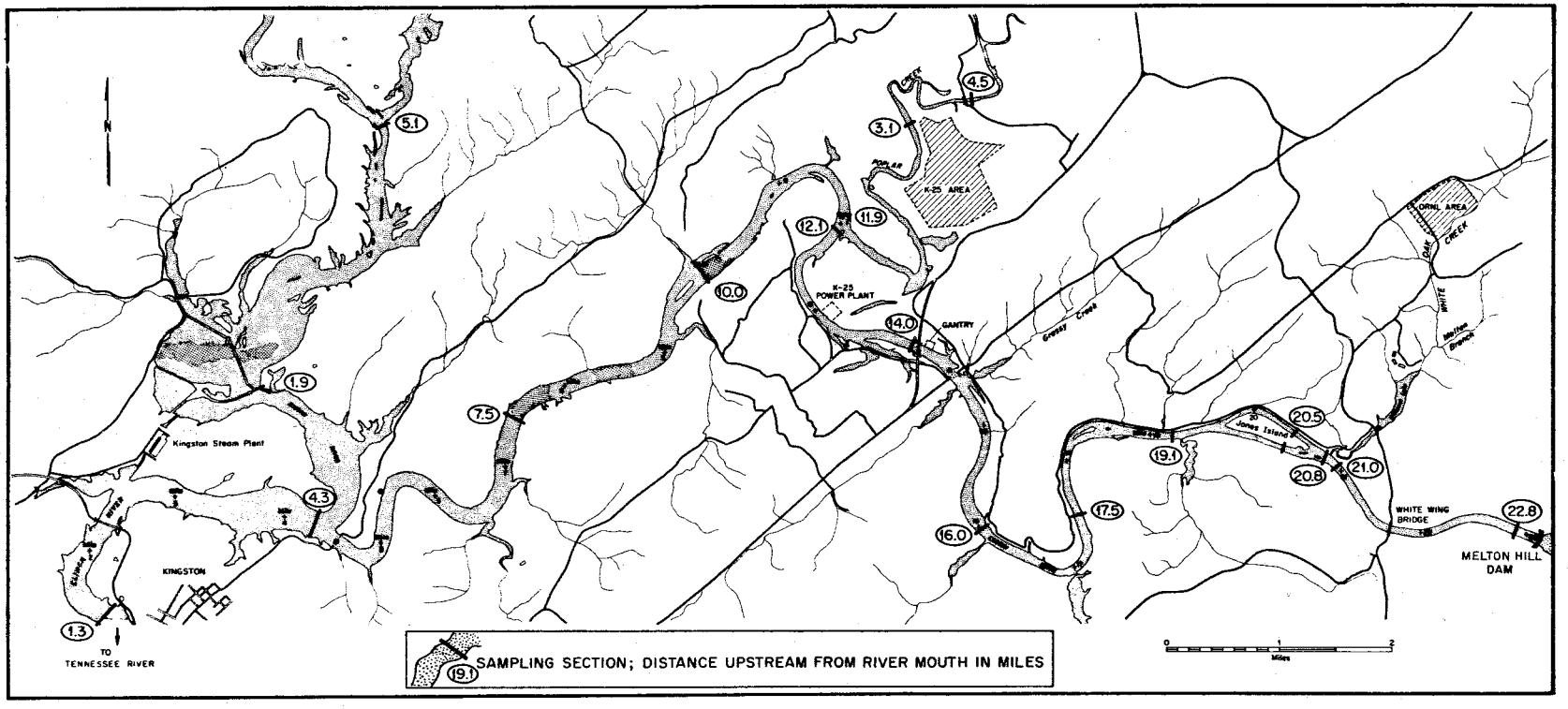

Fig. 2. Map of Clinch River Showing Locations of Bottom Sediment Core Sampling Sections[4]

bottom sediment were investigated through a program of detailed analysis of 23 sediment samples from 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 might be responsible for the presence of each of the radionuclides.

METHODS OF SAMPLE COLLECTION AND ANALYSES

Coring

Samplers.--Relatively undisturbed cores of bottom sediment were obtained by means of the Swedish Foil Sampler. The Swedish Foil Sampler is a 2 1/2 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.

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 produced adequate samples in most cases.[5]

The Swedish Foil Sampler could not be employed in 'soupy' sediments, in sandy sediments, and in sediments consisting mostly of pebbles and gravel. For 'soupy' sediments, SCUBA divers collected samples by driving thin-walled, 2 1/2-inch coring tubes into the sediment. In sandy sediments, sampling was done with a split-barrel sampler containing a basket shoe and plastic sleeve for sample retention. Hand-operated clam shell dredges were used to collect gravel samples.

Sediment cores were transferred in the field from the sample tube to plastic storage tubes, then frozen for ease of handling, to prevent slumping of soft sediment and to suppress biological and chemical action.

Selection of sampling sites.--Locations of sampling sections were based on the longitudinal pattern of radionuclide distribution in the upper portion of bottom sediment in the study reach. Sections were selected to sample reaches of high and low radioactivity, locations at which breaks in the general trend of increasing or decreasing radioactivity occurred, and reaches of transition from one radiation level to another.

Two sections established on the Emory River and on Poplar

Creek (figure 2) were for use in providing subsidiary information to health physicists at ORNL on possible movement of radioactive contaminants from the Clinch River into tributary streams.

Spacing of coring verticals within a sampling section was chosen by combined assessment of transverse distribution of sediment thickness and of radiation levels at the sediment surface. The relation of spacing of verticals to variations in thickness and radiation levels for one sampling section is shown in figure 3.

Results of coring.--Cores were collected at 135 verticals in the 14 sampling sections in the Clinch River and the sections in tributaries to the river (see figure 2). The number of verticals in a section ranged from 4 to 13.

Results of coring at each section are summarized in table 1. Two situations of special significance to the summary of coring results are emphasized by the table: (1) fifteen cores did not completely penetrate the radioactive zone; (2) not all cores or dredge samples contained significant amounts of radionuclides, as revealed by subsequent analyses (these samples were excluded from further direct consideration in computations).

Core Processing

Core processing was divided into four major procedures: (1) measurement of the distribution of gross gamma radiation in all cores and of individual radionuclides in selected cores (core counting), (2) longitudinal cutting, (3) preparation of samples for analysis, and (4) physical and radiochemical analysis of the samples. During the first two processing procedures, the cores (and dredge samples) remained frozen, as they had been since shortly after collection.

Core counting.--Variations of radioactivity with depth in the still-frozen cores were measured with a core scanner[4] (figure 4). For counting of gross gamma radioactivity, the phototube output of the core scanner was routed through a scaler and automatic timer-printout system. The number of seconds required for the accumulation of a pre-set number of counts (usually 2,048 counts) from the 2-inch increment of core opposite the collimator channel was recorded, the

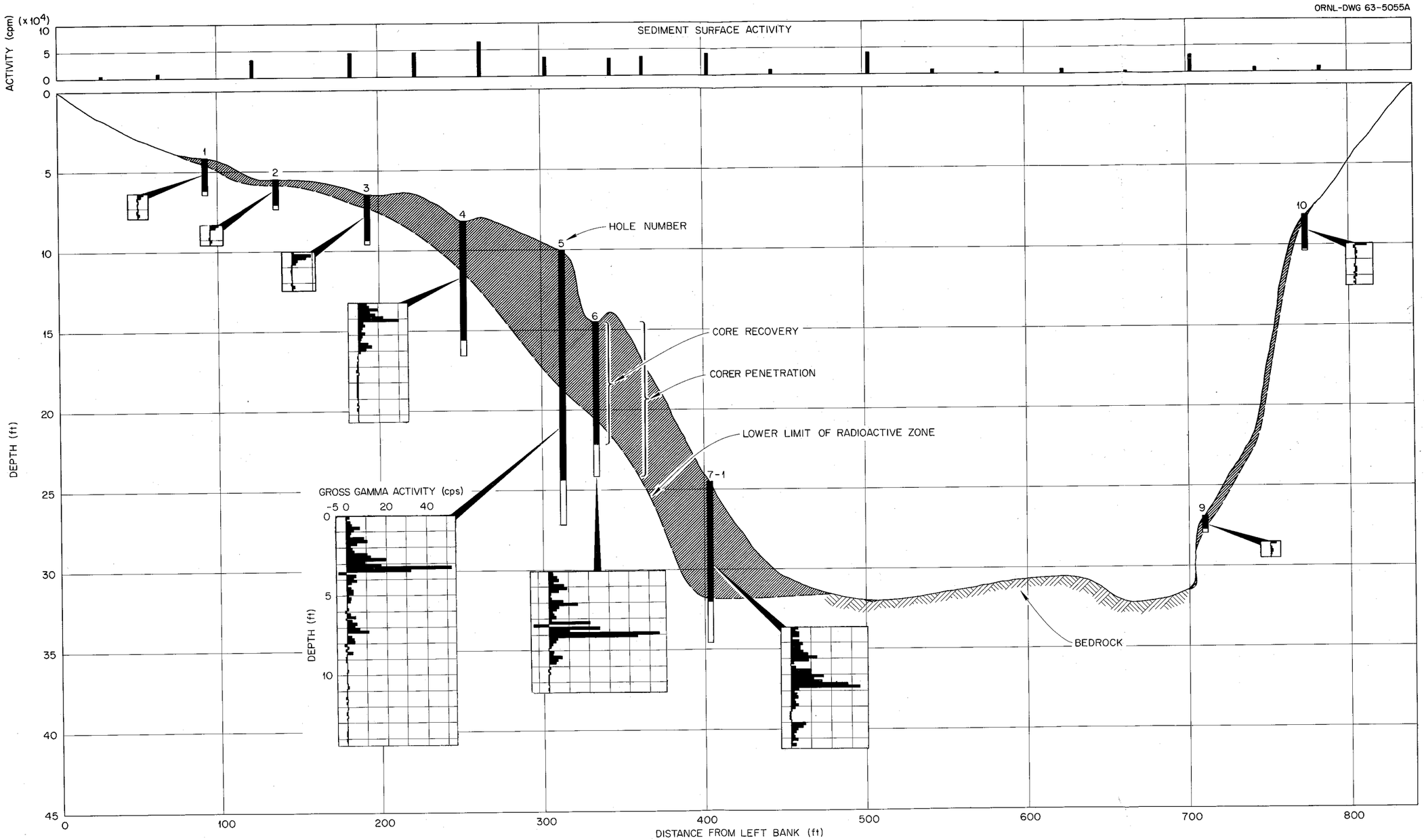

Fig. 3. Section at CRM 7.5 Showing Penetration, Recovery, and Gross Gamma Radioactivity Variation with Depth for 1962 Bottom Sediment Core Samples (Vertical Exaggeration 10:1)[4]

Table 1. Numbers of Sampling Sites, of Samples Used in Computations, and of Truncated Cores[a] Used in Computations for Each Sampling Section

Location of Section Miles Upstream from Mouth	Coring Verticals	Dredge Samples	Used in Computations		
			Cores	Dredge Samples	Truncated Cores
	Clinch River				
1.3	10	0	8	0	3
4.3	13	0	12	0	3
7.5	9	0	9	0	1
10.0	11	0	10	0	2
11.9	11	0	10	0	1
12.1	11	0	11	0	3
14.0	5	0	5	0	0
16.0	7	0	7	0	0
17.5	6	0	6	0	0
19.2	5	0	4	0	0
20.5	8	6	8	6	1
20.8[b]	5	7	3	7	1
21.0	4	0	2	0	0
22.8	5	1	5	1	0
	Emory River				
1.9	7	0	5	0	0
5.1	7	0	0	0	0
	Poplar Creek				
3.1	5	0	0	0	0
4.5	6	0	0	0	0

[a]Cores in which length of core recovered was less than thickness of radioactive zone.

[b]Section 100 feet downstream from mouth of White Oak Creek.

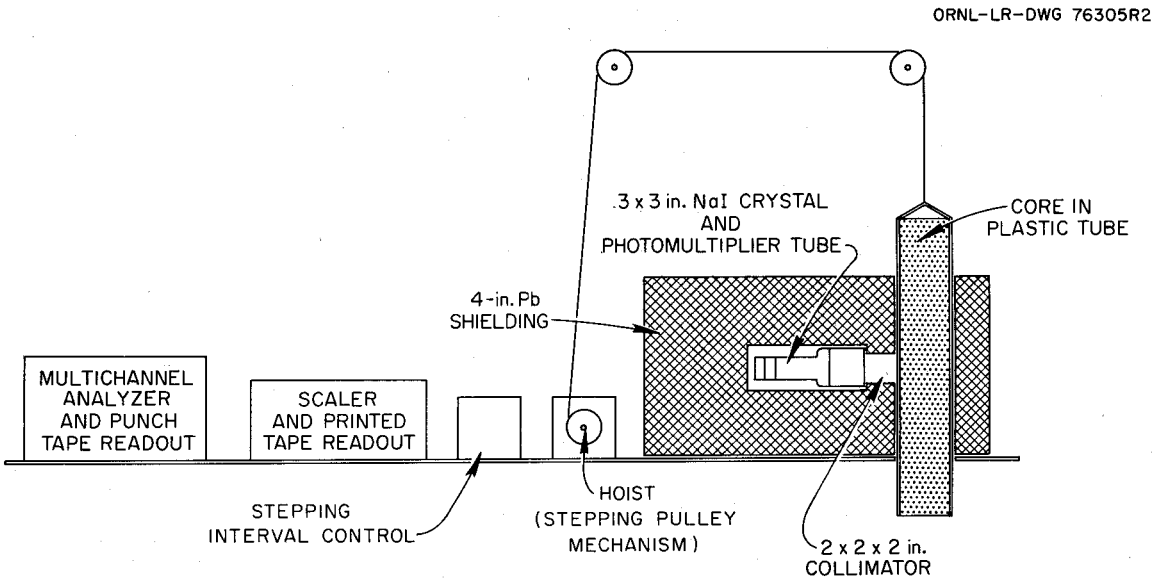

Figure 4. Diagrammatic Representation of Core Scanner[4]

core was automatically raised 2 inches by means of a calibrated hoist, and counting of the next increment of core was begun.

For gamma spectrum scanning, the phototube 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 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 increment of the cores scanned was analyzed through the use of a computer program developed at ORNL[6]. 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 another computer program. The eight radionuclides used in the gamma spectrum analysis included the nuclear-fission products ^{137}Cs, ^{106}Ru, ^{134}Cs, $^{152,154}Eu$, ^{144}Ce, ^{95}Zr-^{95}Nb, the neutron-activation product ^{60}Co, and the naturally-occurring radionuclide ^{40}K.

Each incremental gamma-ray radioactivity measurement was

corrected for the effect of 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.

Corrected counting data for each 2-inch-high slice of core were used to define the thickness of the radioactive zone at each coring vertical (see figure 3). These data were also used as a guide in determining cores in which there was no significant radioactivity.

Compositing of cores for inventory.--After core counting was completed, the cores were cut longitudinally into one half-section and two quarter-sections to provide samples for use in the inventory and for use in studies of the physicochemical characteristics of the sediments. Cutting was done by placing the frozen core in a jig and passing the jig under a carbide-tipped circular saw (figure 5).

After longitudinal sawing, one of the quarter-section cylinders of each core was cut horizontally at the base of the

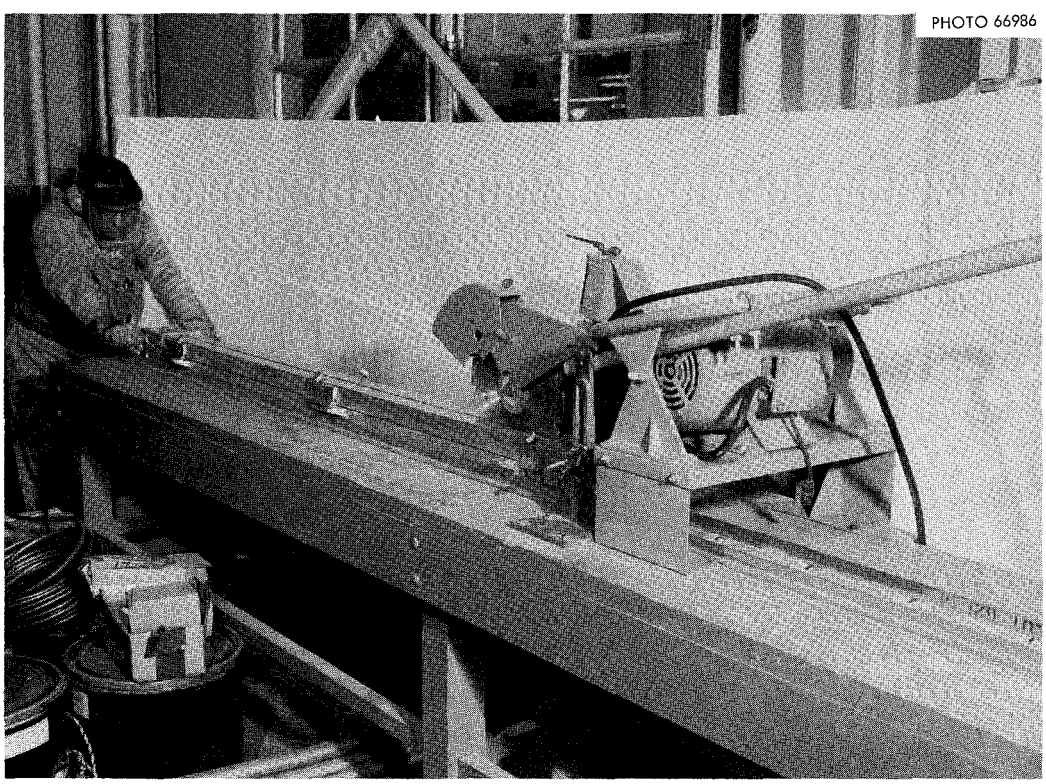

Fig. 5. Masonry Saw for Cutting Frozen Cores

radioactive zone and the non-radioactive part discarded. The radioactive part was retained for use in computing the inventory. Following cutting, the volume and weight of this cylinder of radioactive sediment were measured for determination of specific bulk weight. The volume of the cylinder (including void space) was found by permitting the frozen material to melt in a calibrated vessel. For these volumetric measurements, voids in the sediment were assumed to be completely filled with water.

A homogeneous composite sample of the radioactive quarter-sector was prepared for inventory purposes using an electrically driven egg beater. A 10-gram aliquot was split from the composite for analysis of particle size distribution. Then the remaining composite was weighed, dried at 100° C (Celsius), and reweighed, to provide data for computation of the dry-to-wet-weight ratio of the composite. A 100-gram aliquot ($\pm$ 0.01 gram) for gamma spectrum analysis, and a 20-50 gram aliquot for analyses of strontium-90 and tri-valent rare earth contents, were separated from the dry composite using a Jones soil splitter. Standard methods of chemical extraction, and beta counting of the extracts, were used in analyses[7] for ^{90}Sr and tri-valent rare earths.

Analysis of concentrations of ^{137}Cs, ^{60}Co, and ^{106}Ru in composites of the bottom sediment was made by techniques of gamma spectrometry using a 4-inch high by 4-inch diameter sodium iodide crystal for a detector and a 512-channel pulse-height analyzer. Determination of radionuclide concentrations from gamma spectra recorded on punch-tape was made by means of a computer program which utilized a technique of least-squares resolution of the gamma-ray spectra[6]. Input data for the computer program included the spectrum for the sample, the background associated with the sample, and standard spectra. Standards were prepared for ^{137}Cs, ^{60}Co, ^{106}Ru, ^{144}Ce, ^{95}Zr-^{95}Nb, ^{134}Cs, 152, ^{154}Eu, ^{40}K and uranium and thorium ores.

To assure that results of radionuclide analyses by gamma spectrometry were reasonably accurate, nine selected samples were reanalyzed at the Low-level Counting Facility, ORNL. These samples were

selected as representative of the general range of radioactivity, and of sampling location, for samples collected throughout the study reach. Analysts at this facility found that concentrations of only two radioactive contaminants, ^{137}Cs and ^{60}Co, could be reliably determined by their method of hand computation of gamma spectra data (produced from their pulse-height analyzer). Results for concentrations of ^{60}Co and ^{137}Cs determined by hand computation were slightly greater than those determined by the computer program; 7.8 and 13.8 percent, respectively. These differences are within the range, and in the direction, of error expected from such hand computation.

Physicochemical Analysis of Sediment

Sediment samples for physicochemical analysis were taken from two cores which had radioactive zones several feet thick. The samples consisted of alternate 2-inch slices of sediment from the two cores. Comprehensive particle-size analyses of aliquots of the 23 samples were made by wet seiving and bottom withdrawal tube[8]. The method of size analysis used involves dispersal of the sediment particles by chemical means, which can be expected to break up aggregates of clay minerals which may have 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 ultimate size distribution, which is useful in hydraulic studies of sediment movement and in mineralogical studies of sediment composition.

Particle-size separations of aliquots of the same samples into sand, silt, and clay were performed by wet seiving and centrifugation after the samples had been initially dried. Dispersal was by hand stirring or, for some samples, by ultrasonic agitation. 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 < 2 micron clay-size fraction following centrifugation. De-

terminations of cation exchange capacities and mineralogical composition were made of the three size fractions.

Cation exchange capacities were determined by means of an ammonium chloride leach followed by measurement of the sorbed ammonium 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 hydroxide[9] and colorimetric analysis by the "aluminon" method[10]. Free iron oxides were extracted in a buffered dithionate-citrate solution[11] and analyzed by the potassium thiocyanate method[10]. 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 dioxide evolved through combustion of the sample at 1350° C and the carbonate-derived carbon dioxide as determined by a low-temperature gas-evolution technique[12]. Analysis of the sample for minor and trace elements was performed by flame spectrophotometric methods, using recently-developed precision methods for the determination of strontium[13] and cesium[14]. The content of the radionuclides in each sample was determined by the same method as that used for analysis of composite samples.

Method of Computing Inventory

The volume of radioactive sediments in the Clinch River was divided into subvolumes in computing the inventory. The quantity of a radionuclide in a subvolume is equal to the product of its concentration (curies per unit weight) and weight of sediment in the subvolume. The total quantity of the radionuclide in the study reach is the sum of its quantities in all the subvolumes.

Locations of sampling sections and of coring verticals in the study reach served as controls for dividing the volume of sediment

into subvolumes. Each subvolume is associated with a specific coring vertical.

Each subvolume is assumed to be prismatic in shape; its length extends from midpoint to midpoint between adjacent sampling sections; its width extends from midpoint to midpoint between adjacent coring verticals in the sampling section, and its depth is equal to the thickness of the radioactive zone for the specified core. These definitions of dimensions of a subvolume are modified, first, for the two terminal subreaches and, second, for terminal verticals (where thickness of sediment is zero) in a sampling section.

The widths of radioactive zones in the sampling sections did not in every case extend from one edge of water to the other. Hence, the transverse locations of at least two, and sometimes more, terminal verticals of radioactive zone(s) in a section had to be defined. These terminal verticals were defined on the basis of (1) locations of cores that contained no detectable radioactivity, (2) rates of change of the gamma radiation levels at the surface of the bottom sediments, (3) rates of change of thickness of the radioactive zone, (4) transverse slope of the stream bed, and (5) limits of bedrock areas as determined by probing of the bed material. The influence of some of these factors on location of terminal verticals is indicated in figure 3.

The weight of sediment in a subvolume is the product of its volume and the specific bulk weight and dry-to-wet weight ratio of its associated core. Specific bulk weight is the wet weight per unit volume. Weight of the subvolume was converted to an oven-dry basis by the dry-to-wet weight ratio because oven-dried (100° C) composites were used in radiochemical analyses.

Approximately two years elapsed between collection and radiochemical analyses of cores. During this period decrease in radionuclide content through the process of radioactive decay was appreciable for some radionuclides. Concentrations used in the computations were adjusted to those at the time of sampling except for rare earths. No adjustment for radioactive decay of rare earths was made because they are a mixture of radionuclides having a variety of radioactive half-

lives. Methods of adjustment for effects of radioactive decay on radiation have been described by the U. S. Public Health Service[15].

Computation of the inventory was facilitated through use of a general purpose digital computer. Specific bulk weight, dry-to-wet weight ratio, and area were computed and checked manually prior to use as input data in computer programs.

Thicknesses of radioactive sediment for truncated cores and dredge samples were estimated as follows:

Distribution patterns of gross gamma radiation were discernable in many truncated cores. Patterns of radioactivity distribution in such cores were compared, using correlation analyses (see gross gamma core scans), to patterns for cores which penetrated the entire radioactive zone at nearby coring verticals. From results of these comparisons, the depth to which radioactivity extended could be estimated for truncated cores.

In some instances the truncated length was too short to clearly discern the distribution pattern of radioactivity, in which case the radioactive thickness at the coring vertical was assumed to be equal to the depth of penetration of the coring tool. Fortunately, the fraction of core recovered was large in most cases.

Depth of penetration of hand-operated dredges could not be measured. Depths were computed using information on volume of sample collected and on geometric characteristics of the sampler. Computed depth of sampling was assumed to be the radioactive depth.

INVENTORY

In the course of acquiring necessary data to inventory radioactivity in Clinch River bottom sediment, information has been gathered not only on quantities of radionuclides in the sediment, but also on the distribution of radionuclides in these sediments and the distribution of the radioactive sediments in the river.

Inventory of Identified Radionuclides

On the basis of radionuclide analyses of composite samples of bottom sediment cores from the Clinch River, the following quantities of radionuclides are associated with bottom sediments of the river between CRM 0.0 and CRM 21.0; 150 curies of ^{137}Cs, 18 curies of ^{60}Co, 16 curies of ^{106}Ru, at least 10 curies of rare earths, and 2.9 curies of ^{90}Sr.

All quantities have been adjusted for effects of radioactive decay to the quantities present at the time of sampling (July 1, 1962) except for the rare earths (see Method of Computing Inventory). The quantity of rare earths reported is that present at the time of radiochemical analysis (June 1964).

Total inventoried and identified radioactivity in the river is 200 curies, of which ^{137}Cs constitutes 77 percent; ^{60}Co, 8.7 percent; ^{106}Ru, 7.7 percent; rare earths, 5.1 percent; and ^{90}Sr, 1.5 percent.

About 95 percent of the identified radioactivity is in the reach of river downstream from CRM 15 (table 2). At least 50 percent of the identified radioactivity is downstream from CRM 8.7.

Estimate of accuracy of inventory.--The quantity of a radionuclide in a volume of sediment, R, may be expressed by the equation

$$R = C \times b\ t\ \gamma_s \frac{w_d}{w_w} ,$$

in which the independent variables are, respectively, concentration of a radionuclide, length of subreach, width of sampling section, mean thickness of radioactive sediment in sampling section, specific bulk weight of the sediment, and dry-to-wet weight ratio. The

Table 2. Total Identified Radioactivity and Volume of Radioactive Sediment in Subreaches

Location of Subreach (CRM)		Total Identified Radioactivity, in Curies		Volume in Acre-Feet	
Begin	End	In Subreach	Cumulative	In Subreach	Cumulative
0	2.80	22	22	340	340
2.80	5.90	42	64	380	720
5.90	8.95	54	118	480	1,200
8.95	10.95	46	164	430	1,630
10.95	12.00	9.3	173	93	1,720
12.00	13.05	10	183	85	1,810
13.05	15.00	6.8	191	38	1,850
15.00	16.75	2.2	193	33	1,880
16.75	18.35	4.7	198	39	1,920
18.35	19.85	.1	198	4.7	1,920
19.85	20.65	2.3	200	5.9	1,930
20.65	20.90	.1	200	1.4	1,930
20.90	21.00	.2	200	.3	1,930

following expression is used to compute the level of expected error in R:

$$\sigma_R = R \sum_{1}^{6} \frac{\sigma_z}{Z} ,$$

in which σ_R is the standard deviation for R, σ_z is the standard deviation for each independent variable Z enumerated above, and $\frac{\sigma_z}{Z}$ is the relative error or coefficient of variation, for Z.

The relative error in the measurement of physical properties which appear in the above equation has been estimated by comparing the precision in measuring each property to the magnitude of the property. Errors in measuring specific bulk weight and dry-to-wet weight ratio are less than 5 and 2 percent, respectively. Relative errors in dimensions of sediment volume have been estimated for each subreach downstream from CRM 15. Limitation in considered reach length has been made because 95 percent of the radioactivity is downstream from CRM 15. Coefficients of variation for subreach length, width, and thickness are about 3, 5, and 15 percent, respectively. Other independent sources of error in the determination of radionuclide concentration are accuracy of radionuclide standards, counting errors in analysis, and sampling errors. Error in radionuclide content of standards is about 5 percent. Average levels of counting errors for cesium-137, cobalt-60, ruthenium-106, strontium-90, and rare earths are 2, 8, 23, 4, and 2 percent, respectively.

The estimated errors in the inventory (neglecting sampling error and using the foregoing expression for σ_R) are as follows: 18 percent for ^{137}Cs, ^{90}Sr, and rare earths; 20 percent for ^{60}Co; and 29 percent for ^{106}Ru.

Retention Factors

The retention factor for a radionuclide is defined as the ratio, expressed in percent, of the quantity of the radionuclide in the sediment in July 1962 to that quantity released to the river during the period December 1943 to July 1962.

The magnitude of the retention factor is independent of time

if radioactive decay is taken into account.

$$k_r = \frac{I - O}{I}(100) = \frac{R}{I\,e^{-\lambda T}}(100) \quad ,$$

$$I = \int_0^T i\,dt, \text{ and } O = \int_0^T o\,dt \ ,$$

in which k_r = retention factor, in percent,

I = inflow of radionuclide to study reach during the time period T, in curies,

O = outflow of radionuclide from study reach during time period T, in curies,

R = quantity of radionuclide in bottom sediment of study reach at end of period T, in curies,

λ = radioactive decay constant for radionuclide, in sec^{-1},

T = duration of time period, in seconds,

i = rate of inflow of radionuclide to study reach, in curies per second, a function of time,

o = rate of outflow of radionuclide from study reach, in curies per second, a function of time, and

t = time, in seconds.

R may be expressed as a function of inflow and outflow if the effect of radioactive decay is considered.

$$R = \int_0^T ie^{-\lambda(T - t)}\,dt - \int_0^T oe^{-\lambda(T - t)}dt$$

$$= e^{-\lambda T}\int_0^T i\,dt - e^{-\lambda T}\int_0^T o\,dt$$

$$= e^{-\lambda T}(I - O)$$

Hence, $k_r = \frac{R}{I\,e^{-\lambda T}}(100) = \frac{e^{-\lambda T}(I - 0)}{e^{-\lambda T}\,I}(100) = \frac{I - 0}{I}(100)$

and is independent of time.

Only annual records of inflow are available so it is necessary to use numerical techniques to compute k_r:

$$k_r = \frac{R_m/e^{-\lambda T_2}}{\sum_{n=1}^{T} i_n e^{-\lambda(T-n)}}(100), \quad n = 1, 2, \ldots\ldots, T \text{ years} \qquad (1)$$

in which $R_m = R\,e^{-\lambda T_2}$ = inventory of radionuclide at time of radiochemical analysis, in curies,

λ = radioactive decay constant, in years^{-1},

T_2 = time between collection of cores and radiochemical analysis, in years,

i_n = annual inflow of radionuclide to study reach during n^{th} year, in curies per year.

Using equation 1 the retention factors are computed to be 21 percent for ^{137}Cs, 9 percent for ^{60}Co, 0.4 percent for ^{106}Ru, and 0.2 percent for ^{90}Sr. Retention of the rare earths may approach, and possibly exceeds, 25 percent (see Ref. 16). A good estimate of the retention factor for rare earths has not been obtained because mixtures of radioelements of undetermined composition are involved. The above estimate was derived from a conservative consideration of the ^{144}Ce and other rare earths contents of three samples.

Contributions to Inventory From Reaches Outside of Study Reach

Contributions to the radionuclide content of bottom sediments in the study reach from fallout from weapons tests appear to be negligible. Concentrations of radionuclides in bottom sediment at sampling section 2.0 miles upstream from the study reach, CRM 22.8, are below limits of detection. Concentrations of ^{137}Cs and ^{60}Co in waters

flowing past a water-sampling station at CRM 41.5, upstream from the study reach, have been at or below limits of detection since the study was begun. No rare earths have been detected in water samples collected at this station. Small concentrations of ^{106}Ru and ^{90}Sr have been detected regularly in water samples,[17] but these radionuclides constitute a minor fraction of the total inventory, and their retention factors are low.

Cores were collected in Poplar Creek and Emory River to gage the extent of upstream movement of contaminated waters from the Clinch River. Hydraulic conditions are such in the study reach of the Clinch River that upstream movement of water from the river can occur in any tributary.

Examination of gross gamma radioactivity scans of cores from Poplar Creek and from the section at mile 5.1 on the Emory River indicate that the radionuclide content in bottom sediments at these sampling sites (Fig. 2) is negligible. However, concentrations of radioactivity in some cores from mile 1.9 on the Emory River were above lower limits of detection. Furthermore, Pickering, in a study of radioactivity in bottom sediments of sloughs adjacent to the Clinch River[4], noted that radiation levels in some areas approached levels measured at the surface of Clinch River bottom sediment. This evidence of radioactivity in bottom sediments in the lowermost reaches of tributaries to the Clinch River suggests that supplemental inventories in these areas might be advisable.

DISTRIBUTION OF RADIOACTIVE SEDIMENT

Variation of Radioactivity With Depth in Cores

Gross gamma core scans.--Scanning the bottom sediment cores for variations in gross gamma radioactivity[1] with depth revealed the general pattern of variation in each core and provided a basis for determining the thickness of radioactive sediment at each sample site. The results of the core scanning indicated that the entire thickness of radioactive sediment had been sampled in most cores. The core showing the greatest thickness of radioactive sediment, 8.7 feet, was obtained at CRM 7.5.

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 portion 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). Portions of the cross section showing a water depth of less than 6 feet are not submerged during low winter lake levels. The resulting non-deposition 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 portion of the study reach downstream from CRM 18. The general pattern which they exhibit is strikingly similar to the pattern of annual releases of ^{137}Cs to

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[1] 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.

the Clinch River as measured at White Oak Dam (Table 3). This similarity is illustrated in Figure 6 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 ^{137}Cs were highest -- i.e., 1956[2].

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 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, using a different adjustment factor in each correlation attempt. The output of the program included the coefficients of correlation and of regression for the best correlation attempted with each pair of cores.

Coefficients of correlation resulting from a comparison of the four cores shown in Figure 6 to annual releases of ^{137}Cs to the Clinch River are included in Table 4.

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 to the pattern of annual releases of ^{137}Cs to the Clinch River, corrected for decay to the date of sampling. Coefficients of correlation ranged from 0.60 to 0.90 and had a mean of 0.76. When annual releases of ^{60}Co were added to those of ^{137}Cs, and correction made for the relative gamma ray emission of the two nuclides, the comparison showed improved correlation with the cores, giving a mean coefficient of correlation of 0.80 and a range of 0.64 to 0.98. When the annual releases of ^{106}Ru were added to those of ^{137}Cs and ^{60}Co, after making corrections for relative gamma ray emission, the correlation was not as good, giving a mean coefficient or correlation of 0.68 and a range of 0.44 to 0.97.

The correlation results suggest that the pattern of variation

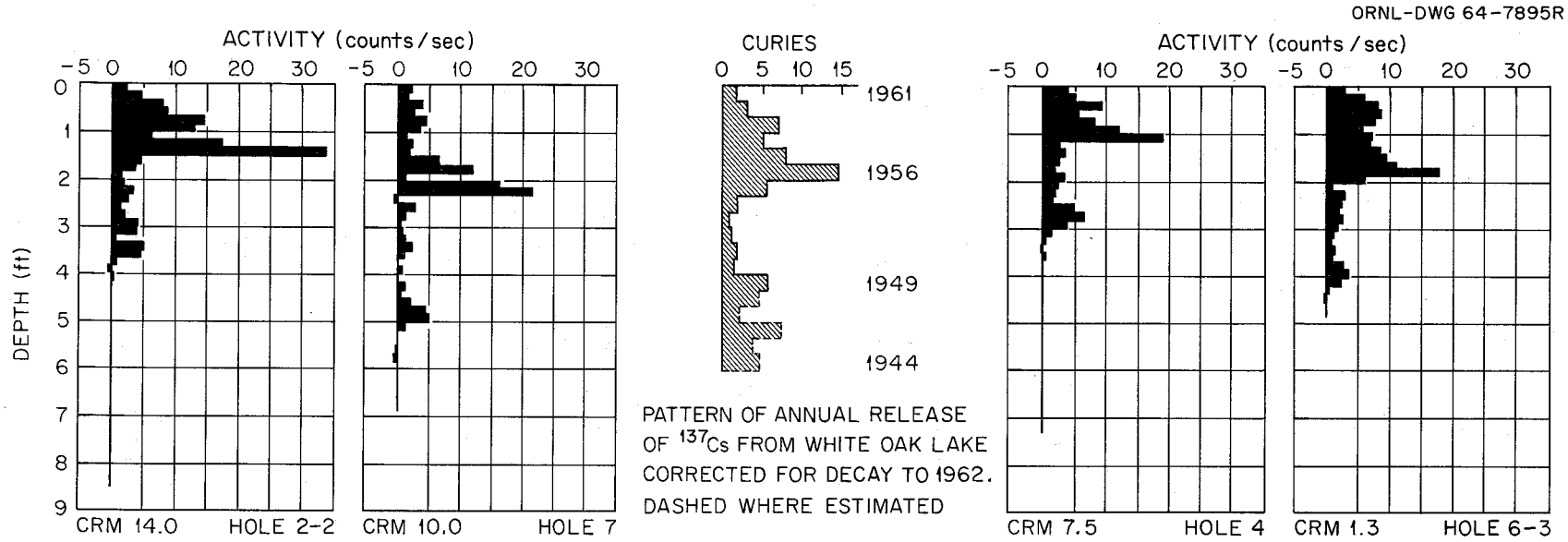

Fig. 6. Comparison of Patterns of Variation with Depth of Gross Gamma Radioactivity in Four Bottom Sediment Cores to Variations in Annual Releases of ^{137}Cs to Clinch River

Table 3. Yearly Discharges of Radionuclides to the Clinch River (curies)[1]

Year	Gross Beta	^{137}Cs	^{106}Ru	^{90}Sr	TRE(-Ce)	^{144}Ce	^{95}Zr	^{95}Nb	^{131}I	^{60}Co
1949	718	77	110	150	77	18	180	22	77	
1950	191	19	23	38	30		15	42	19	
1951	101	20	18	29	11		4.5	2.2	18	
1952	214	9.9	15	72	26	23	19	18	20	
1953	304	6.4	26	130	110	6.7	7.6	3.6	2.1	
1954	384	22	11	140	160	24	14	9.2	3.5	
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	0.5	77
1960	2190	31	1900	28	48	27	38	45	5.3	72
1961	2230	15	2000	22	24	4.2	20	70	3.7	31
1962	1440	5.6	1400	9.4	11	1.2	2.2	7.7	0.36	14
1963	470	3.5	430	7.8	9.4	1.5	0.34	0.71	0.44	14

[1]Calendar year basis, after Cowser and Snyder[30]

Table 4. Coefficients of Correlation for Comparison of Vertical Distribution of Gross Gamma Radioactivity in Sediment Cores with Annual Releases of Radionuclides[a]

Sampling Section (CRM)	Core Number	^{137}Cs[a]	$^{137}Cs + ^{60}Co$[a]	$^{137}Cs + ^{60}Co + ^{106}Ru$
1.3	3	0.64	0.67	0.68
1.3	5	0.78	0.84	0.72
1.3	6-2	0.63	0.72	0.64
1.3	6-3	0.73	0.78	0.78
1.3	7	0.60	0.64	0.65
1.3	7-2	0.88	0.93	0.80
4.3	2-2	0.67	0.83	0.69
4.3	11-2	0.73	0.79	0.63
7.5	4	0.81	0.82	0.62
7.5	5	0.80	0.72	0.51
7.5	6-1	0.81	0.75	0.49
7.5	7-1	0.75	0.79	0.57
7.5	7-2	0.72	0.64	0.50
10.0	2	0.83	0.82	0.61
10.0	3-3	0.77	0.90	0.81
10.0	7	0.83	0.79	0.44
11.9	9	0.76	0.76	----[b]
11.9	10	0.60	0.79	0.71
12.1	2	0.88	0.89	0.80
12.1	6-1	0.66	0.82	0.80
12.1	7	0.79	0.78	0.77
12.1	9	0.89	0.98	0.87
14.0	2-1	0.82	0.85	0.60
14.0	2-2	0.80	0.84	0.62
14.0	3	0.76	0.87	0.97
17.5	3-1	0.90	0.79	0.83
17.5	3-2	0.76	0.66	0.63
Mean		0.76	0.80	0.68
Standard Deviation		0.084	0.083	0.126

[a]Corrected for decay to date of sampling. Where releases are combined, correction has been made for the relative gamma emission of the radionuclides.

[b]Dashes indicate correlation not obtained.

in gross gamma radioactivity exhibited by the cores tested reflects not only the pattern of annual releases of ^{137}Cs to the Clinch River, but to a minor extent, that of annual releases of ^{60}Co as well.

Gamma spectrum core scans.--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 6.

Four gamma-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 ^{137}Cs, ^{60}Co, ^{106}Ru, and ^{40}K. Because of its short half-life (1 year), ^{106}Ru was detected in only the upper portions of most of the cores. The presence of the naturally-occurring radionuclide ^{40}K was not due to releases from ORNL.

Variations in concentrations with depth of the three most abundant gamma-emitting radionuclides in a core from Hole 6, CRM 7.5 are illustrated in Figure 7 (compare with Fig. 3): 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 7 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 ^{137}Cs. The contribution of radioactivity from ^{106}Ru is confined to the upper portion of the core. (The isolated positive values for ^{106}Ru below a depth of 20 inches on the plot are questionable). Similar results obtained for the eight other cores that were gamma spectrum scanned are shown in Table 5. These results compare favorably with results of the inventory of radionuclides in bottom sediments in the Clinch River (see Inventory).

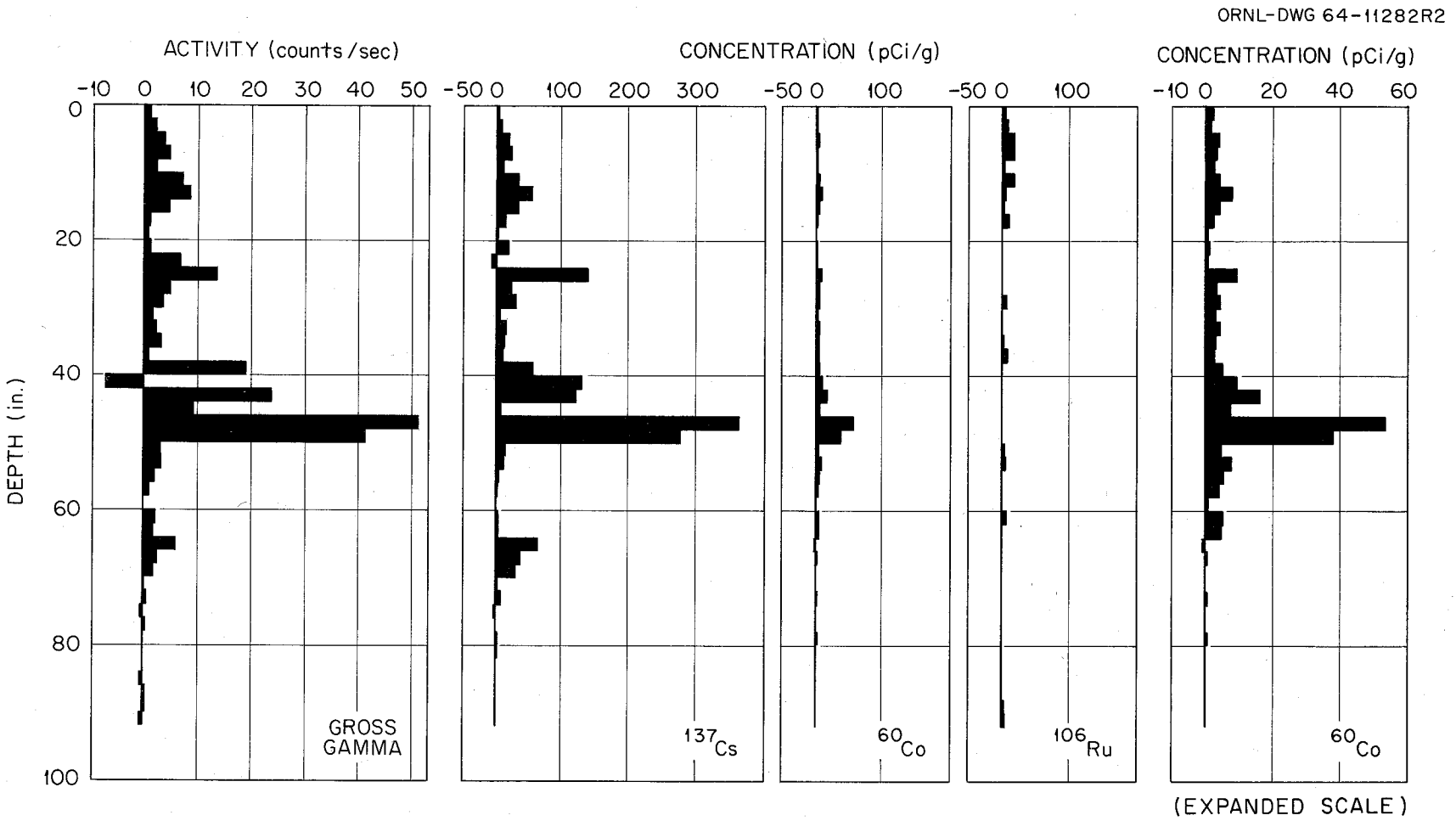

Fig. 7. Variations with Depth in Concentrations of ^{137}Cs, ^{60}Co, and ^{106}Ru in Hole 6, CRM 7.5

Table 5. Fractional Contributions of ^{137}Cs, ^{60}Co, and ^{106}Ru to Total Gamma-Ray Radioactivity of Selected Clinch River Sediment Cores

CRM	Hole Number	^{137}Cs Fraction	^{60}Co Fraction	^{106}Ru Fraction
1.3	3	0.82	0.11	0.07
1.3	6-3	0.77	0.13	0.10
7.5	5	0.81	0.13	0.16
7.5	6	0.82	0.12	0.06
7.5	7-2	0.77	0.13	0.10
10.0	2	0.79	0.10	0.11
14.0	2	0.80	0.13	0.07
14.0	2-2	0.84	0.11	0.05
17.5	3-1	0.86	0.10	0.04
Mean Value		0.81	0.12	0.07

The similarity between the distributions of ^{137}Cs and ^{60}Co with depth in the core can be observed with the aid of the fifth plot in Figure 7, which shows variations in ^{60}Co concentration with depth at an expanded scale. The same relationship was exhibited by the eight other cores gross gamma scanned. Statistical treatment of the relationship between ^{137}Cs and ^{60}Co in all nine cores further emphasizes the similarity in distribution of the two radionuclides (see Table 6).

Relationship of Vertical Distribution to Annual Releases

Results of radiochemical analyses of water samples collected weekly at White Oak Dam for a period of 2 years indicate that on the average nearly 70 percent of the ^{137}Cs present in the water is associated with suspended solids having a diameter greater than 0.7 micron.[17] Cesium is known to be preferentially sorbed by layered aluminosilicate minerals in which the c-axis spacing is 10 angstroms.[18, 19] Such minerals are abundant in White Oak Creek basin.[20, 21] Furthermore,

Table 6. Relationship of ^{60}Co to ^{137}Cs in Selected Bottom Sediment Cores From the Clinch River

CRM	Hole Number	Correlation Coefficient	Regression Coefficient	Ratio of Mean Concentrations (Cs/Co)
1.3	3	0.62	0.09	7.39
1.3	6-3	0.89	0.12	5.83
7.5	5	0.90	0.13	5.77
7.5	6	0.93	0.13	7.34
7.5	7-2	0.91	0.13	5.86
10.0	2	0.87	0.11	7.83
14.0	2	0.90	0.13	6.37
14.0	2-2	0.87	0.13	7.25
17.5	3-1	0.99	0.09	9.11
Mean Value		0.88	0.12	6.97

this sorption reaction is time dependent and only slowly reversible. It is safe to assume, then, that incorporation of ^{137}Cs in Clinch River bottom sediment is primarily a result of sedimentation of suspended radioactive aluminosilicate minerals which enter the river in water from White Oak Creek.

The year of highest release of ^{137}Cs to the Clinch River was 1956. This high release resulted from the draining of White Oak Lake during the latter half of 1955[2], and exposure to erosion 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 higher content of ^{137}Cs in bottom sediment deposited in the river during 1956. Through the operation of processes such as this, variations in annual releases of ^{137}Cs have been recorded as variations in the vertical distribution of ^{137}Cs in the bottom sediment in portions of the river where more or less regular and persistent deposition of sediment has taken place.

The pattern of variation in the content of ^{60}Co in the sediment does not strongly resemble the pattern of annual releases of ^{60}Co to the Clinch River (Table 3). Some effect of annual releases is implied, however, by the improved correlation which resulted when annual ^{60}Co releases were taken into account in the comparison of gross gamma core scan patterns described previously. The most striking feature of the pattern of ^{60}Co variation in the cores gamma spectrum scanned is its great similarity to the pattern of ^{137}Cs variations (see Fig. 7 and Table 6). This similarity suggests that ^{60}Co, like ^{137}Cs, was incorporated in Clinch River bottom sediment primarily by sedimentation of suspended solids which had obtained their content of ^{60}Co before entering the Clinch River. It also suggests that a rather small fraction of the total ^{60}Co released to the river became incorporated in Clinch River bottom sediment; otherwise, distribution of ^{60}Co in the cores would resemble the pattern of annual releases of ^{60}Co to the river. This second suggestion is supported by the results of water sampling which show that only 20 percent of the ^{60}Co released from White Oak Creek is associated with suspended sediment with a diameter greater than 0.7 micron.[17] It is confirmed by the results of the inventory of radionuclides in the bottom sediment (see Retention Factors). Although the results of water sampling do not preclude association of the remaining 80 percent of the ^{60}Co 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 White Oak Creek water also in a second form, such as a dissolved ion. The presence of two chemical forms of ^{60}Co in White Oak Creek water does not conflict with the results of gamma spectrum core scanning if it is assumed that the ratio of dissolved ^{60}Co to solids-associated ^{60}Co in the water did not remain constant during the period in which the sampled sediment was deposited.

The suggestion that some of the ^{60}Co, like most of the ^{137}Cs, is associated with suspended solids when it enters the Clinch River from White Oak Creek does not necessarily imply that the two radionuclides are associated with the same type of solid. It is assumed that ^{137}Cs is primarily associated with layered aluminosilicate

minerals. Leaching experiments[4] have indicated that ^{60}Co may be present in the bottom sediment as a hydrated oxide. The studies further indicate that ^{60}Co, like ^{137}Cs and ^{106}Ru, is not readily removed by leaching after it becomes associated with the bottom sediment. Other studies have indicated that ^{60}Co in the sediment is associated with iron and manganese oxides.[22]

The contribution of ^{106}Ru to gamma radioactivity in the upper portion 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 ^{137}Cs. It can be assumed, then, that a relatively small fraction of the total amount of ^{106}Ru that has been released to the Clinch River has become incorporated in the bottom sediment. This assumption has been confirmed by the results of the inventory of radioactivity in Clinch River bottom sediment (see Retention Factors).

Too little is known about the chemical form of ^{106}Ru in the Clinch River to permit definite inferences concerning the manner in which it is incorporated in Clinch River bottom sediments. The ^{106}Ru content of annual samples of the surface layer of Clinch River bottom sediment has been shown to reflect the amount of ^{106}Ru released to the river during the preceding year.[4] Analyses of water samples from White Oak Lake indicate that less than 10 percent of the ^{106}Ru released to the Clinch River is associated with suspended sediment greater than 0.7 micron in diameter.[17] Even with the benefit of this information, it can be stated only that the mechanism by which ^{106}Ru is incorporated in the sediment may be one or more of the following: (1) sedimentation of radioactive solids suspended in White Oak Creek water, (2) precipitation of a Ru-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[4, 23] that some of the ^{106}Ru in bottom sediments 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 at each of those coring sites. Furthemore, the resemblance of the pattern, which is due primarily to the ^{137}Cs content of the sediment, to variations in the annual release of ^{137}Cs from White Oak Creek to the Clinch River, implies more or less 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 the Tennessee Valley Authority'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 radionuclides to the Clinch River was started by the Oak Ridge National Laboratory.[24]

It should be emphasized that <u>regular</u> and <u>persistent</u> sediment deposition can occur without <u>continual</u> sediment deposition. Because the radioactive portion 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. Sedimentation would be expected to be irregularly distributed throughout the year.

Longitudinal Distribution of Radioactive Sediment

Ninety-five percent of the radioactive bottom sediment in

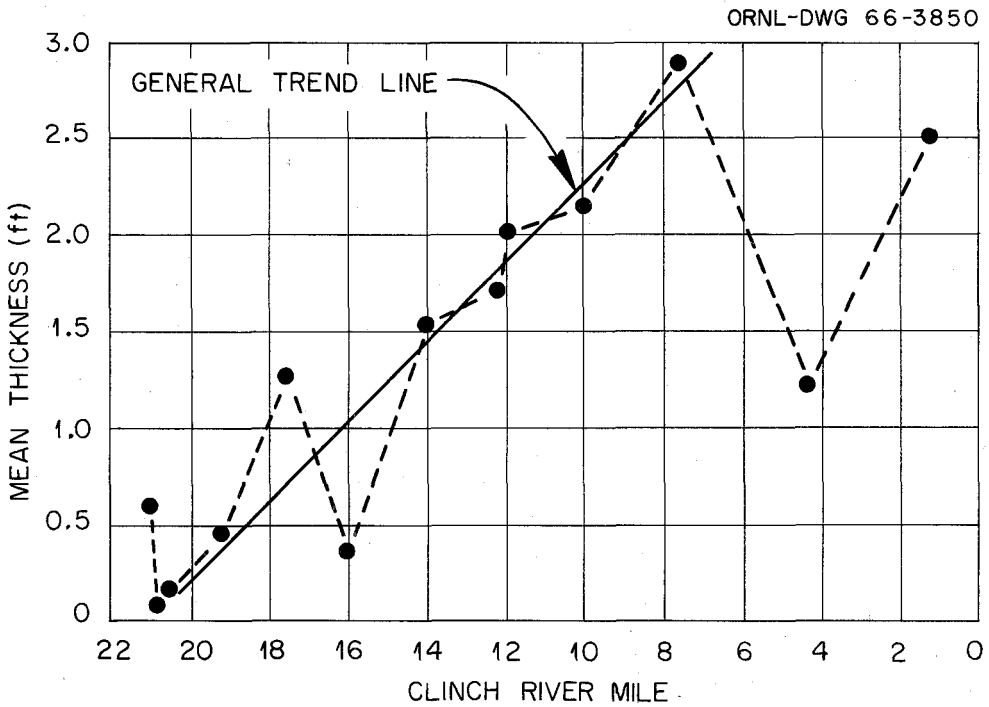

Fig. 8. Variation of Radioactive Sediment Thickness in the Downstream Direction

the Clinch River lies downstream from CRM 15 and at least 50 percent lies downstream from CRM 8.7 (Table 2).

A tendency for mean thickness of radioactive sediment to increase linearly from the head of the study reach to the mouth of the Emory River (CRM 21.0 to 4.5) appears to exist (Fig. 8). The general trend for increasing thickness may be explained by considering hydraulic conditions in the study reach. Accretions to flow in the study reach are almost negligible except inflow from the Emory River.[25] Flow area continuously increases in the downstream direction.[26] Without accretions to flow and with increasing flow area, velocity and intensity of turbulence of the stream decreases, and consequently sediment transport capacity decreases in the downstream direction.

The mean thickness of radioactive sediment is greater at the sampling section upstream from the mouth of Emory River, at CRM 7.5, than it is at the section downstream from the mouth of the river, at CRM 4.3. This difference in sediment thickness may be the result of a localized increase in turbulence downstream from the Emory River due to inflow

of the river into the Clinch River and/or to the diversion of Clinch River water upstream into the Emory River.[27] An increase in turbulence would tend to keep sediment particles in suspension.

At the mouth of Poplar Creek (CRM 12.0), the mean thickness of radioactive sediment sharply increases in relation to upstream thicknesses. Probably the increased thickness, as well as the decrease in radionuclide concentrations observed in this part of the stream bed (see Fig. 10), are due to dilution of radioactive sediment by uncontaminated sediment issuing from the creek.

The section near which sediment transport capacity rapidly decreases is clearly indicated to be CRM 14.0 (Fig. 9). From CRM 21.0

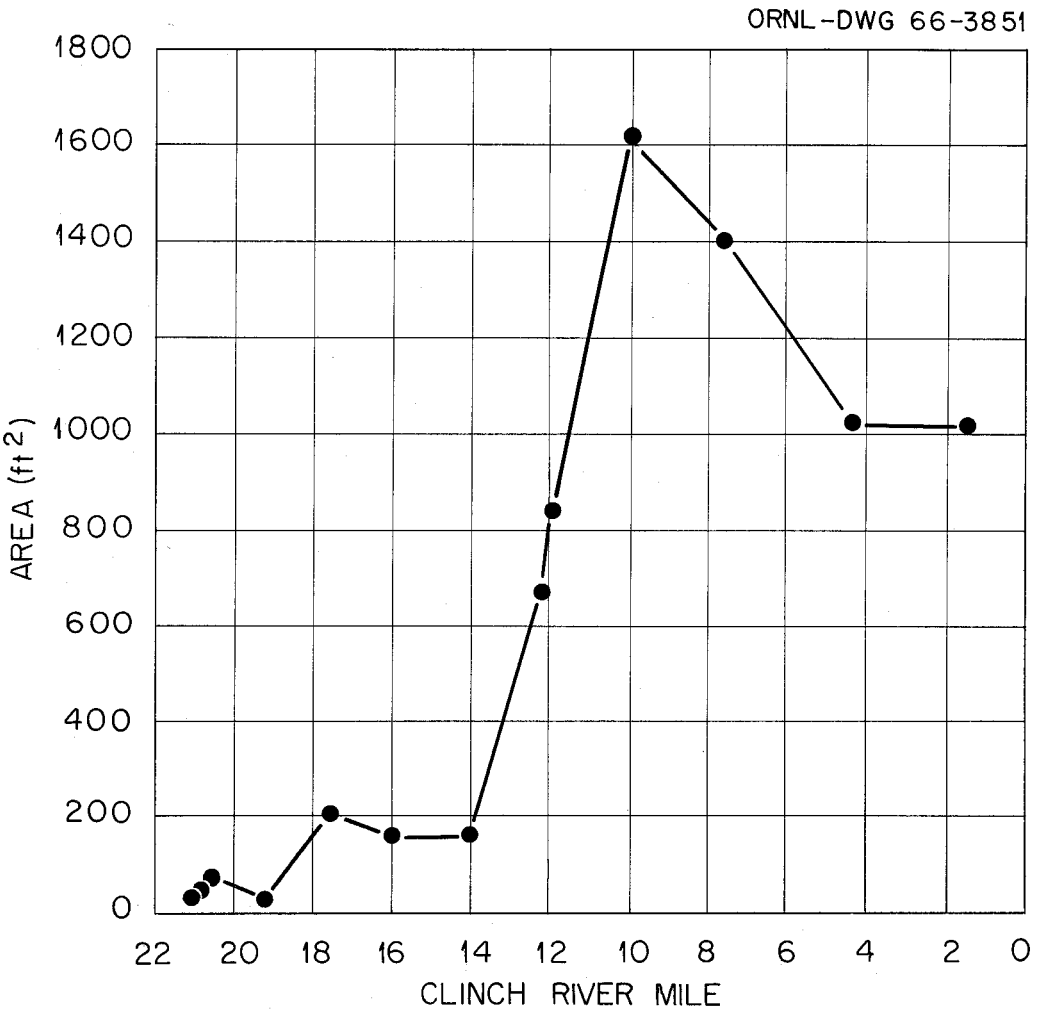

Fig. 9. Variation of Cross-Sectional Area of Radioactive Sediment in the Downstream Direction

to 14.0 deposition is limited to zones of the channel near bank(s) of the river. The sediment transport capacity is greater in the central core of flow in this reach than near the banks. Though transport capacity remains greatest in the central core of flow throughout the study reach, its magnitude relative to that near the banks drops sharply in the vicinity of CRM 14.0.

The cross-sectional area of radioactive sediment is at a maximum at CRM 10.0 (Fig. 9) because of the influence of islands and submerged ridges on the flow pattern at that cross section.

During the 19 1/2-year period of waste disposal by ORNL (December 1943 to July 1962), 64 percent of the stream bed has become covered with radioactive sediment. Areal distribution of radioactive sediments, for selected thicknesses, is listed in Table 7.

Table 7. Areal Distribution of Radioactive Sediments for Selected Thicknesses

Radioactive Sediment Thickness, in Feet	Proportion of Radioactive Surface Area Over Which Indicated Thickness is Equalled or Exceeded
1.0	0.57
1.5	.41
2.0	.31
2.6	.24
4.0	.13

Longitudinal Distribution of Radionuclides

Patterns of longitudinal variation in mean concentration are similar, in most respects, for the principal radionuclides in the sediment (Fig. 10). Highest mean concentrations of the radionuclides are in the two sections immediately downstream from the mouth of White Oak Creek (CRM 20.8 and CRM 20.5). Lowest concentrations generally are in sediments of the next downstream sampling section, at CRM 19.2. Secondary peaks in the distribution (Fig. 10) occurred at CRM 17.5 and 14.0, and for ^{106}Ru and rare earths, at CRM 4.3. Mean sectional concentrations for all but rare earths abruptly decreased between CRM 12.1 and CRM 11.9, immediately upstream and downstream from the mouth of Poplar Creek.

Distribution of several of the principal radionuclides in Clinch River bottom sediment appears to be controlled primarily by sedimentation of suspended radioactive sediment which has passed into the river through White Oak Dam.[28] Patterns of radioactive deposits in the river result from the combined interaction of lateral diffusion, characteristics of the flow pattern, and decreasing downstream intensity of turbulent forces acting to suspend the radioactive sediment particles. Therefore, similarities in concentration distribution are to be expected.

Investigation of the movement of sediment has not been a part of the Clinch River Study. As a consequence, careful documentation of the effects of diffusion, flow distribution, and turbulence on sedimentation is not available. However, the influence of these three characteristics of water movement on the longitudinal distribution of radioactive sediment may be surmised from observation and application of principles of mechanics of fluid motion.

High concentrations of radioactivity in bottom sediment may be expected near the mouth of White Oak Creek. Dispersion of waters and suspended sediments from the creek into the river is restricted because of incomplete lateral diffusion.[3, 28, 29] Consequently, suspended sediments from White Oak Creek, which are rich in sorbed radioactivity, do not immediately become diluted in the rela-

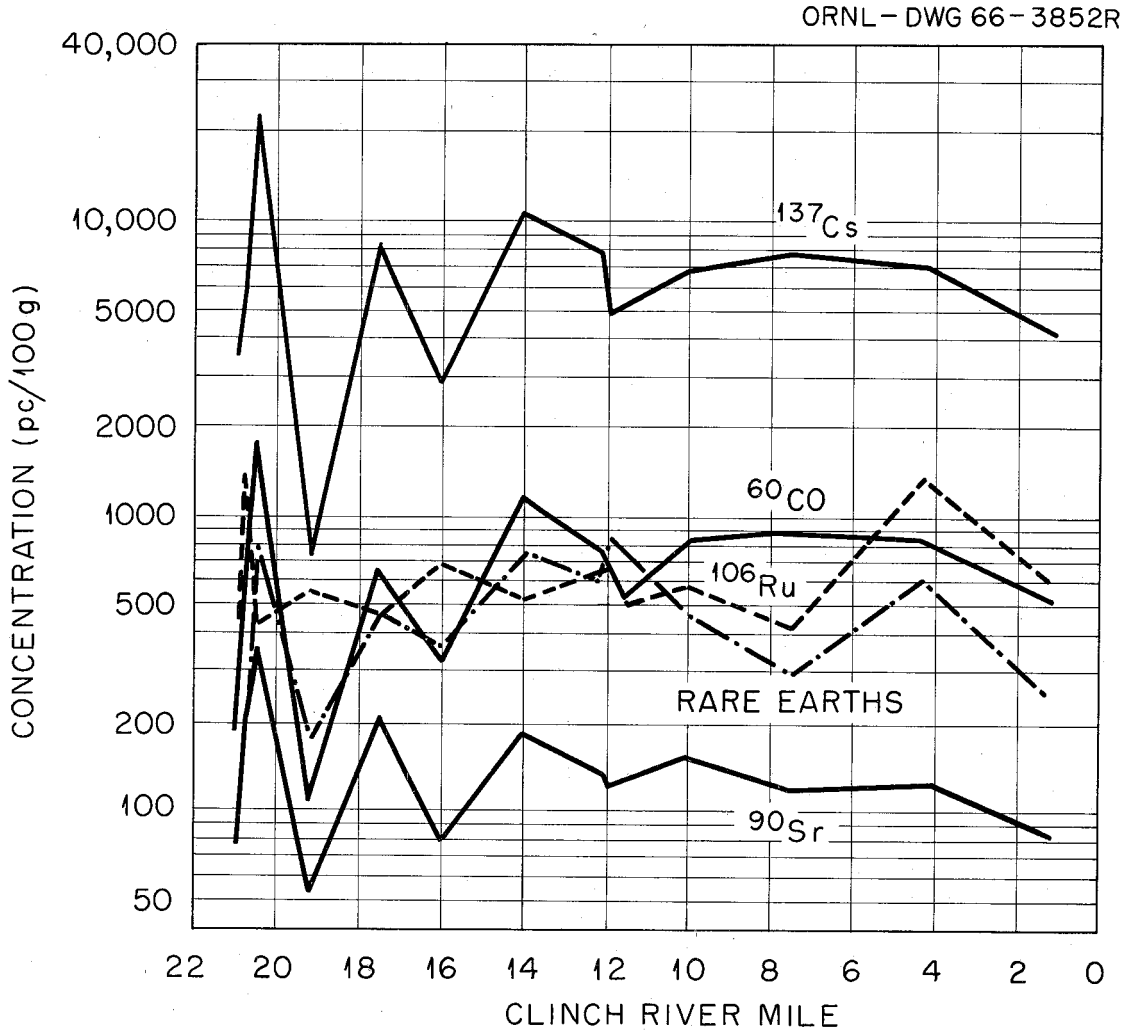

Fig. 10. Longitudinal Distribution of Radionuclides in Bottom Sediment of the Study Reach.

tively uncontaminated river.

Abrupt changes in the momentum of water entering the river from the creek tend to induce deposition. Eddy zones along the right bank of the river immediately downstream from the creek mouth are created as the two streams meet. The eddies and the restricted dispersion of suspended sediment produce immediate deposition of sediment rich in sorbed radioactivity.

As White Oak Creek water moves downstream, suspended sedi-

ment from the creek diffuses outward into the river channel, and the radioactive sediment mixes with sediment carried in the river, thus preventing formation of additional pockets of deposited sediment containing high concentrations of radioactivity.

Slightly farther downstream, turbulence is of sufficient intensity to keep the sediment in suspension through reaches such as those near CRM 19.2.

The abrupt downstream decrease in concentration of radionuclides in bottom sediment at the mouth of Poplar Creek may be the result of dilution of contaminated river sediment by uncontaminated sediment from Poplar Creek.

Relationship of Radionuclide Concentration to Thickness

Radionuclide concentrations are related to sediment thickness and location in the study reach. This relationship is shown in Figure 11. The abscissa in the figure is the ratio of the thickness at a coring vertical to mean thickness in the sampling section (shown in Fig. 8); the ordinate is the ratio of the concentration at a coring vertical to mean concentration in the sampling section (shown in Fig. 10).

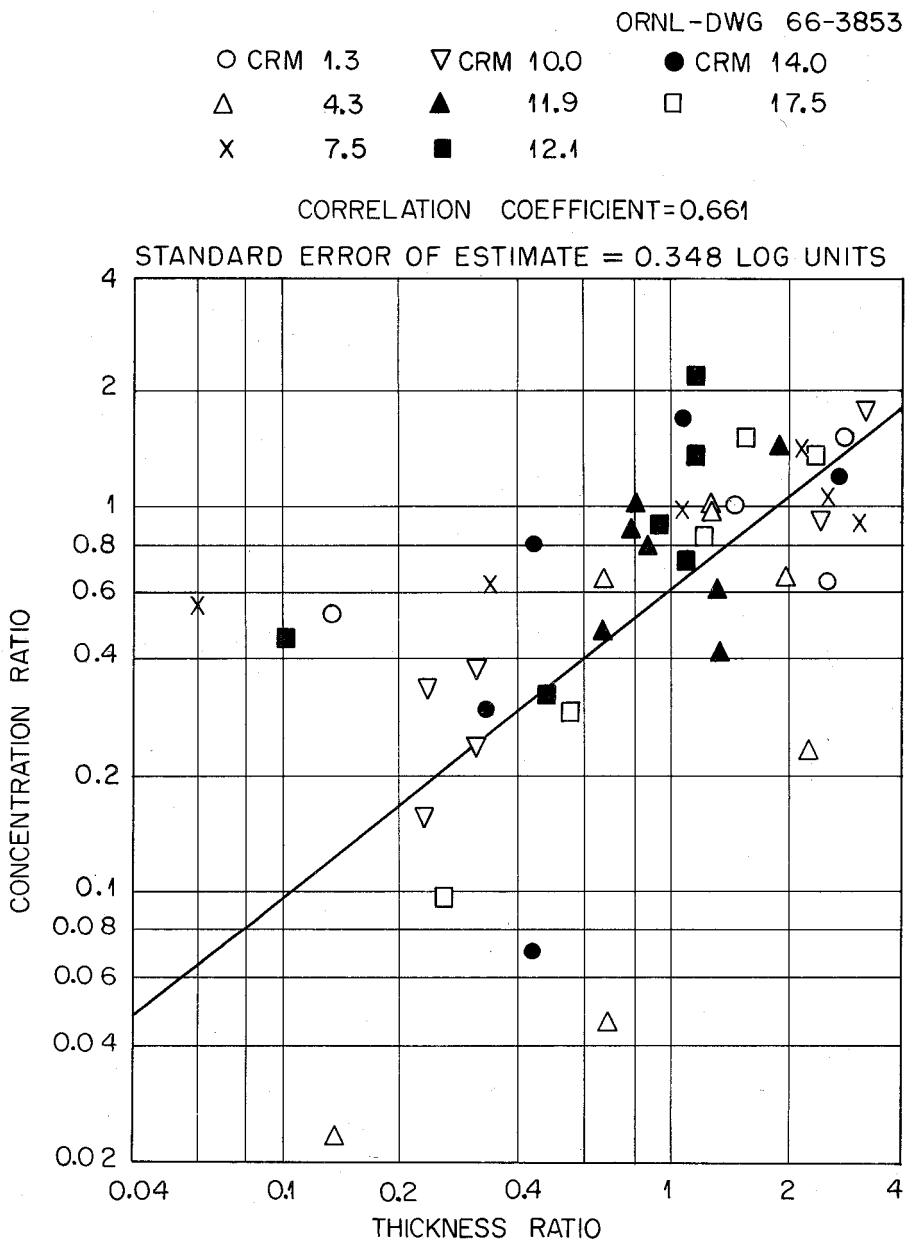

Fig. 11. Relationship of Radionuclide Concentration to Sediment Thickness

FUTURE ESTIMATION OF RADIONUCLIDE LOADING

Continued decline of radionuclide releases to the Clinch River in recent years (see Table 3) has indicated that increasing radiation hazard in the future is unlikely. Cowser and Snyder[30] find that even if releases had continued indefinitely at their highest levels (in 1956 or 1959), bottom sediment would be a minor source of radiation exposure. With continuing decline of releases, further estimation of radionuclide loading in the sediment is not needed.

Possibly waste disposal processes or quantities of radionuclides processed at ORNL will change such that increased releases to the river will result. If releases of ^{137}Cs, ^{60}Co, and/or rare earths should increase substantially above experienced levels for sustained periods, another safety analysis of radiation dosage from bottom sediment would be needed.

Safety analysis may be undertaken at two times: (1) after a period of increased loading, to determine resultant radiation dosage and (2) prior to increased loading to estimate the concentration and duration of releases which will assure safety.

For case (1), knowledge of retention factors would be an aid in estimating the magnitude of radiation dosage resulting from increased releases. The relationship of radiation dose to radioactivity of releases is

$$D = 1/2(\ 51.2\ k_r\ \bar{C}\ E\ f)$$

in which D = dose rate, in rad per day, for beta or gamma radiation from an infinitely thick source uniformly spread over the bed of the study reach,

k_r = retention factor for radionuclide, dimensionless,

$\bar{C}$ = mean radionuclide concentration for releases of known duration through White Oak Dam, in microcuries per gram (10^{-6} curies per gram),

E = effective absorbed radiation of a beta disintegration or maximum energy of gamma ray emission, in million electron volts, and

f = fraction of disintegration of a particular energy, dimensionless.

This equation for dose rate is derived from those proposed by Cowser and Snyder[30] (eqns. (14) and (17)).

D is a nominal dose rate applicable to the study reach as a whole. As such it is an indicator of relative hazard. If D approaches one-third of the critical dose rate, local dose rates may be approaching critical levels. Especially critical areas of river bed are along the right bank between CRM 20 and 21, and also in the vicinity of CRM 4.3, 14.0, and 17.5.

For case (2), safety analysis would seek an estimate of the rate of build-up of radiation to the critical dose rate. Radiation dose at the surface of the sediment increases, in general, as the radioactive sediment becomes thicker. If the radionuclide releases are constant this relationship between radiation dose and thickness is an exponential function. Cottrell[2] indicates that the dose asymptotically approaches a maximum in Clinch River bottom sediment at a thickness of about 2.6 feet; Sayre and Hubbell[31] find this asymptote is somewhat less than 2.6 feet. This thickness (2.6 feet) is called the infinite thickness; its magnitude varies slightly with variations in physicochemical properties of sediment and with energy distribution of gamma ray emissions.

Rate of build-up of radiation dose depends on the rate of accumulation of radioactive deposits; rate of accumulation, in turn, depends on location in the study reach.

Data required to estimate radiation dose would be rate of accumulation, or time to attain infinite thickness, and the interrelationship of radionuclide concentrations in releases and sediments throughout the study reach.

Information on sediment distribution is obtained from Table 7 and from Figure 8. Time required to attain an overburden of infinite thickness may be estimated by assuming the rate of accumulation at a point on the river bed is constant. The rate is assumed to be equal to the ratio of the radioactive thickness observed at the point

in 1962 to the period of radionuclide releases from White Oak Creek, 19.5 years. Using this estimate of the rate of accumulation and the data in Table 7, the time required to attain infinite thickness over various proportions of the river bed may be determined.

The inter-relationship of radionuclide concentrations in releases and in sediment may be derived from the relationship of concentration and thickness (Fig. 11).

PHYSICOCHEMICAL COMPOSITION OF BOTTOM SEDIMENT

The results of physical and chemical analyses of 23 sediment samples from the two cores selected for detailed study are listed in Table 8. In general, radioactive bottom sediment in the Clinch River may be classed, according to particle size and mineralogical composition, as clayey silt[32] 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.

The mean specific bulk weight for all core composites used for the radionuclide inventory was 1.6 g/ml. The mean dry-to-wet weight ratio for all composites was 0.69.

Physical Characteristics

Particle-size distribution.--The results of particle-size analyses of the 23 samples are shown in Tables 8 and 9 and Figures 12 and 13. A diameter of 4 microns was used as the lower limit of silt-size particles 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 8 (columns 2-7) and in constituent correlation studies, because it was believed that additional 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 (columns 2-4 and columns 5-7, table 8) is the result of the use of two different methods of size analysis.

The results of the particle-size analyses by wet seiving and bottom withdrawal tube, plotted on triangular sand-silt-clay diagrams[32], are shown in Figures 12 and 13. Most of the samples fall in the clayey silt portion of the diagram.

Particle-size analyses were made also of composite samples of 45 Clinch River bottom sediment cores. Figure 14 is a plot of the results of these analyses. It is apparent that the particle-size

Table 8. Physical and Chemical Characteristics of Selected Samples of Bottom Sediment from the Clinch River[a]

Depth Interval (in.)	Particle-Size Distribution[b] (%)			Particle-Size Distribution[d] (%)			Adsorbed Water (%)	Cation Exchange Capacity (meq/100 g)	Leachable Cation Content (meq/100 g)		Carbon Content (%)			Free Oxide Content (%)		Minor Element Content (%)				Radionuclide Content (pc/g)[e]			
	Sand	Silt	Clay[c]	Sand	Silt	Clay[c]			Ca	Mg	Mineral	Organic	Total	Fe_2O_3	Al_2O_3	K	Rb	Cs	Sr	^{90}Sr	^{137}Cs	^{60}Co	^{106}Ru
CRM 7.5, Hole No. 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	0.28	2.5	2.75	1.78	0.92	1.47	0.0069	0.00031	0.0097	0.81	57.39	5.370	5.628
18–20	18	66	16	33	48	19	32.4	12.2	19.5	5.6	0.62	1.6	2.22	2.22	1.28	1.98	0.0097	0.00047	0.0105	2.0	58.28	4.682	5.476
22–24	16	63	21	23	58	19	35.1	15.1	23.7	5.6	0.15	2.4	2.50	2.32	1.41	1.96	0.0096	0.00038	0.0118	2.4	153.7	13.55	10.45
30–32	13	68	19	16	58	25	36.2	16.3	15.0	5.4	0.14	2.8	2.97	2.51	1.52	2.02	0.0107	0.00047	0.0090	3.0	305.6	21.46	−0.2331
34–36	6	64	30	21	53	26	35.1	17.9	15.0	5.8	0.21	2.8	3.05	2.48	1.53	1.91	0.0101	0.00048	0.0081	3.0	155.8	18.87	−0.04510
40–42	8	68	24	22	47	31	39.7	15.7	15.3	6.7	0.20	2.8	2.98	2.42	1.54	1.85	0.0096	0.00040	0.0079	2.4	29.14	0.6705	0.6520
44–46	12	63	25	16	58	26	39.3	18.8	14.1	6.0	0.21	2.8	3.02	2.66	1.63	1.82	0.0098	0.00047	0.0088	3.0	34.18	1.212	0.9307
50–52	17	59	24	25	52	23	46.3	23.0	18.6	7.0	0.21	4.2	4.39	2.47	1.33	1.75	0.0096	0.00046	0.0086	1.5	17.08	4.538	0.5210
52–54	18	56	26	25	51	24	36.0	18.0	16.5	5.8	0.22	2.5	2.75	2.28	1.37	1.74	0.0095	0.00038	0.0097	0.81	25.51	3.588	0.1235
56–58	9	68	23	18	56	26	36.9	18.7	12.9	7.5	0.14	2.3	2.40	2.35	1.37	1.85	0.0099	0.00039	0.0118	0.50	60.02	0.2848	0.6987
60–62	4	65	31	5	64	31	35.3	20.0	12.7	5.0	0.08	2.2	2.28	3.03	1.87	2.01	0.0113	0.00065	0.0136	~0.38	34.61	0.2122	0.6145
62–64	6	65	29	11	59	30	36.8	16.9	11.5	7.4	0.08	2.1	2.16	2.99	1.86	1.90	0.0116	0.00046	0.0153	~0.41	51.46	0.04010	0.2155
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	0.15	2.4	2.59	2.85	1.51	2.01	0.0112	0.00039	0.0066	4.91	123.1	15.94	7.211
12–14	6	69	25	18	57	25	40.7	18.4	13.6	8.8	0.12	2.5	2.63	2.56	1.41	1.95	0.0107	0.00038	0.0061	3.2	229.4	13.39	0.8430
16–18	13	63	24	21	41	38	38.1	17.5	15.7	8.7	0.13	2.7	2.80	2.65	1.68	1.87	0.0103	0.00045	0.0063	8.60	605.1	64.63	−2.805
20–22	28	54	18	36	46	18	37.3	14.4	11.9	7.4	0.14	3.2	3.29	2.22	1.17	1.64	0.0083	0.00052	0.0059	2.7	44.16	10.20	0.6095
24–26	13	66	21	24	51	25	39.7	17.3	12.7	7.0	0.25	2.3	2.59	2.31	1.51	1.84	0.0096	0.00041	0.0060	2.2	21.04	8.868	1.289
28–30	13	67	20	31	44	25	37.2	15.0	13.2	4.9	0.22	2.3	2.49	2.22	1.40	1.82	0.0093	0.00040	0.0055	1.3	31.64	16.30	1.045
32–34	15	61	24	32	47	21	35.8	16.4	15.1	10.2	0.36	2.4	2.73	2.30	1.38	1.87	0.0096	0.00037	0.0070	1.2	33.07	4.123	0.6315
36–38	13	64	23	24	53	23	36.1	17.4	11.8	6.7	0.32	2.2	2.53	2.20	1.38	1.74	0.0092	0.00035	0.0054	1.4	82.55	0.4458	0.3147
40–42	13	63	24	23	55	22	33.5	17.9	12.8	8.6	0.33	2.1	2.40	2.23	1.28	1.77	0.0088	0.00038	0.0058	~0.47	39.88	0.2382	0.2847
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

[a]Results are on a dry-weight basis, except in the case of adsorbed water. Analysts, USGS – P. D. Blackmon, H. C. Starkey, H. E. Reeder, E. J. Fennelly, I. C. Frost; ORNL – T. C. Rains.

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

[c]Maximum particle size, 2 μ.

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

[e]Radionuclide content in picocuries per gram of sediment.

Table 9. Results of Particle-Size Analyses of Selected Samples of Bottom Sediment from the Clinch River

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 No. 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 No. 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

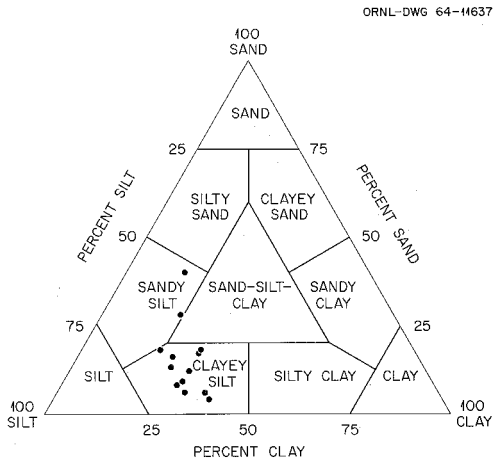

Fig. 12. Results of Particle-Size Analyses of Selected Samples from Hole 7-2, CRM 7.5. Nomenclature after Shepard[32]

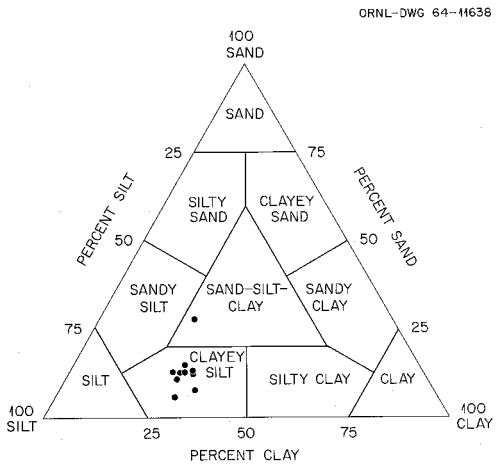

Fig. 13. Results of Particle-Size Analyses of Selected Samples from Hole 2, CRM 14.0. Nomenclature after Shepard[32]

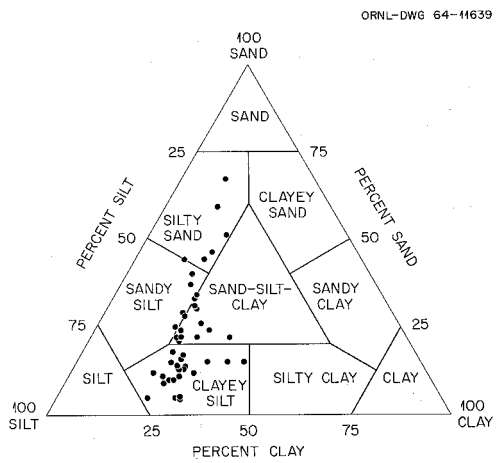

Fig. 14. Results of Particle-Size Analyses of Composite Samples of the Radioactive Portions of 45 Cores of Clinch River Bottom Sediment. Nomenclature after Shepard[32]

distributions in the composite samples, which represent the radioactive portions 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. 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. 12, 13, and 14).

Cation exchange capacity.--The cation exchange capacities of the unsized samples from the two cores studied in detail do not vary greatly (see Table 8). The cation exchange capacities of the clay- and silt-size fractions of the same samples do not vary greatly either (Table 10), suggesting that most of the variation in the unsized samples may be due to variations in content of clay-size 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. This procedure can result in clay-size fragments becoming attached to sand grains and may fail to disperse all aggregates of smaller grains. It can result also in concentration of organic material in the sand- and clay-size fractions. The data reported in Table 10 therefore represent the exchange capacities not of the ultimate particle-size fractions of the sediment, but of the size fractions approximately as they existed in the natural environment. The data in Table 10 are based on the same size separation as the data in columns 5-7, Table 8.

The cation exchange capacity of the clay-size sediment fraction was higher than that of the silt-size fraction in all samples, probably due to the higher content of layered aluminosilicate

Table 10. Cation Exchange Capacities of Clay-, Silt-, and Sand-Size Fractions of Selected Samples of Clinch River Bottom Sediment[1]

Depth Interval (in.)	Sand	Silt	Clay[2]	Depth Interval (in.)	Sand	Silt	Clay[2]
CRM 7.5, Hole No. 7-2				CRM 14.0, Hole 2			
10-12	n.d.[3]	11.7	42.4	4-6	n.d.	12.0	34.8
14-16	n.d.	13.8	33.5	8-10	n.d.	13.5	33.4
18-20	n.d.	9.6	37.1	12-14	n.d.	13.2	35.6
22-24	n.d.	10.2	33.2	16-18	n.d.	15.1	28.1
30-32	n.d.	10.9	33.5	20-22	n.d.	14.2	32.8
34-36	7.8	13.9	33.8	24-26	10.5	12.3	33.2
40-42	n.d.	8.9	39.2	28-30	n.d.	11.0	30.1
44-46	n.d.	14.1	33.4	32-34	n.d.	11.5	35.6
50-52	n.d.	14.9	39.1	36-38	n.d.	12.2	38.6
52-54	n.d.	12.6	30.0	40-42	n.d.	9.3	30.4
56-58	n.d.	11.6	33.4				
60-62	n.d.	10.1	34.2				
62-64	n.d.	11.1	32.2				

1 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.

2 Maximum diameter, 2 microns.

3 n.d., not determined.

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 capacity of these two sediment fractions.

Mineralogical Characteristics

Semi-quantitative determinations of the mineralogical compositions of the clay-, silt-, and sand-size fractions were made for all 23 sediment samples (Table 11). The size fractions studied were separated by wet seiving 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 the 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[34] for discussion of the structures of the clay minerals). Calcite was reported in three samples.

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

Table 11. Mineralogical Composition of Selected Samples of Clinch River Bottom Sediment[a]

Sampling Section	Hole Number	Depth Interval (in.)	Size of Fraction	Mica	Vermiculite (Trioctahedral)	Mica-Vermiculite (Mixed-Layered)	Montmorillonite	Chlorite	Aluminum-Interlayered Vermiculite (Dioctahedral)	Kaolinite	Quartz	Feldspar	Dolomite	Calcite
CRM 7.5	7-2	10–12	Clay[b]	20	10–15	10	Trace	Trace	10–15	20	10–15	–	–	–
			Silt[c]	<10	Trace	Trace	–	–	Trace	Trace	70	<10	<10	–
			Sand[d]	<10	–[e]	–	–	–	–	–	80	<10	10	–
		14–16	Clay	20	10–15	<10	Trace	Trace	20	20	10–15	–	–	–
			Silt	10	Trace	Trace	–	Trace	–	Trace	70	Trace	Trace	Trace
			Sand	<10	–	–	–	–	–	–	80	10	<10	–
		18–20	Clay	20	10–15	<10	–	Trace	20	20	10–15	Trace	–	–
			Silt	10	Trace	Trace	–	–	Trace	<10	60–65	<10	Trace	Trace
			Sand	<10	–	–	–	–	–	–	70	10	10–15	–
		22–24	Clay	10–15	10–15	10	–	Trace	20	20	10–15	–	–	–
			Silt	<10	<10	<10	–	–	<10	<10	60–65	Trace	Trace	–
			Sand	<10	–	–	–	–	–	<10	70	<10	<10	–
		30–32	Clay	20	10–15	<10	–	Trace	20	20	10–15	Trace	–	–
			Silt	10	–	<10	–	–	Trace	Trace	60–65	10	–	–
			Sand	<10	–	–	–	–	–	–	70–75	10	10	–
		34–36	Clay	20	10–15	<10	–	Trace	10–15	20	20	Trace	–	–
			Silt	10–15	–	Trace	–	–	Trace	<10	70	<10	–	–
			Sand	Trace	–	–	–	–	–	–	70–75	10–15	10	–
		40–42	Clay	20	20	10	Trace	–	10	20	20	–	–	–
			Silt	10	–	–	–	–	–	<10	70	<10	Trace	–
			Sand	Trace	–	–	–	–	–	–	70–75	10	10	–
		44–46	Clay	20	10–15	10–15	–	Trace	10	20	10–15	–	–	–
			Silt	10	–	<10	–	–	–	Trace	70	<10	Trace	–
			Sand	<10	–	–	–	–	–	Trace	80	<10	Trace	–
		50–52	Clay	10–15	10–15	10–15	–	Trace	20	10–15	10–15	–	–	–
			Silt	10	–	–	–	10	–	Trace	70–75	Trace	Trace	–
			Sand	<10	–	–	–	–	–	–	70	10	10–15	–
		52–54	Clay	20	10–15	<10	–	Trace	20	20	10–15	–	–	–
			Silt	10	–	–	–	–	10	Trace	60–65	10	Trace	–
			Sand	<10	–	–	–	–	–	–	70–75	10	10	–
		56–58	Clay	20	10–15	10	Trace	–	10–15	20	10–15	–	–	–
			Silt	10	<10	–	–	–	<10	–	70	<10	Trace	–
			Sand	Trace	–	–	–	–	–	–	80	<10	<10	–
		60–62	Clay	20	10	10	Trace	–	20	20	10–15	–	–	–
			Silt	10	–	Trace	–	–	<10	Trace	70	<10	Trace	Trace
			Sand	Trace	–	–	–	–	–	Trace	70–75	10–15	Trace	–
		62–64	Clay	20	10	10	–	–	20	20	20	–	–	–
			Silt	10	–	–	–	–	<10	Trace	70	10	Trace	–
			Sand	10	–	–	–	–	–	Trace	70–75	<10	<10	–

Table 11 (continued)

Sampling Section	Hole Number	Depth Interval (in.)	Size of Fraction	Mica	Vermiculite (Trioctahedral)	Mica-Vermiculite (Mixed-Layered)	Montmorillonite	Chlorite	Aluminum-Interlayered Vermiculite (Dioctahedral)	Kaolinite	Quartz	Feldspar	Dolomite	Calcite
CRM 14	2	4–6	Clay	20	10	10–15	–	Trace	10–15	20	10–15	–	–	–
			Silt	10	–	Trace	–	–	Trace	Trace	70–75	10	–	–
			Sand	<10	–	–	–	–	–	–	80	10	Trace	–
		8–10	Clay	20	10–15	10	–	Trace	10–15	20	10–15	–	–	–
			Silt	10–15	–	Trace	–	–	Trace	<10	70	<10	–	–
			Sand	<10	–	–	–	–	–	<10	70	10	<10	–
		12–14	Clay	20	10	10–15	–	Trace	10–15	20	10–15	–	–	–
			Silt	10	–	Trace	–	–	Trace	<10	70	<10	Trace	–
			Sand	<10	–	–	–	–	–	–	80	<10	<10	–
		16–18	Clay	20–25	10	10	Trace	Trace	10	10–15	20–25	–	–	–
			Silt	10	–	<10	–	–	<10	Trace	70	<10	Trace	–
			Sand	<10	–	–	–	–	–	–	80	10–15	–	–
		20–22	Clay	10–15	10–15	10–15	Trace	Trace	10–15	20	10–15	–	–	–
			Silt	<10	–	Trace	–	–	Trace	<10	70	<10	Trace	–
			Sand	Trace	–	–	–	–	–	–	80–85	10	<10	–
		24–26	Clay	20	10–15	10–15	–	–	10–15	20	10–15	–	–	–
			Silt	<10	–	Trace	–	–	Trace	Trace	70	10	Trace	–
			Sand	<10	–	–	–	–	–	–	70	10–15	10–15	–
		28–30	Clay	10–15	10–15	10–15	–	–	10–15	10–15	20	–	–	–
			Silt	<10	–	Trace	–	–	Trace	<10	70	<10	Trace	–
			Sand	Trace	–	–	–	–	–	Trace	70	10–15	<10	–
		32–34	Clay	10–15	10	10–15	–	–	10–15	20	20	<10	–	–
			Silt	10	–	Trace	–	–	Trace	Trace	70	<10	<10	–
			Sand	Trace	–	–	–	–	–	Trace	70–75	10	<10	–
		36–38	Clay	10–15	10–15	10–15	Trace	–	10–15	20	10–15	–	–	–
			Silt	10	Trace	Trace	–	–	Trace	Trace	70	10	Trace	–
			Sand	Trace	–	–	–	–	–	–	70–75	10	10–15	–
		40–42	Clay	10–15	10–15	10–15	–	–	20	20	10–15	–	–	–
			Silt	10	–	Trace	–	–	Trace	–	70–75	10	<10	–
			Sand	10	–	–	–	–	–	–	70–75	10	<10	–

[a]Determined by x-ray diffraction method. Because of inherent inaccuracies of method, contents have been rounded to nearest 10%, ±5%, except where noted. Analyst, P. D. Blackmon.

[b]<2 μ in diameter.

[c]2 to 62 μ in diameter.

[d]>62 μ in diameter.

[e]Dash indicates mineral not detected.

present in the silt-size fractions of all but 4 of the 23 samples (Table 11). 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 case 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/gram (picocuries per gram) $^{103-106}Ru$ and 10.1 pc/gram $^{90}Sr^{35}$. This radionuclide content is within the range of radioactivity commonly found in suspended sediment in Clinch River water downstream from the mouth of White Oak Creek. In the case 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 mineral carbon was very low compared to the content of organic carbon (Table 8). The presence of dolomite as sand- and silt-size particles suggests that much of the mineral carbon content was derived from the coarser sediment fractions (Table 11). Thus, it appears that poorly-crystallized calcium carbonate is not abundant in the finer size fractions of Clinch River bottom sediment. However, its presence even in very small amounts may be significant in retention of radionuclides by the sediment.

Tamura[19] has described the strong affinity of ^{90}Sr 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 aluminum oxide in the sediment samples are listed in Table 8. 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 8, also. The variation in content of organic carbon between samples is small.

Chemical Characteristics

Concentrations of one minor element and three trace elements in the sediment samples are shown in Table 8. Concentrations of potassium and rubidium were measured as possible indicators of the content 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 (^{137}Cs) for cation exchange sites on clay-type minerals. In the case of strontium, not only does radiostrontium (^{90}Sr) compete with stable strontium for exchange sites, but both forms compete with the much more abundant calcium. Nelson[36] measured the radiostrontium : stable strontium ratio in clam shells in the Clinch River and the Tennessee River, and reported that the ratio decreased with distance downstream from the mouth of White Oak 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 radiostrontium and stable strontium for some of the calcium ions in the calcium carbonate of the clam shell 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 8). This observation suggests that interstitial water contained in the sediment, and minerals dissolved by the leaching solution, as well as sorbed ions displaced by ammonium ions from the leaching solution, contributed to the cation content of the leachate. 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 the case of 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[34].

Results of radiochemical analyses of the 23 sediment samples are shown in Table 8.

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 12 and 13) confirm the inter-relationships 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 essentially all clay minerals in the clay-size sediment fraction.

Mutual correlations of the contents of ^{90}Sr, ^{137}Cs, and ^{60}Co, which occur in both cores, are best in Hole 2. For Hole 7-2, a correlation of ^{137}Cs and ^{60}Co concentrations with content of particles less than 2 microns in diameter was detected after application of a correction for variations in annual releases of ^{137}Cs to the river (see Relationship of Vertical Distribution to Annual Releases). The correction was possible for only the upper five samples out of the thirteen 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 concentrations of ^{137}Cs and ^{60}Co correlate with cation exchange capacity and the contents of free

Table 12. Coefficients of Correlation for Constituent Pairs, Hole 7-2, CRM 7.5

	Particle-Size Distribution[a]			Particle-Size Distribution[c]			Adsorbed Water	Cation Exchange Capacity	Leachable Cation Content		Carbon Content			Free Oxide Content		Minor Element Content				Radionuclide Content					
	Sand	Silt	Clay[b]	Sand	Silt	Clay[b]			Ca	Mg	Mineral	Organic	Total	Fe_2O_3	Al_2O_3	K	Rb	Cs	Sr	^{90}Sr	^{137}Cs	^{60}Co	^{106}Ru	^{137}Cs Corrected	^{60}Co Corrected
Sand		−0.84	−0.85	0.91	−0.81	−0.93	−0.23	−0.57	0.71	0.33	0.52	−0.07	0.05	−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	−0.84		0.43	−0.70	0.71	0.60	0.06	0.21	−0.53	−0.36	−0.25	−0.09	−0.16	0.56	0.64	0.69	0.74	0.23	0.21	0.28	0.36	0.13	−0.37	0.55	0.49
Clay[b]	−0.85	0.43		−0.83	0.87	0.90	0.32	0.74	−0.67	−0.20	−0.63	0.20	0.07	0.84	0.86	0.45	0.79	0.50	0.31	−0.01	−0.10	−0.10	−0.66	0.68	0.74
Sand	0.91	−0.70	−0.83		−0.97	−0.91	−0.29	−0.67	0.64	0.25	0.58	−0.09	0.04	−0.92	−0.94	−0.78	−0.96	−0.58	−0.37	−0.17	−0.18	−0.002	0.59	−0.77	−0.76
Silt	−0.81	0.62	0.75	−0.97		0.77	0.23	0.64	−0.48	−0.28	−0.53	0.06	−0.07	0.87	0.88	0.83	0.92	0.59	0.42	0.27	0.25	0.08	−0.42	0.71	0.73
Clay[b]	−0.93	0.71	0.87	−0.91	0.77		0.36	0.63	−0.79	−0.16	−0.58	0.14	0.01	0.87	0.91	0.59	0.87	0.49	0.26	−0.04	0.003	−0.16	−0.76	0.81	0.75
Adsorbed water	−0.23	0.06	0.32	−0.29	0.23	0.36		0.74	−0.16	−0.29	−0.35	0.88	0.83	0.29	0.20	−0.09	0.19	0.04	−0.27	−0.27	−0.25	−0.20	−0.45	0.89	0.93
Cation exchange capacity	−0.57	0.21	0.74	−0.67	0.64	0.63	0.74		−0.47	−0.001	−0.59	0.66	0.55	0.64	0.55	0.23	0.56	0.43	0.05	−0.13	−0.14	−0.12	−0.66	0.97	0.99
Calcium	0.71	−0.53	−0.67	0.64	−0.48	−0.79	−0.16	−0.47		0.01	0.51	0.02	0.13	−0.69	−0.69	−0.25	−0.64	−0.39	−0.30	0.11	0.05	0.26	0.86	−0.49	−0.34
Magnesium	0.33	−0.36	−0.20	0.25	−0.28	−0.16	0.29	−0.001	0.01		−0.01	0.09	0.08	−0.18	−0.26	−0.35	−0.29	−0.26	0.04	−0.53	−0.38	−0.41	−0.02	−0.40	−0.38
Mineral carbon	0.52	−0.25	−0.63	0.58	−0.53	−0.58	−0.35	−0.59	0.51	−0.01		−0.32	−0.09	−0.58	−0.58	−0.17	−0.47	−0.17	−0.34	0.12	−0.19	−0.04	0.42	−0.71	−0.81
Organic carbon	−0.07	−0.09	0.20	−0.09	0.06	0.14	0.88	0.66	0.02	−0.09	−0.32		0.97	0.06	−0.01	−0.22	−0.006	−0.06	−0.52	−0.05	0.01	0.18	−0.35	0.74	0.77
Total carbon	0.05	−0.16	0.07	0.04	−0.07	0.01	0.83	0.55	0.13	0.08	−0.09	0.97		−0.07	−0.14	−0.26	−0.11	−0.08	−0.65	−0.004	−0.02	0.19	−0.29	0.71	0.69
Fe_2O_3	−0.83	0.56	0.84	−0.92	0.87	0.87	0.29	0.64	−0.69	−0.18	−0.58	0.06	−0.07		0.97	0.68	0.92	0.72	0.51	0.04	0.03	−0.14	−0.58	0.79	0.76
Al_2O_3	−0.89	0.64	0.86	−0.94	0.88	0.91	0.20	0.55	−0.69	−0.26	−0.58	−0.01	−0.14	0.97		0.72	0.94	0.64	0.46	0.12	0.10	−0.08	−0.57	0.77	0.75
Potassium	−0.67	0.69	0.45	−0.78	0.83	0.59	−0.09	0.23	−0.25	−0.35	−0.17	−0.22	−0.26	0.68	0.72		0.82	0.62	0.32	0.40	0.46	0.30	−0.19	0.56	0.53
Rubidium	−0.90	0.74	0.79	−0.96	0.92	0.87	0.19	0.56	−0.64	−0.29	−0.47	−0.006	−0.11	0.92	0.94	0.82		0.59	0.47	0.17	0.24	0.06	−0.56	0.69	0.65
Cesium	−0.44	0.23	0.50	−0.58	0.59	0.49	0.04	0.43	−0.39	−0.26	−0.17	−0.06	−0.08	0.72	0.64	0.62	0.59		0.30	0.06	0.02	−0.04	−0.36	0.30	0.20
Strontium	−0.31	0.21	0.31	−0.37	0.42	0.26	−0.27	0.05	−0.30	0.04	−0.34	−0.52	−0.65	0.51	0.46	0.32	0.47	0.30		−0.30	−0.13	−0.35	0.07	−0.19	−0.07
^{90}Sr	−0.15	0.28	−0.01	−0.17	0.27	−0.04	−0.27	−0.13	0.11	−0.53	0.12	−0.05	−0.004	0.04	0.12	0.40	0.17	0.06	−0.30		0.69	0.72	0.11	0.78	0.76
^{137}Cs	−0.15	0.36	−0.10	−0.18	0.25	0.003	−0.25	−0.14	0.05	−0.38	−0.19	0.01	−0.02	0.03	0.10	0.46	0.24	0.02	−0.13	0.69		0.90	0.03	0.99	0.96
^{60}Co	−0.01	0.13	−0.10	−0.002	0.08	−0.16	−0.20	−0.12	0.26	−0.41	−0.04	0.18	0.19	−0.14	−0.08	0.30	0.06	−0.04	−0.35	0.72	0.90		0.12	0.99	0.98
^{106}Ru	0.61	−0.37	−0.66	0.59	−0.42	−0.76	−0.45	−0.66	0.86	−0.02	0.42	−0.35	−0.29	−0.58	−0.57	−0.19	−0.56	−0.36	0.07	0.11	0.03	0.12		−0.54	−0.39
^{137}Cs corrected[d]	−0.62	0.55	0.68	−0.77	0.71	0.81	0.89	0.97	−0.49	−0.40	−0.71	0.74	0.71	0.79	0.77	0.56	0.69	0.30	−0.19	0.78	0.99	0.99	−0.54		0.98
^{60}Co corrected[d]	−0.60	0.49	0.74	−0.76	0.73	0.75	0.93	0.99	−0.34	−0.38	−0.81	0.77	0.69	0.76	0.75	0.53	0.65	0.20	−0.07	0.76	0.96	0.98	−0.39	0.98	

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

[b]Maximum particle diameter, 2 μ.

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

[d]Five values only.

Table 13. Coefficients of Correlation for Constituent Pairs, Hole 2, CRM 14.0

	Particle-Size Distribution[a]			Particle-Size Distribution[c]			Adsorbed Water	Cation Exchange Capacity	Leachable Cation Content		Carbon Content			Free Oxide Content		Minor Element Content				Radionuclide Content					
	Sand	Silt	Clay[b]	Sand	Silt	Clay[b]			Ca	Mg	Mineral	Organic	Total	Fe_2O_3	Al_2O_3	K	Rb	Cs	Sr	^{90}Sr	^{137}Cs	^{60}Co	^{106}Ru	^{137}Cs Corrected	^{60}Co Corrected
Sand		−0.91	−0.69	0.80	−0.52	−0.40	−0.43	−0.63	−0.42	−0.26	0.07	0.69	0.78	−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	−0.91		0.33	−0.74	0.51	0.32	0.37	0.43	0.43	−0.06	−0.20	−0.71	−0.83	0.46	0.56	0.22	0.65	−0.67	−0.13	−0.03	0.12	0.02	0.44	0.09	0.02
Clay[b]	−0.69	0.33		−0.53	0.28	0.34	0.32	0.68	0.20	0.68	0.17	−0.34	−0.32	0.45	0.38	0.66	0.67	−0.56	0.50	0.33	0.43	0.15	−0.16	0.36	0.14
Sand	0.80	−0.74	−0.53		−0.72	−0.42	−0.55	−0.77	−0.63	−0.38	0.41	0.46	0.60	−0.79	−0.62	−0.73	−0.77	0.44	−0.08	−0.33	−0.45	−0.20	−0.58	−0.41	−0.19
Silt	−0.52	0.51	0.28	−0.72		−0.32	0.41	0.67	0.43	0.34	−0.14	−0.49	−0.54	0.45	−0.02	0.44	0.39	−0.49	−0.01	−0.29	−0.22	−0.50	0.66	−0.27	−0.50
Clay[b]	−0.40	0.32	0.34	−0.42	−0.32		0.20	0.19	0.31	0.07	−0.36	−0.005	−0.13	0.50	0.87	0.41	0.52	0.05	0.12	0.82	0.90	0.91	−0.05	0.91	0.91
Adsorbed water	−0.43	0.37	0.32	−0.55	0.41	0.20		0.77	0.45	0.32	−0.60	0.12	0.009	0.77	0.44	0.71	0.80	0.002	0.35	0.47	0.30	0.20	0.37	0.27	0.20
Cation exchange capacity	−0.63	0.43	0.68	−0.77	0.67	0.19	0.77		0.54	0.59	−0.23	−0.30	−0.35	0.77	0.42	0.74	0.78	−0.36	0.39	0.33	0.29	0.05	0.41	0.24	0.05
Calcium	−0.42	0.43	0.20	−0.63	0.43	0.31	0.45	0.54		0.50	−0.45	−0.19	−0.34	0.82	0.57	0.67	0.64	−0.16	0.46	0.31	0.38	0.30	0.84	0.35	−0.30
Magnesium	−0.26	−0.06	0.68	−0.38	0.34	0.07	0.32	0.59	0.50		−0.04	0.05	0.07	0.58	0.19	0.54	0.52	−0.14	0.85	0.31	0.33	0.10	0.18	0.27	0.09
Mineral carbon	0.07	−0.20	0.17	0.41	−0.14	−0.36	−0.60	−0.23	−0.45	−0.04		−0.41	−0.24	−0.68	−0.33	−0.31	−0.46	−0.50	−0.004	−0.59	−0.53	−0.52	−0.45	−0.53	−0.52
Organic carbon	0.69	−0.71	−0.34	0.46	−0.49	−0.005	0.12	−0.30	−0.19	0.05	−0.48		0.96	0.003	−0.24	−0.35	−0.19	0.89	0.18	0.45	0.27	0.37	−0.26	0.29	0.37
Total carbon	0.78	−0.83	−0.32	0.60	−0.54	−0.13	0.009	−0.35	−0.34	0.07	−0.24	0.96		−0.17	−0.35	−0.46	−0.31	0.83	0.22	0.33	0.12	0.23	−0.40	0.15	0.23
Fe_2O_3	−0.55	0.46	0.45	−0.79	0.45	0.50	0.77	0.77	0.82	0.58	−0.68	0.003	−0.17		0.67	0.81	0.88	−0.05	0.46	0.64	0.65	0.48	0.60	0.61	0.48
Al_2O_3	−0.60	0.56	0.38	−0.62	−0.02	0.87	0.44	0.42	0.57	0.19	−0.33	−0.24	−0.35	0.67		0.67	0.73	−0.19	0.28	0.68	0.72	0.70	0.27	0.72	0.70
Potassium	−0.84	0.22	0.66	−0.73	0.44	0.41	0.71	0.74	0.67	0.54	−0.31	−0.35	−0.46	0.81	0.67		0.95	−0.46	0.55	0.36	0.40	0.25	0.40	0.34	0.24
Rubidium	−0.79	0.65	0.67	−0.77	0.39	0.52	0.80	0.78	0.64	0.52	−0.46	−0.19	−0.31	0.88	0.73	0.95		−0.34	0.49	0.55	0.57	0.39	0.36	0.52	0.39
Cesium	0.76	−0.67	−0.56	0.44	−0.49	0.05	0.002	−0.36	−0.16	−0.14	−0.50	0.89	0.83	−0.05	−0.19	−0.46	−0.34		−0.009	0.40	0.20	0.40	−0.14	0.25	0.41
Strontium	−0.12	−0.13	0.50	−0.08	−0.01	0.12	0.35	0.39	0.46	0.85	−0.004	0.18	0.22	0.46	0.28	0.55	0.49	−0.009		0.35	0.27	0.20	0.05	0.22	0.20
^{90}Sr	−0.12	−0.03	0.33	−0.33	−0.29	0.82	0.47	0.33	0.31	0.31	−0.59	0.45	0.33	0.64	0.68	0.36	0.55	0.40	0.35		0.94	0.92	−0.05	0.94	0.92
^{137}Cs	−0.28	0.12	0.43	−0.45	−0.22	0.90	0.30	0.29	0.38	0.33	−0.53	0.27	0.12	0.65	0.72	0.40	0.57	0.20	0.27	0.94		0.92	−0.009	0.99	0.92
^{60}Co	−0.08	0.02	0.15	−0.20	−0.50	0.91	0.20	0.05	0.30	0.10	−0.52	0.37	0.23	0.48	0.70	0.25	0.39	0.40	0.20	0.92	0.92		−0.08	0.94	~1.00
^{106}Ru	−0.27	0.44	−0.16	−0.58	0.66	−0.05	0.37	0.41	0.84	0.18	−0.45	−0.26	−0.40	0.60	0.27	0.40	0.36	−0.14	0.05	−0.05	−0.009	−0.08		−0.02	−0.08
^{137}Cs corrected	−0.23	0.09	0.36	−0.41	−0.27	0.91	0.27	0.24	0.35	0.27	−0.53	0.29	0.15	0.61	0.72	0.34	0.52	0.25	0.22	0.94	0.99	0.94	−0.02		0.94
^{60}Co corrected	−0.08	0.02	0.14	−0.19	−0.50	0.91	0.20	0.05	0.30	0.09	−0.52	0.37	0.23	0.48	0.70	0.24	0.39	0.41	0.20	0.92	0.92	~1.00	−0.08	0.94	

[a]Separation of sand-size particles by wet sieving of wet samples. Determination of content of silt- and clay-size particles by bottom withdrawal tube.

[b]Maximum particle diameter, 2 μ.

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

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 ^{106}Ru 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 showed a strong negative correlation with the content of silt-size particles, emphasizing the previously-noted small variation in content of clay-size particles in the composite samples. No correlation of particle-size distribution with the content of individual radionuclides in the samples was noted. A strong correlation of ^{137}Cs concentration with ^{60}Co concentration observed in the composite samples is consistent with the correlation of those two radionuclides observed in other samples of Clinch River bottom sediment.

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[34], usually increases as the mean particle size of the sediment decreases; (2) in some materials, including several of the layered aluminosilicate minerals and organic matter, sorption capacity varies directly with surface area, which increases as particle size decreases[34, 37].

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 studied in detail exhibited patterns of variation in gross gamma radioactivity with depth which were similar to the pattern of annual releases of ^{137}Cs to the Clinch River from ORNL. Because ^{137}Cs and ^{60}Co account for most of the radioactivity in the sediment, the effect of annual releases must be corrected for 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 concentrations of the individual slices by an amount proportional to the amount of ^{137}Cs released during the years which the sample is believed to represent; (2) in the case of the composite samples of the sediment cores, by using cores from portions of the river in which there has been a great preponderance of sediment deposition over sediment erosion, thus obtaining an integrated sample representing sediment deposited during the entire period since 1943.

Another complicating factor in a study of the relationship of sediment composition to radionuclide content is the effect of dilution of radioactive sediment by non-radioactive sediment. If incorporation of ^{137}Cs and ^{60}Co 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 non-radioactive sediment deposited with the radioactive sediment. Because the proportions of the two kinds of 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 ^{137}Cs and ^{60}Co content 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 which were applied to the analyses for the two radionuclides may not have been adequate. Effects of dilution of radioactive sediment by non-radioactive sediment may have been considerable. Particle-size ranges (sand, silt, clay) used in the comparisons may not have been detailed enough to demonstrate particle size-radionuclide content relationships. 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.

Concentrations 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 White Oak Creek have indicated that of the four most important radionuclides in Clinch River bottom sediment (^{90}Sr, ^{137}Cs, ^{60}Co, ^{106}Ru), only ^{90}Sr was held primarily by simple ion exchange[4]. Evidence that some of the ^{90}Sr was associated with calcium carbonate in the sediment was obtained also.

The content of ^{90}Sr 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 ^{90}Sr. Measured concentrations of ^{90}Sr could not be corrected for variations in annual releases as were concentrations of ^{137}Cs and ^{60}Co.

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 10). 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 fraction 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[4] that the content of calcium carbonate is higher in bottom sediment from White Oak Lake than in bottom sediment from near the mouth of White Oak Creek, 0.6 mile downstream, and that the fraction of the total content of ^{90}Sr desorbed from White Oak Lake sediment through ion exchange mechanisms is less than in the case of sediment from near the mouth of the creek. The tendency of strontium to substitute for calcium in the lattices of calcium carbonate minerals[36]; the observed inverse relationships between the calcium carbonate content of White Oak Creek bottom sediment and the amount of ^{90}Sr held through ion exchange; the incorporation of both ^{90}Sr and ^{106}Ru in calcium carbonate that precipitated in the Clinch River, and the correlation of ^{106}Ru 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.

The content of organic matter (organic carbon) appears to add to the cation exchange capacity of the sediment in Hole 7-2 (see Table 12). The reported high cation exchange capacities of a number of organic materials[38] 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 feldspars, 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 its content of layered aluminosilicate minerals.

The mutual correlations between the contents of ^{90}Sr, ^{137}Cs, and ^{60}Co in the two cores (quite strong correlation in Hole 2) suggest that the ^{90}Sr content of Clinch River bottom sediment may be related in some way to the process of sedimentation of suspended matter in the river.

CONCLUSIONS

The inventory of radionuclides in Clinch River bottom sediment provides strong insight to the fate of radioactive waste released to the Clinch River. The inventory is a direct and principal measure of the residual between 20-year input and output loads of the radionuclides in the study reach of the river.

In the study reach at least 20 percent of the ^{137}Cs and rare earths released to the river are retained in the bottom sediment. Retention of ^{60}Co releases is 9 percent. Less than 1 percent each of ^{106}Ru and ^{90}Sr releases are retained in the bottom sediment. Most of the radioactive sediment, 95 percent, is in that portion of the channel bed between CRM 0 and CRM 15. Because of downstream decrease in the turbulence of flow, the thickness, cross-sectional area, and volume per unit length of radioactive sediment are generally greater in the downstream parts of the reach than in the upstream parts. Of these three geometric variables, only thickness varies in a regular manner, showing a linear increase in the downstream direction.

The total inventory of principal radionuclides in bottom sediment (^{137}Cs, ^{60}Co, and rare earths) in the Tennessee River basin is greater than that measured for the main channel of the Clinch River. Radionuclides are associated with bottom sediment of sloughs and mouths of streams tributary to the river. Downstream from the mouth of the Clinch River in Watts Bar Lake a large volume of sediment deposits exist and significant concentrations of the principal radionuclides are found in the upper strata of the bottom sediment (see ref. 29).

Variations in gross gamma radioactivity with depth in Clinch River bottom sediment largely reflect variations in the content of ^{137}Cs in the sediment. A similar pattern of variations in the content of ^{137}Cs in the sediment at several coring sites in the reach of river downstream from CRM 18 indicates more or less regular, persistent deposition of sediment at those particular sites, and thus longterm net accumulation of sediment at those sampling sections.

Incorporation of ^{137}Cs 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: (a) the similarity of the patterns of variations with depth in ^{137}Cs content of bottom sediment cores to the pattern of annual releases of ^{137}Cs from the Laboratory to the Clinch River, (b) the knowledge that at least 70 percent of the ^{137}Cs released from the Laboratory to the Clinch River was associated with suspended solids, and (c) the known preferential sorption of cesium by certain layered aluminosilicate minerals.

The similarity of the distribution pattern of ^{60}Co to that of ^{137}Cs in Clinch River bottom sediment suggests that ^{60}Co may be incorporated in the sediment by deposition of suspended solids entering the Clinch River from White Oak Creek. The two radionuclides are not necessarily associated with the same solids, however. The fact that the distribution pattern of ^{60}Co does not reflect the pattern of annual releases of ^{60}Co from White Oak Creek implies that most of the ^{60}Co 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 ^{60}Co released to the river becomes incorporated in Clinch River bottom sediment.

The relatively small contribution of ^{106}Ru to the gross gamma radioactivity of the upper portion of Clinch River bottom sediment supports the results of the radionuclide inventory, which has shown that only a small fraction of the total amount of ^{106}Ru released to the river is incorporated in the bottom sediment. The manner of incorporation of ^{106}Ru in the sediment is not known.

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 measureable quantities.

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 non-radioactive 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.

Two methods of aiding future safety analyses are suggested: (1) estimation of dose rates for individual radionuclides incorporated in bottom sediment by consideration of mean concentration of the radionuclides in releases and of the retention factor, and (2) consideration of (a) rate of sediment thickness build-up in various areas of the study reach, (b) relation of concentration to sediment thickness, and (c) inter-relationship of concentrations in releases and in sediments.

Future use of the river for radioactive liquid waste disposal is predicated on the safety of such practices. It has been concluded that past and present disposal practices have been safe[31], and that continued use of the river for such disposal is permissible. A projected decline in release of radionuclides to the river suggests that retention of radionuclides in bottom sediment will not seriously limit the river's usefulness in the immediate future, either. However, the possibility of an increase in radionuclide releases and, consequently, the possibility of increased radiation hazard, must be recognized. If it is assumed that retention of some radionuclides in bottom sediment will reach 10 to 20 percent, or more, the incorporation of radionuclides from a substantial increase in releases might be a factor leading to limited use of the stream for disposal of radioactive waste. Limitations resulting from such an increase in the radionuclide content of bottom sediment can be determined only through a safety analysis in which all avenues of radiation exposure are considered.

Water-sampling stations have value in obtaining continuous and current records of radionuclide concentrations at a site. However, measurement of small but significant losses or gains in radionuclide loads occurring between stations of a network is difficult and costly. Inventory of accumulated radioactivity in sediment is inherently more accurate than determining a small residual between

large radionuclide loads measured at two water-sampling stations. Cost of such an inventory would be comparable to the annual cost of operating a two-station water-sampling network in the Clinch River study reach. The techniques of radioactive sediment inventory developed in this investigation can be used for surveillance of long-term effects of the release of radioactive material. The method can be used in lieu of water-sampling networks.

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