

THE USE OF FALLOUT CESIUM-137
AS A TRACER OF SEDIMENT MOVEMENT AND DEPOSITION ^{1/}

by

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INTRODUCTION

The origin and movement of sediment may be determined by identifying certain sediment particles as being specifically associated with a given area and by following the movement of "tagged" sediment particles. These "tracer" particles may be naturally occurring or they may be artificially introduced into the system. The ideal tracer is one which [1] moves, or has moved, exactly as the population as a whole and which [2] is identified readily and its concentration determined. Dyes and radioactivity have been used to tag sediment particles and to follow their movement. The mineralogical and radiological identification of particles also can be used to determine the origins of various sediments.

During the past 20 years, man's activity associated with testing of nuclear bombs has provided tagging for many sediment particles. All regions of the world have received radioactive fallout, although this fallout has varied in intensity and in composition of the radioactivity with time and place. The rate of release of radioactivity into the atmosphere at Birmingham, Alabama (5) ^{2/} has decreased in the past several years (Figure 1).

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^{2/} Numbers in parentheses refer to reference citations.

Two radioisotopes, strontium-90 and cesium-137, comprise much of the longer lived radioactive fallout that has occurred. Biologically, strontium-90 is of greater concern because of its greater radiological hazard to life. However, strontium-90 is a beta emitter and as such is more difficult to measure accurately. Cesium-137, occurring in a ratio of 1.6 to 1.0 for strontium-90 (10), is much more easily determined because of its energetic gamma emission of 0.662 million electron volts (Mev). With strontium-90, the analyst is faced with extraction and concentration problems prior to radiological analyses. With cesium-137, the field sample, after a minimal treatment, can be analyzed directly using a scintillation detector, a gamma differential spectrometer and a multi-channel analyzer for fast, complete analyses of the gamma-ray energy spectrums.

Low and Edvarson (7), Walton (13), and Franklin *et al.* (3) have shown that most of the fallout cesium-137 remains in the upper 2.5 cm of the soil, the soil layer most susceptible to erosion. In general, the fixation of cesium-137 in soils and sediments depends on the ion-exchange capacity of the medium (1). Clays absorb a greater amount of cesium-137 than do other soil particles (12) and hold cesium-137 strongly against removal by leaching (2,9). Once the cesium-137 becomes attached to a clay particle, it probably will remain with the particle through the sedimentation cycle. By determining the cesium-137 concentration in the eroding material, in the material being transported, and in the deposited material, we can follow the movement of the radioactive cesium-137 and sediment through the watershed.

Past radioactive fallout can be used as a tracer of sediment movement and as an indicator of sediment age and origin. Within a given watershed, fallout is redistributed when erosion removes the surface soils. The probability of concentrating this radioactivity within a watershed, or drainage basin, is high if areas of sediment deposition, such as reservoirs, lakes, stream channels or estuaries, exist.

The purpose of this paper is to describe the use of radioactive fallout, as cesium-137, as a tracer of erosion, sediment transport, and sediment deposition within a watershed.

METHODS AND MATERIALS

1. Field Collection

Soil samples are collected from watersheds that drain into a reservoir or some other type of water impoundment. These watersheds are selected to represent a wide variety of environmental, land use, topographic, and geologic characteristics. Samples are taken that are representative of the various soils and vegetation types found in the watershed, as well as of the erosion present. Soil samples are taken to a depth of 10 cm in either 2.5 or 5.0-cm increments. Where possible, the samples are of a known volume and area.

Sediment samples are taken either as "grab" samples or as volume samples. Grab samples give an indication of radioactivity per unit weight while volume samples give an indication of the concentration and the vertical distribution of radioactivity in the sediment. The depth to which the volume sediment samples are taken depends on the depth of the accumulated sediment and on the accessibility of the sampling site.

2. Laboratory Procedures

The soil and sediment samples are brought into the laboratory and dried at 50° centigrade for 48 hours. The samples are then sieved through a 12-mm screen and further dried at 50°C. After removal from the oven, the samples are weighed, and a subsample taken for determination of dry weight (heated at 105°C). The bulk sample is used for gamma-ray spectrometric analyses.

3. Gamma-ray Spectrometric Analysis

Gamma-ray spectrometric analyses are made using a 1024-channel analyzer. The main component of the analyzer is a Nuclear Data ^{3/} Model 2200 series system analyzer. Accessory equipment includes a preamplifier, a linear amplifier and a test pulser, all made by Canberra; a Harshaw "integral-line" detector with a 10- x 12.5-cm thallium-activated sodium iodide [NaI(Tl)] crystal; and a Teletype Model 33 ASCII printer, paper tape punch and reader. A lead cave contains the scintillation crystal, preamplifier and the sample. The lead cave has inside dimensions of 61 x 61 x 61 cm with 10-cm thick walls and is lined with 0.038-cm copper and 0.076-cm cadmium sheets (6).

A Marinelli beaker (Figure 2) was designed and fabricated of kydex which allows 2.5-cm of sample to be placed around the circumference and over the top of the NaI(Tl) crystal. The Marinelli beaker will hold between 2250 and 2750 grams of sample when filled to the 2.5-cm depth. The samples are packed into the beaker, weighed, and stored 2 weeks before counting.

^{3/} Names of products or companies are provided for informational purposes only and do not constitute an endorsement or preferential use by the U.S. Department of Agriculture.

Standards containing cesium-137, thorium, uranium and potassium are prepared to simulate the soil and sediment samples. Known quantities of each of the radionuclides are mixed with sodium phosphate ($\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$) to make a total of 2500 grams for each standard. Sodium phosphate is used as the matrix for carrying the radionuclides because it has a density and proton number (Z) similar to soil (4). Gamma-ray analyses showed no gamma radioactivity in the sodium phosphate above background. Analyzed ore samples of thorium (NBL-79) and uranium (NBL-73) from the National Bureau of Standards are used to prepare the standards. Potassium chloride, reagent grade, is used as the standard for potassium.

Pulses emitted from the NaI(Tl) crystal with energies from 0 to 2.82 Mev are sorted into 512 channels of the 1024-channel analyzer for each sample. A background count is stored in the other 512 channels of the analyzer. The counting time was 8000 seconds for both samples and standards. Upon completion of counting, the stored background count is subtracted from the gross count and the net count (count due to sample) is punched on paper tape and printed.

The data on the paper tape are transferred to magnetic tape using a computer program developed at the University of Mississippi Computer Center. The gamma-ray spectra on the magnetic tape are resolved to give concentrations of radionuclides using a computer program developed by E. Schonfeld (11) of the Oak Ridge National Laboratory, and modified by R. B. Wilson of the USDA Sedimentation Laboratory for compatibility with the IBM-360-40 computer at the University of Mississippi Computer Center. The computer program determines concentration of radionuclides by least-squares resolution of gamma-ray spectra using a library of standards. Our library of standards contains cesium-137, thorium, uranium, and potassium.

EXPERIMENTAL RESULTS AND DISCUSSION

Soil and sediment samples were collected in 1969 from watersheds in nine states, including Mississippi. Sampling in Mississippi was confined almost entirely to the Little Tallahatchie River Watershed above Sardis Dam (Figure 3). A number of sediment samples was taken in Sardis Reservoir as well as from smaller upstream reservoirs. The data in this report are confined to those sediment and soil samples taken upstream of Sardis Dam. The reported results are neither complete nor final but are illustrative of the material and information being collected both in Mississippi and other states.

A small dam and reservoir identified as Power Line Dam is located in the Little Tallahatchie River Watershed. A number of sediment surveys have been made on this reservoir including one reported before this conference in 1969 (8). Soil samples were taken in the watershed above Power Line Dam at nine sites; three each under dominant oak, pine, and grass cover. Volume samples were collected to a depth of 10 cm in 2.5-cm increments. The intensity of the measured cesium-137 fallout is summarized in Table 1.

The greater accumulation of cesium-137 in the immediate surface soil (0-2.5 cm) under forest cover as contrasted to grass cover is due to the greater organic matter under the forest cover acting as a ion exchange site to retain the cesium-137 (1). The depth of movement of cesium-137 is greater under the grass cover, giving the total accumulation in the soil of cesium-137 to be grass > oak > pine cover. The difference in total accumulation under the three stands is an indication of the amount of litter under each cover type. The pine cover had the most litter and the grass cover did not have any litter. All of these sites were relatively undisturbed. Where erosion does occur, fallout activity should be less because the removal of the fallout by eroding sediment. For example, in two other samples from above Power Line Dam, an uneroded area contained 1.50 ± 0.02 nanocuries (nCi) cesium-137 per kilogram and an adjacent eroded area contained 0.04 ± 0.01 nCi cesium-137 per kilogram. Error values are standard error, i.e., one sigma.

A number of sediment profiles were sampled in Sardis Reservoir in the fall of 1969. Because the water level in the reservoir was low, samples were taken in 2.5-cm increments until the original soil surface was reached. The observed cesium-137 concentrations for these samples are summarized in Table 2.

The amount, distribution, and depth of fallout cesium-137 in Sardis Reservoir sediment samples varied with location and type of sediment. Coarse-textured sediments, as the Clear Creek profile, contained little cesium-137. This was true for upland soils of sandy texture also. Cesium-137 is most associated with clays and organics because of its absorption by the exchange complex (1). For instance a sandy deposited soil above Power Line Dam contained only 0.02 ± 0.003 nCi cesium-137 per kilogram as compared to the 1.50 nCi/kg mentioned above for an uneroded area or even 0.04 nCi/kg for eroded areas of medium-textured soil.

The water level in Sardis Reservoir varies in a more or less cyclic manner. Generally, the parts of the reservoir near the river channel are inundated longer and hence will have the greater accumulations of fine-textured sediment. The three sites lakeward (toward river channel) from the Coontown boat ramp show increasingly greater depths of accumulated sediment (C > B > A). Also as sediment accumulation increases (C > B > A) the depth of maximum cesium-137 concentration is progressively lower in the profile as one proceeds to the lower lake elevation (A = 0. to 2.5 cm; B = 2.5 to 10.0 cm; C = 7.5 to 15.0 cm).

If one assumes the maximum cesium-137 fallout occurred with that for strontium-90 (Figure 1) in 1963, one can calculate that the minimum rate of sediment accumulation from 1963 to 1969 is less than 1.5 cm per year at Coontown B site, less than 0.5 cm per year at Coontown A site and about 1.5 cm per year at Coontown C site. The rates at the Sardis Dam Park and Hurricane Creek sites are in the 1.0 to 1.5 cm per year range. The two peak concentrations of cesium-137 occurring in the Hurricane Creek profile (at 5.0 - 7.5 and 15.0 - 17.5 cm) may correlate with the two known peaks of fallout, 1959 and 1963 (Figure 1). If so, the sediment accumulation rate from 1959 to 1963 was about twice (2.5 cm/yr) that from 1963 to 1969 (1.25 cm/yr).

The large differences in concentrations of cesium-137 in the different sediment profiles are noteworthy. Some difference can be assigned to differences in texture of the sediment, as with the Clear Creek sample. The higher concentration of cesium-137 in the Hurricane Creek sediments remains unexplained. The observed cesium-137 concentrations in sediment are of the same order of variation as for soils (Table 1). It seems likely that sediments deposited in the Hurricane Creek area are derived from soils high in clay and/or organic matter.

SUMMARY AND CONCLUSIONS

Radioactive fallout tags soil particles with identifiable radioisotopes so that movement of these particles can be followed through the sedimentation process. The content of cesium-137, a gamma emitter (0.662 Mev) and a fallout product, was determined in a number of selected soils and sediments from the Little Tallahatchie River Watershed above Sardis Dam. The variation in measured concentrations of cesium-137 in sediment profiles can be related to periods of maximum fallout. The age of the upper sediments and the rate of deposition can be estimated. A greater depth and rate of sediment accumulation were found in the deeper parts of the reservoir area sampled. Data indicate the recent (1963-69) rate of sedimentation is less than in prior years (1959-63).

Soils under forest cover contain more cesium-137 in the surface layer than those under grass. Where soils have eroded, cesium-137 content is less. The cesium-137 content is also less in sandy soils than in finer textured soils because of the differences in ion exchange capacity of the soils. Concentrations of cesium-137 in some sediments are greater than in the contributing upland soils, undoubtedly associated with the greater percentage of finer particles.

The sampling techniques and methods of radiological analyses of soils and sediments for cesium-137 by gamma scintillation spectrometry are described.

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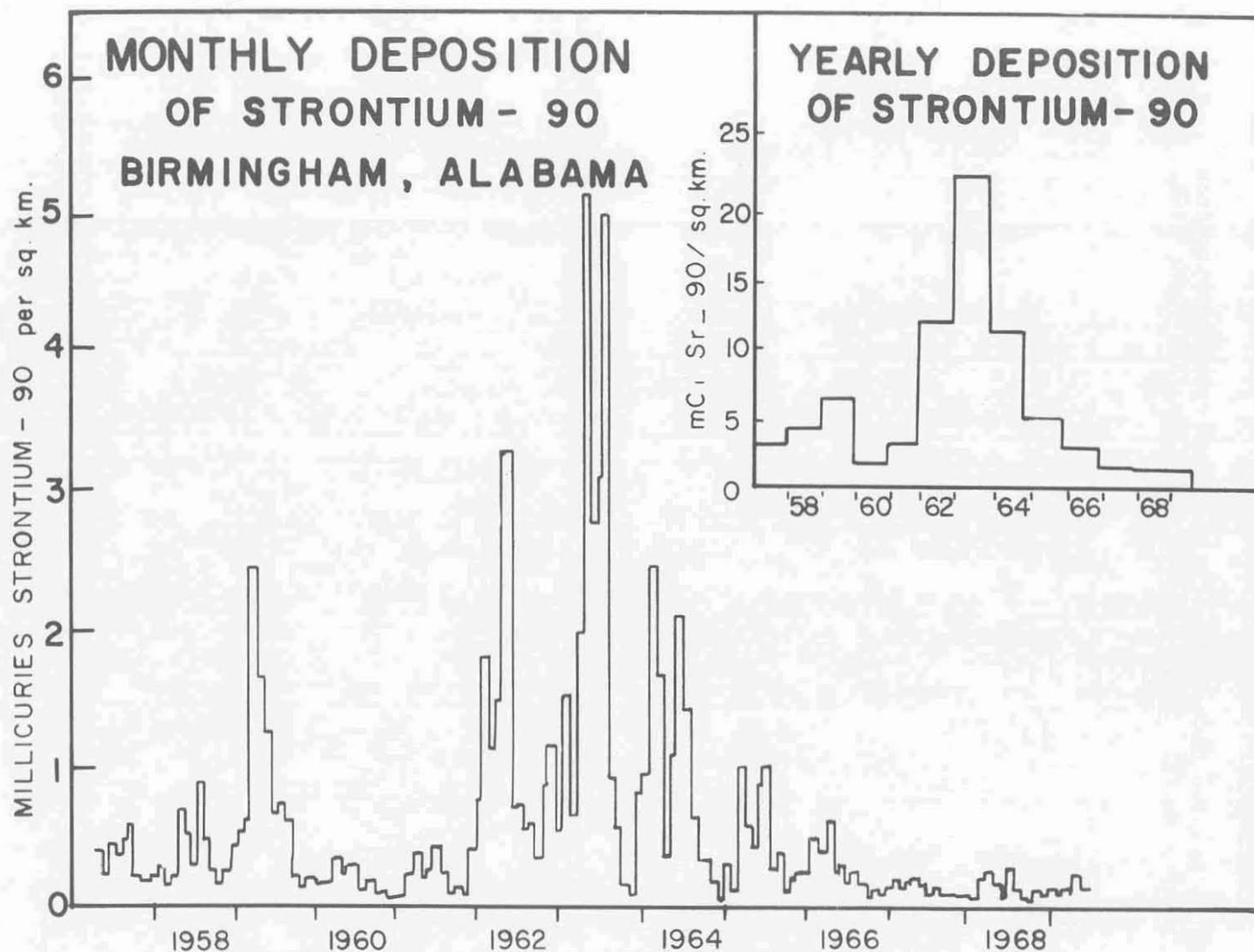


Figure 1.--The monthly and yearly distribution of fallout strontium-90 at Birmingham, Alabama.

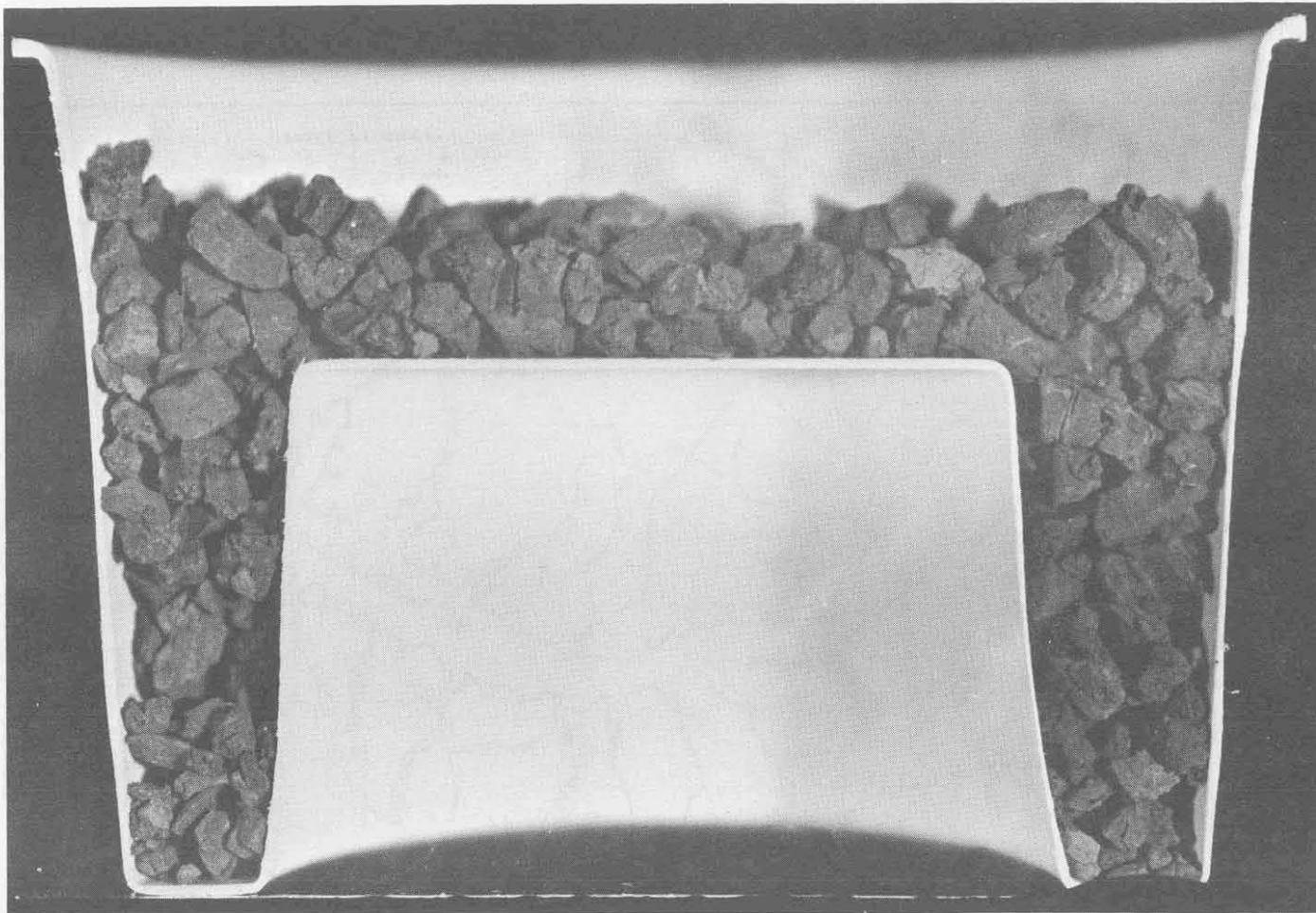


Figure 2.--Cutaway view of kydex Marinelli beaker showing soil around circumference and across the face of the NaI crystal. Marinelli beaker, with soil packed as illustrated, is placed over the 10 x 12.5-cm NaI(Tl) crystal for counting.

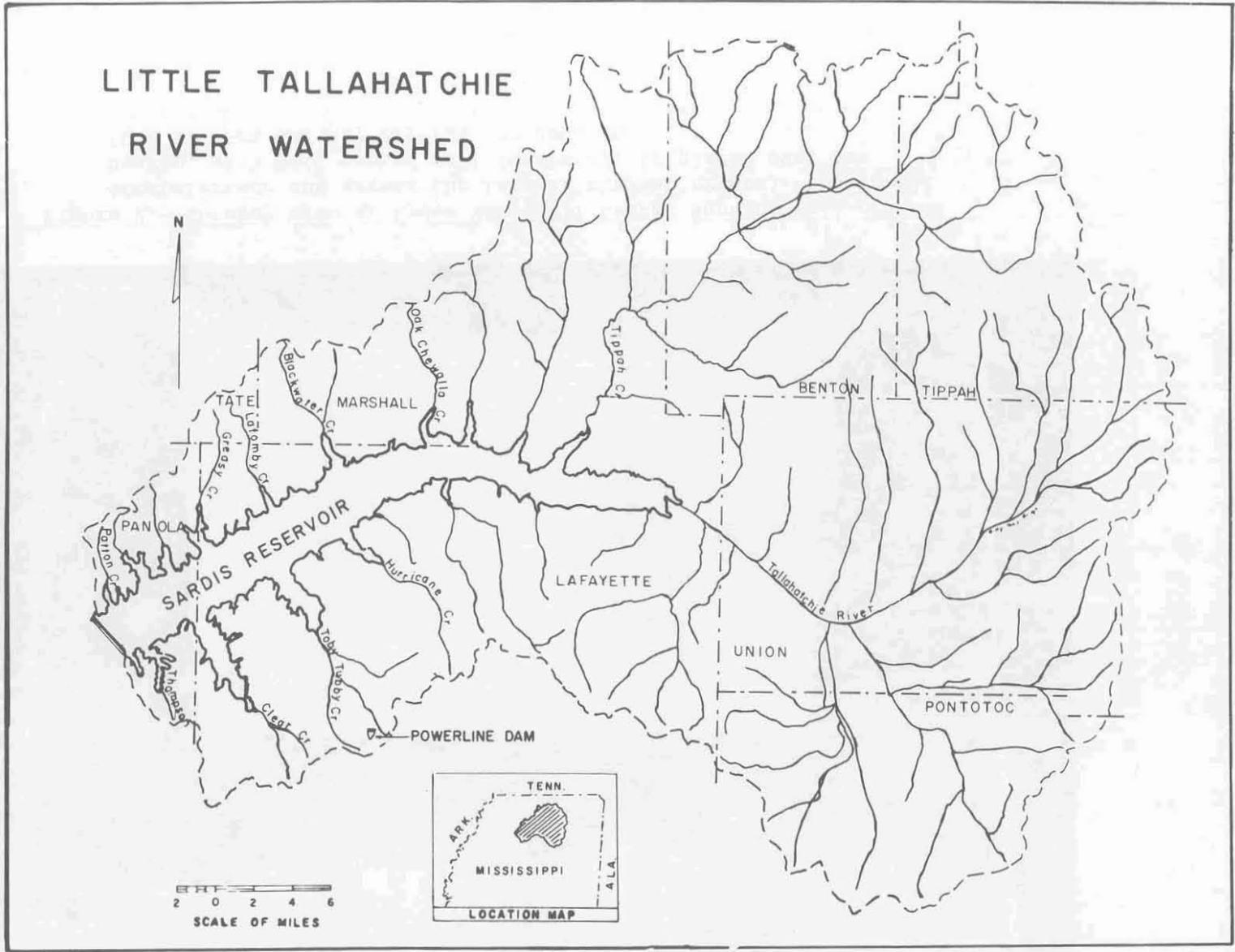


Figure 3.--Map of the Little Tallahatchie River Watershed and Sardis Dam and Reservoir.

Table 1. Concentration of fallout cesium-137 in soils under different vegetative cover, Power Line Dam, Lafayette County, Mississippi, 1969

Cover	Cesium-137 Concentration			
	Depth of sample, centimeters			
	0 - 2.5	2.5 - 5.0	5.0 - 7.5	7.5 - 10.0
	nanocurie/kilogram (millicurie/hectare)			
Oak A	5.58	0.67	0.20	0.09
	(1.27)*	(0.21)	(0.07)	(0.03)
Oak B	6.31	0.84	0.30	0.16
	(1.27)	(0.28)	(0.11)	(0.06)
Oak C	5.02	0.60	0.19	0.11
	(1.38)	(0.28)	(0.07)	(0.05)
Pine A	5.00	0.60	0.15	0.13
	(1.26)	(0.22)	(0.06)	(0.05)
Pine B	4.79	0.66	0.27	0.14
	(1.17)	(0.27)	(0.11)	(0.10)
Pine C	4.40	0.67	0.25	0.13
	(1.13)	(0.24)	(0.10)	(0.05)
Grass A	3.20	0.90	0.36	0.14
	(0.89)	(0.42)	(0.10)	(0.10)
Grass B	3.80	1.57	0.62	0.32
	(0.85)	(0.67)	(0.27)	(0.14)
Grass C	3.32	1.47	0.61	0.20
	(0.95)	(0.71)	(0.24)	(0.08)

* To convert mCi/ha to mCi/acre, divide by 2.471. The result will be the activity per acre-inch. Multiply mCi/ha by 100 to find mCi/km².

Table 2. Distribution of fallout cesium-137 in sediment profiles taken from Sardis Reservoir, Mississippi, 1969

Depth (cm)	Cesium-137 Concentration (nanocuries/kilogram)					
	Coontown A 0.3 mi. from boat ramp	Coontown B 1.2 mi. from boat ramp	Coontown C 1.5 mi. from boat ramp	Clear Creek Sandy delta	Sardis Dam Park, east boat ramp	Hurricane Creek, 0.5 mi. from boat ramp
0 - 2.5	2.32	1.33	0.92	0.24	0.53	3.25
2.5 - 5.0	0.50	2.12	1.01	0.18	1.11	5.40
5.0 - 7.5	0.19	1.91	1.11	0.15	1.57	6.00
7.5 - 10.0	0.20	2.61	2.49	0.13	0.83	2.25
10.0 - 12.5	0.17	1.53	2.45	0.06	0.53	1.77
12.5 - 15.0	0.17	0.97	2.13	0.03	0.17	1.74
15.0 - 17.5	*	1.06	1.09	0.02	0.05	2.02
17.5 - 20.0		0.92	1.03	0.01	0.06	1.49
20.0 - 22.5		*	0.89	0.02	0.04	0.87
22.5 - 25.0			0.84	*	0.06	0.33
25.0 - 27.5			1.03		0.04	0.14
27.5 - 30.0			0.71		0.03	0.16
30.0 - 32.5			0.22		*	*
32.5 - 35.0			0.15			
35.0 - 37.5			0.10			
37.5 - 40.0			0.03			

* Depth of sediment sampled at location.