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Environmental Restoration Division
Clinch River Environmental Restoration Program

Transport and Accumulation of Cesium-137 and Mercury in the Clinch River and Watts Bar Reservoir System

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Note to Recipients of ORNL/ER-7

The draft version of this report was distributed for comments in March 1990. Only minor changes were made for this final version.

Environmental Restoration Program
Martin Marietta Energy Systems, Inc.

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ACRONYMS

ACD	Analytical Chemistry Division
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
DOE	Department of Energy
EFPC	East Fork Poplar Creek
EMSL-LV	Environmental Monitoring Systems Laboratory, Las Vegas
EPA	Environmental Protection Agency
FFA	Federal Facility Agreement
GIS	Geographic Information System
Hg	mercury
HSWA	Hazardous and Solid Waste Amendments
KCP	a core taken near Kingston City Park
NBS	National Bureau of Standards
ORNL	Oak Ridge National Laboratory
ORR	Oak Ridge Reservation
ORTF	Oak Ridge Task Force
Pb	lead
PCBs	polychlorinated biphenyls
Pu	plutonium
RCRA	Resource Conservation and Recovery Act
SARA	Superfund Amendments and Reauthorization Act of 1986
SRM	standard reference material
TVA	Tennessee Valley Authority
WOD	White Oak Dam
WOL	White Oak Lake

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EXECUTIVE SUMMARY

Operations and waste disposal activities on the U.S. Department of Energy's Oak Ridge Reservation have introduced cesium-137 (^{137}Cs) and mercury (Hg) into local streams that ultimately drain into the Clinch River and Watts Bar Reservoir system. The highest discharges for both ^{137}Cs and Hg occurred during the mid-1950s. Measurements of the partitioning of ^{137}Cs and Hg between dissolved and particulate phases in the reservoir water column indicate that both contaminants have particle-to-water sorption ratios of about 10^5 and are therefore strongly associated with particles in this aquatic system. About 190 surface-sediment grab samples and more than 60 sediment cores were collected in Watts Bar Reservoir to (1) determine the extent of downstream contamination and (2) document particle and particle-associated contaminant accumulation patterns. The vertical distributions of ^{137}Cs and Hg in these sediment cores are strongly correlated ($r^2 = 0.87$), and both contaminants exhibit a large subsurface peak coincident with their peak discharge histories. Concentrations of ^{137}Cs and Hg as high as 80 pCi/g (3.0 Bq/g) and 47 μg/g, respectively, occur in this subsurface peak. A preliminary screening of the contaminants that may contribute to human health and environmental risk (Hoffman et al. 1991;* Suter 1991†) showed that these ^{137}Cs and Hg concentrations do not pose an imminent risk, especially if deep sediments are not disturbed. Further study, however, is warranted to determine the need for remediation. The sediment depth of this subsurface peak and the thickness of contaminated sediment varies with location in the reservoir and depends on the rate of sediment accumulation. The total accumulation of ^{137}Cs in Watts Bar Reservoir sediments has been estimated by measuring the ^{137}Cs inventory in each sediment core and extrapolating these data spatially with the ARC:INFO Geographic Information System software package. Results indicate that about 304 Ci (1.12×10^{13} Bq) of ^{137}Cs now reside in the reservoir sediments. Discharge records indicate that a decay-corrected total of about 335 Ci (1.24×10^{13} Bq) of ^{137}Cs have been released into the river system between 1949 and 1986. Some ^{137}Cs was released before 1949, but discharges prior to 1949 were not monitored for ^{137}Cs. Sediment core profiles of ^{137}Cs in the Clinch River and Watts Bar Reservoir suggest that the annual amounts discharged from White Oak Dam before 1949 were considerably less than the amount discharged in 1949. The comparison between measured and discharged ^{137}Cs indicates that almost 91% of the total ^{137}Cs released to the Clinch River and Tennessee River system has been retained by accumulation in Watts Bar Reservoir sediments. Using the strong correlation between the vertical distribution of ^{137}Cs and Hg in sediment cores, it has been estimated that about 76 metric tons of Hg have also accumulated in the sediments of Watts Bar Reservoir. The vertical distribution of ^{137}Cs and Hg in dated sediment cores was also used to document levels of contamination in the reservoir water column during the past 40 years.

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*F. O. Hoffman, B. G. Blaylock, M. L. Frank, L. A. Hook, E. L. Etnier, and S. S. Talmage. 1991. *Preliminary Screening of Contaminants in the Off-Site Surface Water Environment Downstream of the U.S. Department of Energy Oak Ridge Reservation*. ORNL/ER-9. Martin Marietta Energy Systems, Inc., Oak Ridge National Laboratory, Oak Ridge, Tenn.

†G. W. Suter II. May 1991. *Screening Level Risk Assessment for Off-Site Ecological Effects in Surface Waters Downstream from the U.S. Department of Energy Oak Ridge Reservation*. ORNL/ER-8. Martin Marietta Energy Systems, Inc., Oak Ridge National Laboratory, Oak Ridge, Tenn.

INTRODUCTION

Operations and waste disposal activities at the Oak Ridge Y-12 Plant, the Oak Ridge National Laboratory (ORNL), and the Oak Ridge K-25 Site (formerly the Oak Ridge Gaseous Diffusion Plant) on the U.S. Department of Energy (DOE) Oak Ridge Reservation (ORR) have introduced a variety of airborne, liquid, and solid wastes into the surrounding environment. Some of these wastes may affect off-site areas by entering local streams, which ultimately drain into the Clinch and Tennessee river system (Fig. 1). Previously reported concentrations of radionuclides, metals, and organic compounds in water, sediment, and biota of the Clinch River and Watts Bar Reservoir suggest the presence of a variety of contaminants of possible concern to the protection of human health and the environment.

DOE has initiated a comprehensive waste management and environmental restoration effort to achieve the comprehensive remediation of releases of hazardous substances, pollutants, or contaminants from the Oak Ridge Reservation (Jones et al. 1990). This effort has been undertaken in accordance with a draft Federal Facility Agreement (FFA) between the Environmental Protection Agency (EPA) Region IV, the Tennessee Department of Environment and Conservation, and DOE. The FFA requires that the cleanup of the ORR and environs be conducted in compliance with both the Resource Conservation and Recovery Act of 1976 (RCRA), as amended by the Hazardous and Solid Waste Amendments of 1984 (HWSA), and the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) of 1980, as amended by the Superfund Amendments and Reauthorization Act of 1986 (SARA).

The work reported here represents part of the initial scoping phase for the Clinch River RCRA Facility Investigation. In this work, the distribution of ^{137}Cs is used to identify contaminant accumulation patterns and potential problem, or "hot-spot," areas with regard to environmental hazard or human health. Radiocesium was chosen for this scoping effort because (1) its history of release into the Clinch River is reasonably well documented, (2) it is easy and inexpensive to measure by gamma spectrometry, and (3) it is rapidly sorbed to particulate matter and thus serves as a cost-effective tracer for identifying the transport and accumulation patterns of many other particle-reactive contaminants, such as mercury (Hg), lead (Pb), plutonium (Pu), and polychlorinated biphenyls (PCBs) (Olsen et al. 1989a).

HISTORY OF CONTAMINANT RELEASES

During the mid-1950s and early 1960s, relatively large quantities of ^{137}Cs and Hg were released into the Clinch River in association with nuclear energy research at ORNL and weapons components production at the Y-12 Plant, respectively (Fig. 2). Some of the ^{137}Cs and other radioactive wastes generated at ORNL enter surface streams that drain into White Oak Lake (WOL) (Fig. 1). Discharges from WOL into the Clinch River are controlled and monitored at White Oak Dam (WOD). Annual discharges of radioactivity from ORNL via WOD are summarized in Table 1. These discharges were calculated by (1) analyzing radionuclide concentrations in weekly flow-proportional samples, (2) multiplying this concentration by the total weekly flow, and (3) integrating these weekly samples for a year. Approximately 665 Ci (2.5×10^{13} Bq) of ^{137}Cs was released from WOL into the Clinch River and Watts Bar Reservoir system between 1949 and 1986. Some ^{137}Cs was released into the

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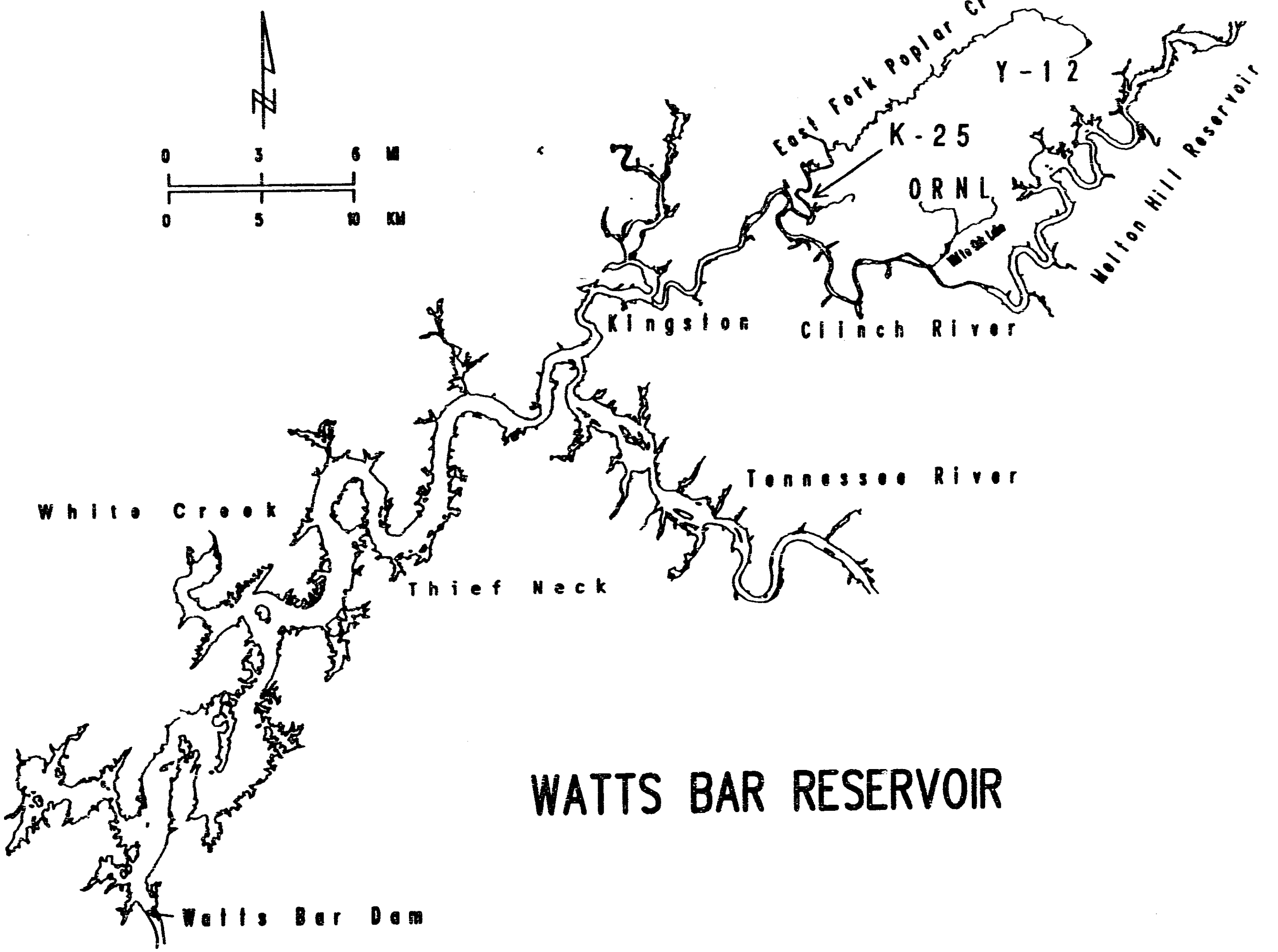

Fig. 1. Map showing the Clinch River and Tennessee River tributaries to Watts Bar Reservoir and the location of the three DOE facilities on the ORR.

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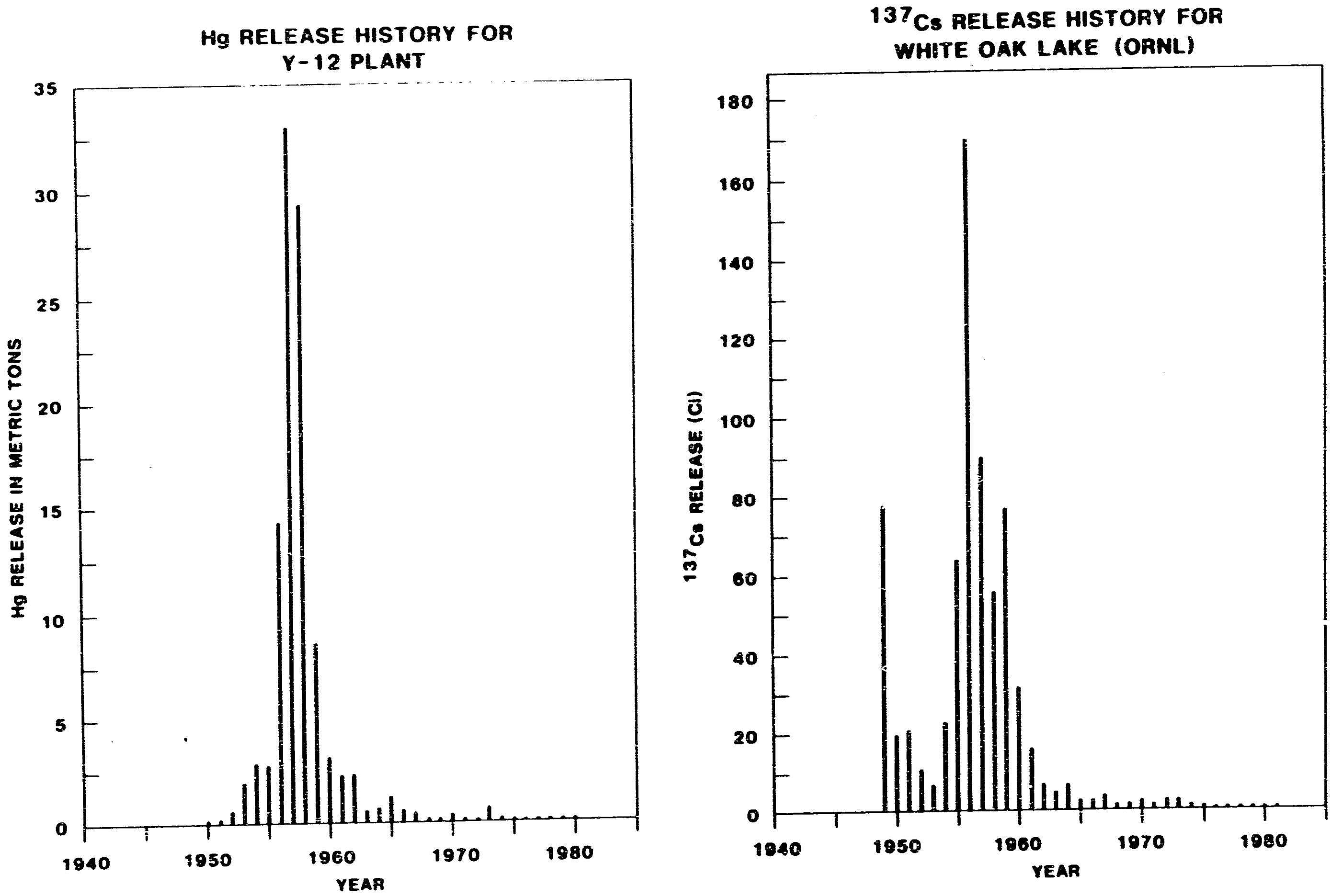

Fig. 2. Aquatic discharge histories for Hg from the Y-12 Plant and for ^{137}Cs from ORNL. A comparison of these discharge histories indicates a near coincidence in the peak release of ^{137}Cs in 1956 (with the draining of WOL) and that of Hg in 1957–58 and sharp declines in both releases after 1959.

Table 1. Annual discharges (Ci) of radionuclides from White Oak Lake to the Clinch River, 1944 to 1984[a]

Year	Beta	^{137}Cs	^{106}Ru	^{89}Sr	^{90}Sr	TRE - Ce[b]	^{144}Ce	^{95}Zr	^{95}Nb	^{131}I	^{60}Co	^{3}H	TRU[c]
1944[d]	600												
1945[d]	500												
1946[d]	900												
1947[d]	200												
1948[d]	494												
1949		77	110		150	77	18	180	22	77		NA[e]	0.04
1950		19	23		38	30	NA	15	42	19			0.04
1951		20	18		29	11	NA	5	2	18			0.08
1952		10	15		72	26	23	19	18	20			0.03
1953		6	26		130	110	7	8	4	2			0.08
1954		22	11		140	160	24	14	9	4	NA		0.07
1955		63	31		93	150	85	5	6	[illegible]	7		0.25
1956		170	29		100	140	59	12	15	4	46		0.28
1957		89	60		83	110	13	23	7	1	5		0.15
1958		55	42	NA	150	240	30	6	6	8	9		0.08
1959		76	520	0.3	60	94	48	27	30	1	77		0.68
1960		31	1,900	1.9	28	48	27	38	45	5	72		0.19
1961		15	2,000	2.0	22	24	4	20	70	4	31		0.07
1962		6	1,400	1.7	9	11	1	2	8	0.4	14		0.06
1963		4	430	1.0	8	9	2	0.3	0.7	0.4	14		0.17
1964		6	190	0.8	7	13	0.3	0.2	0.1	0.3	15	1,900	0.08
1965		2	69	0.6	3	6	0.1	0.3	0.3	0.2	12	1,200	0.50
1966		2	29	0.9	3	5	0.1	0.7	0.7	0.2	7	3,100	0.16
1967		3	17	0.7	5	9	0.2	0.5	0.5	0.9	3	13,300	1.03
1968		1	5	0.6	3	4	0.03	0.3	0.3	0.3	1	9,700	0.04
1969		1	2	0.3	3	5	0.02	0.2	0.2	0.5	1	12,200	0.20
1970		2	1	0.3	4	5	0.06	0.02	0.02	0.3	1	9,500	0.40
1971		1	0.5	0.2	3	3	0.05	0.01	0.01	0.2	1	8,900	0.05
1972		2	0.5	NA	6	5	0.03	0.01	0.01	0.3	1	10,600	0.07
1973		2	0.7		7	NA	0.02	0.05	0.05	0.5	1	15,000	0.08
1974		1	0.2		6		0.02	0.02	0.02	0.2	0.6	8,600	0.02
1975		0.6	0.3		7		NA	NA	NA	0.3	0.5	11,000	0.02
1976		0.2	0.2		5					0.03	0.9	7,400	0.01
1977		0.2	0.2		3					0.03	0.4	6,200	0.03
1978		0.3	0.2		2					0.04	0.4	6,300	0.03
1979		0.2	0.1		2.4					0.04	0.4	7,700	0.03
1980		0.6	0		1.5					0.04	0.4	4,600	0.04
1981		0.2	0.1		1.5					0.04	0.7	2,900	0.04
1982		1.5	0.2		2.7					0.06	1.0	5,400	0.03
1983		1.2	0.2		2.1					0.004	0.3	5,600	0.05
1984		0.6	0.2		2.6					0.05	0.2	6,400	0.03

[a] Data from Ohnesorge (1986).
[b] Total rare earths minus cerium.
[c] TRU = total transuranic elements.
[d] Individual radionuclide data not available.
[e] No analysis performed.
Source: Clinch River RCRA Facility Investigation, Table 4.8.

Clinch River before 1949, but discharges from WOL prior to 1949 were not monitored for ^{137}Cs. Sediment core profiles of ^{137}Cs in the Clinch River and Watts Bar Reservoir (see below) suggest that the annual amounts of ^{137}Cs discharged from WOD before 1949 were considerably less than the amount discharged in 1949. Because most of this release occurred in the mid-1950s in association with the draining of WOL in 1956 (Fig. 2) and because the half-life of ^{137}Cs is 30 years, the total decay-corrected amount of ^{137}Cs discharged as of June 1986 was about 335 Ci (1.24×10^{13} Bq).

Although the history of radionuclide releases from each of the three DOE facilities on the ORR is reasonably well documented (Table 1 in DOE 1988), quantitative information on releases of most other contaminants is either absent or incomplete. During the 1950s and early 1960s, relatively large quantities of metallic Hg were released to surface waters (Fig. 2) in association with the production-scale lithium-isotope separation process initiated at the Y-12 Plant in 1953. In this process, lithium isotopes are separated as they are transferred between two chemical phases. One of these phases is a solution of lithium in Hg, and, as a result, millions of kilograms of inorganic Hg were used in this separation project. Floor drains were installed in the process building to collect spilled Hg into special tanks in the basement. However, some of this Hg escaped these collection drains and entered into East Fork of Poplar Creek (EFPC) (Fig. 1).

Measurement of aquatic discharges of Hg from the Y-12 Plant began in April 1954. Annual releases of Hg (Fig. 2) are characterized by (1) a sharp increase in 1956 when full-scale lithium isotope processing began; (2) peak releases of 33 and 29 metric tons of inorganic Hg in 1957 and 1958, respectively; and (3) a sharp decline in Hg releases after 1958. Process changes in 1958 resulted in declining releases, and all production had ceased by 1963. The total Hg release to the environment, including estimates for the 1950-to-1954 period, has been estimated to range from about 75 to 150 metric tons (Turner et al. 1985).

PREVIOUS STUDIES

The first comprehensive program to identify the transport, accumulation, and fate of contaminants released to off-site areas from the ORR began in the mid 1950s in conjunction with the draining of WOL (Cottrell 1959). In this program, a gamma-radiation survey of surface sediments was conducted with a submersible Geiger-Mueller counting system. Most of the gamma radioactivity (^{137}Cs, ^{60}Co, rare-earth isotopes, and short-lived ^{103}Ru) was found to have been deposited in the sediments of Watts Bar Reservoir, but some could be traced down the Tennessee River system to the Chickamauga Reservoir and beyond.

A second study was conducted over a 5-year period between 1960 and 1964. This "Clinch River Study" was a comprehensive physical, chemical, biological, and sedimentological investigation to determine the environmental fate, ecological effects, and impact on man of radionuclides released to the Clinch River from ORNL (results are summarized by Struxness et al. 1967). Unfortunately, this intensive study was restricted primarily to the Clinch River itself, and the extent of contamination further downstream in Watts Bar Reservoir was not examined. Results indicated that soluble contaminants (e.g., ^{3}H and ^{90}Sr) released from WOL were rapidly diluted in the Clinch River and flushed downstream without accumulating to any great extent (Cowser et al. 1966). Results from ^{90}Sr analysis of water samples and the shells of freshwater clams showed that concentrations of ^{90}Sr were detectable in the Tennessee

River up to 500 miles downstream from the release point (WOD) and could be accurately predicted on the basis of dilution (Nelson 1969).

Only about 21% of the ^{137}Cs, 9% of the ^{60}Co, and less than 1% of the ^{90}Sr previously released from WOL had accumulated in the sediments of the Clinch River (Pickering et al. 1966; Struxness et al. 1967). As a consequence, Parker et al. (1966) concluded that very little of the radioactive material introduced into the Clinch River remained there in either the bottom sediments or in the biota. Struxness et al. (1967) concluded that the Clinch River functioned much like a pipe, transporting contaminants to sites farther downstream.

A third set of studies of the Clinch River sediments was conducted in the late 1970s to reevaluate the distribution of ^{137}Cs and to document levels of plutonium in the Clinch River sediments near the site proposed for the Clinch River Breeder Reactor (Oakes et al. 1982). Those results indicated that much of the ^{137}Cs previously deposited in the Clinch River had been eroded from the river sediments and transported downstream. Oakes et al. (1982) also reported that ^{239,240}Pu activities in the sediment were as high as 2 pCi/g (75 mBq/g) in the Clinch River near the proposed reactor site. These ^{239,240}Pu concentrations in sediments do not pose a risk to human health that would warrant remediation (Hoffman et al. 1991).

To document levels of contamination in the sediments and soils near the proposed construction site of the New Blair Road Bridge across Poplar Creek, Olsen and Cutshall (1985) measured the vertical distribution of ^{137}Cs, Hg, and ^{238}U in a sediment core collected within the creek and in a soil core collected on its floodplain. Contaminant concentrations in the Poplar Creek sediment core are presented in Table 2 and indicate that Hg levels exceeded 450 μg/g. Olsen and Cutshall (1985) attributed this high Hg concentration to discharges from the Y-12 Plant via EFPC. In addition, Olsen and Cutshall (1985) suggested that the Y-12 Plant may also be a source of uranium to off-site areas because this sediment core was collected upstream of any K-25 discharge sites to Poplar Creek.

Before uranium enrichment operations at the K-25 Site were halted, Ashwood et al. (1986) collected approximately 180 surface sediment samples and three sediment cores from the Poplar Creek and Clinch River system to identify contaminant source areas around the K-25 Site. Contaminant concentration levels in the three sediment cores are illustrated in Fig. 3. From these data, Ashwood et al. (1986) concluded that Poplar Creek sediments upstream of the K-25 Site were contaminated with Hg, uranium, and ^{60}Co and suggested that the Y-12 Plant was a significant source of the Hg and uranium contamination and that releases from the Oak Ridge Sewage Treatment Facility were responsible for the ^{60}Co. They also indicated that inputs of ^{137}Cs and ^{60}Co to off-site areas from the Y-12 Plant and from the City of Oak Ridge Sewage Treatment Plant via EFPC were relatively insignificant compared with the release of these two radionuclides from ORNL via WOL. As a consequence, much of the ^{137}Cs and ^{60}Co in the sediments near the K-25 Site were introduced via the Clinch River during periods of backflow into Poplar Creek.

The declassification of information on Hg discharged and unaccounted for from the Y-12 Plant in 1983 led to increased scrutiny of downstream areas for Hg contamination (Elwood 1984) and to the organization of an interagency task force [Oak Ridge Task Force (ORTF)] to evaluate the associated threats of off-site contamination to human health, fish, and wildlife. Most of the ORTF investigative efforts were focused on the Hg contamination of EFPC and its floodplain. A total of 1526 water, sediment, and aquatic biota samples were collected by the Tennessee Valley Authority (TVA) to assess off-site mercury contamination derived from

Table 2. Vertical distribution of mercury, ^{137}Cs, and ^{238}U in a Poplar Creek sediment core collected near Blair Road bridge (Olsen and Cutshall 1985)

Sediment depth (cm)	Organic carbon (%)	Mercury (μg/g)	^{137}Cs (pCi/g)	^{238}U (pCi/g)
0–2	1.5	6.3	1.11 ± 0.03	≤2.8
2–4	2.4	4.2	1.26 ± 0.03	≤2.8
4–8	2.5	2.2	1.07 ± 0.02	3.0 ± 1.1
8–12	1.7	5.6	0.33 ± 0.02	≤2.8
12–16	1.6	6.8	0.23 ± 0.01	3.5 ± 0.9
16–20			0.18 ± 0.01	≤2.8
20–24			0.30 ± 0.04	≤2.8
24–28			0.34 ± 0.04	4.1 ± 2.3
28–32			0.38 ± 0.05	≤2.8
32–36	1.3	14.0	0.79 ± 0.06	≤2.8
36–40	1.3	22.6	2.63 ± 0.11	≤2.8
40–44			1.33 ± 0.08	≤2.8
44–48			0.68 ± 0.05	8.3 ± 2.6
48–52	1.6	18.0	0.90 ± 0.07	≤2.8
52–56			1.33 ± 0.08	12.2 ± 4.0
56–60			1.10 ± 0.08	≤2.8
60–64	1.4	38.3	0.82 ± 0.04	7.5 ± 2.5
64–68	0.7	54.4	1.33 ± 0.08	10.2 ± 2.9
68–72			0.87 ± 0.07	4.3 ± 5.9
72–76			1.08 ± 0.08	29.8 ± 5.3
76–80			1.01 ± 0.06	15.5 ± 2.5
80–84	1.1	460.0	1.06 ± 0.07	8.8 ± 3.2
84–88	1.1	220.0	1.53 ± 0.08	8.0 ± 3.4
88–92	0.9	40.0	1.71 ± 0.08	3.8 ± 2.2
92–96	1.0	56.0	4.64 ± 0.13	7.4 ± 3.1
96–98			2.81 ± 0.11	≤2.8

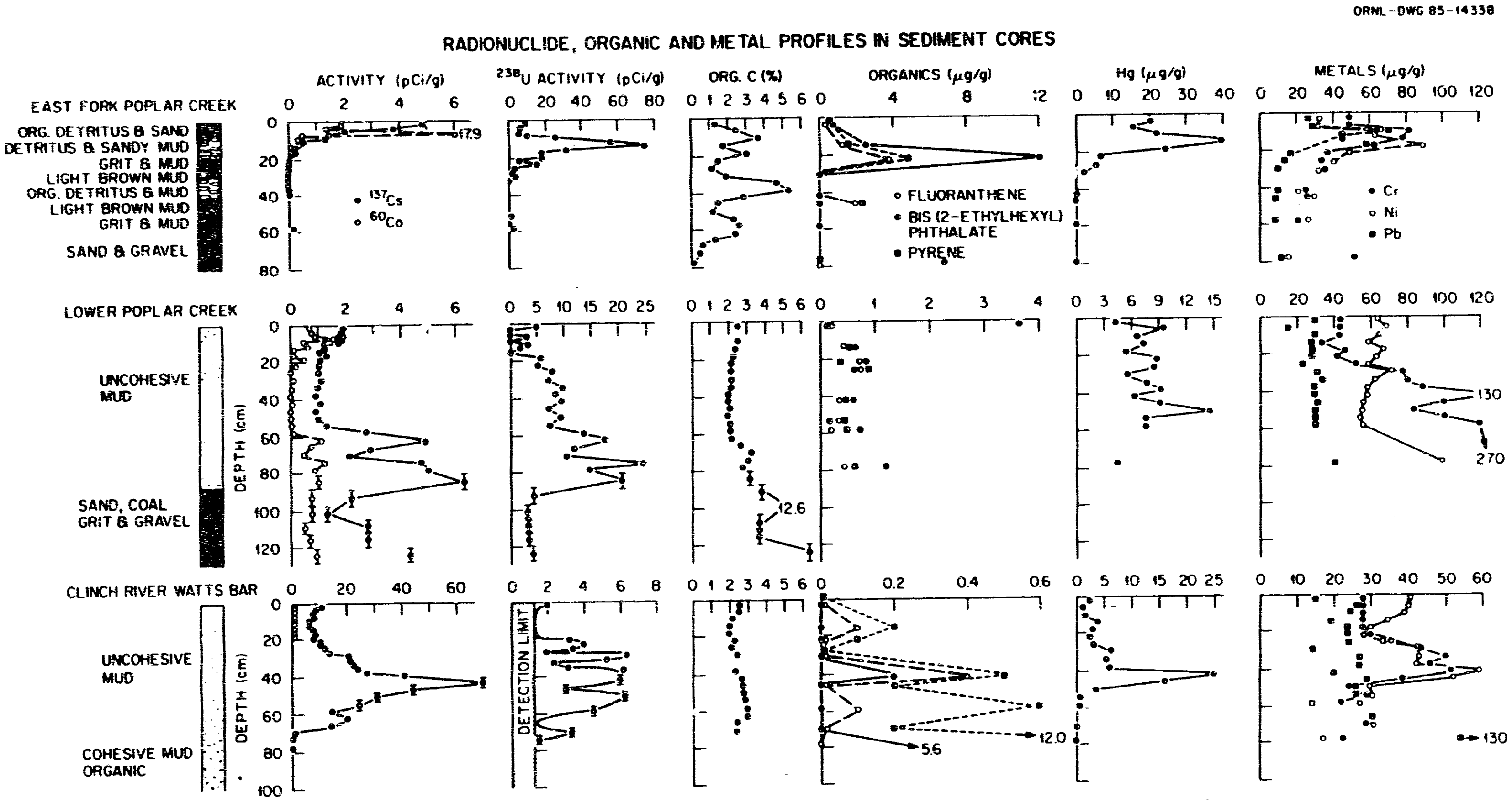

Fig. 3. Vertical distribution of ^{137}Cs, Hg, ^{238}U, and several other contaminants in a sediment core collected at the junction of East Fork Poplar Creek and Poplar Creek, a sediment core collected in Poplar Creek just downstream of the K-25 Site, and a sediment core collected near the mouth of the Clinch River at Kingston (Ashwood et al. 1986).

the Y-12 releases (TVA 1986). This ORTF-supported study indicated that about 170,000 lb (75 metric tons) of Hg had accumulated in the EFPC floodplain and that about 500 lb (0.2 metric tons) were annually exported from EFPC to off-site areas.

In another ORTF-supported study, Turner et al. (1985) collected a total of seven sediment cores from Watts Bar and Chickamauga reservoirs to determine the downstream extent of the Hg contamination. They found that concentrations of Hg and ^{137}Cs were strongly correlated in sediment cores collected in Watts Bar Reservoir, exhibiting a peak concentration at sediment depths ranging from about 40 to 100 cm (Fig. 4). The highest concentrations of Hg (47 μg/g) and of ^{137}Cs [152 pCi/g (5.6 Bq/g)] were found to occur in the core CRM-1, obtained at the mouth of the Clinch River near Kingston, Tennessee (Fig. 4). Hoffman et al. (1991) evaluated the risk to human health of these Hg and ^{137}Cs concentrations and found that, as long as the sediments remained in place, there was no imminent risk to human health. Further study is required to determine the need for remediation. Sediment cores collected from the lower Chickamauga Reservoir, however, contained Hg profiles that were more complex (in part a result of additional Hg inputs to the reservoir from a chloralkali plant located on the Hiwassee River) but contained ^{137}Cs profiles that were very similar to those in Watts Bar Reservoir (Turner et al. 1985).

On the basis of the results presented in Turner et al. (1985) and in Ashwood et al. (1986) and because most of the previous studies have been focused on the transport and fate of contaminants in EFPC, Poplar Creek, and the Clinch River downstream from the ORR, a need existed for a thorough sampling of Watts Bar Reservoir. All of the above previous studies have clearly shown that Poplar Creek and the Clinch River serve as pipelines for contaminants released from the ORR and that Watts Bar Reservoir serves as the major zone for contaminant accumulation. This conclusion is consistent with work in other river-reservoir systems, which have also indicated that reservoirs are very efficient traps for river-borne particles, nutrients, and contaminants and are sites of rapid sediment and contaminant accumulation (Dendy 1973; Ritchie, Hawks, and McHenry 1975; Olsen et al. 1981; Kimmel and Groeger 1986; Olsen et al. 1989a).

To address this need for characterizing the nature and extent of contamination in Watts Bar Reservoir, we have measured the vertical distribution of ^{137}Cs in more than 60 sediment cores (Fig. 5) and the concentration of ^{137}Cs in more than 190 surface sediment samples (Fig. 6) collected from Watts Bar Reservoir. The surface sediment samples were used to develop a map of sediment characteristics (Fig. 7) and to identify sites best suited for sediment coring. The objectives of this scoping study were to (1) use ^{137}Cs to evaluate the extent of contaminant accumulation in the reservoir sediments, (2) preliminarily identify highly contaminated off-site areas that could constitute potential risks to human health or the environment, and (3) estimate the retention efficiency of the reservoir for ^{137}Cs and, thereby, for other particle-associated contaminants.

As stated previously, this work represents the initial phase of the Clinch River RCRA Facility Investigation and provides important information for characterizing the nature and extent of ORR-derived contamination in off-site areas.

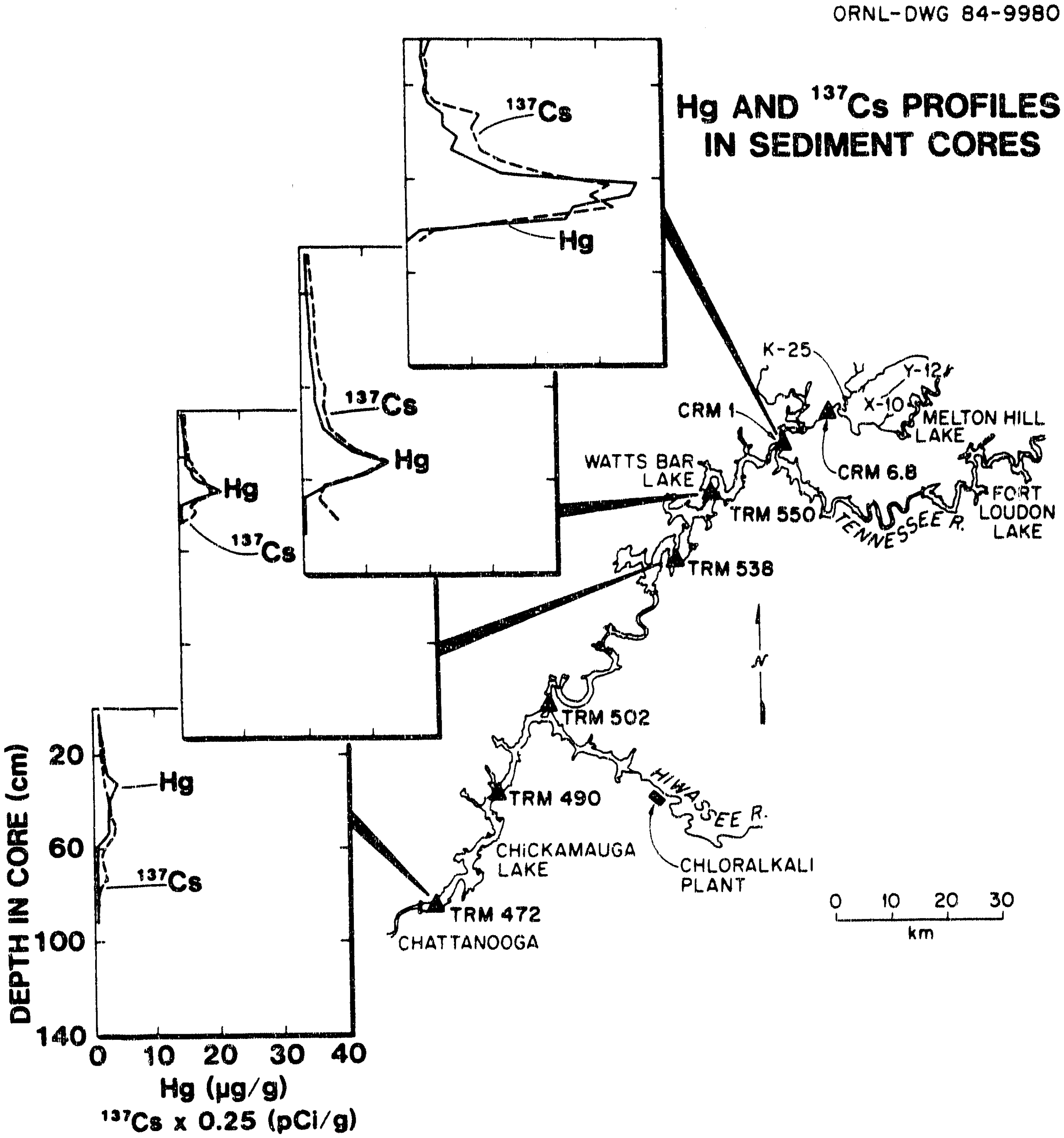

Fig. 4. Vertical profiles of ^{137}Cs and Hg for selected sediment cores collected in the Tennessee River Reservoir system (Turner et al. 1985).

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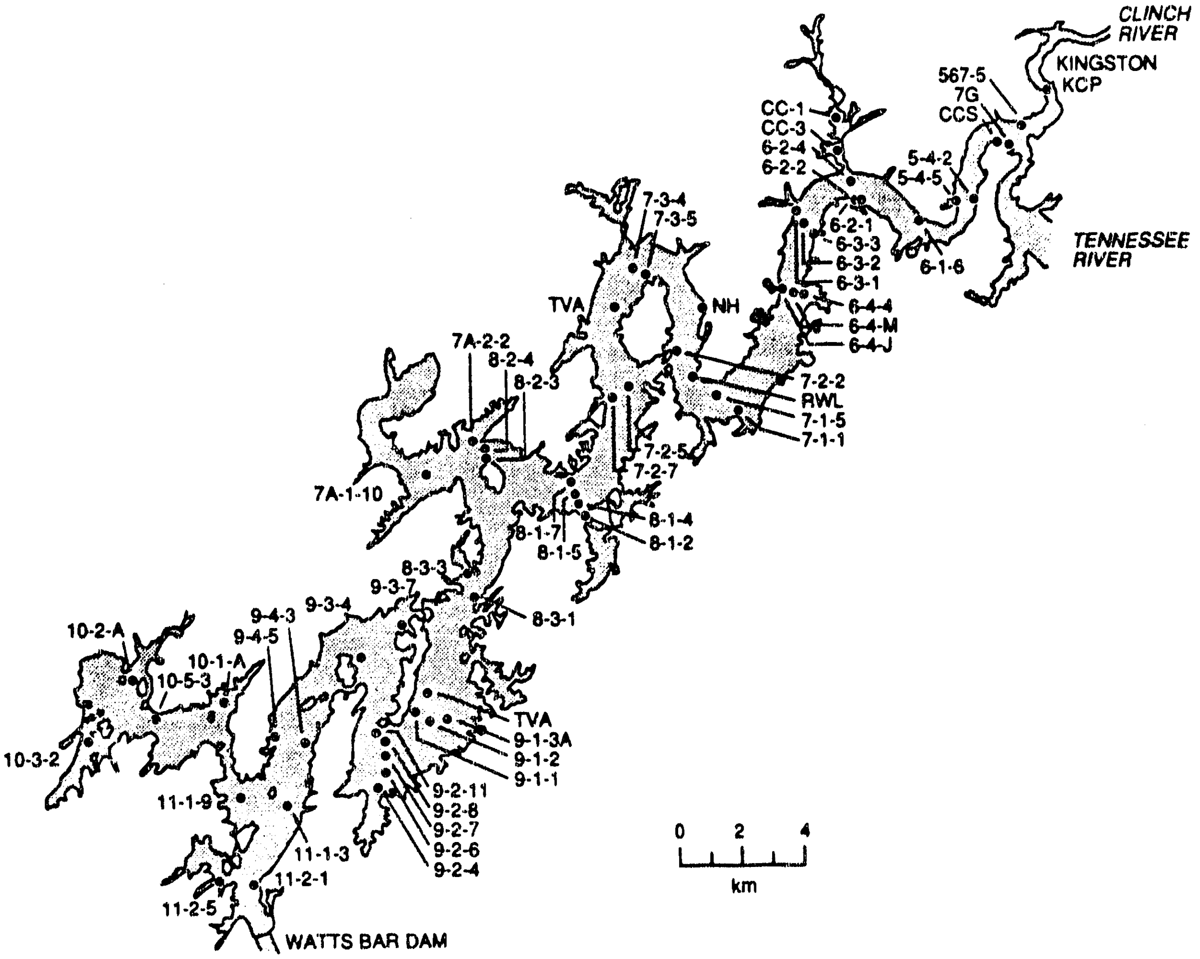

Fig. 5. Map of Watts Bar Reservoir indicating the locations and identifying the sediment cores collected in this study.

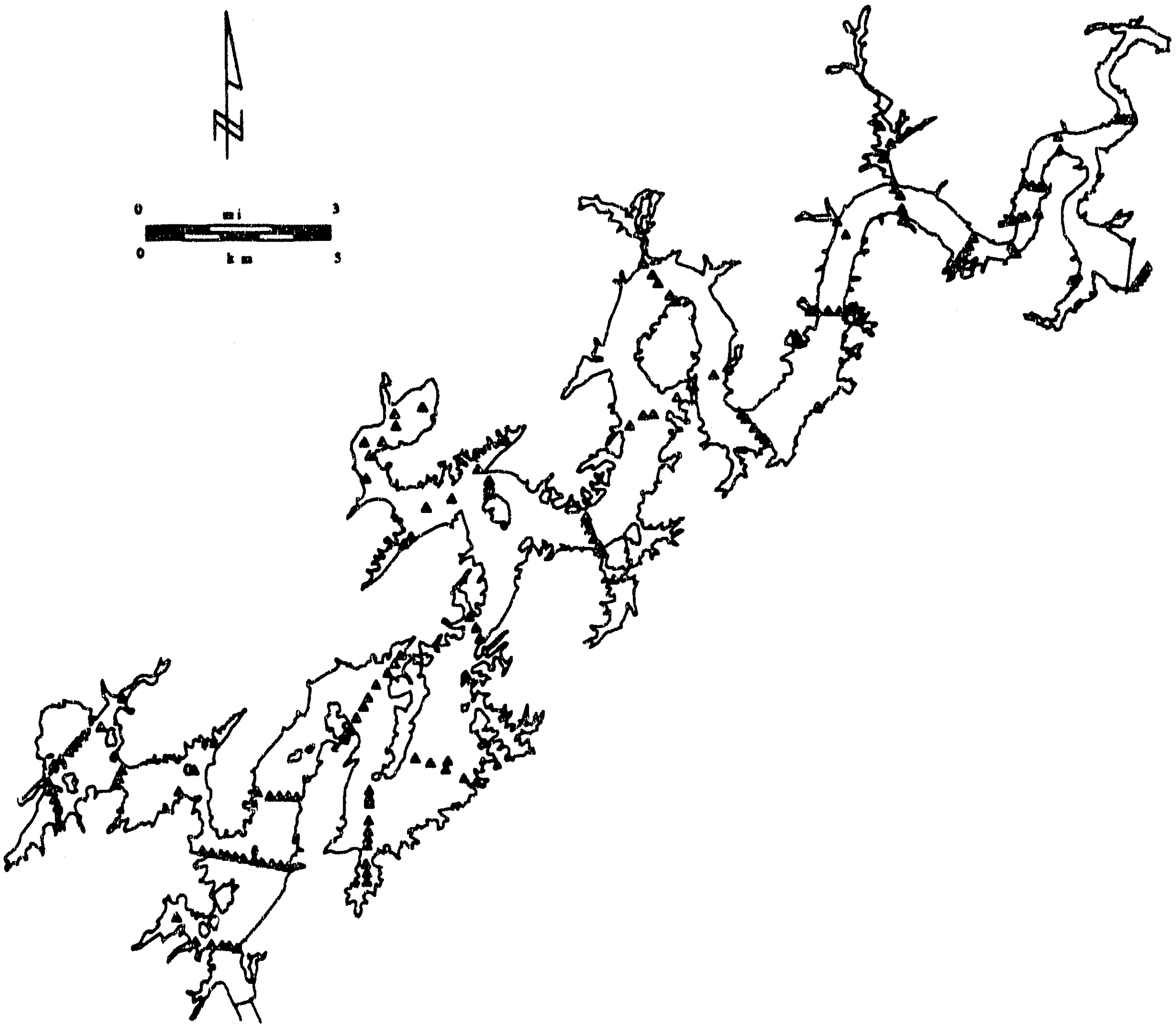

Fig. 6. Map of Watts Bar Reservoir indicating the locations of the surface grab samples collected in this study.

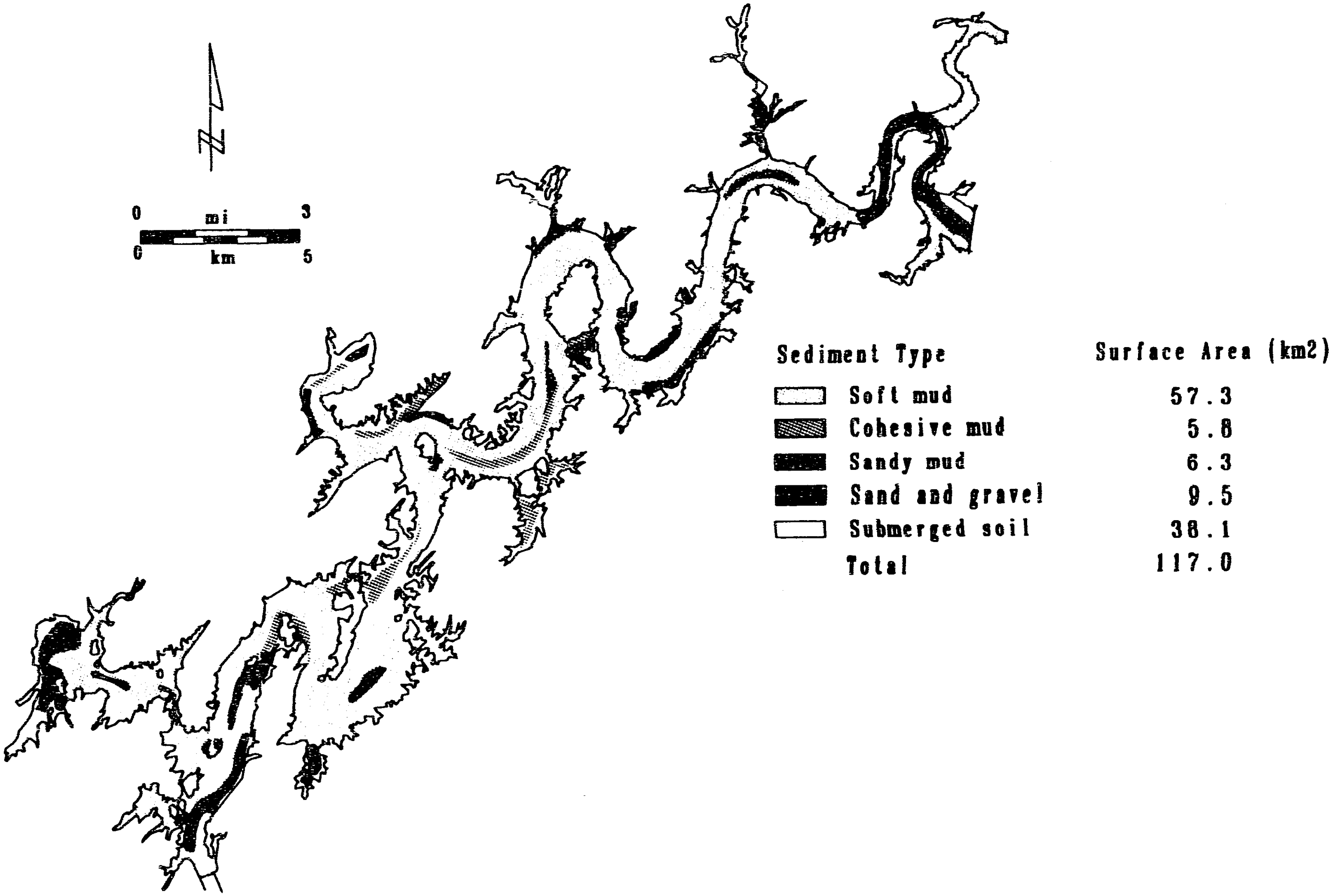

Fig. 7. Map of the surface sedimentary characteristics based on the visual textural analysis of more than 190 surface sediment samples collected in Watts Bar Reservoir.

MATERIALS AND METHODS

Sampling locations in the Clinch River and Tennessee River system were identified using TVA Navigation Charts with corresponding shore markers and/or channel buoys. In addition, prominent shore features (e.g., TVA power lines, bridges, and barge moorings) were logged for each sampling location. Surface sediment samples were collected using a Ponar bottom-grab sampler (17 × 10 × 7 cm). The retrieved surface grab samples were immediately placed into labeled Marinelli beakers, which were placed directly on gamma detectors for radionuclide (^{137}Cs) analysis. These samples were used to characterize sediment types and distributions (gravel, sand, mud, and soil detritus) and to develop a map of surfacial contaminant concentrations.

Two types of coring devices were also used to obtain sediment profiles: a gravity corer and a vibracorer. The free-fall gravity corer (Wildco KB) was equipped with a plastic liner that was 120 cm long and 4.7 cm in diameter. The corer was attached to a cable on a reel and allowed to free-fall during descent. This coring device was primarily used in areas where the reservoir water depth was greater than 10 m. Upon retrieval, the plastic liner containing sediment was capped and then removed from the core barrel. The core was extruded from the liner and sectioned into either 1-, 2-, or 4-cm depth increments. These sections were sealed into plastic-lined aluminum cans and returned to the laboratory for radionuclide analysis.

The vibracorer consists of a vibrating head attached to an aluminum irrigation pipe, typically 7.2 cm in diameter. The vibrating head allows for greater penetration of the core pipe by thixotropic action. Sediment penetration by vibracoring is usually much greater than that obtainable by gravity coring and also minimizes compression of the sediment during sampling. Because pipe lengths greater than the depth of the water column are required in this operation, vibracores were collected only in areas where the water depth was less than 10 m. After penetration into the sediment, the top of the core was plugged, and the entire core pipe was brought to the surface. The bottom was then plugged, and the excess core pipe was removed to facilitate handling and sediment extrusion. During extrusion the sediment core was sectioned into either 2- or 4-cm depth increments and sealed into labeled aluminum cans, as described previously.

SEDIMENT ANALYSES

Initially, the sediment samples collected in off-site areas were analyzed and screened for ^{137}Cs in our laboratory by gamma spectrometry. A few selected samples were also analyzed for ^{60}Co and naturally occurring ^{210}Pb, which was used to estimate sediment accumulation rates and to determine the age of the sediments.

The samples were radiochemically analyzed using germanium solid state detectors. A Nuclear Data 6700 microprocessor and later a Nuclear Data 9900 microprocessor acquisition system with spectra acquired in 4096 channels were used to record ^{137}Cs decays. Counting times for each sample ranged from 60 to 1000 min or longer, depending on the activity level present and the degree of precision desired.

Each detector used for the initial screening was calibrated for photon energy versus channel number using isotopes of known gamma-ray energy (e.g., ^{133}Ba, ^{137}Cs, and ^{60}Co). Efficiency calibrations for the various geometries were performed using National Bureau of Standards (NBS) reference sources (e.g., Amersham's QCY 46 mixed gamma solution). A description of the calibration procedures has been presented elsewhere (Larsen and Cutshall 1981). After counting, the sample was weighed, oven-air dried (60°C) for several days, and weighed again to determine both the wet and dry weight. The 60°C drying temperature was selected to prevent volatilization of other types of contaminants (e.g., Hg and PCBs.) Wet and dry weights of the samples were used to calculate porosity and activity concentrations.

Various techniques were used to provide quality assurance/quality control for the radionuclide measurements. Detector performance was evaluated weekly by counting a source of known activity and comparing the value obtained with the reported value. These values were then plotted on a control chart to keep a track record of detector performance. In addition, we routinely participated in the Quality Assurance/Quality Control Radioanalytical Program at the EPA Environmental Monitoring Systems Laboratory in Las Vegas, Nevada (EMSL-LV). Samples received from the program are routinely analyzed by gamma ray spectrometry as cross-checks or blind samples. Table 3 illustrates the performance for each of our three detectors (A, B, and C) in the April 20, 1987, laboratory intercomparison. In addition, certified reference materials from the NBS were also analyzed. Table 4 illustrates the analysis of standard reference material (SRM) NBS SRM 4353 Rocky Flats Soil contained in our aluminum can geometry.

Table 3. EPA EMSL-LV Intercomparison Study, April 1987, Marinelli beaker geometry
(pCi/L ± 1σ)

	Environmental Sciences Division detector				EPA standard value	Mean for participating labs
	A	B	C	Mean		
^{60}Co	8.0	8.0	7.0	7.7 ± 0.6	8.0 ± 5.0	9.0 ± 2.0
^{134}Cs	18.0	18.0	16.0	17.3 ± 1.2	20.0 ± 5.0	18.2 ± 2.6
^{137}Cs	15.0	15.0	14.0	14.7 ± 0.6	15.0 ± 5.0	15.7 ± 2.2

After initial screening by gamma spectrometry, selected samples (hermetically sealed in plastic-lined aluminum cans) were sent, through an appropriate chain of custody, to the ORNL Analytical Chemistry Division (ACD) for analysis of total Hg using ACD Preparation Method 10915 and ACD Analytical Method 1214922. In addition to routine quality assurance/quality control procedures used by ACD, an SRM (NBS SRM 1646, Estuarine Sediment) was analyzed with one batch of core samples. The results (0.065 and 0.067 μg/g, respectively, for the two duplicates) were in good agreement with the certified value (0.063 ± 0.012 μg/g) for this material.

Table 4. National Bureau of Standards SRM 4353 Rocky Flats soil, aluminum can geometry[a]
(pCi/g ± 1σ)

	Environmental Sciences Division detector				NBS standard value
	A	B	C	Mean	
^{137}Cs	0.46 ± 0.03	0.45 ± 0.05	0.49 ± 0.04	0.47 ± 0.02	0.48 ± 0.01
^{40}K	18.8 ± 0.4	18.6 ± 0.5	19.4 ± 0.6	18.9 ± 0.6	19.5 ± 0.6

[a] Decay corrected to Dec. 15, 1980.

The vertical distribution of ^{90}Sr was also measured in one of the sediment cores (core 567.5 in Fig. 5) collected at the mouth of the Clinch River near Kingston. The samples were radiochemically analyzed following the procedures established at the DOE Environmental Measurements Laboratory (E-SR-01). A ^{85}Sr tracer was added to each sediment sample for yield determinations. The ^{90}Sr activity was measured with a low-background gas-flow proportional beta counter, and accuracy was assessed with a ^{90}Sr standard supplied by EPA.

WATER ANALYSES

To quantity the distribution of ^{137}Cs and Hg between dissolved and particulate phases, several large-volume (400- to 800-L) water samples were collected at various locations in Watts Bar Reservoir. Suspended particles (>0.45 μm) were removed from these large-volume samples by continuous-flow centrifugation. The suspended matter was dried, weighed, and analyzed for ^{137}Cs by the procedures described above and analyzed for total Hg by the ACD.

After centrifugation, each large-volume water sample was acidified with HCl to a pH of approximately 2, and stable Cs and Fe (and occasionally Pb, Co, Be, and ^{242}Pu) were added as carriers and yield tracers. The yield tracers were allowed to equilibrate for 6 to 8 h, and dissolved radiocesium was removed from the large-volume water sample by sorption on a cation-exchange resin (ammonium molybdophosphate). The resin was added on the same day of sample collection and was allowed to settle out of the sample overnight. For selected samples the water was then transferred to another 1000-L tank, and the pH was adjusted to about 10 with NaOH to allow the iron to precipitate. Dissolved ^{60}Co, ^{7}Be, ^{210}Pb, and plutonium isotopes were removed from these samples by coprecipitation with or sorption on the $Fe(OH)_3$ precipitate. Quantification of the yield tracers by atomic absorption spectrometry indicated that 70 to 100% of the Cs, Be, Pb, and Co could be recovered by these procedures. The radionuclide activities for each sample were yield corrected according to the actual recovery.

The dissolved and particulate plutonium analyses were conducted by M. Thein (ORNL Environmental Compliance and Health Protection Division). These analyses involved dissolution with HCl, coprecipitation with calcium oxalate, radiochemical separation with ion-exchange columns, electrodeposition onto stainless steel disks, and alpha spectrometry with silicon surface-barrier detectors. The samples were alpha counted for about 21 d, and yields were evaluated using a ^{242}Pu tracer. Plutonium-239 and -240 activities are collectively reported because the energies of the alpha particles produced by the decay of ^{240}Pu (6580-year half-life) cannot be resolved from those produced by the decay of ^{239}Pu (24,400-year half-life) by alpha spectrometry.

Two 500-mL water samples were collected at the same time and at two locations in Watts Bar Reservoir to measure the concentration of dissolved Hg in the water column. These samples were filtered through 0.2-μm filters, and the filtrates were analyzed for total Hg by ACD.

RESULTS AND DISCUSSION

Some contaminants and radionuclides (such as ^{3}H, ^{90}Sr, and ^{131}I) are relatively soluble in freshwater systems, and consequently their transport and biogeochemical fate are mediated by water movements and biological uptake from the water phase. Most contaminants (e.g., Hg, ^{137}Cs, ^{60}Co, and ^{239,240}Pu), however, are chemically and biologically reactive and rapidly become associated with particles in freshwater systems. Consequently, the transport and biogeochemical fate of these contaminants are primarily governed by particle dynamics.

The tendency for a contaminant to become associated with particles in aquatic systems is expressed quantitatively by the distribution coefficient (K_d), defined as

$$K_d = \frac{C_p}{C_w} ,$$

where C_p is the concentration of a specific contaminant associated with a given weight of particles (μg/g) and C_w is the concentration of the contaminant in an equal weight of water (μg/mL). Ideally, this ratio is a measure of the reversible equilibrium partitioning of a contaminant between dissolved and particulate phases and would be a constant. Because most natural environments (including Watts Bar Reservoir) are affected by short-term physical, chemical, and biological processes, chemical equilibrium is continually adjusting and rarely attained.

CONTAMINANTS IN THE WATER COLUMN

Dissolved and particulate concentrations for Hg, ^{137}Cs, ^{60}Co, ^{7}Be, ^{239,240}Pu, and ^{238}Pu in the water column of Watts Bar Reservoir and their calculated particle-to-water distribution coefficients are listed in Table 5. These ^{137}Cs, ^{60}Co, ^{239,240}Pu, and Hg concentrations are comparable to values found in previous studies in the Clinch River. Hoffman et al. (1991) have shown these concentrations to pose no imminent risk to human health, especially if the deep sediment is not subjected to dredging. The particle-to-water distribution coefficients for both ^{137}Cs and Hg range between 1×10^5 to 5×10^5 (Table 5). This indicates that both contaminants are particle-reactive and that the dissolved concentrations of ^{137}Cs and Hg are about 10,000 times lower than the concentration on suspended particles and surface sediments.

The data presented for the large-volume water samples collected at the mouth of the Clinch River on December 1, 5, and 17, 1986 (Table 5), were obtained to (1) provide information on the partitioning of these contaminants between dissolved and particulate phases and (2) determine whether the abnormally high concentrations of ^{60}Co that were measured by others in WOL on November 25-26, 1986, could be traced into Watts Bar Reservoir.

On December 1, the dissolved concentration of ^{60}Co was 0.024 pCi/L (0.9 mBq/L) near Kingston City Park, and the concentration of ^{60}Co on the particulate matter was 1.1 pCi/g (41 mBq/g) (Table 5). Because the ^{60}Co concentration on bottom sediments in this area ranges from about 0.8 to 1.2 pCi/g (30 to 45 mBq/g), the ^{60}Co concentration measured on the particulate matter (1.1 pCi/g) is typical for resuspended bottom sediments and does not reflect

Table 5. Contaminant distributions between aqueous and particulate phases

Date	Nuclide	Suspended load (mg/L)	Dissolved (fCi/L)	Particulate (pCi/g)	Distribution[a] K_d
		City of Kingston—Mouth of the Clinch River			
12/1/86	^{60}Co	14	24	1.1	5×10^4
	^{137}Cs		35	6.8	2×10^5
	^{7}Be		92	5.4	6×10^4
	^{239,240}Pu		0.38	0.04	1×10^5
	^{238}Pu		0.11	0.005	5×10^4
12/5/86	^{60}Co	11	250	4.3	2×10^4
	^{137}Cs		49	14.6	3×10^5
	^{7}Be		65	5.3	8×10^4
	Hg (ppb)[b]		0.005	2360	5×10^5
12/17/86	^{60}Co	7	215	7.3	3×10^4
	^{137}Cs		103	26.5	3×10^5
	^{7}Be		76	8.0	1×10^5
		Thief Neck—Watts Bar Reservoir			
12/22/86	^{60}Cs	7	12	0.9	8×10^4
	^{137}Cs		17	5.1	3×10^5
	^{7}Be		78	8.7	1×10^5
		Mouth of White Creek—Watts Bar Reservoir			
3/9/89	Hg (ppb)[b]	21	0.004	510	1×10^5

[a] Particle-to-water distribution

$$K_d = \frac{\text{concentration per kilogram of particles}}{\text{concentration per liter of water}}.$$

[b] ppb = parts per billion or μg/L.

any recent additional inputs. Likewise, the concentrations of ^{137}Cs, Hg, and plutonium in the water and on the particulate matter were not abnormally higher than the values expected from the resuspension of river-reservoir sediments, primary productivity, and equilibrium particle-to-water distributions.

On December 5 (about 10 d after the ^{60}Co release was observed in WOL), the dissolved concentration of ^{60}Co increased by an order of magnitude and particulate concentrations of ^{60}Co increased by a factor of 4 (Table 5). On December 17, the dissolved concentration of ^{60}Co began to decrease but particulate ^{60}Co concentrations continued to increase by another factor of 2 (Table 5). These data indicate that it takes about 2 to 3 weeks before the ^{60}Co released into White Oak Creek is transported via the Clinch River into Watts Bar Reservoir. In addition, the time-delayed increase in the particulate ^{60}Co and ^{137}Cs concentrations at Kingston (Table 5) imply that particle deposition and resuspension processes cause the maximum water column concentration of particle-reactive radionuclides to occur at Kingston about 1 month after release into WOL. This delay may be even longer during the summer and fall, when rainstorm resuspension events occur less frequently.

Comparison of the suspended-particulate ^{137}Cs and Hg concentration data measured at the mouth of the Clinch River with respective data collected at Thief Neck and White Creek (Table 5) indicate that the concentrations for both of these contaminants are reduced by a factor of about 3. This decrease is also apparent in the ^{137}Cs and Hg concentrations in surface sediments (Appendix A). Concentrations of ^{137}Cs in soft-mud surface sediments at the mouth of the Clinch River average about 7.0 pCi/g (260 mBq/g), whereas ^{137}Cs concentrations in soft-mud surface sediments below the confluence of the Tennessee River average about 2.5 pCi/g (90 mBq/g). This trend probably reflects the dilution of Clinch River particulate material with particles from other sources (primarily the Tennessee River).

Finally, it should be noted that concentrations of ^{239,240}Pu and ^{238}Pu in the water column of Watts Bar Reservoir near Kingston are also reported in Table 5. These dissolved and particulate plutonium concentrations are about a factor of 5 higher than respective concentrations that have been measured by us and others in other river-reservoir systems along the east coast of the United States, including the Savannah River downstream from the DOE Savannah River Plant (Olsen et al. 1989b). In addition, the ratio of ^{238}Pu to ^{239,240}Pu on the suspended particles in Watts Bar Reservoir (0.13) is about a factor of 3 higher than the ^{238}Pu to ^{239,240}Pu ratio in global fallout (0.045) delivered to mid-latitude areas of the Northern Hemisphere. This suggests that some of the plutonium disposed of or stored on the ORR is being transported into off-site areas. Preliminary screening assessments, however, indicate that the measured concentrations of ^{239,240}Pb and ^{238}Pu in water and sediment are far below any established level of risk that would be of concern for the protection of human health (Hoffman et al. 1991).

CONTAMINANTS IN BOTTOM SEDIMENTS

Sorption onto suspended particles and sediment deposition are the principal mechanisms by which many chemically reactive contaminants (such as ^{137}Cs and Hg) are removed from the water column and accumulated in the bottom sediments. Although burial in sediments helps to isolate these contaminants from human and biotic contact, contaminant burial may be disturbed in some areas by sediment resuspension, sediment mixing, or diagenetic remobilization processes. Unfortunately, however, quantitative measurements of the extent of removal, burial, and remobilization in any field system are extremely difficult because of the complex physical, chemical, and biological interactions that affect contaminant fate and their extreme variability with space and time. One of the tools available for tracing and quantifying these interactions is the distribution of a radionuclide with a known source and history of input into the system. In this scoping study, we have used the distribution of ^{137}Cs as a cost-effective tracer to identify where sediments and particle-reactive contaminants are accumulating in the Clinch River and Watts Bar Reservoir system. This approach will allow for more-efficient characterization of contamination in off-site areas and, therefore, reductions in time and costs.

RADIONUCLIDE DISTRIBUTIONS

The distribution of ^{137}Cs concentrations in the surface sediments of Watts Bar Reservoir is illustrated in Fig. 8. A comparison of this figure with the map of sedimentary characteristics (Fig. 7) indicates that the concentrations of ^{137}Cs are highest in the soft-mud areas and lowest in the sand/gravel and submerged soil areas of the reservoir.

The vertical distribution of ^{137}Cs in Watts Bar Reservoir sediment cores is strongly correlated with the historical record of ^{137}Cs discharges from WOL, exhibiting a large subsurface peak coincident with the draining of WOL in the mid-1950s (Fig. 9). The depth of this subsurface peak and the thickness of ^{137}Cs-contaminated sediment vary with the rate of sediment accumulation. In areas of rapid sediment accumulation, such as in the upper portion of the reservoir (core 567.5 in Fig. 9) and along the old river channel (cores 6-2-1, 8-1-4, and 9-4-3 in Fig. 9), the highest ^{137}Cs concentrations occur at sediment depths as great as 80 cm below the surface. In areas of slower sediment accumulation, such as along the reservoir margins (cores KCP, 8-2-3 and 11-2-1 in Fig. 9), the highest ^{137}Cs concentrations can often occur much nearer the sediment surface. Hoffman et al. (1991) evaluated the risk to human health of ^{137}Cs concentrations of this magnitude and found that, as long as the sediments remained in place, there was no imminent risk to human health. Further study, however, is required to determine the need for remediation.

To document the fact that the ^{137}Cs peak in Watts Bar Reservoir sediments actually reflects the draining of WOL in the mid-1950s, the rate of sediment accumulation (and thus the age of the sediment at various depths) was independently determined using the ^{210}Pb chronological technique. Lead-210, a naturally occurring radionuclide that has a 22-year half-life, has been extensively used for dating sediment and soil samples deposited during the past 100 years. Although ^{210}Pb is produced in sediments from the decay of ^{226}Ra, much of the ^{210}Pb in surface sediments is a result of its removal from the atmosphere via precipitation scavenging and washout. By measuring ^{210}Pb and ^{226}Ra profiles in sediment cores, chronological information can be obtained from the decline (by radioactive decay) of atmospherically derived ^{210}Pb (termed excess ^{210}Pb) in the sediment.

Vertical profiles of ^{137}Cs, ^{60}Co, and excess ^{210}Pb with sediment depth are illustrated for core 567.5 in Fig. 10. The slope of the line through the data for excess ^{210}Pb indicates a sediment accumulation rate of about 2.7 cm/year.

Because this core was collected in August 1986, such a sedimentation rate implies that the 80- to 84-cm depth increment and ^{137}Cs peak were deposited around 1955–1956, which is coincident with the draining of WOL. These data also imply that particle-associated

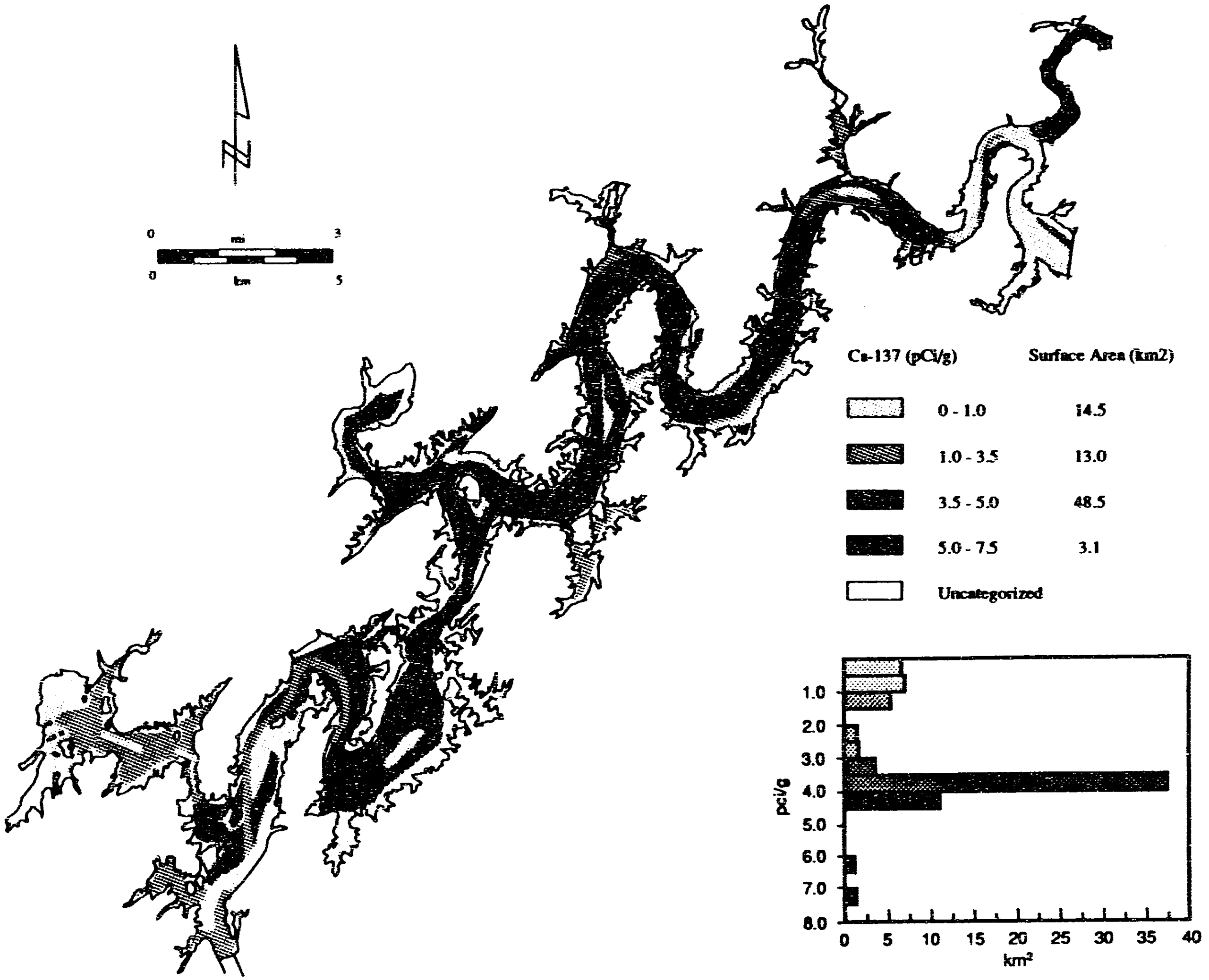

Fig. 8. Map of Watts Bar Reservoir illustrating the average ^{137}Cs concentration (pCi/g) in the top 16 cm of sediment.

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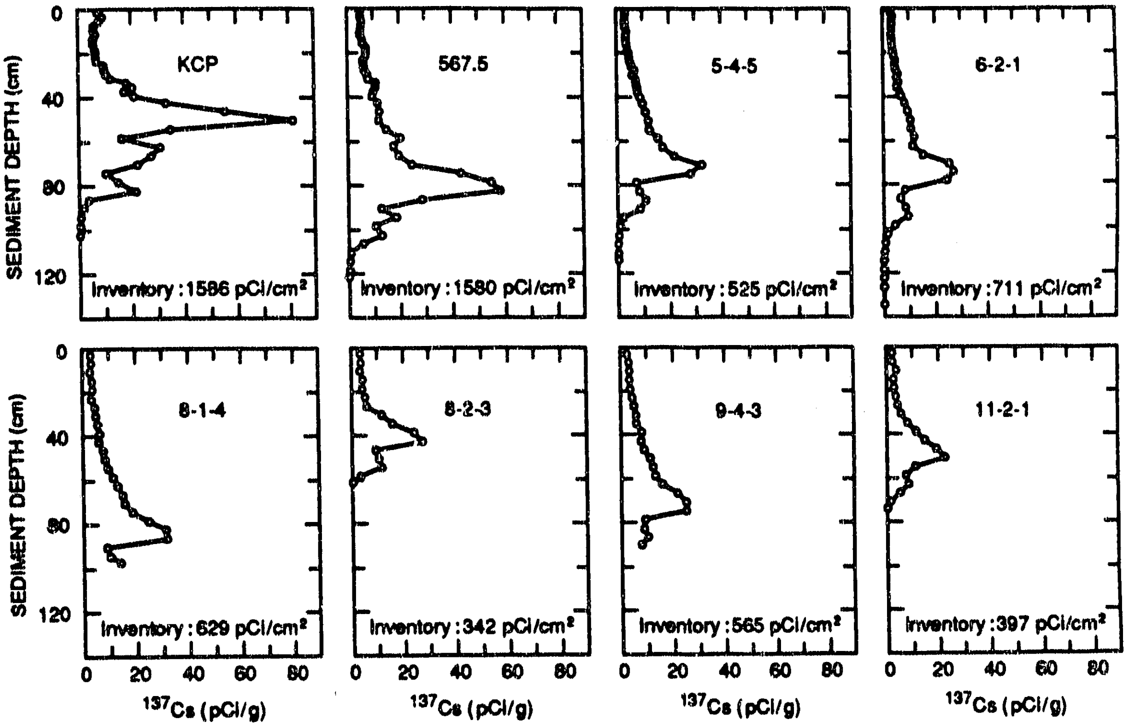

Fig. 9. Vertical distribution of ^{137}Cs with sediment depth in cores KCP, 567.5, 5-4-5, 6-2-1, 8-1-4, 8-2-3, 9-4-3, and 11-2-1. Cores KCP and 567.7 were collected near the mouth of the Clinch River, and the others were collected in a downstream sequence to Watts Bar Dam (see Fig. 5 for core locations).

radionuclides released from WOL are transported to and deposited in Watts Bar Reservoir within a year after their discharge. Because of the short half-life of ^{60}Co (about 5 years), its vertical profile in the sediments is different from that of ^{137}Cs. Most of the ^{60}Co deposited with sediments in 1955–1956 has decayed, and consequently ^{60}Co concentrations are highest in the recently deposited surface sediments.

The vertical distribution of ^{90}Sr in sediment core 567.5 is illustrated in Fig. 11. This ^{90}Sr profile shows a peak concentration of 580 pCi/kg (22 Bq/kg) at a sediment depth of 80 to 84 cm and a secondary peak [325 pCi/kg (12 Bq/kg)] at 36 to 40 cm. A comparison of this profile with the vertical distribution of ^{137}Cs (Fig. 10) indicates that the ^{90}Sr peak at about 80 to 84 cm is coincident with a peak in the ^{137}Cs concentration but that the secondary ^{90}Sr peak at about 40 cm occurs at a sediment depth where the ^{137}Cs profile is relatively uniform. A sediment accumulation rate of 2.7 cm/year (Fig. 10) suggests that another substantial release of ^{90}Sr occurred on the ORR during 1972–1973.

The total amount of ^{137}Cs that has accumulated at the core 567.5 site can be estimated by summing the vertical distribution of ^{137}Cs over the diameter of the sediment core. This calculation indicates that about 1580 pCi/cm², or 15,800 mCi/km², has accumulated at the mouth of the Clinch River into Watts Bar Reservoir. This inventory is about 165 times

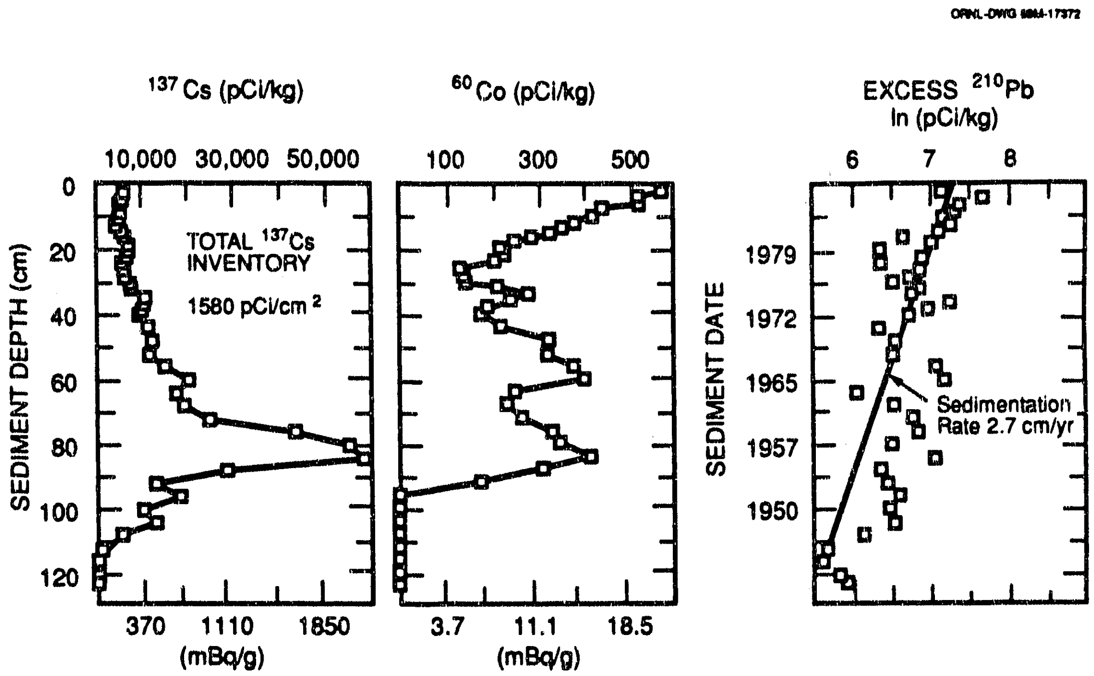

Fig. 10. Vertical profiles for ^{137}Cs, ^{60}Co, and excess ^{210}Pb in sediment core 567.5. The line drawn through the ^{210}Pb data has a slope corresponding to the sedimentation rate determined by the ^{137}Cs peak.

greater than the ^{137}Cs inventory expected from global fallout (about 95 mCi/km^2) in association with the atmospheric testing of nuclear weapons in the mid-1960s.

For comparison, the vertical profile and inventory of ^{137}Cs in a sediment core collected from Norris Reservoir are presented in Fig. 12 Norris is the reservoir farthest upstream on the Clinch River and drains a relatively pristine area of the Cumberland Mountains. The rate of sediment accumulation (as determined from the excess ^{210}Pb profile for this core) is 1.8 cm/year. Although the vertical ^{137}Cs profile in this core also exhibits a peak, it occurs at a sediment depth that corresponds to the 1962-1964 maximum in fallout ^{137}Cs delivery (Olsen et al. 1989a). The total inventory of ^{137}Cs in this core is about 49 pCi/cm^2, or 490 mCi/km^2, which is about five times greater than the inventory expected from global fallout. The inventory of excess ^{210}Pb in this Norris sediment core is also about five times greater than the level expected from its atmospheric flux (Olsen et al. 1989a). Consequently, it is suspected that sediments eroded from other areas of the upstream Clinch River (which contain fallout ^{137}Cs and excess ^{210}Pb) are being focused during accumulation at this site. Even with sediment focusing, it is apparent that the inventory of ^{137}Cs in Watts Bar Reservoir is at least 30 times greater than the inventory expected from atmospheric fallout.

The total burden of ^{137}Cs in Watts Bar Reservoir sediments was estimated by measuring the inventory of ^{137}Cs in each sediment core (Appendix A) and integrating these data over the entire reservoir using the ARC:INFO Geographic Information System (GIS). Logistically,

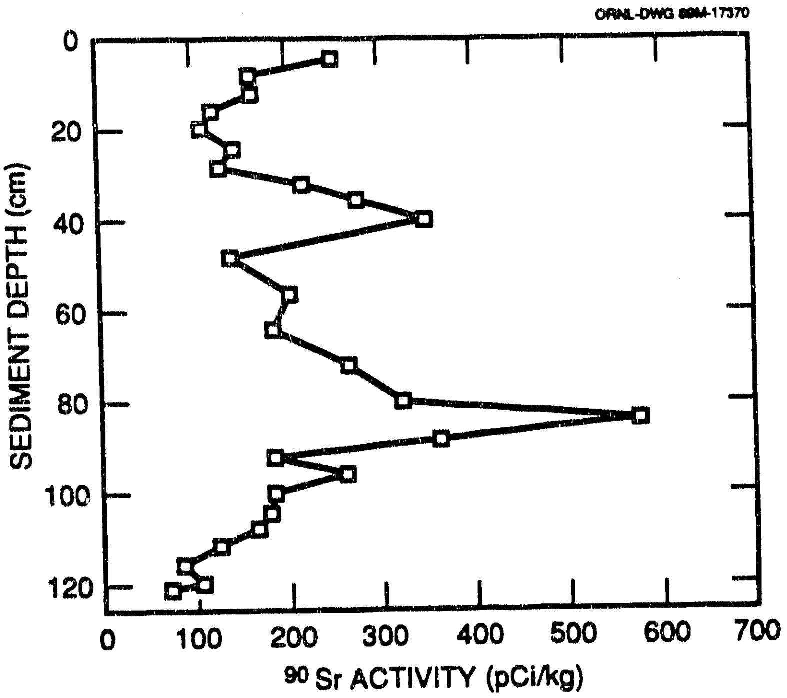

Fig. 11. Vertical distribution of ^{90}Sr in sediment core 567.5.

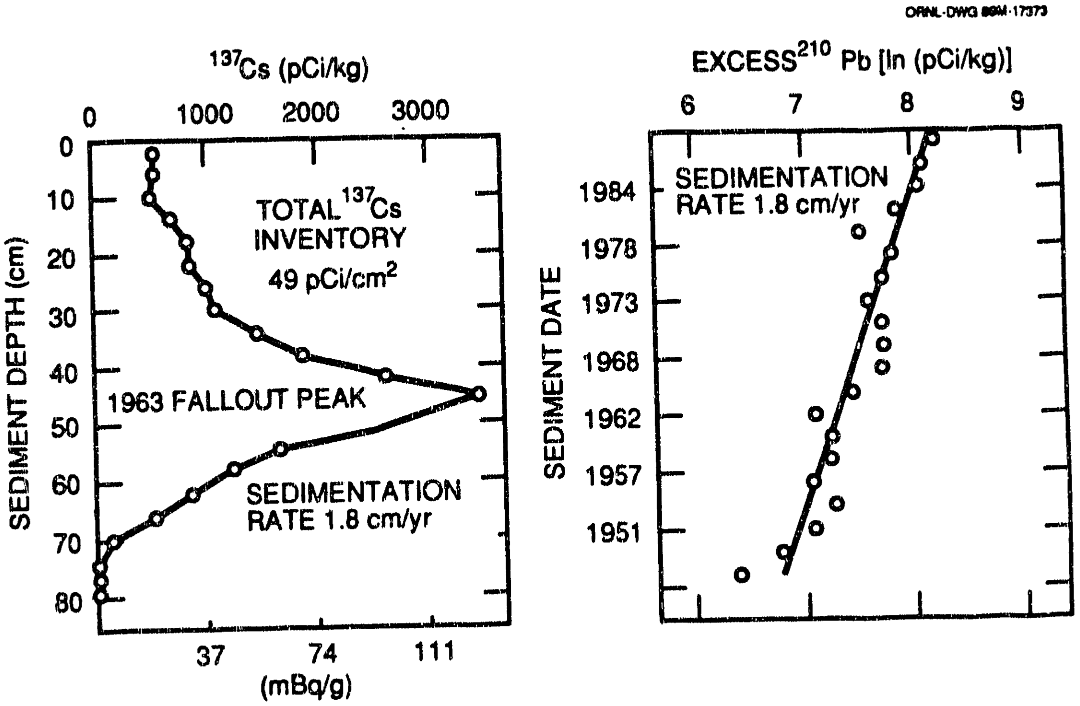

Fig. 12. Vertical profiles for ^{137}Cs and excess ^{210}Pb in the sediments of Norris Reservoir.

the reservoir surface area (from the city of Kingston to Watts Bar Dam) was subdivided into polygons (Fig. 13) on the basis of (1) proximity to the mouth of the Clinch River, (2) sedimentary characteristics, and (3) the ^{137}Cs concentration in surface-sediment samples (Appendix A). A ^{137}Cs inventory was then calculated for each polygonal area (Fig. 14). In areas where no sediment cores were collected, the ^{137}Cs inventory was estimated as an average calculated from the inventories for cores collected within the same vicinity and with the same sedimentary characteristics.

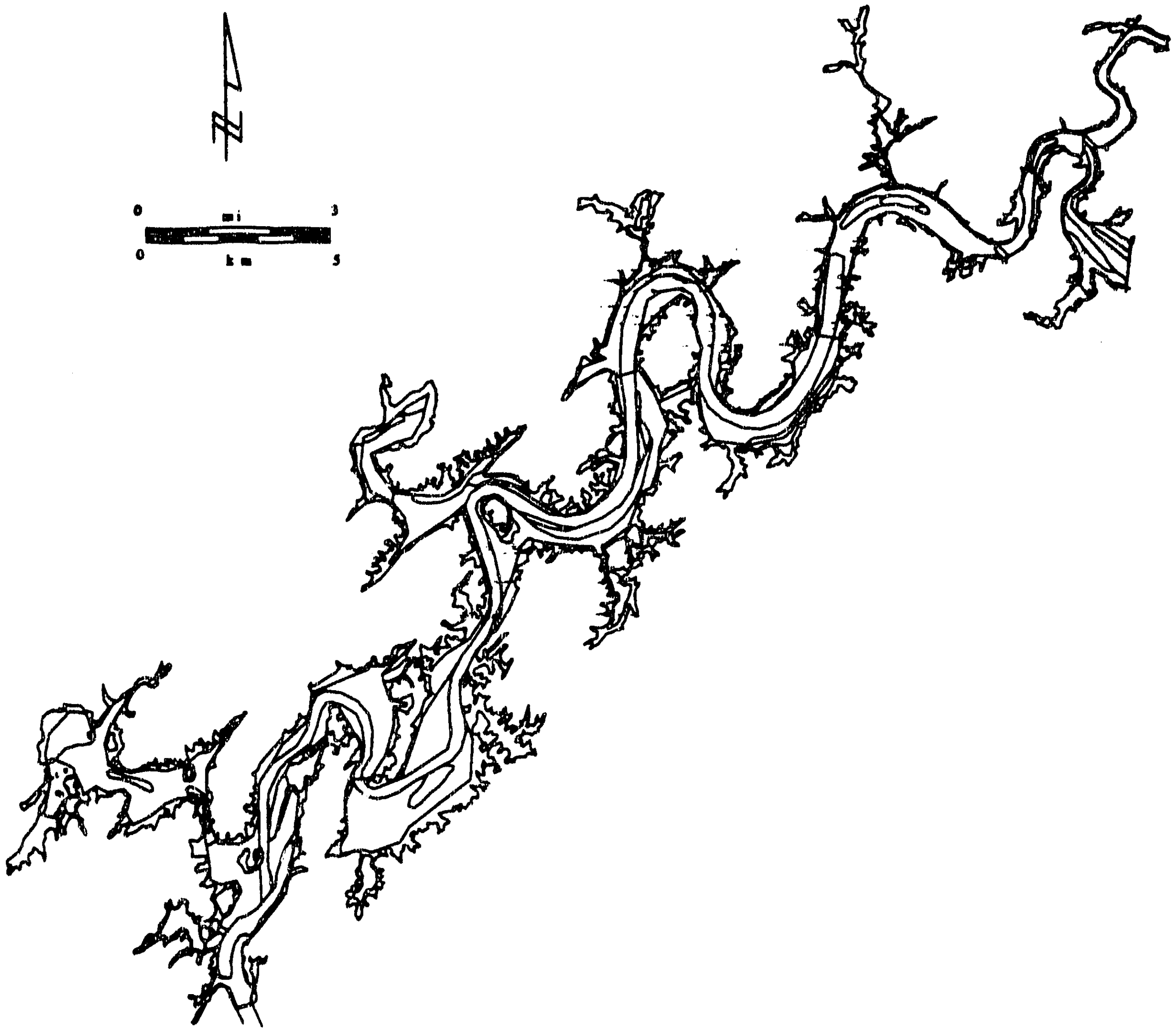

Fig. 13. Map of Watts Bar Reservoir illustrating the GIS polygonal areas that were used for integrating the sediment core data over the entire reservoir.

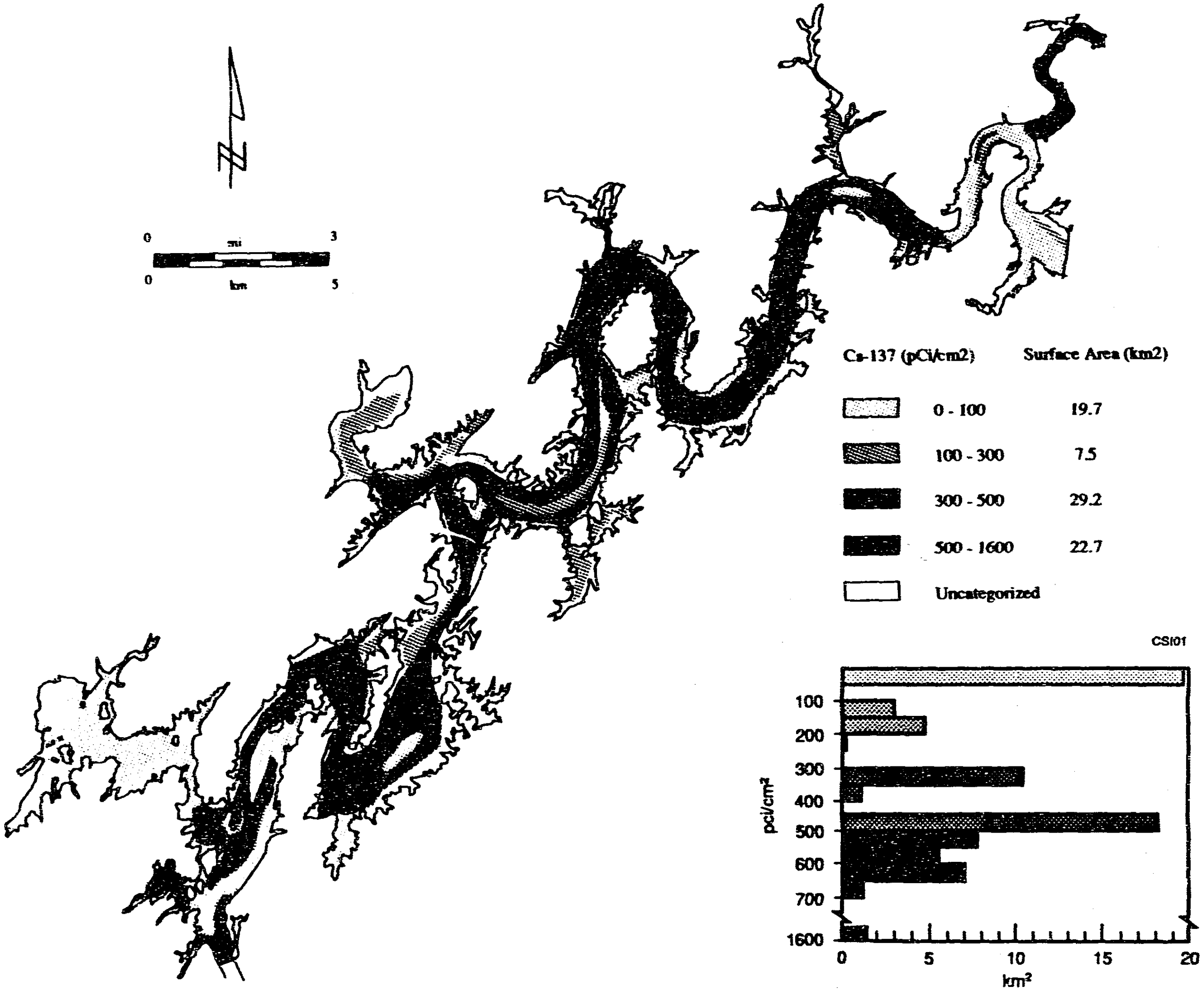

Fig. 14. Map of Watts Bar Reservoir illustrating ^{137}Cs accumulation patterns and total inventories (pCi/cm^2).

The three main points illustrated in Fig. 14 are that

1. the highest ^{137}Cs inventories occur along the old Clinch River and Tennessee River channels (deepest portions of the reservoir), where the impoundment of water has reduced currents and induced rapid accumulation of sediment and particle-associated contaminants;

2. scouring and little or no ^{137}Cs accumulation is occurring in areas where river currents are still strong and relatively unaffected by Watts Bar Dam; and

3. the sediments in marginal coves appear to contain relatively minor amounts of ^{137}Cs accumulation, suggesting local sediment sources rather than particles derived from the Clinch or Tennessee rivers.

Although total ^{137}Cs inventories are greatest along the impounded river channel, ^{137}Cs concentrations and inventories in near-surface (0- to 50-cm and 0- to 16-cm) sediments appear to be highest along the shallower channel margins (Figs. 15 and 16). This is because peak ^{137}Cs concentrations occur closer to the sediment surface in areas affected by relatively lower rates of sediment accumulation (Figs. 9 and 15). This has important environmental and ecological implications because (1) epibenthic fauna are generally confined to the top 16 cm of sediment (biologically active layer) and (2) game fish are often caught in shallower marginal habitats. The risk to human health of these peak ^{137}Cs concentrations was preliminarily evaluated by Hoffman et al. (1991), who found that there was no imminent risk to human health, as long as the sediments remained in the reservoir without being disturbed.

The total ^{137}Cs inventory calculated from the sum of the polygons was 304 Ci (1.12×10^{13} Bq). Because a decay-corrected total of 335 Ci of ^{137}Cs has been released into the Clinch River via WOL between 1949 and 1986, it appears that about 91% of the ^{137}Cs released from WOD has been trapped within the sediments of Watts Bar Reservoir. The remaining 9% may reflect error in the estimate, unmonitored discharges before 1949, or uncertainty in the ^{137}Cs release history, or it may indicate the relative amount of ^{137}Cs retained in floodplain areas or transported downstream past Watts Bar Dam.

MERCURY AND ^{137}Cs CORRELATIONS

A comparison of the aquatic discharge histories of ^{137}Cs from ORNL's WOL and inorganic Hg from the Y-12 Plant (Fig. 2) shows the near coincidence of the peak release of ^{137}Cs in 1956 with that of Hg in 1957–1958 and sharp declines in both releases after 1959. Because of this near coincidence in peak release years and because of the high chemical affinity of both Hg and ^{137}Cs for particulate matter, the vertical profiles of these two contaminants in sediment cores collected throughout Watts Bar Reservoir were strongly correlated (Fig. 17).

This relationship is illustrated in Fig. 18, and the resultant coefficient of determination (r^2) is 0.87. This r^2 value indicates that 87% of the variation in the mercury data can be accounted for by the ^{137}Cs data via a linear-regression model. The corresponding correlation coefficient (r) for the Hg-^{137}Cs relationship is 0.93. The amount of Hg in a sediment sample from Watts Bar Reservoir can be estimated fairly accurately by multiplying the ^{137}Cs inventory by 0.25 (Fig. 18).

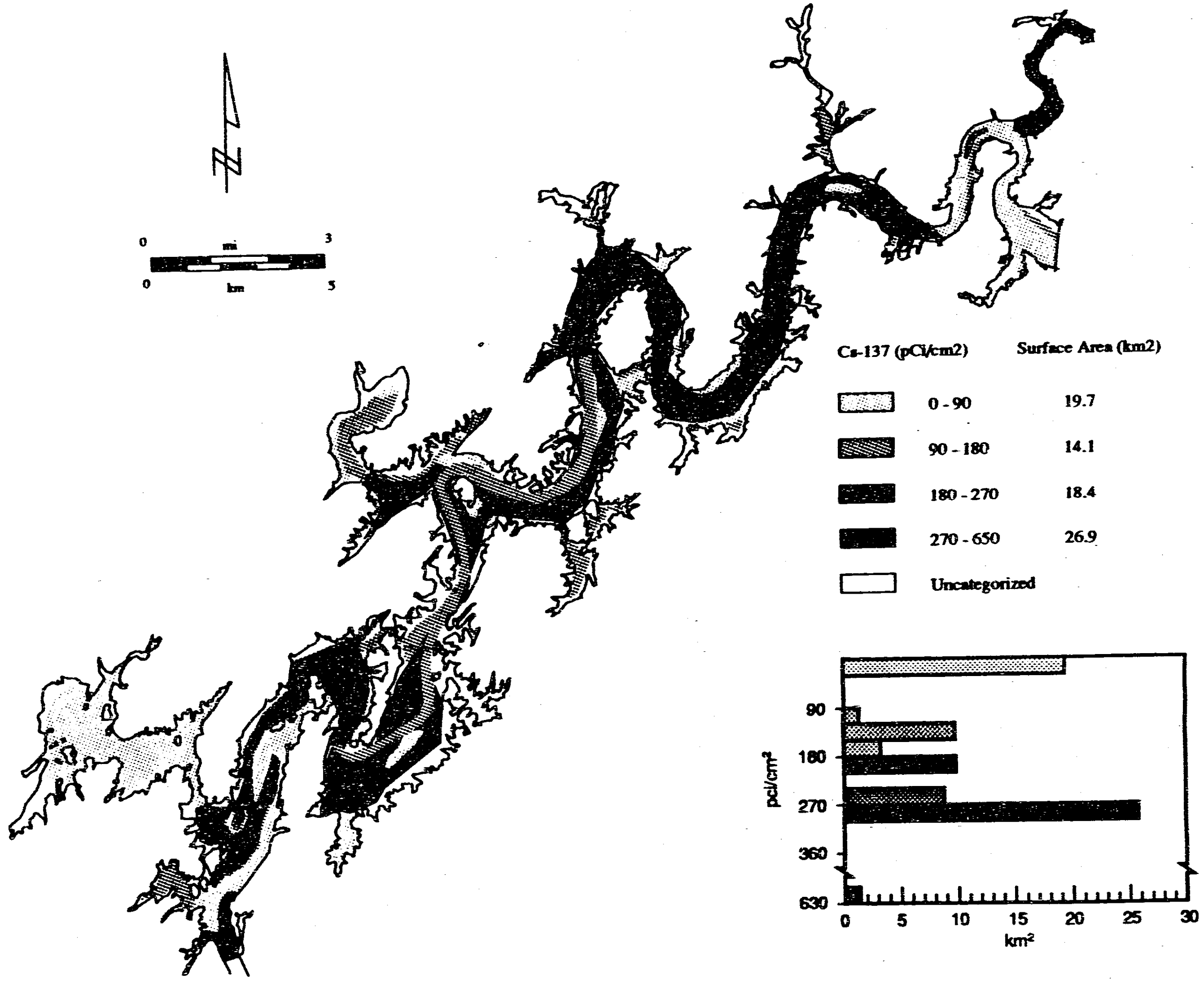

Fig. 15. Map of Watts Bar Reservoir illustrating the inventory (pCi/cm^2) of ^{137}Cs in the top 50 cm of sediment.

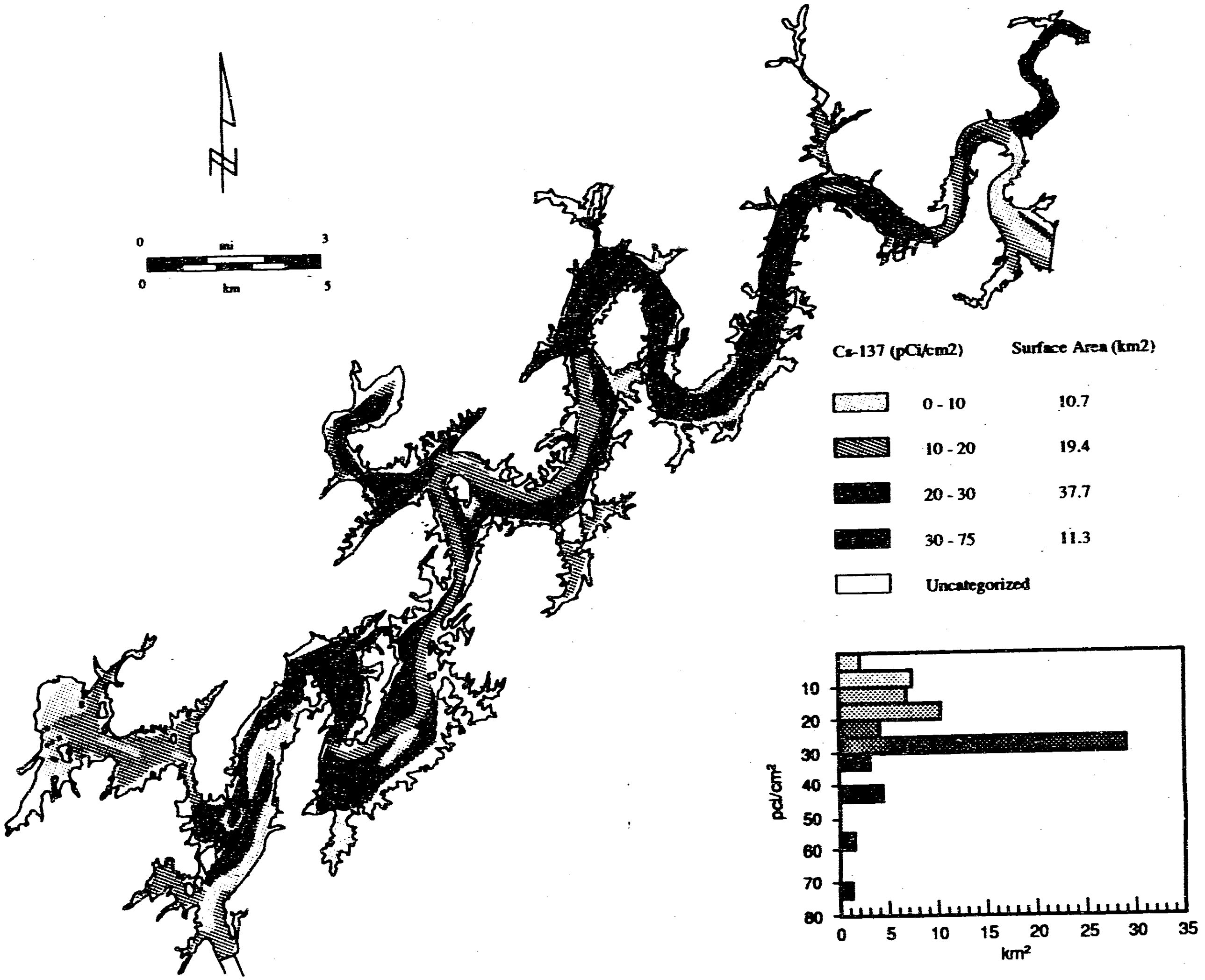

Fig. 16. Map of Watts Bar Reservoir illustrating the inventory (pCi/cm^2) of ^{137}Cs in the top 16 cm of sediment.

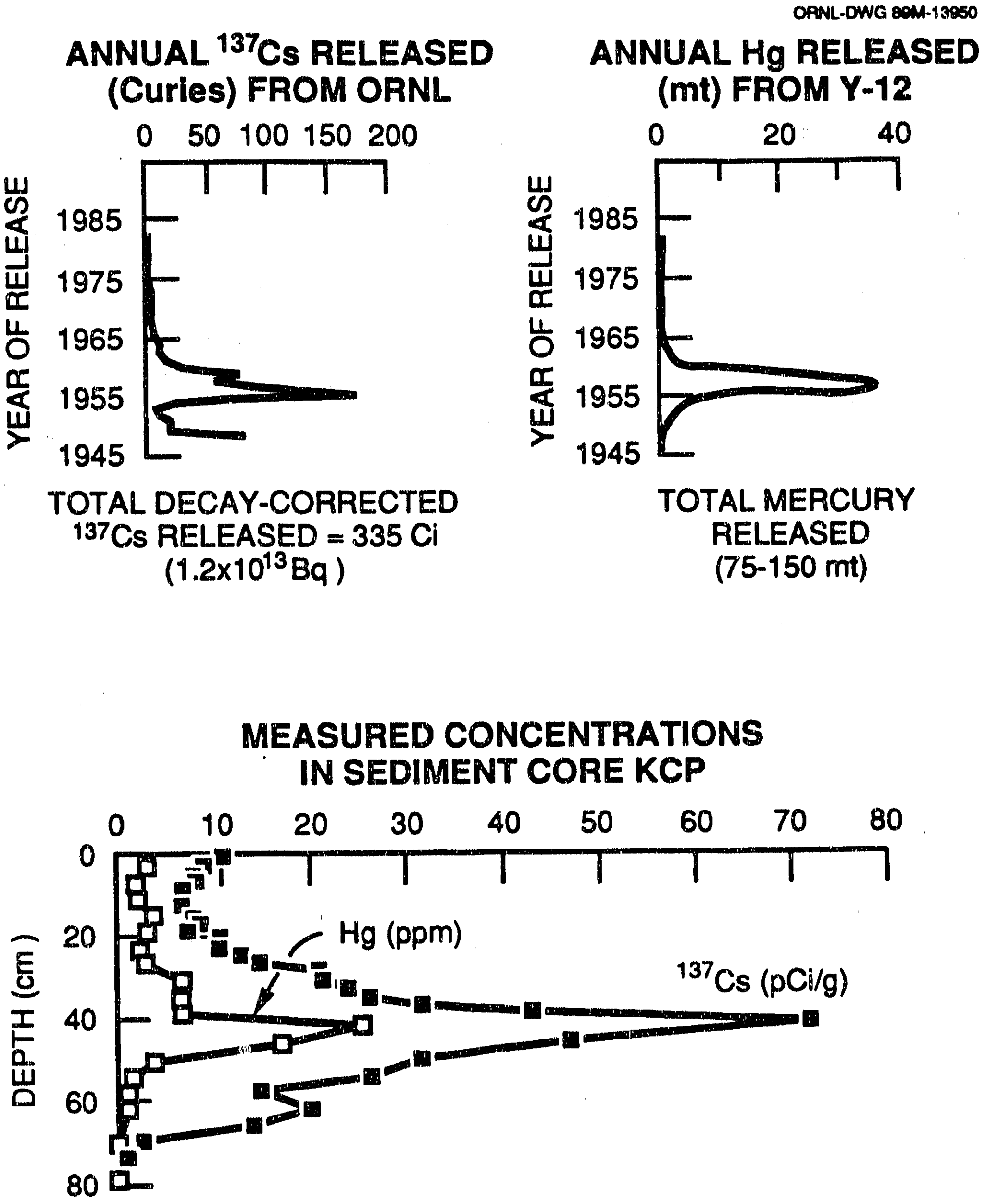

Fig. 17. The history of ^{137}Cs releases from ORNL and Hg releases from Y-12 compared with the vertical distribution of ^{137}Cs and Hg in the sediments of core KCP.

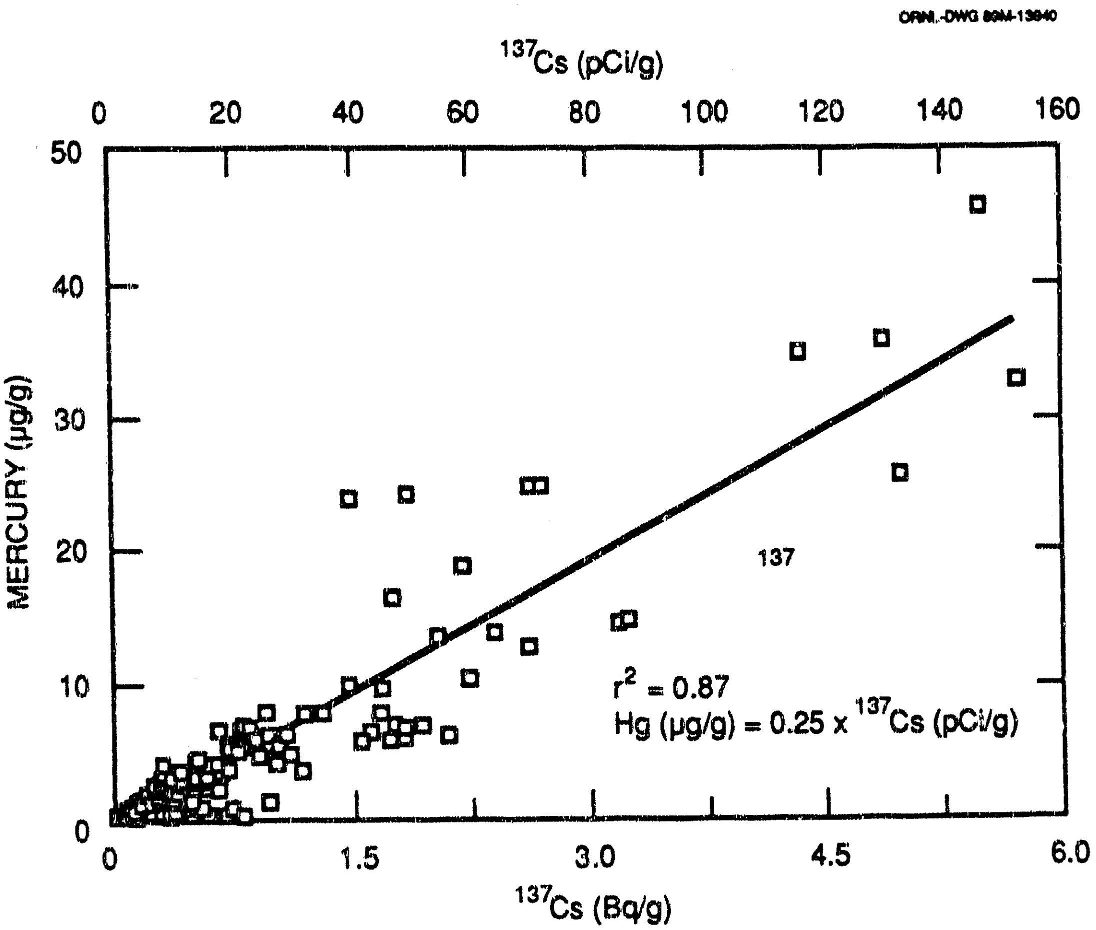

Fig. 18. Illustration of the relationship between the concentration of ^{137}Cs and the concentration of Hg in the sediments of Watts Bar Reservoir.

Using information on the concentration and distribution of ^{137}Cs as an indicator of the concentration and distribution of Hg, we have generated GIS maps that illustrate total Hg inventories (Fig. 19) and concentrations (Figs. 20 and 21) in the sediments of Watts Bar Reservoir. In addition, we estimate that about 76 metric tons of Hg has accumulated within the reservoir sediments.

$$\text{Hg } (\mu g/g) = 0.25 \times {}^{137}\text{Cs (pCi/g)}$$

or

$$\text{Hg (mt)} = 0.25 \times {}^{137}\text{Cs (Ci)}$$

$$\text{Hg (mt)} = 0.25 \times 304 \text{ Ci}$$

$$\text{Hg (mt)} = 76.0 \,.$$

Because of the skewed distribution of both the ^{137}Cs and Hg data, a log-log transformation was also performed and a linear model fit to the transformed data. The functional form of the model is Hg = $(a)(\text{Cs}^b)$, where a is the intercept and b is the slope. An analysis of

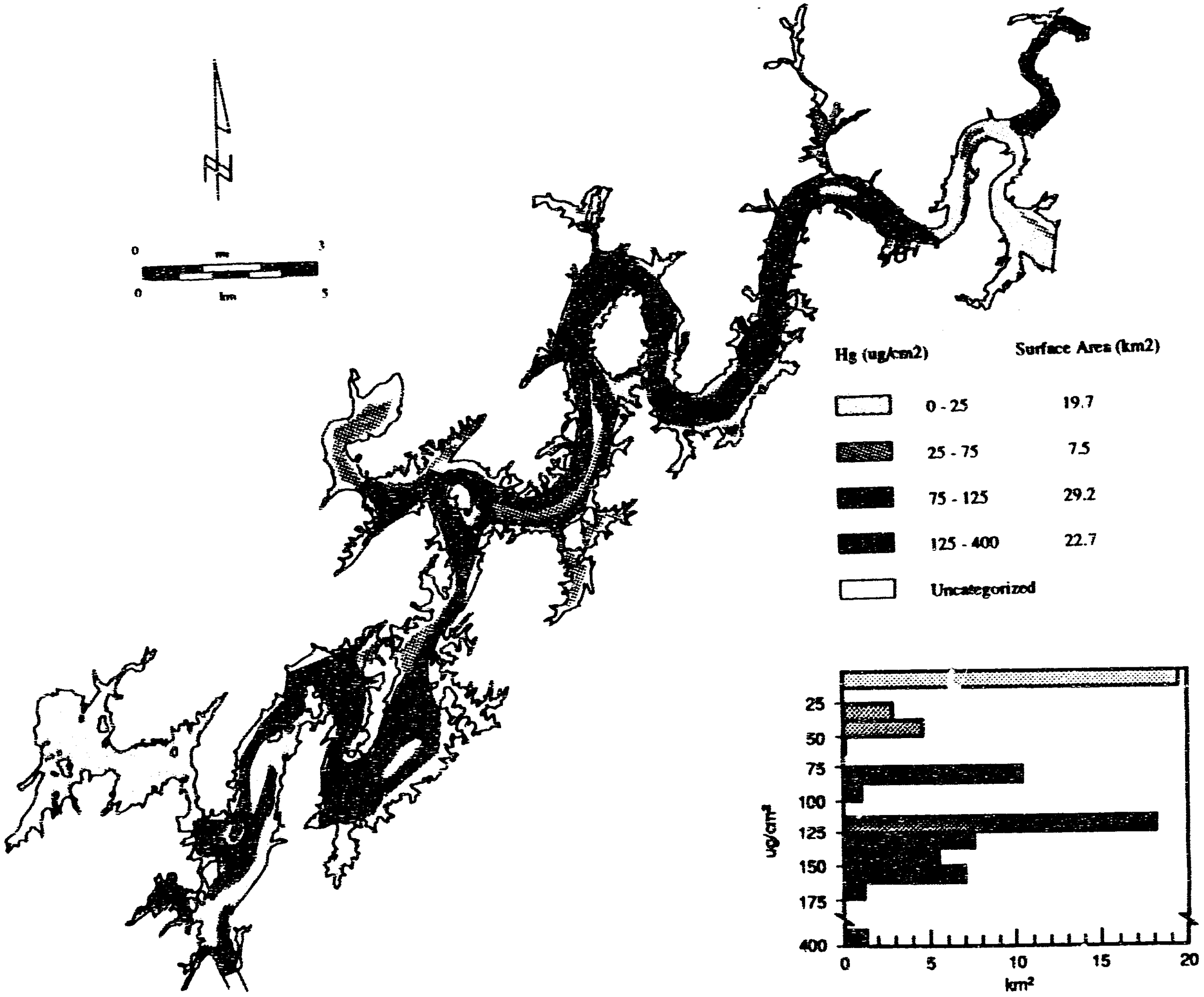

Fig. 19. Map of Watts Bar Reservoir illustrating Hg accumulation patterns and total inventories ($\mu g/cm^2$).

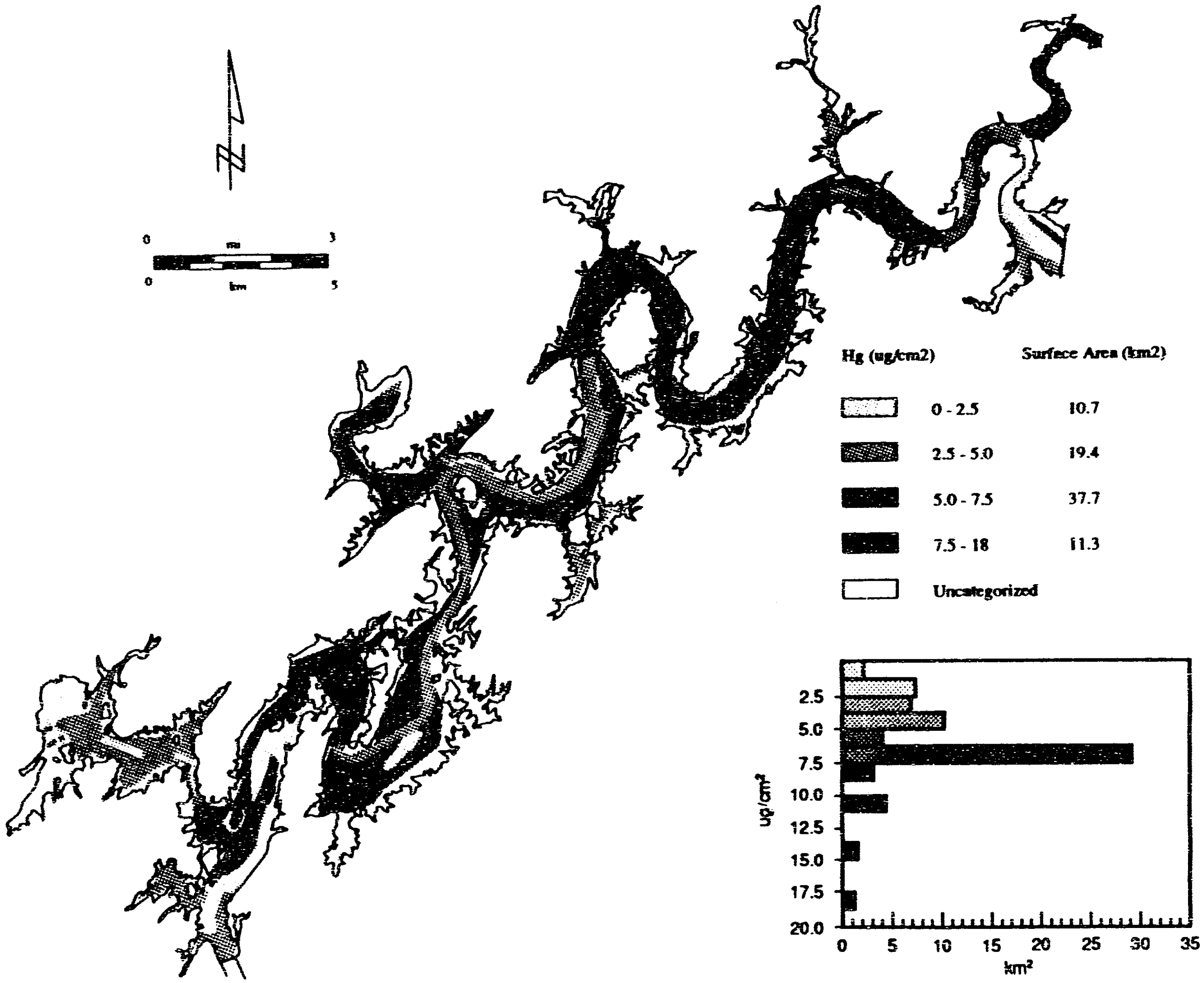

Fig. 20. Map of Watts Bar Reservoir illustrating the Hg inventory ($\mu g/cm^2$) in the upper 16 cm of sediment.

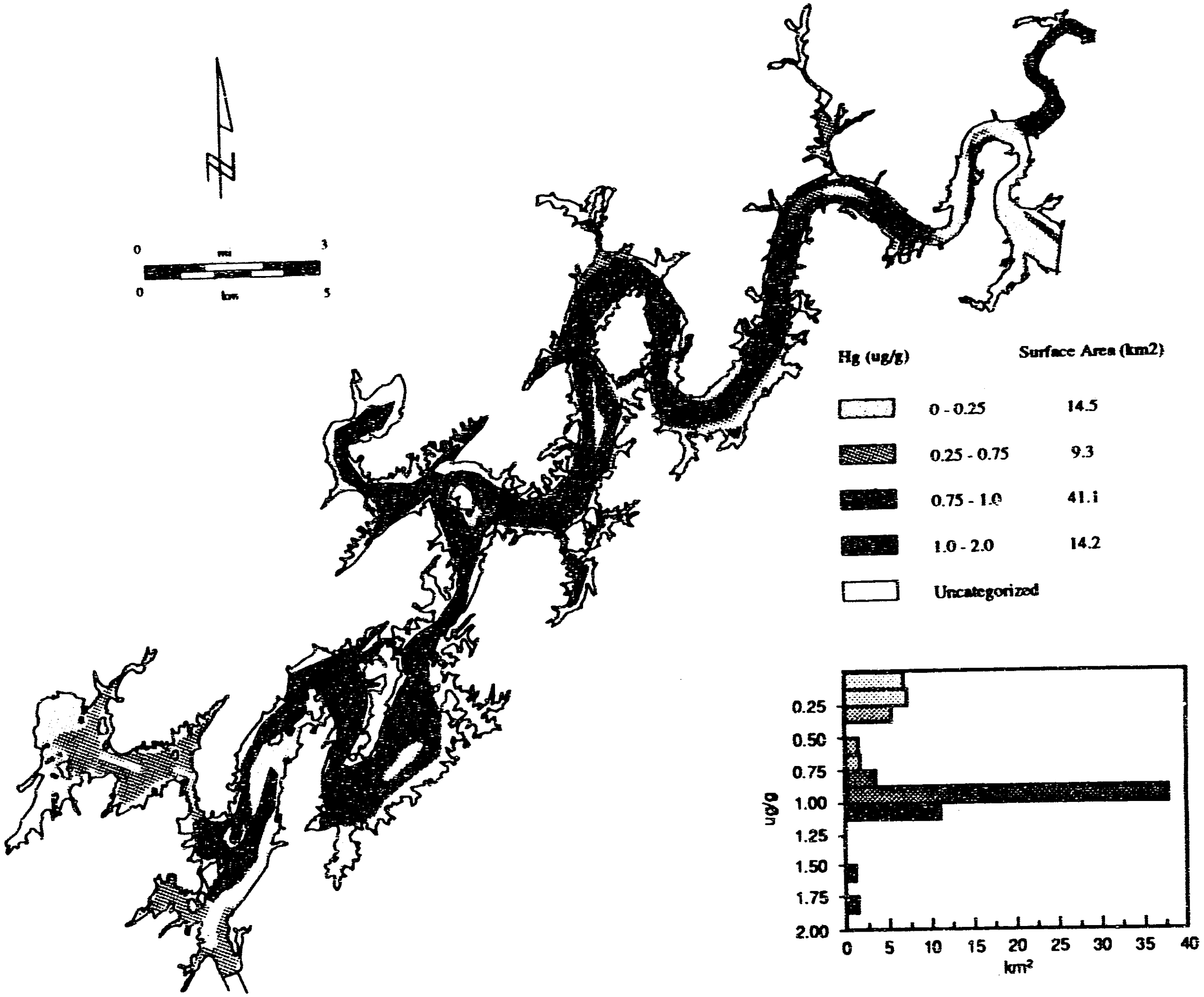

Fig. 21. Map of Watts Bar Reservoir illustrating the average Hg concentration (μg/g) in upper 16 cm of sediment.

covariance revealed that a separate model was needed for the Clinch River and Watts Bar cores. For both the Clinch River and Watts Bar data, we found that 87% ($r^2 = 0.87$) of the variation in the mercury data can be explained by the ^{137}Cs data. The model parameter estimates determined in this analysis are

Location	*a*	*b*
Clinch River	0.335	0.884
Watts Bar	0.265	0.889

Based on these models, we estimate that 62 metric tons of mercury is retained in the system. This model together with the linear model described above provides an estimate on the range of total mercury in Watts Bar.

HISTORICAL CONCENTRATIONS OF DISSOLVED MERCURY IN WATTS BAR RESERVOIR

Because sorption onto suspended particles and sediments is the principal mechanism by which many chemically reactive contaminants (such as ^{137}Cs and Hg) are removed from aquatic systems, the history of contamination in Watts Bar Reservoir has been recorded in the sediments (Fig. 15). Consequently, vertical profiles of ^{137}Cs or Hg in a chronologically dated sediment core can be used to estimate the age of the sediment and the concentration of Hg in surface sediments during previous years. In addition, by assuming that the particle-to-water distribution of Hg (about 1×10^5 in Table 5) has been relatively constant at a specific core site throughout the past, we can estimate past levels of dissolved Hg from the dated levels of Hg in the sediment cores.

Estimates for the historical concentrations of dissolved Hg near the mouth of the Clinch River and in Watts Bar Reservoir are presented in Tables 6 and 7. It is evident from the data in Table 7 that the levels of dissolved Hg (0.005 μg/L) estimated from the Hg concentration in the dated surface sediments are similar to the dissolved levels measured in the water column (0.004 ppb in Table 5). At the mouth of the Clinch River, however, before its confluence with the Tennessee River, estimated dissolved Hg concentrations are about three times higher. The highest predicted dissolved Hg levels (0.224 ppb, or μg/L) occurred between 1957 and 1959 at this site (Table 6). At the mouth of White Creek near the midsection of Watts Bar Reservoir (Fig. 1), the highest predicted dissolved Hg concentrations were about 0.06 μg/L (Table 7), and these levels also occurred during 1957–1958. These Hg concentrations are similar to those measured elsewhere in the Clinch River/Watts Bar Reservoir system and, according to Hoffman et al. (1991), do not pose an imminent risk to human health.

This information is critical for assessing the environmental and health risks associated with the past discharges of contaminants into aquatic systems. Historical estimates for dissolved contaminants with additional information on contaminant bioaccumulation in organisms are needed to predict contaminant levels in fish caught and consumed in prior years. Once particle-to-water distributions and vertical sedimentary profiles for other contaminants of concern are measured in Watts Bar Reservoir, it will be possible to obtain relatively accurate estimates for their past concentrations in drinking water or edible tissues.

Table 6. Watts Bar sediment core 567.5 (August 22, 1986)

Sediment depth (cm)	Dry weight (g)	^{137}Cs sediment (pCi/g ± 1σ)	Hg sediment (μg/g)	Hg dissolved[a] (μg/L)	Historical time period[b] (years)
0–4	72.58	5.26 ± 0.06	1.52	0.015	1986
4–8	119.62	5.68 ± 0.07	1.36	0.014	1984–1985
8–12	127.51	5.06 ± 0.08	1.77	0.018	1983
12–16	140.85	6.05 ± 0.19	2.42	0.024	1981–1982
16–20	147.80	7.39 ± 0.07	3.16	0.032	1980
20–24	144.13	6.51 ± 0.06	2.32	0.023	1978–1979
24–28	138.34	6.61 ± 0.06	2.32	0.023	1977
28–32	109.97	7.85 ± 0.06	3.33	0.033	1975–1976
32–36	101.34	11.01 ± 0.09	3.44	0.034	1974
36–40	133.20	9.81 ± 0.06	2.98	0.030	1972–1973
40–48	144.17	12.03 ± 0.24	2.91	0.029	1969–1971
48–56	149.98	13.56 ± 0.11	4.12	0.041	1966–1968
56–64	161.15	19.02 ± 0.15	5.10	0.051	1963–1965
64–72	156.36	22.24 ± 0.11	6.80	0.068	1960–1962
72–80	145.92	49.08 ± 0.21	24.40	0.244	1957–1959
80–84	70.62	58.36 ± 0.17	19.00	0.190	1955–1956
84–88	78.68	28.66 ± 0.11	6.31	0.063	1952–1954
88–92	81.22	13.01 ± 0.07	1.24	0.012	1950–1951
92–96	79.15	18.59 ± 0.08	0.36	0.004	1948–1949
96–100	77.05	10.64 ± 0.07	0.35	0.003	1946–1947
100–104	76.68	13.11 ± 0.07	0.31	0.003	
104–108	75.71	5.84 ± 0.05	0.34	0.003	
108–112	81.48	0.87 ± 0.02	0.12	0.001	
112–116	118.11	0.25 ± 0.03	0.09	0.001	
116–120	119.10	0.27 ± 0.05	0.05	0.001	
120–122	125.07	0.00 ± 0.00			

[a] Estimated using a particle-to-water distribution ratio of 1 × 10^5.

[b] Estimated using a sediment accumulation rate of 1.4 cm/year.

Table 7. Watts Bar sediment core 8-2-3 (October 17, 1986)

Sediment depth (cm)	Dry weight (g)	^{137}Cs sediment (pCi/g ± 1σ)	Hg sediment (μg/g)	Hg dissolved[a] (μg/L)	Historical time period[b] (years)
0–4	13.81	3.74 ± 0.10	0.52	0.005	1984–1986
4–8	18.10	3.80 ± 0.10	0.53	0.005	1981-1983
8–12	26.93	3.71 ± 0.33	0.52	0.005	1978–1980
12-16	31.17	4.43 ± 0.24	0.64	0.006	1975-1977
16–20	31.26	4.22 ± 0.30	0.72	0.007	1972–1974
20–24	32.77	5.19 ± 0.25	0.78	0.008	1969-1971
24–28	37.01	5.84 ± 0.35	1.07	0.011	1966–1968
28–32	39.95	11.76 ± 0.40	1.75	0.018	1964–1965
32-36	43.53	15.87 ± 0.53	2.30	0.023	1961-1963
36–40	44.33	24.34 ± 0.47	4.69	0.047	1958-1960
40-44	42.08	27.57 ± 0.50	5.46	0.055	1956–1958
44–48	42.64	9.45 ± 0.42	1.08	0.011	1953-1955
48–52	46.06	10.53 ± 0.30	0.47	0.005	1950–1952
52–56	48.91	11.67 ± 0.41	0.29	0.003	1947-1949
56–60	63.41	3.33 ± 0.19	0.14	0.001	1944–1946
60–62	53.50	0.29 ± 0.07	0.06	0.001	

[a] Estimated using a particle-to-water distribution ratio of 1 × 10^5.
[b] Estimated using a sediment accumulation rate of 1.4 cm/year.

SUMMARY

Operations and waste disposal activities on the DOE ORR have introduced ^{137}Cs and Hg into local streams that ultimately drain into the Clinch River and Tennessee River systems. Previous work has shown that (1) the highest discharges for both ^{137}Cs and Hg occurred during the mid-1950s; (2) contaminants introduced into the Clinch River have not remained there in either the bottom sediments or in the biota but instead have been flushed downstream; and (3) Watts Bar Reservoir (like other reservoirs on river systems) serves as a very efficient trap for riverborne particles, nutrients, and contaminants and is therefore a site of rapid sediment and contaminant accumulation.

To address the need for characterizing the nature and extent of contamination in Watts Bar Reservoir, we have measured the vertical distribution of ^{137}Cs in over 60 sediment cores and the concentration of ^{137}Cs in more than 190 surface-sediment samples. This work represents the initial scoping phase for the Clinch River RCRA Facility Investigation and uses the distribution of ^{137}Cs to identify contaminant accumulation patterns and potential problem, or "hot-spot," areas with regard to environmental hazard or human health. Radiocesium was chosen for this scoping effort because (1) its history of release into the Clinch River is reasonable well documented, (2) it is easy and inexpensive to measure by gamma spectrometry, and (3) it is rapidly sorbed to particulate matter and thus serves as a cost-effective tracer for identifying the transport and accumulation patterns of many other contaminants that are strongly associated with particulate matter, such as Hg, Pb, Pu, PCBs, other metals, and synthetic organic compounds.

The results from this study indicate that both ^{137}Cs and Hg are strongly associated with particles in Watts Bar Reservoir and have particle-to-water sorption ratios with values about 10^5. Vertical distributions of ^{137}Cs and Hg in the reservoir sediments are also strongly correlated ($r^2 = 0.87$), with both contaminants exhibiting a large subsurface peak coincident with their peak discharge histories. The sediment depth of this subsurface peak and the thickness of contaminated sediment varies with location in the reservoir and depends on the rate of sediment accumulation. A preliminary screening of the contaminants that may contribute to human health and environmental risk (Hoffman et al. 1991; Suter 1991) showed that these ^{137}Cs and Hg concentrations did not pose an imminent risk. Further study, however, is warranted to determine the need for remediation.

The total accumulation of ^{137}Cs in Watts Bar Reservoir sediments was estimated by measuring the ^{137}Cs inventory in each sediment core and extrapolating these data spatially with the ARC:INFO software package. These results indicate that about 304 Ci (1.12×10^{13} Bq) of ^{137}Cs now reside in the reservoir sediments. Discharge records indicate that a decay-corrected total of about 335 Ci (1.24×10^{13} Bq) of ^{137}Cs was released into the Clinch River system since 1949. This indicates that almost 91% of the total ^{137}Cs released has been retained by accumulation in the reservoir sediments.

The strong correlation between the vertical distribution of ^{137}Cs and Hg in the reservoir sediments was used to estimate that about 76 metric tons of Hg also reside in the sediments of Watts Bar Reservoir. In addition, the historical record of Hg accumulation in dated sediment cores was used to document levels of contamination in the reservoir water-column during the past 40 years. The highest level of dissolved Hg predicted from these data is about 0.224 ppb, which occurred between 1957 and 1959.

This initial scoping study was conducted to provide a preliminary characterization of the spatial extent of ORR-derived contamination in off-site areas, to help design cost-effective sampling strategies in environmental risk and human health assessments, and to identify specific off-site areas requiring more detailed work. Additional characterization of contaminant concentrations and distributions in the sediments, soils, water, and biota of off-site surface water environments will be conducted as part of the Clinch River RCRA Facility Investigation. These additional and more extensive site characterization data will be used to (1) further define the nature and extent of off-site contamination occurring downstream of the ORR, (2) estimate potential risks to human health and to the environment that may be associated with the occurrence of off-site contaminants, and (3) evaluate the need for remediation activities designed to reduce human-health and environmental risks.

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Appendix A

RADIONUCLIDE CONCENTRATIONS IN WATTS BAR RESERVOIR SEDIMENTS

Note: ^{40}K concentrations are reported, along with ^{137}Cs concentrations and inventories. ^{40}K is a naturally occurring, gamma-emitting radionuclide that is present in the environment, particularly in potassium-rich rocks, soils, and sediments.

Radionuclide concentrations in Watts Bar Reservoir sediments

Sample identification	Sample type[a]	Depth (ft)	^{137}Cs (pCi/g)	^{137}Cs (pCi/cm^2)	^{40}K (pCi/g)
C-KCP	soft sed		7.76 ± 0.06	1586	14.3 ± 0.1
5-1	soft sed	5	no sample		
5-2	soft sed	24	5.99 ± 0.05		12.4 ± 0.3
5-3	soft sed	50	10.65 ± 0.08		14.6 ± 0.4
5-4	soft sed	15	4.67 ± 0.04		12.0 ± 0.3
C-567.5	soft sed	20	6.38 ± 0.07	1580	17.7 ± 0.6
WH-1	sandy mud	7	0.37 ± 0.01		10.5 ± 0.1
WH-2	soft sed	10	1.35 ± 0.02		14.4 ± 0.2
C-GCS	soft sed	10	1.67 ± 0.04	59	14.8 ± 0.5
C-7GC	clay mud	10	1.09 ± 0.02	148	14.1 ± 0.3
5A-1	soft sed	35	0.29 ± 0.02		13.5 ± 0.4
5A-2	sand	6	0.12 ± 0.02		13.2 ± 0.3
5A-3	sand	9	0.24 ± 0.02		13.0 ± 0.4
5A-4	sand	9	0.14 ± 0.02		12.1 ± 0.4
5A-5	sandy mud	24	0.29 ± 0.01		13.2 ± 0.3
5A-6	soft sed	30	0.41 ± 0.06		13.6 ± 0.8
5-3-1	soil gravel	6	0.67 ± 0.01		8.6 ± 0.2
5-3-2	sandy mud	20	0.61 ± 0.01		12.1 ± 0.2
5-3-3	sandy mud	60	1.37 ± 0.02		7.6 ± 0.2
5-4-1	soft sed	8	no sample		
C-5-4-2	sandy mud	10	1.26 ± 0.03	18	14.0 ± 0.3
5-4-3	sandy mud	30	0.45 ± 0.01		12.0 ± 0.1
5-4-4	sand gravel	65	0.58 ± 0.01		7.9 ± 0.2
5-4-5	soft sed	18	4.44 ± 0.04		21.3 ± 0.3
C-5-4-5	soft sed	18	3.48 ± 0.08	823	18.9 ± 0.6
5-5-1	sandy mud	65	1.63 ± 0.02		11.1 ± 0.2
5-5-2	sandy mud	54	2.57 ± 0.03		11.5 ± 0.3
5-5-3	sand gravel	18	0.11 ± 0.01		9.9 ± 0.2
6-1-1	sand gravel	12	1.98 ± 0.03		13.2 ± 0.3
6-1-2	sandy mud	16	2.78 ± 0.04		13.5 ± 0.3
6-1-3	sand gravel	6	no sample		
6-1-3A	sandy mud	12	2.64 ± 0.04		12.9 ± 0.3
6-1-4	soft sed	60	2.66 ± 0.05		11.6 ± 0.4
6-1-5	soft sed	45	2.61 ± 0.04		13.3 ± 0.3
6-1-6	soft sed	12	3.57 ± 0.04		13.4 ± 0.3
C-6-1-6	soft sed	12	3.89 ± 0.06	406	14.7 ± 0.5
CC1	sandy mud	20	0.90 ± 0.03		18.5 ± 0.4
C-CC1	sandy mud	20	1.15 ± 0.10	91	12.0 ± 1.2
CC2	sandy mud	20	0.87 ± 0.02		20.3 ± 0.3
CC3	sandy mud	28	1.02 ± 0.03		10.7 ± 0.3
C-CC3	sandy mud	21	0.86 ± 0.04	109	13.3 ± 0.5
C-6-2-1	soft sed	18	3.04 ± 0.06	711	14.3 ± 0.6

Radionuclide concentrations (continued)

Sample identification	Sample type[a]	Depth (ft)	^{137}Cs (pCi/g)	^{137}Cs (pCi/cm^2)	^{40}K (pCi/g)
6-2-2	soil sed	35	2.36 ± 0.05		10.9 ± 0.4
GC-6-2-2	soil sed	35	2.94 ± 0.04	>181	12.6 ± 0.5
6-2-3	sandy mud	27	1.28 ± 0.03		12.1 ± 0.3
6-2-4	soft sed	45	2.81 ± 0.06		12.9 ± 0.5
GC-6-2-4	soft sed	45	3.44 ± 0.07	>218	13.2 ± 0.6
6-2-5	hard clay mud	20	2.20 ± 0.03		15.4 ± 0.3
6-3-1	sandy mud	30	2.39 ± 0.03		11.8 ± 0.2
C-6-3-1	sandy mud	22	2.48 ± 0.04	315	13.8 ± 0.3
6-3-2	soft sed	39	3.06 ± 0.06		13.3 ± 0.5
GC-6-3-2	soft sed	39	3.78 ± 0.07	>471	14.8 ± 0.5
C-6-3-3	soft sed	30	3.11 ± 0.07	542	17.1 ± 0.7
6-4-1	sand gravel	20	0.89 ± 0.02		7.3 ± 0.2
6-4-2	sand gravel	20	1.38 ± 0.03		6.9 ± 0.2
6-4-3	sand gravel		no sample		
6-4-4	soft sed	25	2.61 ± 0.03		13.7 ± 0.3
C-6-4-4	soft sed	26	2.85 ± 0.07	532	15.1 ± 0.1
6-4-5	soft sed	25	2.89 ± 0.05		14.1 ± 0.4
6-4-6	soft sed	50	2.92 ± 0.06		13.9 ± 0.5
GC-6-4-M	soft sed	50	3.37 ± 0.08	>403	12.6 ± 0.5
6-4-7	soft sed	50	3.16 ± 0.05		14.5 ± 0.4
6-4-8	soft sed	30	2.87 ± 0.08		13.3 ± 0.6
6-4-9	soil sed	25	2.21 ± 0.03		9.1 ± 0.3
GC-6-4-9	soil sed	25	3.42 ± 0.07	>102	14.5 ± 0.5
7-1-1	soft sed	35	2.34 ± 0.04		12.9 ± 0.4
C-7-1-1	soft sed	30	2.64 ± 0.05	308	11.6 ± 0.9
7-1-2	sand gravel	5	no sample		
7-1-3	sandy mud	21	0.58 ± 0.01		11.9 ± 0.2
7-1-4	soft sed	50	3.01 ± 0.05		12.8 ± 0.4
7-1-5	soft sed	50	2.85 ± 0.06		14.2 ± 0.5
GC-7-1-5	soft sed	50	3.97 ± 0.10	>311	15.2 ± 0.7
GC-7-1-M[b]	soft sed	52	3.97 ± 0.12		19.5 ± 2.0
7-1-6	sand gravel		no sample		
7-2-1	soft sed	22	2.01 ± 0.05		11.0 ± 0.5
7-2-2	soft sed	55	2.98 ± 0.06		11.8 ± 0.5
GC-7-2-2	soft sed	57	4.30 ± 0.09	>439	14.9 ± 0.8
7-2-3	sand gravel	20	no sample		
7-2-4	sand gravel	16	0.91 ± 0.03		9.7 ± 0.3
7-2-5	soft sed	36	2.55 ± 0.06		12.1 ± 0.5
GC-7-2-5	soft sed	40	3.98 ± 0.10	>508	16.3 ± 0.8
7-2-6	soft sed	52	3.05 ± 0.06		14.0 ± 0.5
7-2-7	clay mud	53	2.86 ± 0.05		14.5 ± 0.4
GC-7-2-7	clay mud	53	4.02 ± 0.11	>205	16.5 ± 0.7

Radionuclide concentrations (continued)

Sample identification	Sample type[a]	Depth (ft)	^{137}Cs (pCi/g)	^{137}Cs (pCi/cm^2)	^{40}K (pCi/g)
ROCKWD LAND	soil sed	15	1.25 ± 0.04	84	11.6 ± 0.6
NEW HOPE	sand gravel	12	0.18 ± 0.02	4	2.6 ± 0.2
7-3-1	soft sed	35	2.23 ± 0.04		12.9 ± 0.4
7-3-2	clay mud	25	2.68 ± 0.02		13.4 ± 0.2
7-3-3	soft sed	50	3.14 ± 0.06		13.3 ± 0.5
GC-7-3-4	soft sed	50	4.08 ± 0.04	>634	15.6 ± 0.4
7-3-4	soft sed	50	2.15 ± 0.03		11.0 ± 0.3
7-3-5	soft sed	23	4.46 ± 0.05		15.3 ± 0.3
C-7-3-5	soft sed	23	3.36 ± 0.05	440	16.7 ± 0.7
7-3-6	soil sed	17	no sample		
7A-1-1	sandy mud	12	0.51 ± 0.03		7.6 ± 0.4
7A-1-2	soft sed	16	0.60 ± 0.02		6.7 ± 0.4
7A-1-3	clay mud	15	0.57 ± 0.02		8.9 ± 0.2
7A-1-4	clay mud	26	0.71 ± 0.02		11.0 ± 0.3
7A-1-5	soft sed		1.17 ± 0.03		11.5 ± 0.3
7A-1-6	soft sed	40	1.04 ± 0.03		12.2 ± 0.3
7A-1-7	sandy mud	39	0.62 ± 0.02		4.7 ± 0.2
7A-1-8	soft sed	25	1.29 ± 0.03		12.9 ± 0.4
7A-1-9	clay mud	32	1.36 ± 0.03		12.4 ± 0.3
7A-1-10	soft sed	45	2.14 ± 0.04		13.0 ± 0.4
GC-7A-1-10	soft sed	33	2.48 ± 0.05	124	13.5 ± 0.5
7A-1-11	sandy mud	30	0.86 ± 0.02		10.5 ± 0.3
7A-2-1	gravel		no sample		
7A-2-2	clay mud	20	2.06 ± 0.02		12.5 ± 0.2
GC-7A-2-2	soft mud	25	2.52 ± 0.04	>153	14.3 ± 0.4
8-1-1	soft sed	30	2.05 ± 0.03		15.6 ± 0.3
8-1-2	soft sed	35	2.36 ± 0.05		18.4 ± 0.5
GC-8-1-2	clay mud	35	2.05 ± 0.06	158	16.5 ± 0.7
8-1-3	soil gravel	33	no sample		
8-1-4	soft sed	55	2.73 ± 0.05		14.7 ± 0.5
GC-8-1-4	soft sed	55	3.44 ± 0.07	>629	14.6 ± 0.7
8-1-5	clay mud	32	3.95 ± 0.05		12.5 ± 0.4
GC-8-1-5	clay mud	32	3.20 ± 0.07	163	15.0 ± 0.7
8-1-6	soft mud	55	3.63 ± 0.07		15.9 ± 0.6
8-1-7	soft mud	57	3.38 ± 0.07		15.1 ± 0.6
GC-8-1-7	soft sed	55	3.79 ± 0.10	180	13.6 ± 1.0
8-2-1	soil gravel	45	no sample		
8-2-2	soft sed	60	2.90 ± 0.06		13.4 ± 0.5
8-2-3	soft sed	65	2.57 ± 0.05		12.5 ± 0.5
GC-8-2-3	soft sed	60	3.84 ± 1.00	342	14.7 ± 1.1
8-2-4	sandy mud	33	1.64 ± 0.02		13.7 ± 0.2
GC-8-2-4	sandy mud	30	1.76 ± 0.04	12	16.4 ± 0.5

Radionuclide concentrations (continued)

Sample identification	Sample type[a]	Depth (ft)	^{137}Cs (pCi/g)	^{137}Cs (pCi/cm^2)	^{40}K (pCi/g)
8-3-1	soft sed		2.84 ± 0.06		13.6 ± 0.5
GC-8-3-1	soft sed		4.24 ± 0.05	>271	15.9 ± 0.5
8-3-2	sand gravel		no sample		
8-3-3	soft sed	20	2.02 ± 0.03		11.4 ± 0.3
GC-8-3-3	soil sed	20	1.71 ± 0.02	134	10.9 ± 0.2
9-1-1	soft sed	20	4.29 ± 0.05		15.7 ± 0.4
GC-9-1-1	sandy mud	20	2.52 ± 0.06	266	14.5 ± 0.6
9-1-2	soft sed	50	no sample		
GC-9-1-2	soft sed	50	2.55 ± 0.05	>505	11.7 ± 0.4
9-1-3	sandy soil	4	no sample		
9-1-3A	sandy soil	10	0.48 ± 0.01		5.4 ± 0.2
GC-9-1-3A	sandy soil	10	0.64 ± 0.02	6	6.8 ± 0.3
9-1-4	soil gravel	15	0.31 ± 0.02		11.3 ± 0.3
9-1-5	soil gravel		1.17 ± 0.03		10.2 ± 0.3
9-2-1	soil gravel	15	no sample		
9-2-2	soil gravel	18	no sample		
9-2-3	soft sed	30	2.17 ± 0.02		12.8 ± 0.2
9-2-4	soft sed	40	1.58 ± 0.02		9.4 ± 0.3
GC-9-2-4	soil sand	20	0.33 ± 0.03	1	6.4 ± 0.4
9-2-5	soft sed	35	2.46 ± 0.05		12.3 ± 0.5
9-2-6	soft sed	30	3.04 ± 0.05		11.5 ± 0.4
GC-9-2-6	soft sed	30	2.00 ± 0.07	>446	14.4 ± 0.7
9-2-7	soft sed	45	1.17 ± 0.05		11.0 ± 0.4
GC-9-2-7	soft sed	25	2.75 ± 0.07	289	13.9 ± 0.7
9-2-8	soft sed	40	3.16 ± 0.01		15.0 ± 0.6
GC-9-2-8	soft sed	30	3.31 ± 0.10	248	15.2 ± 0.9
9-2-9	soft sed	48	2.59 ± 0.02		14.9 ± 0.1
9-2-10	sandy mud	45	0.72 ± 0.01		12.2 ± 0.2
9-2-11	sandy mud	65	1.17 ± 0.05		11.7 ± 0.4
GC-9-2-11	sandy mud	25	0.18 ± 0.01	5	9.2 ± 0.2
9-3-1	clay sed	25	1.45 ± 0.03		9.9 ± 0.3
9-3-2	sand gravel		no sample		
9-3-3	clay sed	48	6.00 ± 0.05		12.6 ± 0.3
9-3-4	soft sed	69	2.65 ± 0.04		14.5 ± 0.3
GC-9-3-4	soft sed	63	2.79 ± 0.05	341	10.7 ± 0.3
9-3-5	soft sed	30	4.18 ± 0.05		16.1 ± 0.4
9-3-6	soft sed	39	2.65 ± 0.06		14.0 ± 0.5
9-3-7	clay sed	36	2.75 ± 0.04		12.9 ± 0.4
GC-9-3-7	soft sed	36	1.89 ± 0.06	339	12.1 ± 0.6
9-3-8	soil gravel	30	0.19 ± 0.02		6.3 ± 0.2
9-3-9	soft sed	36	1.92 ± 0.05		10.8 ± 0.4
9-4-1	clay sed	18	0.59 ± 0.02		5.1 ± 0.2
9-4-2	sandy mud	20	0.62 ± 0.01		4.5 ± 0.1

Radionuclide concentrations (continued)

Sample identification	Sample type[a]	Depth (ft)	^{137}Cs (pCi/g)	^{137}Cs (pCi/cm^2)	^{40}K (pCi/g)
9-4-3	soft sed	48	2.46 ± 0.06		13.9 ± 0.5
GC-9-4-3	soft sed	42	2.51 ± 0.10	565	12.6 ± 1.1
9-4-4	sandy mud	42	2.04 ± 0.02		11.3 ± 0.2
9-4-5	soft sed	75	2.51 ± 0.06		13.0 ± 0.6
GC-9-4-5	soft sed	70	2.34 ± 0.04	122	13.2 ± 0.7
10-1-1	sandy mud	27	0.69 ± 0.03		5.9 ± 0.3
GC-10-1-2	soft sed	30	1.12 ± 0.04	108	10.8 ± 0.5
10-1-3	sandy mud	12	0.17 ± 0.01		2.6 ± 0.1
10-1-4	clay mud	15	0.40 ± 0.02		6.9 ± 0.3
10-2-1	soft sed	27	0.80 ± 0.02		10.5 ± 0.3
10-2-2	soft sed	22	0.73 ± 0.04		7.3 ± 0.4
GC-10-2-A	soft sed	30	0.82 ± 0.05	21	10.2 ± 0.5
10-3-1	sand gravel	17	0.05 ± 0.01		0.9 ± 0.1
10-3-2	sandy mud	24	0.64 ± 0.03		7.7 ± 0.3
GC-10-3-2	sandy mud	24	0.74 ± 0.06	9	8.3 ± 0.7
10-3-3	soft sed	24	0.59 ± 0.03		6.3 ± 0.3
10-3-4	soft sed	24	0.65 ± 0.01		7.0 ± 0.3
10-4-1	sandy mud	15	0.16 ± 0.01		3.1 ± 0.1
10-4-2	sandy mud	14	0.16 ± 0.02		3.0 ± 0.2
10-4-3	soil sed	10	0.32 ± 0.02		6.6 ± 0.3
10-4-4	soil sed	9	0.28 ± 0.02		5.9 ± 0.3
10-4-5	soil sed	15	0.27 ± 0.02		4.6 ± 0.3
10-4-6	sandy mud	15	0.18 ± 0.02		2.6 ± 0.2
10-5-1	soft sed	33	0.69 ± 0.01		5.8 ± 0.1
10-5-2	sand gravel	33			
10-5-3	soft sed	36	1.44 ± 0.04		1.0 ± 0.4
GC-10-5-3	soft sed	36	1.01 ± 0.05	22	10.8 ± 0.6
11-1-1	sand	30	0.43 ± 0.01		3.6 ± 0.1
11-1-2	soft sed	45	2.89 ± 0.04		12.7 ± 0.3
11-1-3	soft sed	45	2.42 ± 0.02		12.7 ± 0.2
GC-11-1-3	soft sed	30	2.61 ± 0.06	250	12.8 ± 0.5
11-1-4	soft sed	75	2.76 ± 0.04		16.0 ± 0.4
11-1-5	sand gravel	15	0.45 ± 0.02		5.1 ± 0.2
11-1-6	soft sed	40	2.42 ± 0.02		11.4 ± 0.2
11-1-7	soft sed	40	2.78 ± 0.01		12.1 ± 0.3
11-1-8	soft sed	63	1.93 ± 0.03		11.1 ± 0.4
11-1-9	soft sed	55	2.45 ± 0.05		12.8 ± 0.5
GC-11-1-9	soft sed	55	2.44 ± 0.06	289	12.2 ± 0.5
11-1-10	soft sed		2.42 ± 0.05		11.0 ± 0.5
11-1-11	soft sed	45	1.83 ± 0.05		8.9 ± 0.4
11-2-1	soft sed	75	2.71 ± 0.01		15.0 ± 0.4
GC-11-2-1	soft sed	70	2.38 ± 0.08	397	11.7 ± 0.7
11-2-2	sandy mud	33	1.10 ± 0.01		13.0 ± 0.2

Radionuclide concentrations (continued)

Sample identification	Sample type[a]	Depth (ft)	^{137}Cs (pCi/g)	^{137}Cs (pCi/cm^2)	^{40}K (pCi/g)
11-2-3	sand gravel	45	0.67 ± 0.01		11.9 ± 0.1
11-2-4	sand gravel	18	0.16 ± 0.01		6.1 ± 0.2
11-2-5	soft sed	33	1.96 ± 0.04		8.9 ± 0.3
GC-11-2-5	soft sed	35	2.32 ± 0.19	321	15.7 ± 2.3
11-2-5B	soft sed	30	1.69 ± 0.02		7.1 ± 0.1

[a] Sample types: soft sed = recently deposited soft mud; clay mud = cohesive clayey mud; sandy mud = sandy mud or fine sands; sand = sand and gravel; soil gravel = submerged soil and eroded bank materials.

[b] No inventory made: core only 24 cm long.

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