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Mound Laboratory

Environmental Plutonium Study

1974

D. R. Rogers

September 15, 1975

MOUND
Miamisburg, Ohio 45342

operated by
MONSANTO RESEARCH CORPORATION
a subsidiary of Monsanto Company

for the
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MONSANTO RESEARCH CORPORATION
A Subsidiary of Monsanto Company

MOUND LABORATORY
Miamisburg, Ohio 45342
operated for

UNITED STATES ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

U.S. Government Contract No. E-33-1-GEN-53

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SUMMARY

In 1974, Mound Laboratory found that the sediment in certain waterways near the laboratory site appeared to exhibit plutonium-238 concentrations higher than the expected baseline levels. As a result, Mound Laboratory initiated a comprehensive environmental plutonium-238 study to determine the full extent of the contamination, the cause and mechanisms of the release, and the health and safety impact of these deposits on the public.

During the plutonium-238 environmental survey program, over 1750 soil, sediment, biota, water, and air samples were collected in the off-site areas and analyzed for plutonium-238. From these data, it was determined that about 5.2 curies of plutonium-238 are deposited in these waterways, mostly buried under up to 3 ft of sediment.

The plutonium-238 was found to be strongly sorbed and fixed onto the sediment. Autoradiographic analysis indicated very little, if any, particulate forms of plutonium. The solubility of the plutonium/sediment in the natural surface water is very low; only about one part per one hundred thousand parts of the plutonium is soluble in canal water. The maximum concentration in the water sampled from the waterways is about 0.00001 nCi/g. The highest subsurface sediment concentration is 4.56 nCi/g at a 3 or 4 ft depth in a localized area. The plutonium-238 concentration in samples of the biota was found to be very low.

The plutonium-238 concentrations in land areas contiguous to the waterways are at or below baseline levels ($<$0.0004 nCi/g).

An intensive investigation identified the cause of the plutonium-238 deposits and the mechanisms of the release, transport and deposition into these off-site waterways. Experimental laboratory studies and field observations were used to verify these mechanisms.

In January, 1969, an underground pipeline carrying plutonium-238 waste solution from the Plutonium Processing (PP) Building to the Waste Disposal Facility (WD) ruptured. Acidic waste solution containing plutonium-238 was released to the soil adjacent to the pipe. The plutonium was quickly and strongly sorbed by the soil where it was immobilized. During the excavation and repair operations, when the contaminated soil was most susceptible to erosion, the weather warmed, and intense rain was experienced for two days. This heavy rain eroded the exposed surface of the contaminated soil causing the soil particles to be carried off-site. These erosion products, suspended in the moving water, settled according to normal sedimentation processes in the waterways adjacent to Mound Laboratory. Water sampling performed during this occurrence failed to detect this movement because the plutonium was in the sediment.

The health and safety aspects of the plutonium-238 sediment deposits were evaluated under the prevailing conditions and under credible worst-case future conditions.

The evaluation under prevailing conditions was performed considering the measured concentrations of plutonium-238 in air, water, vegetation, fish, soil and sediment and the physical conditions and circumstances prevalent in this specific area. The air and water data were compared with existing Radioactivity Concentration Guides (RCG) for plutonium-238. The biota was evaluated by determining the amount of each of the materials which would have to be ingested to receive 1/70 of a permissible body burden per year. It was concluded that the air and water concentrations are at safe levels (substantially below RCG). Due to the physical and chemical properties of the area and the sediment, the present air and water concentrations are not likely to be significantly higher in the future under prevalent conditions. The amount of the other materials which would have to be ingested to lead to a potential uptake of 1/70 of a permissable body burden per year is too large to be of concern. Overall, these plutonium-238 deposits, therefore, were evaluated and found to present no hazard to the public under the prevalent conditions which presently existed in this area.

Anticipating that future conditions may change, comprehensive pathway analyses were performed, assuming credible worst-case conditions associated with each of the several ingestion, absorption, and inhalation pathways considered. From these pathway analyses, *Sediment Concentration Decision Guides* were estimated using methods and philosophies similar to those used for RCG deviations. The maximum available, potentially available, and worst-case credible plutonium-238 sediment/soil concentrations found in and around these waterways were compared with these decision guides.

On the basis of this analysis, the concentrations of plutonium in the sediment are not expected to present a hazard to the public in the future.

I. INTRODUCTION

Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Energy Research and Development Administration (ERDA), formerly the U. S. Atomic Energy Commission (AEC). The laboratory, constructed in 1947, is located in Miamisburg, Ohio, overlooking the old abandoned Miami-Erie Canal and the Great Miami River, as shown in Figure 1. Since 1959 the Laboratory has been the major research and development site for fabrication of radioisotopic heat sources used for peaceful space and terrestrial applications.

Radioisotopic Thermoelectric Generators (RTG) powered by Mound Laboratory plutonium-238 heat sources have been used to provide electrical energy for many space satellites from the SNAP-3 RTG in 1961 to the SNAP-19 RTG in the Nimbus Weather Satellite in 1969 and the Transit Navigational Satellite in 1972.

Heat sources provided heat and electrical power for the scientific experimental packages left on the moon during the Apollo missions (1967-1972) as shown in Figure 2. Recently, Mound-prepared plutonium-238 heat sources were used to provide electrical energy for the Pioneer-Jupiter Missions (1972 and 1973) and will be used for the Viking Mars Lander in 1975 and 1976.

Experimental plutonium-238 sources also have been developed to supply energy for experimental heart pacemakers shown in Figure 3, and experimental artificial hearts. Miniature experimental plutonium-238 X-ray sources were used to rapidly determine minute amounts of lead in the blood of children by a micro X-ray fluorescence technique.

Because plutonium is a potentially hazardous material, extensive precautions have been exercised since plutonium operations began at Mound Laboratory to carefully maintain strict control of the plutonium and to prevent significant amounts from entering the environment. These precautions include elaborate facility and equipment design criteria, scientific expertise, experience, personnel training, management and operational control systems, and environmental monitoring.

In spite of these precautions, in early 1974, core samples collected and analyzed by the Mound Laboratory Environmental Monitoring Group as a part of an upgrading program, established that plutonium-238

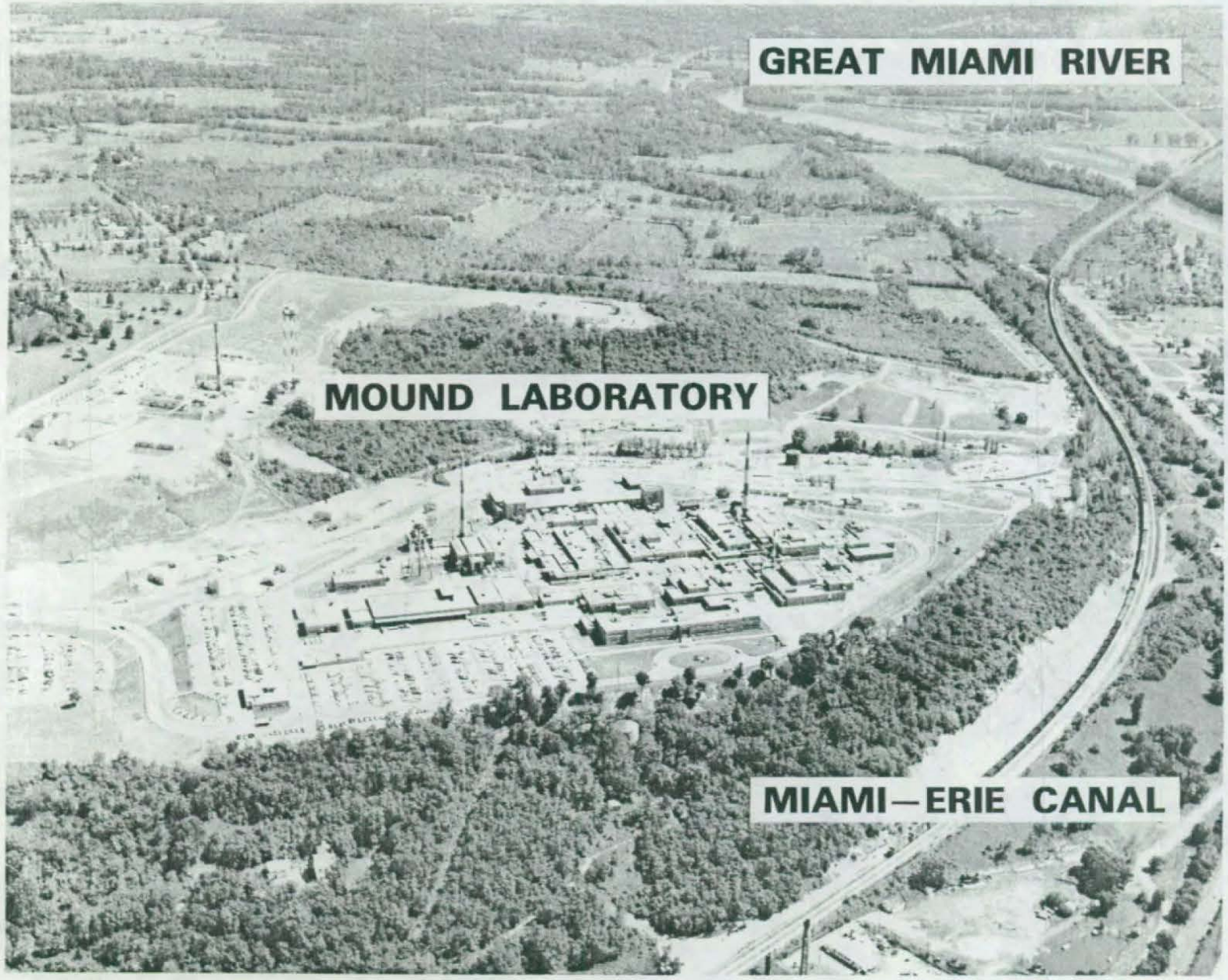


FIGURE 1 - Aerial view of Mound Laboratory looking South.

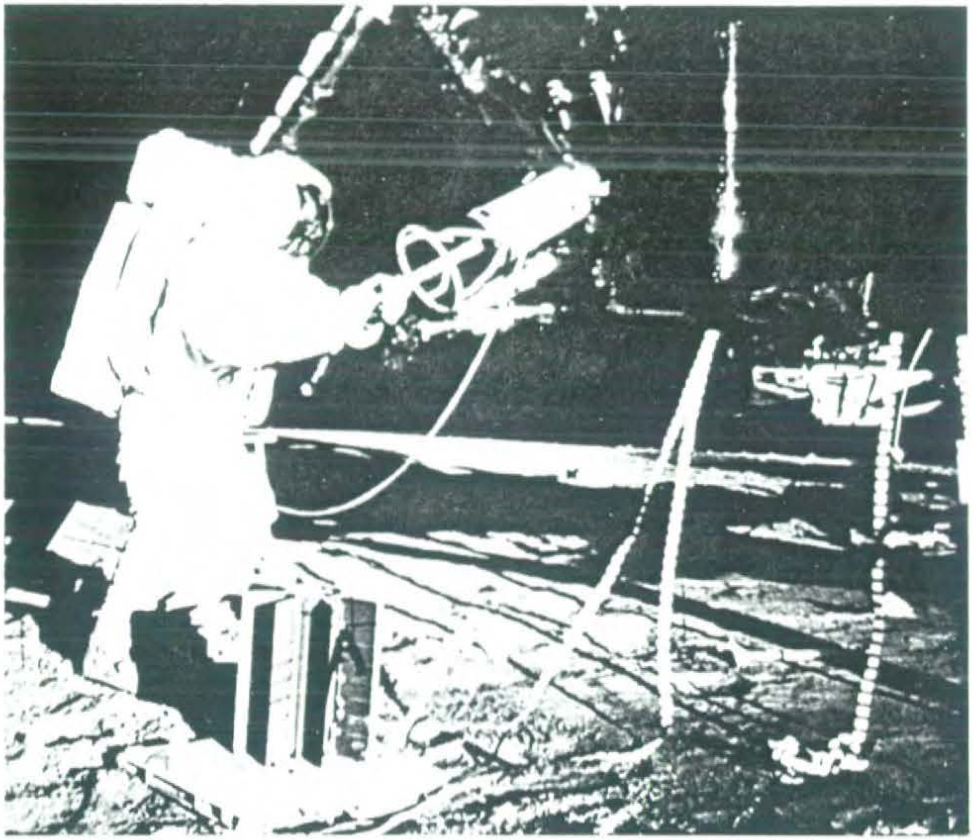


FIGURE 2 - ^{238}Pu SNAP-27 Heat Source being removed from the LEM on the lunar surface on the Apollo-12 mission in 1969.

FIGURE 3 - ^{238}Pu powered experimental model Cardiac Pacemaker.

concentrations in the sediment of certain waterways adjacent to the site were above the baseline levels expected (<0.0004 nCi/g). These plutonium-238 deposits presented no immediate hazard to the general population in the area as indicated by the air and water concentrations which were well within accepted Radioactivity Concentration Guides (RCG) for plutonium-238. These preliminary findings were reported to ERDA, to local, state and federal government representatives and agencies and to the public through the news media.

Mound Laboratory launched the Mound Laboratory Environmental Plutonium Study to fully investigate the extent of the contamination, the source of plutonium, how it was transported and deposited in these waterways and what potential hazards these deposits might present to the general public now and in the future. A special scientific and management team was organized to participate and direct this study. The members of this team and their primary responsibilities are listed in Table 1.

A Monsanto Research Corporation advisory group, consisting of Mound Laboratory department directors, also reviewed plans, goals, schedules, and accomplishments during the programs.

A number of outside consultants were used to augment and review plans and help interpret the data obtained. The consultants' specialties are:

- Hydrology
- Sedimentation
- Soil Chemistry
- Geology
- Ecology
- Biology

The consulting organizations are:

- Georgia Institute of Technology
- Emory University
- Wright State University
- Dames and Moore Company
- United States Geological Survey
- Miami Conservancy District
- United States Department of Agriculture.

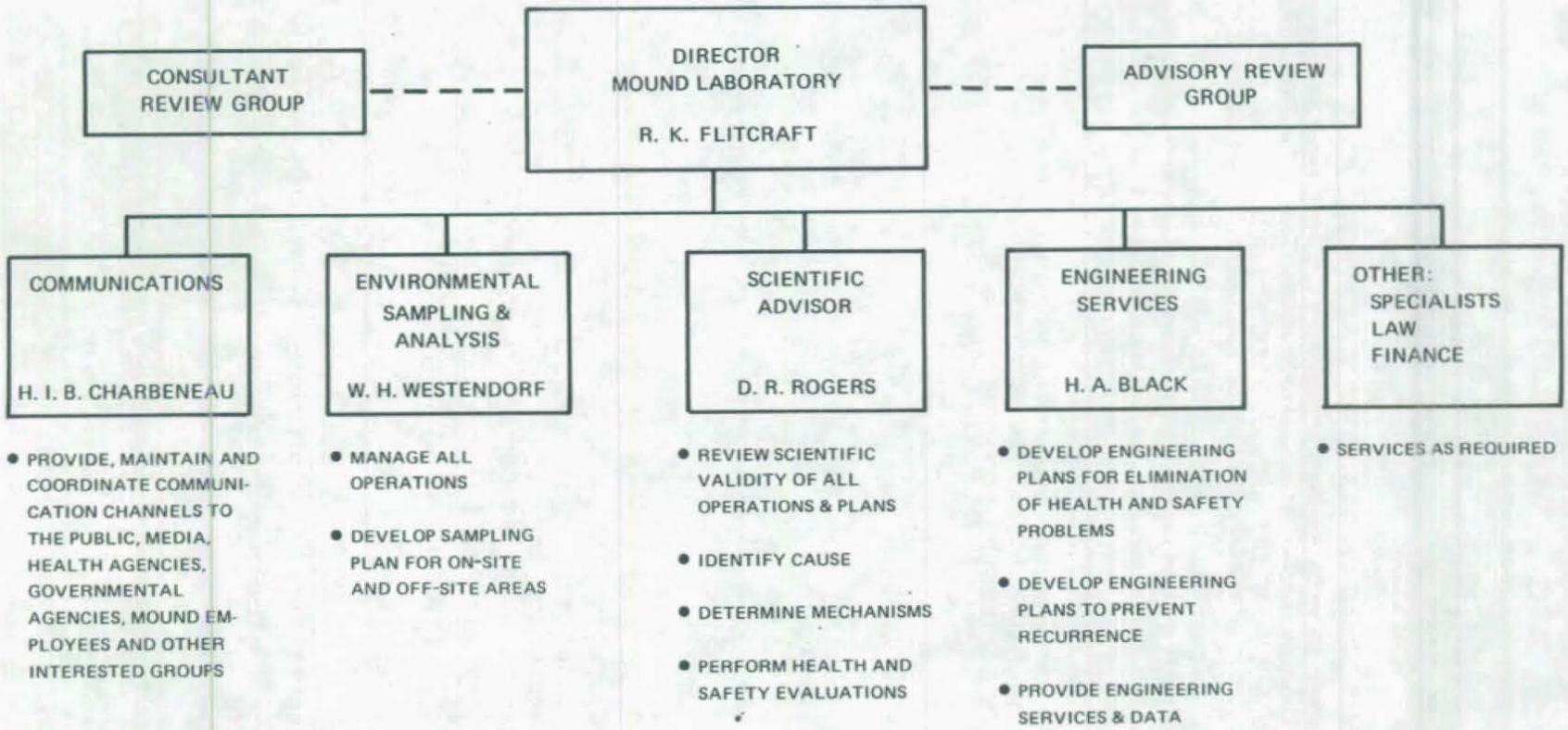
The overall objectives of the Mound Laboratory Environmental Plutonium Study were to:

- Perform a comprehensive Plutonium-238 *Environmental Survey* of the entire affected area to define the spatial extent, distribution, concentrations, and total inventory of the plutonium in and around these waterways;

- Perform an intensive and thorough *Investigation* and *Cause Analysis* to determine the source of the plutonium, the cause of the release, and the mechanisms of transport and deposit in the waterways;

Table 1

MOUND LABORATORY ENVIRONMENTAL PLUTONIUM STUDY TEAM AND RESPONSIBILITIES



- Perform a comprehensive *Health and Safety Analysis* to evaluate credible potential health hazards to the general public.

During the course of this program, interim reports were periodically submitted and reviewed with health, environmental and governmental agencies to keep them informed as the environmental results became available.

The findings of the Mound Laboratory Environmental Study, essentially completed in September 1974, were orally presented to interested local, state and federal government officials, and health and environmental agencies prior to a press conference held at Mound Laboratory on October 2, 1974.

The purpose of this report is to summarize the results and conclusions of the Mound Laboratory Environmental Plutonium Study, concentrating on the details of the health and safety analyses which were performed.

II. TOPOGRAPHY, HYDROLOGY AND DESCRIPTION OF THE AREA

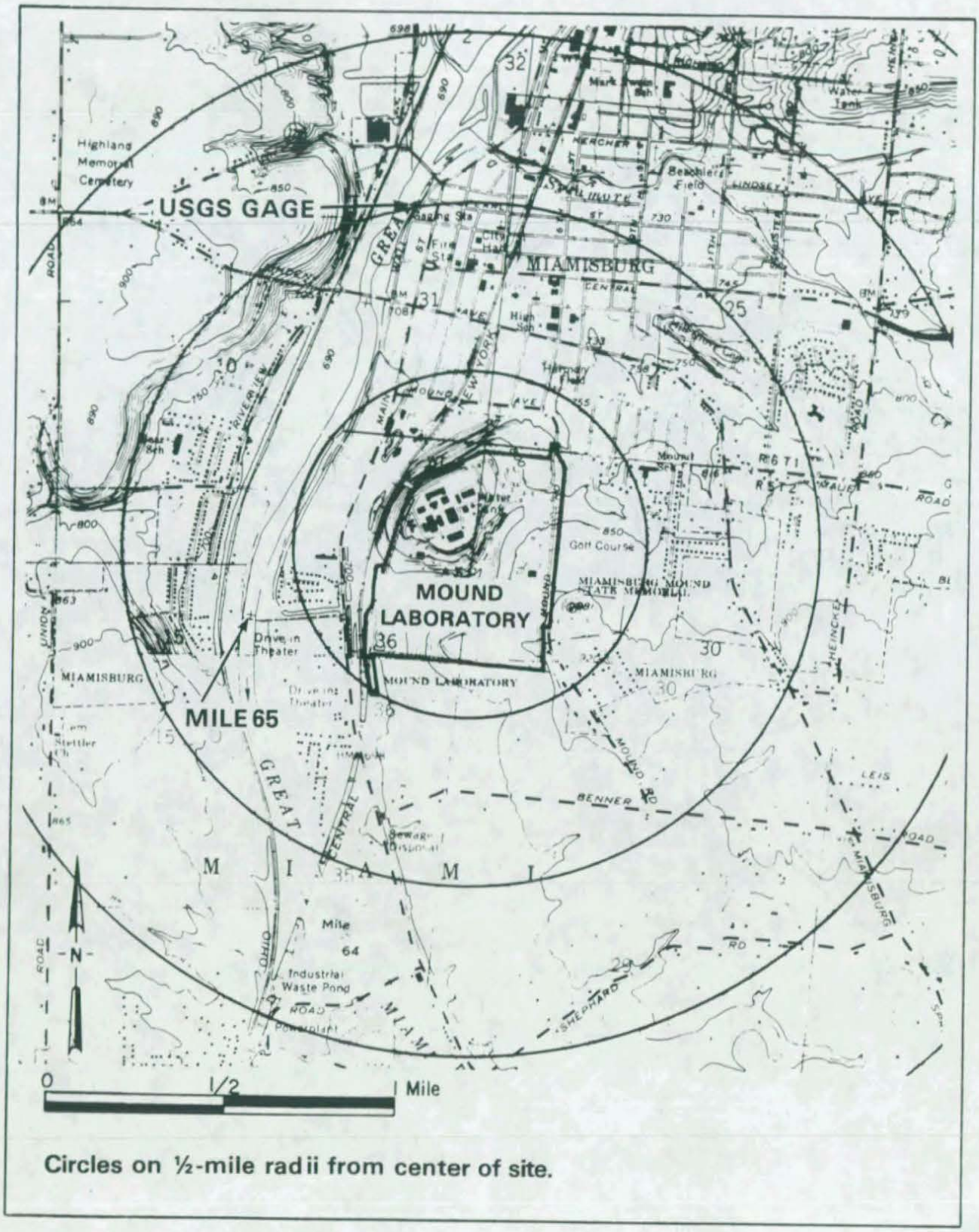
Mound Laboratory is situated on a topographically high area overlooking Miamisburg, the Great Miami River, and the river plain area to the west. Figure 4 shows the topography in the general area.

The 180-acre laboratory site is basically located on two hills of about 880 ft elevation and a valley between with an elevation of about 705 ft. The site topography and facilities are shown in Figure 5. The Plutonium Processing Facility (SM-PP) is on the southeast hill while the Plutonium Research Facility (R Building) and the Waste Disposal (WD) and Sewage Disposal (SD) facilities are on the northwest hill.

A drainage ditch flows continuously through the on-site valley generally from east to west and is the major surface hydrological artery for carrying surface run-off water from the site (Figure 6). This drainage ditch flows off the site on the western side through a culvert under a raised railroad grade which runs generally north-south along the western boundary of the laboratory. Since 1971, Mound Laboratory has had an automatic flow measurement weir and an environmental sampling station on the drainage ditch just before it flows off-site (Figure 7). After the drainage ditch passes under the railroad grade, it flows to an abandoned section of the old Miami-Erie Canal. Part of the water is diverted north through pipes under an earthen dam into the North Canal while the remainder of the water flows around a make-shift dam into the South Canal (Figures 8, 9, and 10). These two sections of the old Miami-Erie Canal extend north and south (2500 ft north and 2700 ft south) of the drainage ditch/canal confluence as shown in Figure 11. The canal bed is approximately 40 ft wide and 5 to 10 ft deep relative to the bank height. It was constructed in the 19th Century as a commercial transportation barge canal and abandoned in 1913.

The North Canal, immediately north of the earthen dam, is a high sedimentation area and contains 5 ft or more of sediment. Turbulent water, heavily laden with erosion products from the drainage ditch, passes through the pipes in the earthen dam and encounters calm water and a heavy growth of cattail reeds which tends to cause laminar flow (Figure 12). Under the less turbulent flow condition, a large percentage of the erosion products settle out and deposit. In a short distance, ~100 to 200 ft, the canal gets wider and deeper. At the northern end of the North Canal (Figure 13), the water is again diverted by an earthen dam and an underground pipe into the South Pond (Figure 14). The water flows north from the South Pond (which consists of a north and south basin) and into the North Pond (Figure 15) where the excess is carried off through a standpipe drain into the underground Mound Street storm sewer which carries the water directly to the river.

Under very high flow conditions, water in the North Canal flows through a notch in the earthen weir and can be released to the Mound Street storm sewer directly by opening a sewer gate at the north end of the canal (Figure 13). The North Canal and ponds remain under water at all times.



Circles on ½-mile radii from center of site.

FIGURE 4 - Topology of Mound Laboratory and neighboring areas.

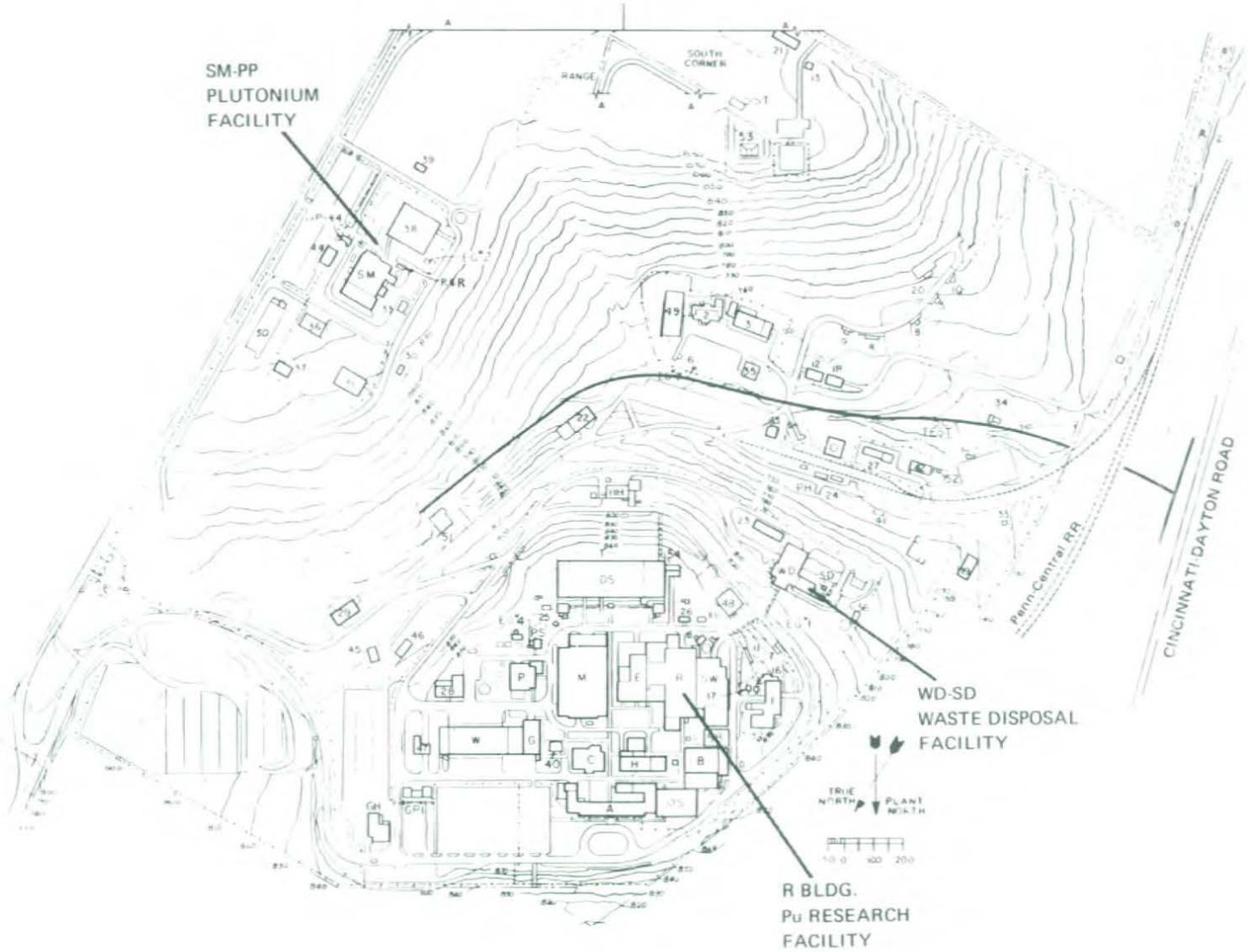


FIGURE 5 - Topology of Mound Laboratory site.

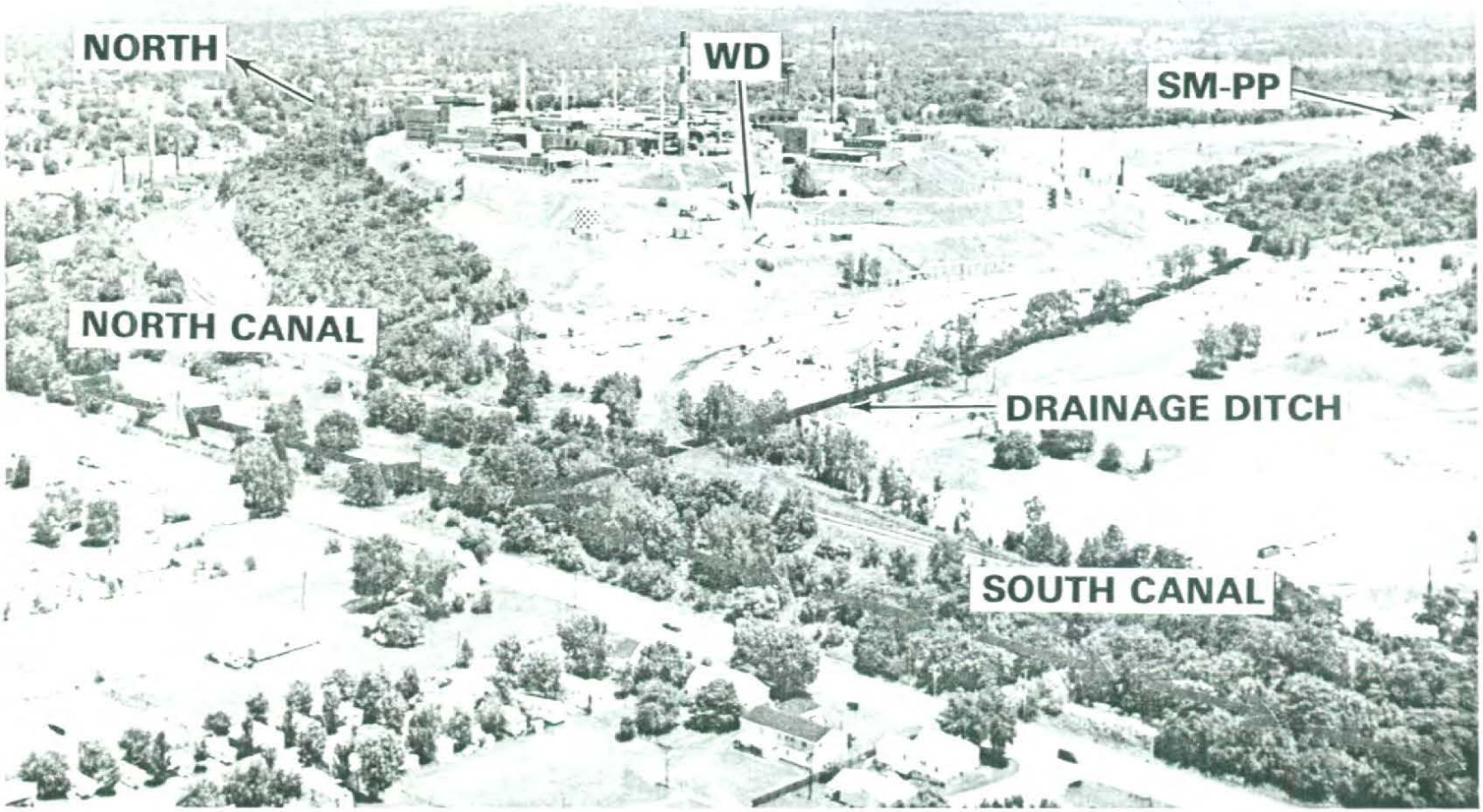


FIGURE 6 - Aerial View (from the west) of Mound Laboratory showing major surface hydrological arteries.

FIGURE 7 - Environmental monitoring station and automatic flow measurement weir on the drainage ditch.

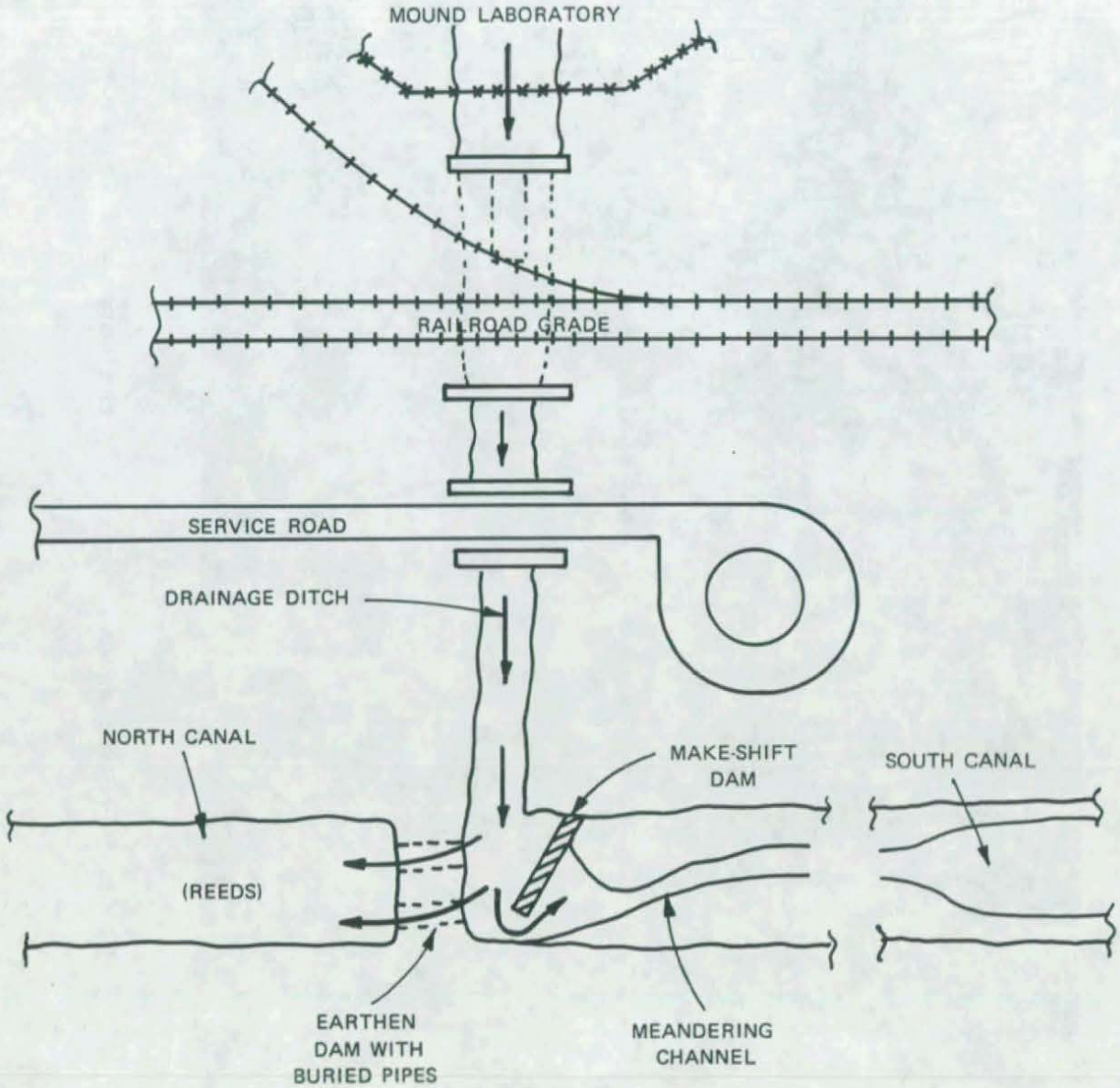


FIGURE 8 - Schematic diagram of the drainage ditch/Miami-Erie Canal confluence area.

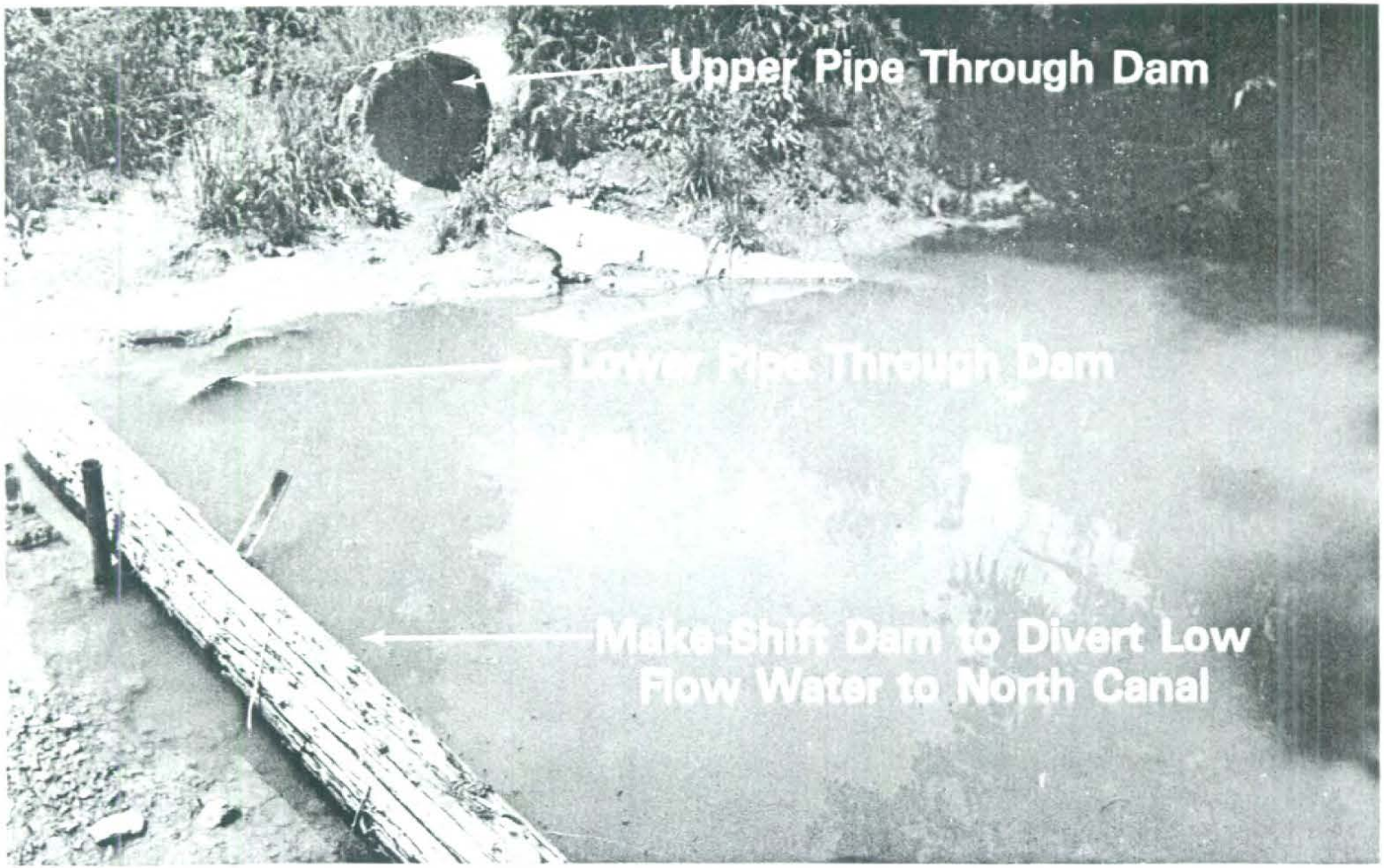


FIGURE 9 - Earthen dam separating the North and South Canals.

FIGURE 10 - South Canal viewed from the top of the earthen dam.

FIGURE 11 - Aerial view (from the north) of off-site waterways.

FIGURE 12 - North Canal, viewed from the top of the earthen dam, showing growth of cattail reeds.

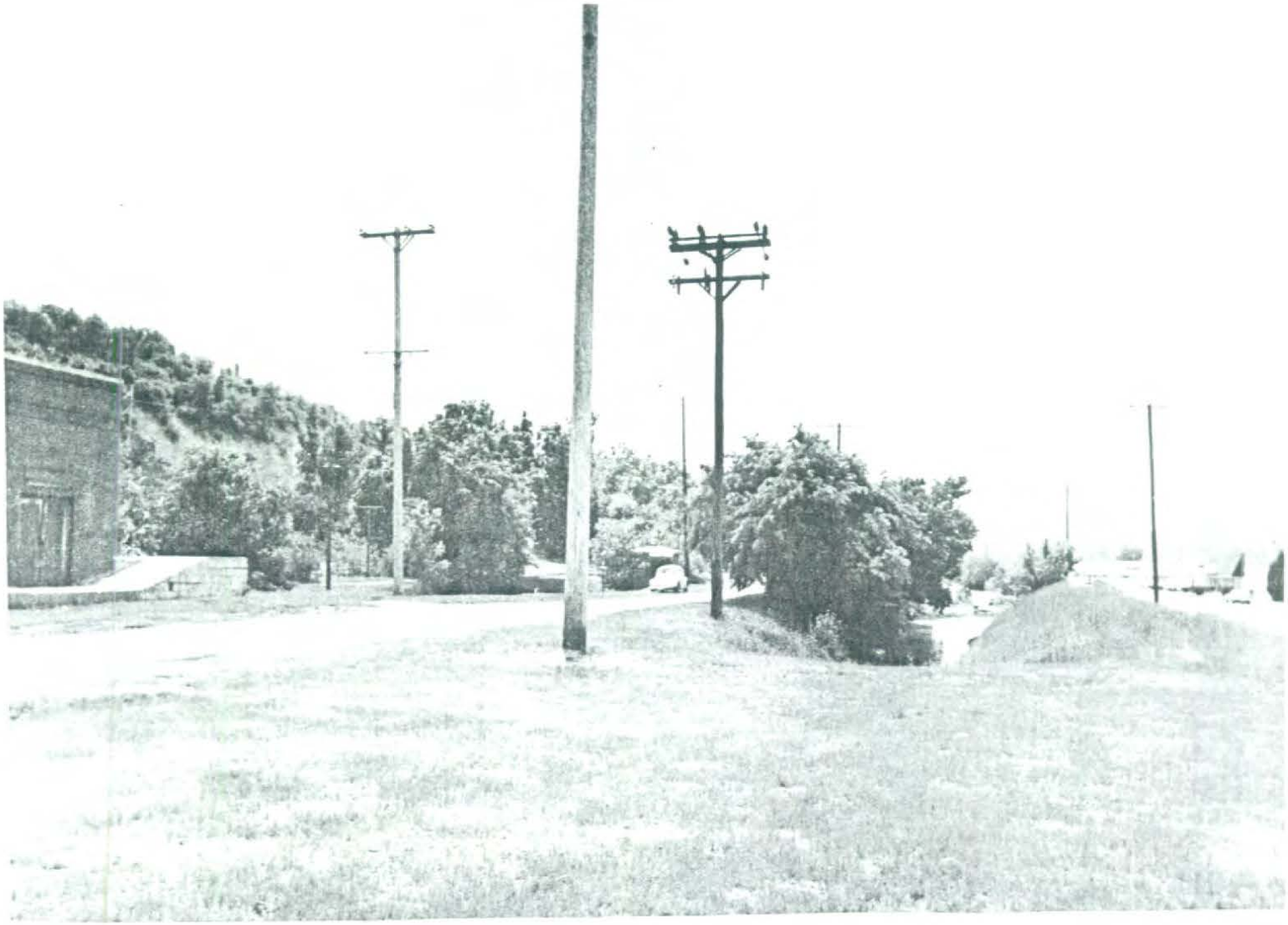


FIGURE 13 - North Canal viewed from the extreme northern end.

FIGURE 14 - South Pond viewed from the south.

FIGURE 15 - North Pond viewed from the northeast.

The South Canal is essentially overgrown with vegetation except for a meandering channel within the old canal bed (Figure 10). Under normal flow conditions, only a small amount of water flows past the make-shift dam into the South Canal and this water is carried by the meandering channel. Under high flow conditions, the entire South Canal channel fills and contains the water flow. There is a high sedimentation area which extends from approximately 600 to 1500 ft south of the earthen dam. This area is wide and also has a heavy stand of cattail reeds which impedes the water flow and induces laminar flow conditions. At the extreme south end of the South Canal, the water flows under a railroad bridge, then over a concrete weir spillway (Figure 16) to a culvert under the Cincinnati-Dayton Road, then to an overflow creek which carries the water to the river (Figure 17).

The banks of the North and South Canals are high and quite definite except at the extreme southern end. In the section adjacent to the overflow weir, the bank to the east is lower. Water flows north to this area from a drainage ditch that extends to the south. During periods of heavy flow, water backs up and overflows the eastern bank and floods an area about 400 ft long and 30 ft wide. The grade of the land beyond this prevents further flooding. The area immediately behind the overflow creek spillway is considered only a moderate sedimentation area since it is already nearly full of sediment.

The overflow creek was used, when the canal was in operation, to carry the excess water from the canal during periods of high accumulation. This channel is 15 to 25 ft wide and is quite rocky. Its outfall on the river is immediately south of the railroad river bridge.

The area to the east of the North Canal, west of the raised railroad bed, and north of the drainage ditch, is a grassy area owned by the City of Miamisburg. Its present use is as a utility area for storage of street maintenance supplies. North of the canal pond area is a municipal swimming pool and an abandoned (Miamisburg) power plant. It is anticipated that this area will be improved to become a city park. The run-off hollow, a very narrow strip of land between the railroad grade and the Mound Laboratory site, is the Penn Central Railroad right of way.

The area south of the drainage ditch, east of the South Canal and west of the railroad grade, is an undeveloped strip of land that is heavily overgrown with vegetation. The area adjacent to the canal is owned by the Miami Conservancy District; the land further to the east is the Penn Central Railroad right of way. It is anticipated that this land will ultimately be improved and utilized as a park. All of the area from the drainage ditch to and beyond the overflow weir with contamination greater than 0.001 nCi/g is owned by the Miami Conservancy District.

The overflow creek flows across the Miamisburg Sewage Treatment Plant property. To the southeast is a large grass field used by the city for sanitary sewage sludge disposal. To the northwest is the raised railroad grade (Penn Central Railroad). It is anticipated that the treatment plant property will maintain its present use for a considerable time, but will be ultimately used as a park.

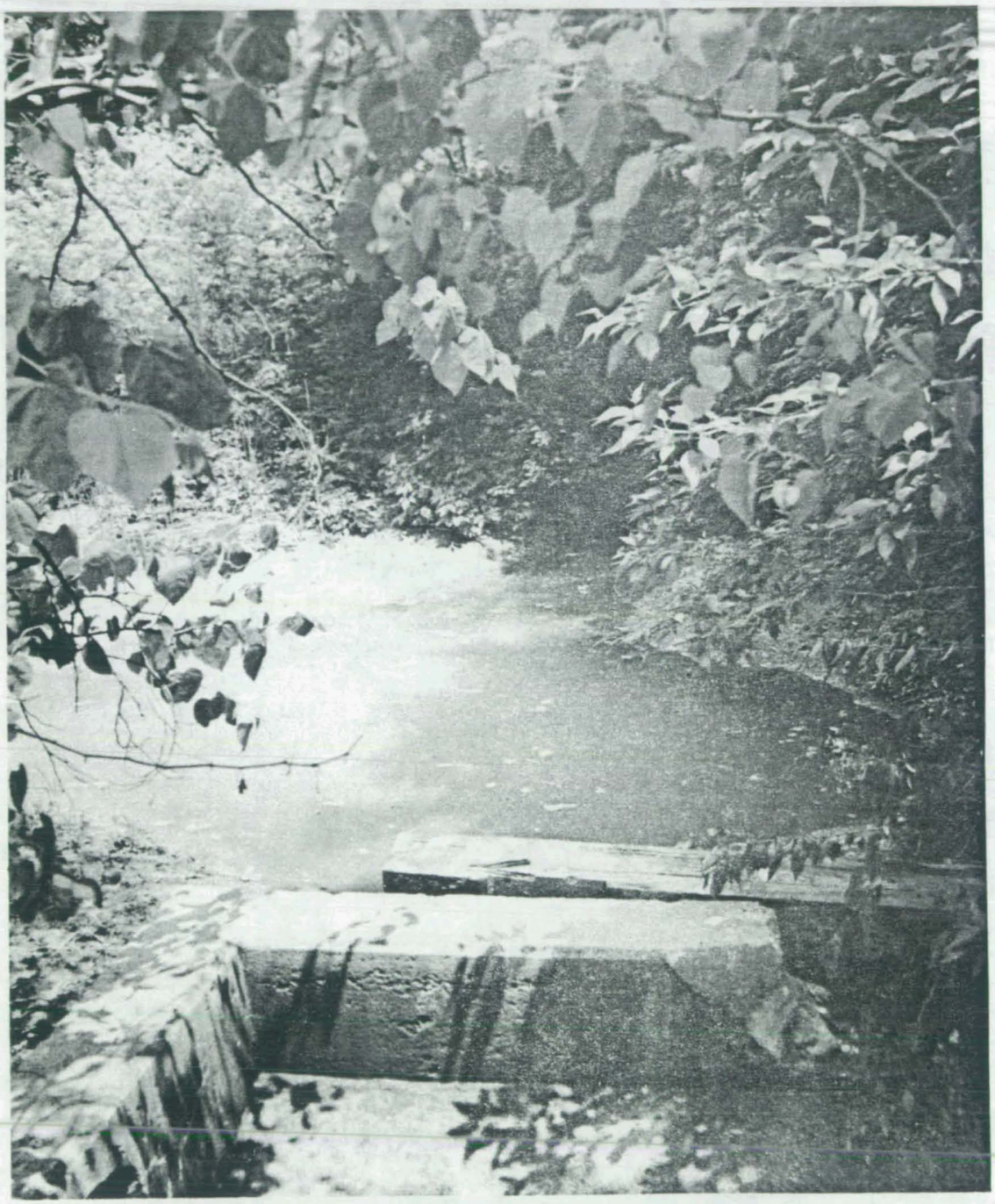


FIGURE 16 - Water in the South Canal as it enters the overflow creek as viewed from the Cincinnati Dayton Road.

FIGURE 17 - Overflow creek on the west side of the highway.

The Cincinnati-Dayton Road parallels the North and South Canals, approximately 50 to 75 ft to the west of these waterways. This is a two-lane paved road which essentially follows the river to Franklin, Ohio, and on to Cincinnati. The area between Cincinnati-Dayton Road and the river is relatively flat and grassy and is used presently as a commercial and residential section of the city. The area immediately adjacent to the river is owned by the Miami Conservancy District and includes grassy flood dikes and the flood plain between the dikes and the river.

III. ENVIRONMENTAL PLUTONIUM STUDY

A. Sampling and Analysis

Based on fragmentary information obtained by preliminary sampling, a comprehensive ^{238}Pu environmental sampling plan was developed for the waterways and adjacent areas to determine the full extent of the contamination. This proposed sampling plan was reviewed by the Monsanto Company, ERDA (formerly AEC), the Montgomery County Health Department, the Miami Conservancy District, the Ohio Department of Health, the Ohio EPA, the U. S. EPA, and the consultants from the USDA, universities and private companies. The plan called for plutonium-238 analysis of over 1340 samples of the following materials:

- Soil (core, plug and scoop samples)
- Sediment (core samples)
- Water (both canal and well water)
- Solids suspended in water
- Vegetation
- Fish
- Air

As was expected at the outset, the plan was modified subsequently as the data or other information became available and on the advice of expert consultants. In all, about 1750 samples were taken and analyzed for ^{238}Pu. These analytical results[1] have been reported previously to governmental, health and environmental agencies.

Figures 18, 19 and 20 show the major sediment core sampling locations in and around the drainage ditch, runoff hollow, North Canal, ponds, South Canal and overflow creek. At most of the sampling locations, multiple core samples were taken as shown in Figures 21 and 22. These multiple samples were taken to establish the distribution of concentration across the waterways and on the banks, and to establish local variability. The cores were generally 5 ft deep and were divided into five equal sections to determine concentration as a function of depth. Typical sampling techniques are shown in Figures 23 and 24. Additional scoop and 2-inch plug samples were taken at frequent distance intervals in the land areas near the waterways to determine the level of ^{238}Pu contamination on the surface.

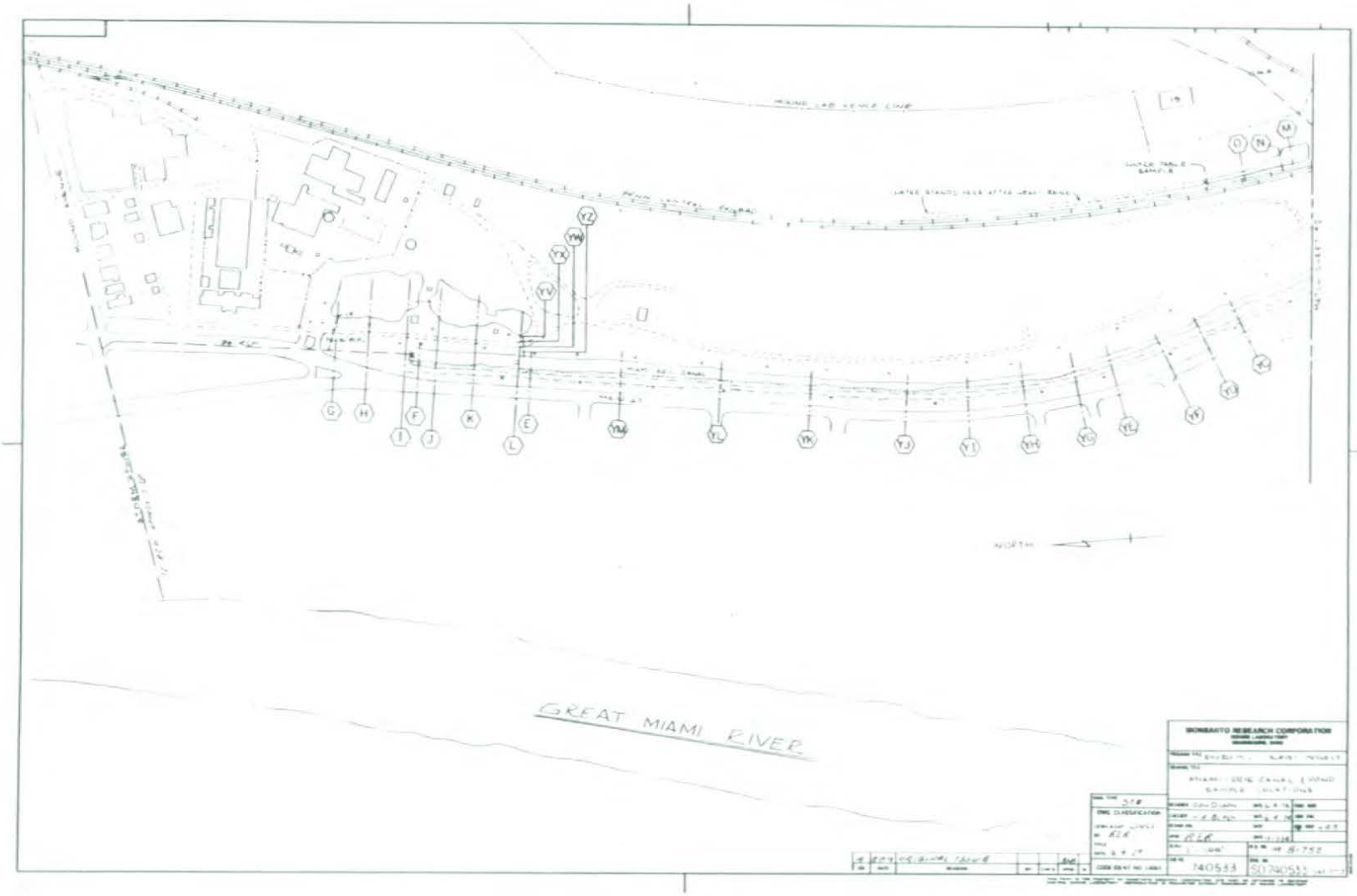


FIGURE 18 - Core sampling locations: North Canal and pond area.

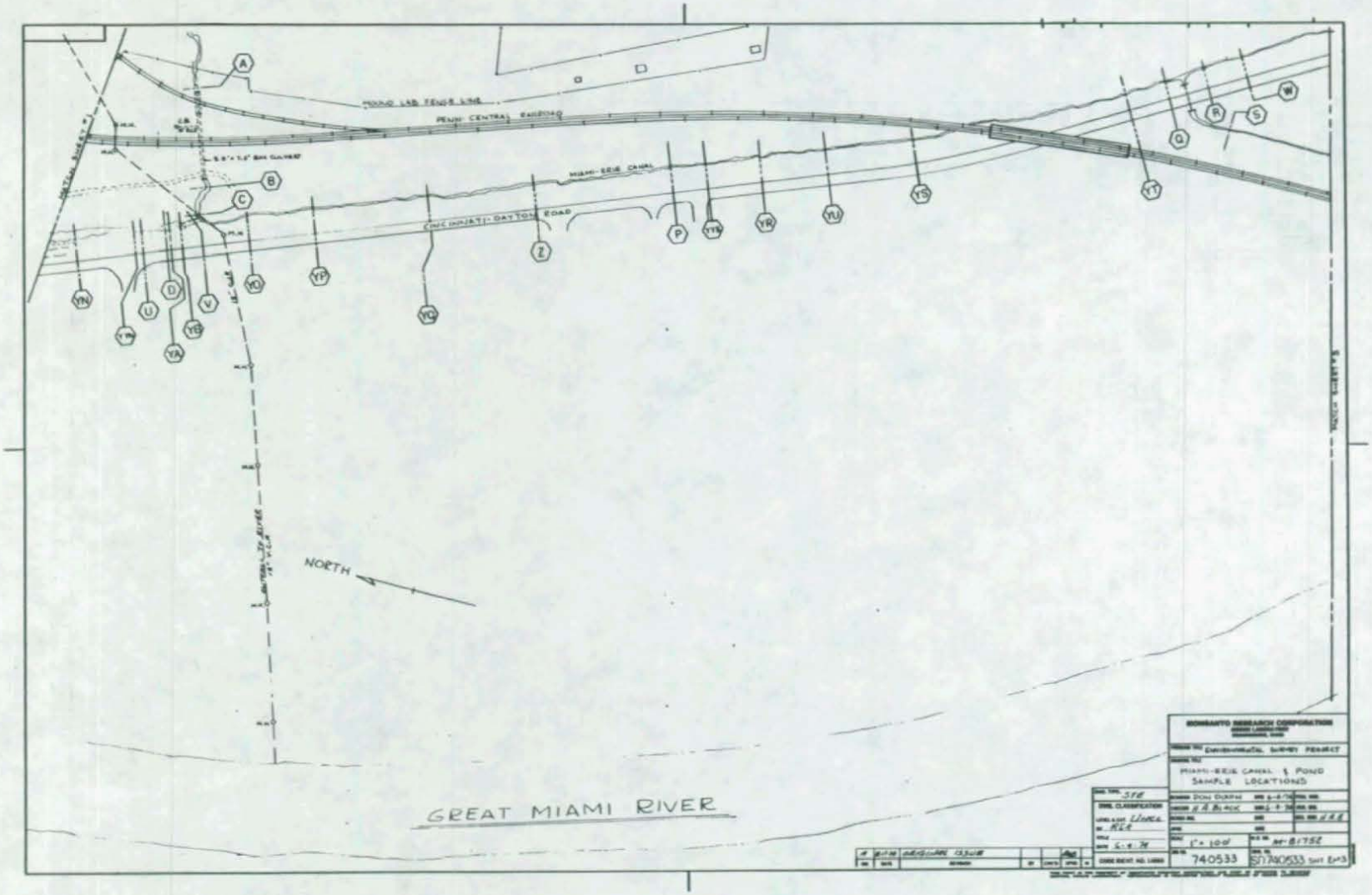


FIGURE 19 - Core sampling locations: drainage ditch and south canal area.

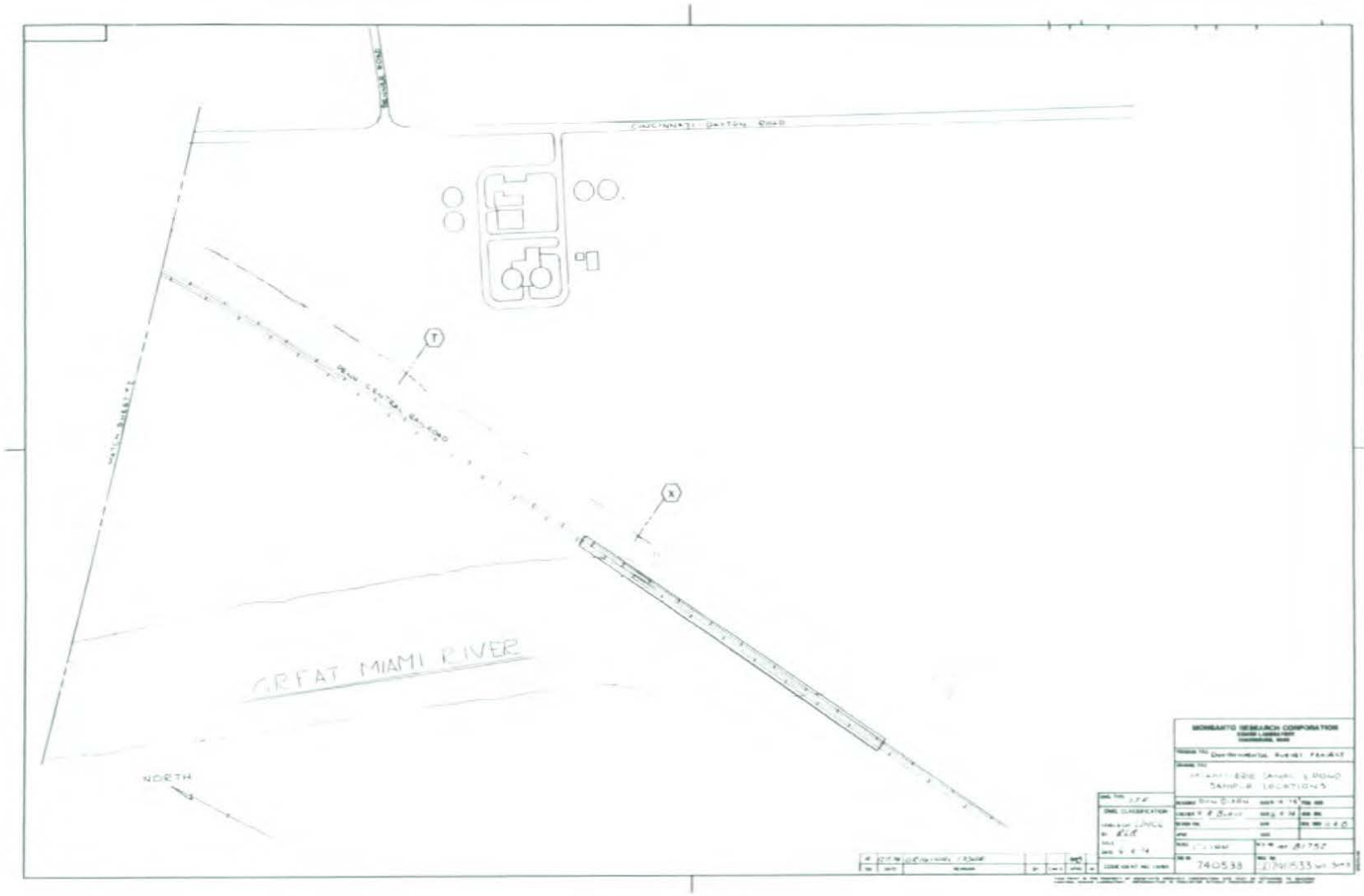


FIGURE 20 - Core sampling locations: overflow creek area.

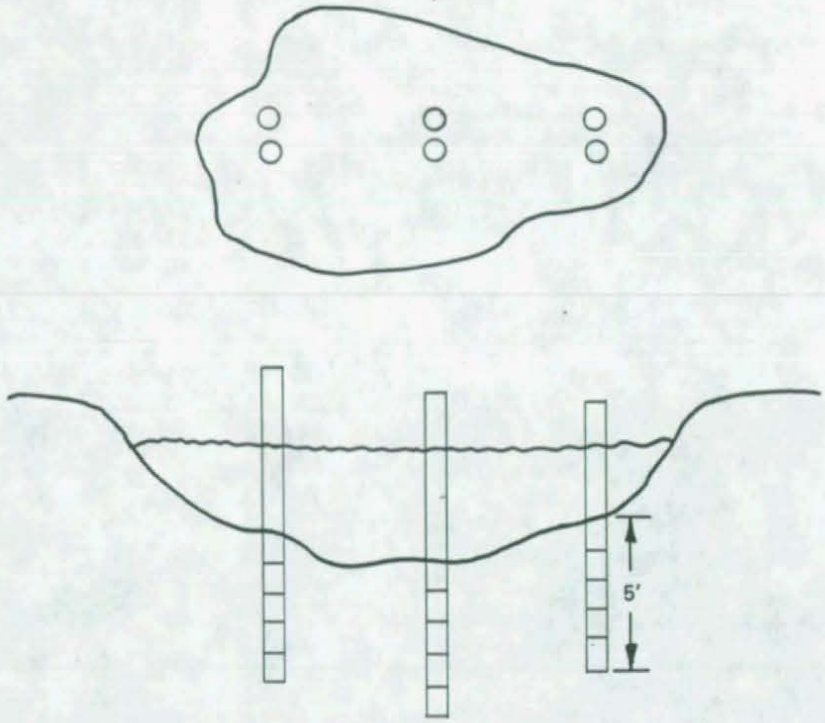


FIGURE 21 - Typical distribution of samples taken at pond location.

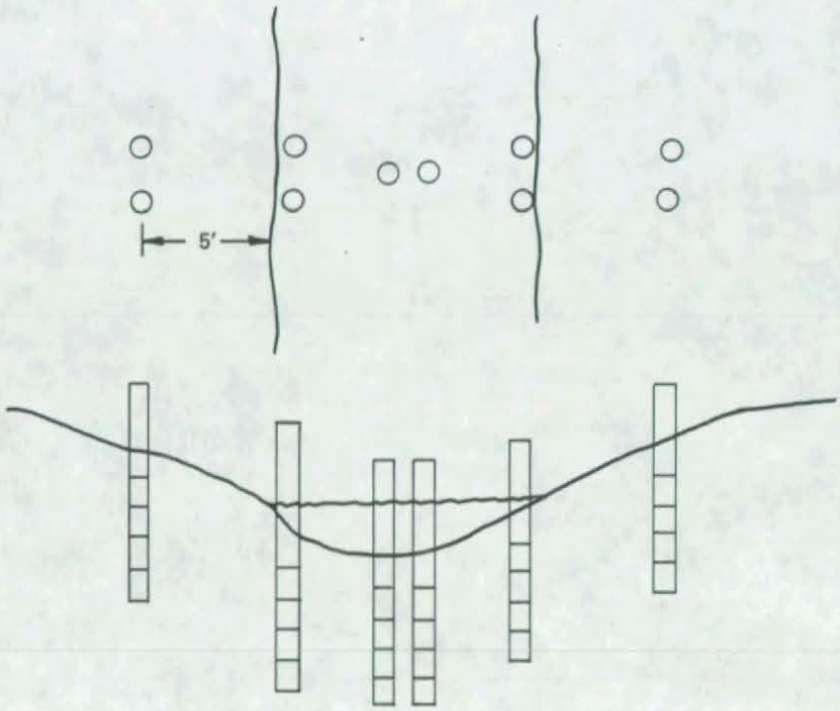


FIGURE 22 - Typical distribution of samples taken at canal locations.

FIGURE 23 - Sampling tube being carried to North Canal sampling site.

FIGURE 24 - Typical sampling techniques.

The Great Miami River sediment was core-sampled upstream from the laboratory and at several locations downstream for a distance of 10 miles. Core samples were collected near the Mound Avenue storm sewer (pond) outfall, near the outfall of the closed pipeline carrying effluent from the laboratory sewage and waste disposal operations to the river and near the overflow creek outfall. These sampling locations are shown in Figure 25. Typical spatial distribution of samples taken at a particular location are presented in Figures 26 and 27. The river core samples were collected by an independent company, Bowser Morner Testing Laboratory, Dayton, Ohio, utilizing a sampling rig shown in Figure 28.

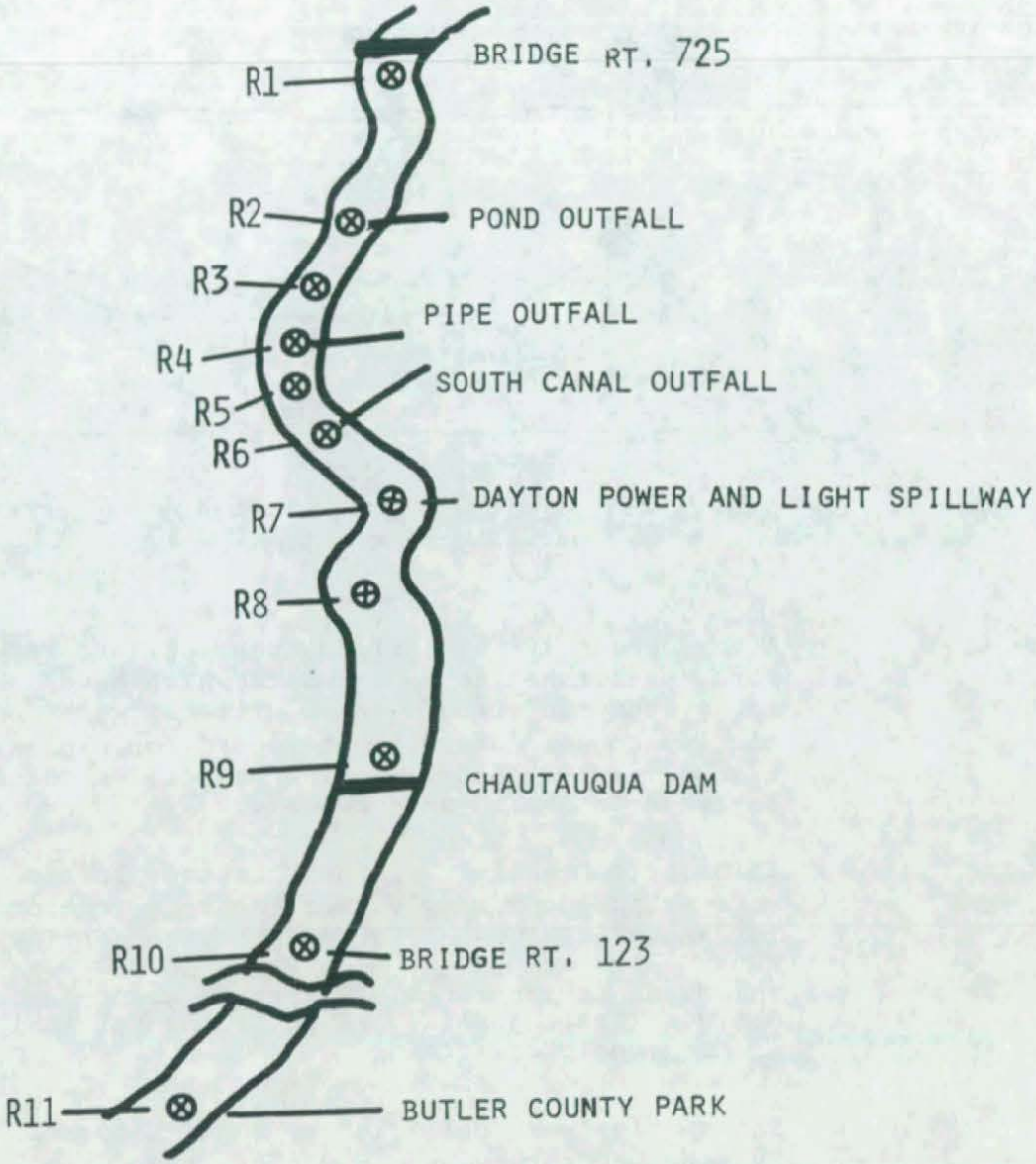
Considerable care was exercised during sampling, analyses, and data evaluation to ensure the reliability of the results. Figure 29 shows a schematic flow sheet of the analytical and data control methods. Quality control methods routinely applied for Mound Laboratory products for space applications were implemented on this program to ensure sampling and analytical process control, personal accountability, data processing and review, and record keeping.

Operation sheets, shown in Figure 30, which described the sampling and analytical procedures in step-by-step detail were used for each sample. These operation sheets required the sampling team and analytical chemists to record data and verify strict adherence to procedure by initialing each step as it was completed. A permanent operation sheet record package was then maintained on each sample taken and analyzed. Quality control inspectors made unannounced inspections in the field and in the laboratories to further check for adherence to procedures and good analytical practices.

Blank soil or sediment samples were processed with each group of samples to indicate laboratory sample contamination levels. The blank results were never subtracted from the sample results, but were used for reliability reference.

Approximately 135 replicate analyses on samples containing a wide range of plutonium were performed periodically during the program and used to develop statistics for the analytical operations. Figure 31 shows the relationship established between the precision of the analytical determination (standard deviation) and the plutonium concentration in the soil or sediment.

SAMPLE LOCATIONS
GREAT MIAMI RIVER



APPROXIMATELY 10 MI. DOWNSTREAM OF MOUND LABORATORY

FIGURE 25 - Great Miami River core sampling sites.

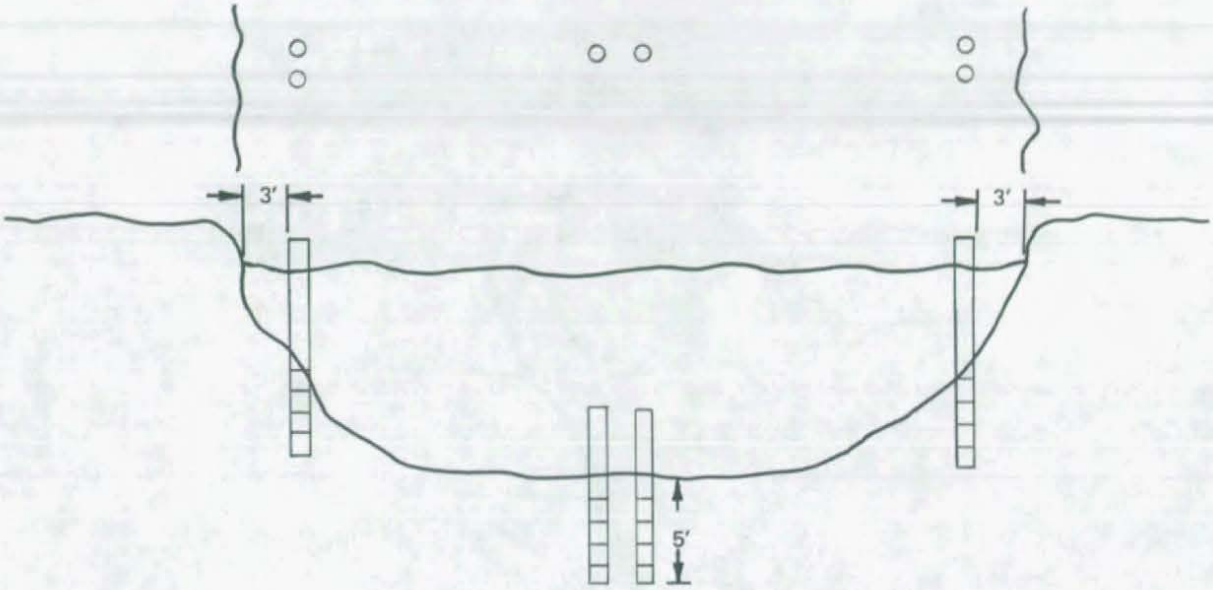


FIGURE 26 - Typical distribution of samples taken at river locations.

After exhaustive evaluation, several data rejection criteria were established to remove data which have a high potential for systematic error. These criteria were applied systematically to all data without regard for plutonium concentration or sample location. The rejected samples were generally reanalyzed and valid data reported.

In order to verify that the measured plutonium concentrations were valid, many samples were sent to independent laboratories for analysis. These independent laboratories were: the U. S. ERDA Health and Safety Laboratory in New York, New York (HASL), the U. S. Environmental Protection Agency Laboratory in Atlanta, Georgia (EPA), and the LFE Environmental Analysis Laboratory in Richmond, California.

The samples were collected by Mound Laboratory, Bowser Morner Testing Laboratory, and U.S. EPA, then dried and ground to less than 20-mesh particle size. The samples were homogenized and aliquots from these samples were riffled and sent to the independent laboratories for analysis.

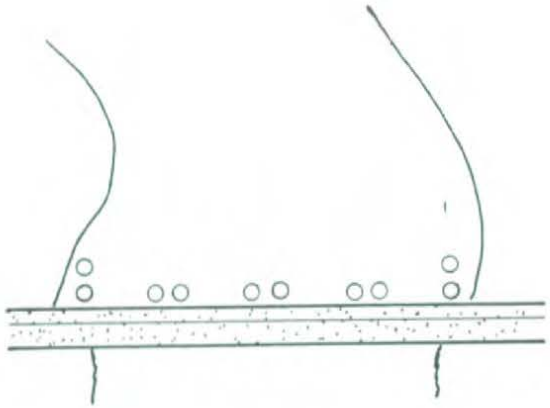


FIGURE 27 - Typical distribution of samples taken behind dams in the river.

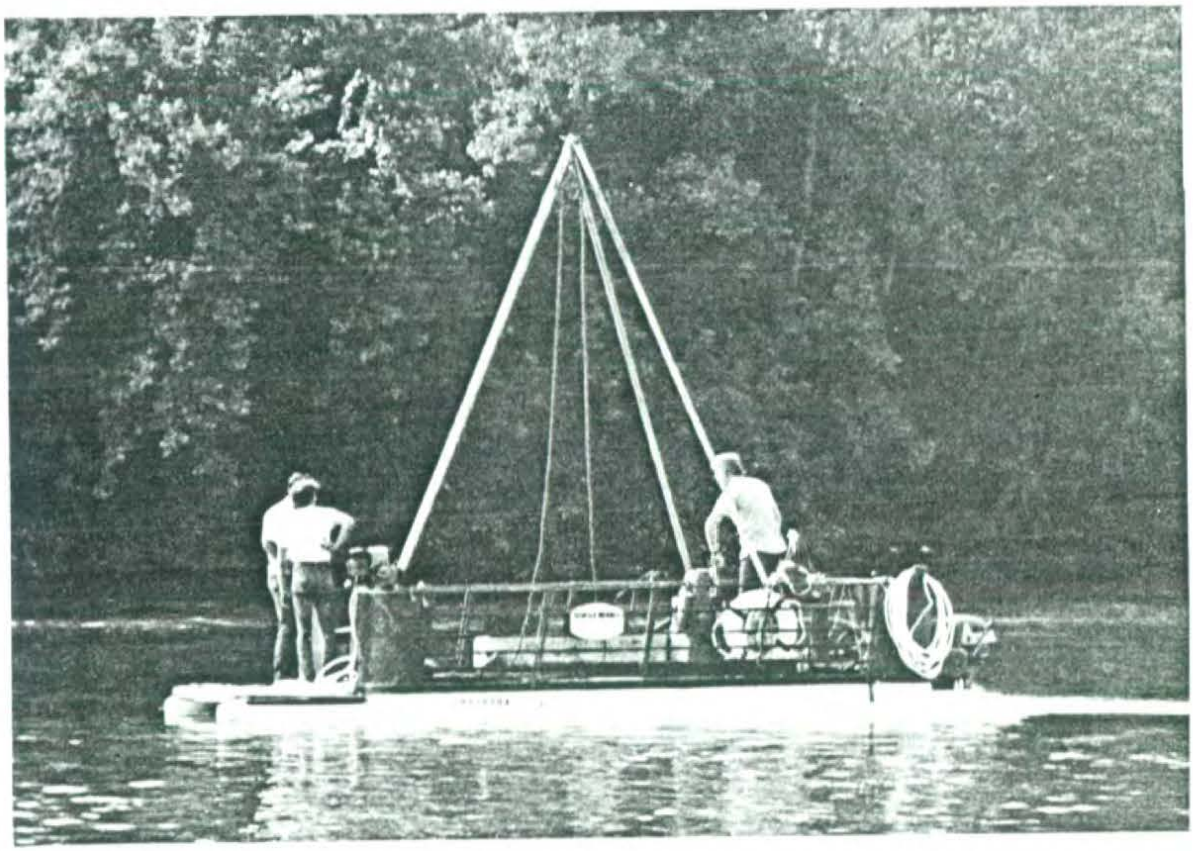


FIGURE 28 - Rig used for river sampling by Bowser Morner.

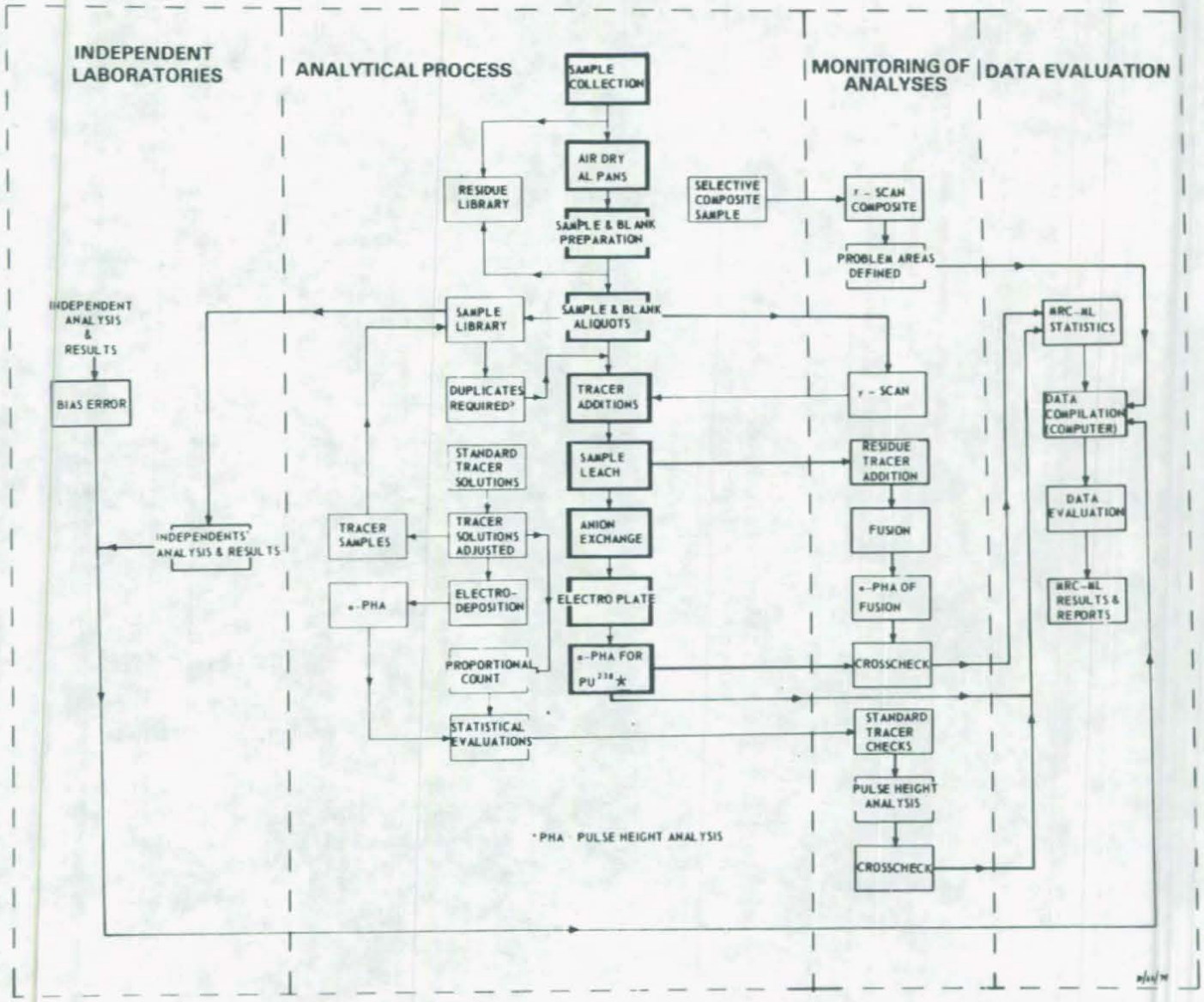
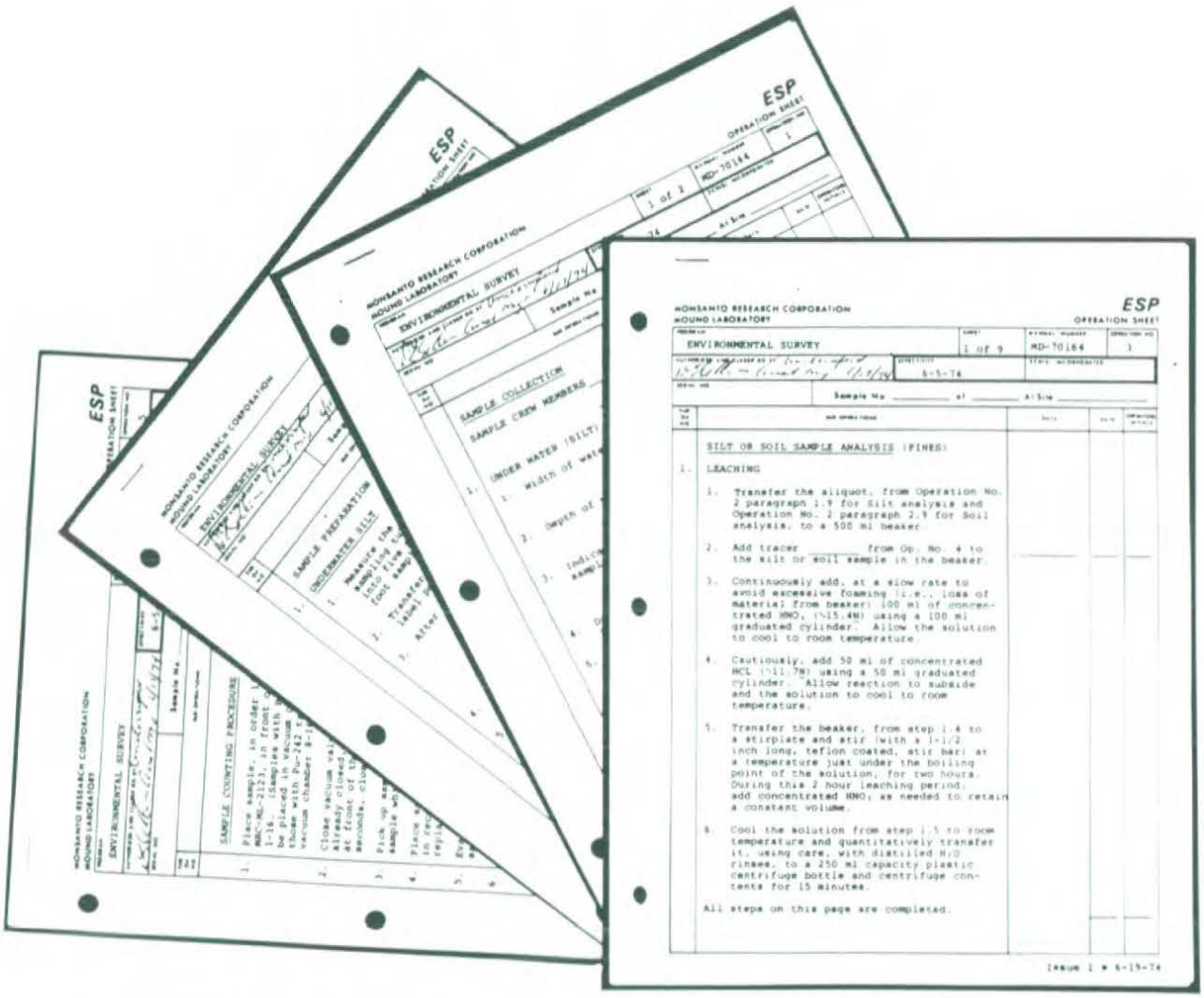


FIGURE 29 - Schematic flow sheet of analytical and data control methods.



MONSANTO RESEARCH CORPORATION
MOUND LABORATORY

ESP
OPERATION SHEET

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Sample No _____ at _____ A/Site _____

SILT OR SOIL SAMPLE ANALYSIS (FINES)

1. LEACHING

1. Transfer the aliquot, from Operation No. 2 paragraph 1.9 for Silt analysis and Operation No. 2 paragraph 2.9 for Soil analysis, to a 500 ml beaker.

2. Add tracer _____ from Op. No. 4 to the silt or soil sample in the beaker.

3. Continuously add, at a slow rate to avoid excessive foaming (i.e., loss of material from beaker) 100 ml of concentrated HNO_3 (~15.4M) using a 100 ml graduated cylinder. Allow the solution to cool to room temperature.

4. Cautiously, add 50 ml of concentrated HCL (~11.7M) using a 50 ml graduated cylinder. Allow reaction to subside and the solution to cool to room temperature.

5. Transfer the beaker, from step 1.4 to a stirplate and stir (with a 1-1/2 inch long, teflon coated, stir bar) at a temperature just under the boiling point of the solution, for two hours. During this 2 hour leaching period, add concentrated HNO_3 as needed to retain a constant volume.

6. Cool the solution from step 1.5 to room temperature and quantitatively transfer it, using care, with distilled H_2O rinses, to a 250 ml capacity plastic centrifuge bottle and centrifuge contents for 15 minutes.

All steps on this page are completed.

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FIGURE 30 - Operation sheets for analytical quality control.

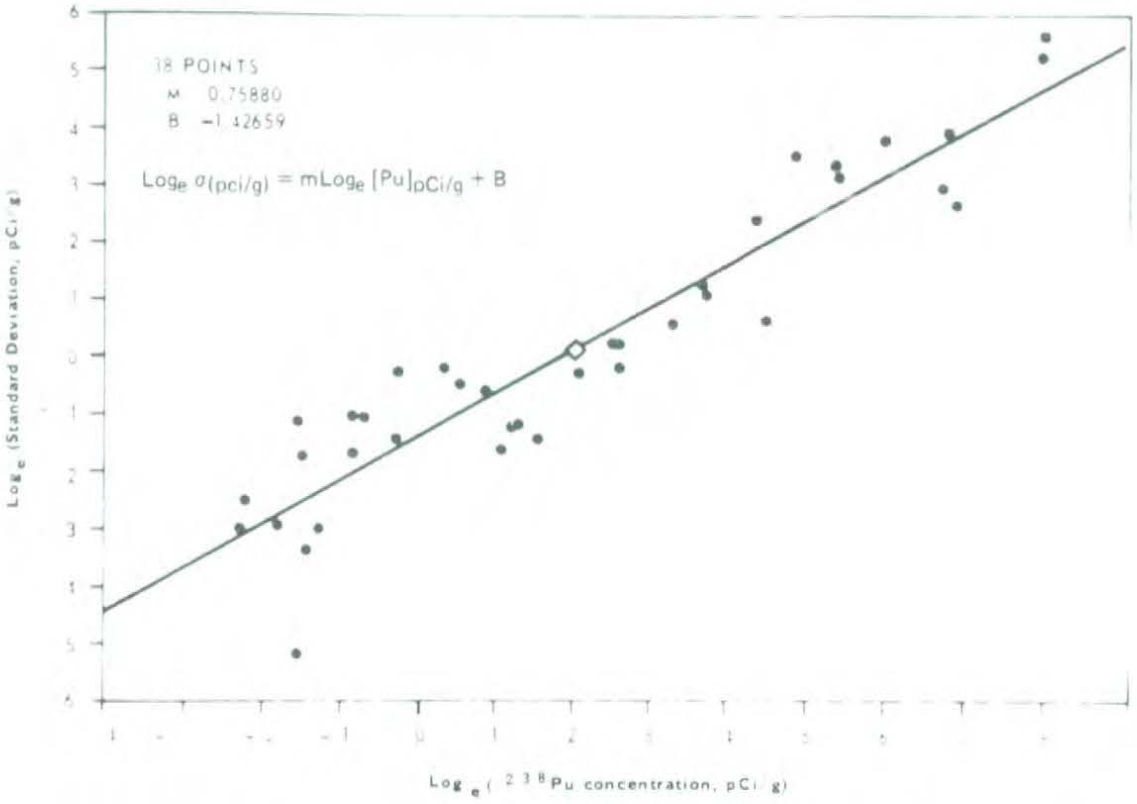

FIGURE 31 - Standard deviation as a function of the plutonium-238 concentration in soil and sediment samples.

Interlaboratory analytical agreement was generally good at all concentration levels.[1] EPA results tended to be slightly higher than the Mound values while LFE results tended to be slightly lower. The Mound and HASL results were almost identical except at very low concentrations where HASL values tended to be lower.

In addition to the analytical variance, the variance due to the actual distribution of plutonium in and around these waterways was estimated by taking duplicate local variability samples 1 to 3 ft apart in many of the sampling locations.

The plutonium deposition in the areas, based on the local variability samples, exhibited log-normal distribution characteristics. The geometric standard deviation of the 5-ft composite samples representing variation of total deposition within a local area was found to be 1.77. This value did not vary significantly with plutonium concentration or location.

The local variability on individual 1-ft sections of the cores representing the local variability with respect to depth was considerably higher ($\sigma_g \approx 10$). These variances are believed to arise from the way in which the plutonium is deposited rather than to sampling or analytical techniques.

Samples, independently collected in and around these waterways and analyzed by HASL, U.S. EPA, Ohio EPA, and HNL (Holifield National Laboratory) showed agreement with Mound samples taken nearby.[1]

B. Summary of Results

The individual analytical values of each of the samples taken have been reported previously[1] to governmental, health and environmental agencies. In this summary, the data are ordered and presented on the basis of plutonium availability to man in order to permit their use later in the report to evaluate the potential health and safety hazards to the public. The data are divided into three categories:

- Plutonium-238 concentration available to man
- Plutonium-238 concentration potentially availabe to man
- Inventory of plutonium deposited in waterways

1. Plutonium-238 Concentration Available to Man

The plutonium-238 available to man in this case is that which is deposited on the "very surface" of the soil or sediment, is dissolved in water, or is present in biota used for food. The dry land "very surface" contamination is available to be suspended in air under certain conditions where it might be inhaled or is available to be ingested directly. The "very surface" of sediment, which is under water, is subject to easy suspension in water or if the sediment dries, then it could be subject to suspension in air.

a. "Very Surface" ^{238}Pu concentrations in sediment and soil.- The "very surface" concentrations of ^{238}Pu in the sediment were measured in most cases by collecting and analyzing the solids suspended in the natural water by *in situ* vigorous agitation of the water near the sediment interface. These solids were usually found to be in the particle size range less than 50 μm (silt and clay), although some samples contain small amounts of fine sand. These concentrations should approximate the plutonium-238 concentration of the air-suspendible dust should the sediment dry.

The maximum sediment "very surface" values found in each of the waterways is presented in Table 2. As can be seen, the values vary from 0.02 to 0.45 nCi/g depending on the location. This range of values was found to be in agreement with shallow surface scoop samples taken by Mound Laboratory, U. S. EPA, and HASL in sediment areas not covered with water.

Table 2

MAXIMUM "VERY SURFACE" ^{238}PU CONCENTRATION OF SEDIMENT IN WATERWAYS NEAR MOUND LABORATORY

Waterway	Maximum "Very Surface" Concentration (nCi/g ± 2σ)
Runoff Hollow	0.0286 ± 0.0061
North Pond	0.0223 ± 0.0051
South Pond	
North Basin	0.0653 ± 0.0114
South Basin	0.208 ± 0.028
North Canal	0.267 ± 0.033
Drainage Ditch	0.450 ± 0.050
South Canal	0.395 ± 0.045
Overflow Creek	0.270 ± 0.034

The maximum "very surface" concentrations along the immediate banks of the waterways which are subject to occasional flooding are presented in Table 3 for each of the waterways. The values were taken from shallow surface soil samples and tended to range from 0.002 to 0.06 nCi/g.

Table 3

MAXIMUM ^{238}Pu "VERY SURFACE" CONCENTRATIONS IN SOIL ALONG THE IMMEDIATE BANKS OF THE WATERWAYS WHICH ARE OCCASIONALLY FLOODED

Waterways	Maximum "Very Surface" Concentration (nCi/g ± 2σ)
Ponds (Composite)	0.0017 ± 0.0007
North Canal	
East Bank	0.0446 ± 0.0086
West Bank	0.0540 ± 0.0099
Earthen Dam/Ditch Area	0.0542 ± 0.0099
South Canal	
East Bank	0.0609 ± 0.0109
West Bank	0.0540 ± 0.0099
Overflow Creek	
East Bank	0.0116 ± 0.0031
West Bank	0.0021 ± 0.0008

The "very surface" values in the land areas (Table 4) adjacent to the waterways, which for physical reasons are not likely to be exposed to flooding from the waterways, were found to be in the 0.0002 to 0.001 nCi/g concentration range.

b. ^{238}Pu concentrations in water.– The maximum soluble ^{238}Pu concentration in water samples taken in each of the waterways is presented in Table 5. The value varied from less than 0.000001 to 0.000014 nCi/ml.

Table 4

"VERY SURFACE" ^{238}Pu CONCENTRATION IN AREAS ADJACENT TO THE WATERWAYS WHICH ARE NOT SUBJECT TO FLOODING

Land Area	"Very Surface" Concentrations (nCi/g ± 2σ)
North Canal - Pond Area (Composite)	0.00023 ± 0.00005
South Canal - Overflow Creek Area (Composite)	0.00075 ± 0.00020

Table 5

CONCENTRATION OF ^{238}Pu IN WATER TAKEN FROM THE WATERWAYS

Waterway	Water Concentration (nCi/ml ± 2σ)
Runoff Hollow	<0.000001
North Pond	<0.000001
South Pond	
North Basin	<0.000001
South Basin	<0.000001
North Canal	0.000005 ± 0.000003
Off-Site Ditch	0.000006 ± 0.000003
South Canal	0.000014 ± 0.000006
Overflow Creek	0.000003 ± 0.000002
River (at Outfall)	0.000001 ± 0.000001

c. Distribution of ^{238}Pu between water and sediment. - Water samples which were allowed to remain in contact with suspendible solids tend to attain a constant relationship between the water and solid ^{238}Pu concentration. When the two phases were separated, the ratio of the ^{238}Pu concentration between the natural water and sediment samples (distribution ratio, $D_{w/s}$) was found to approach 1×10^{-5}. This distribution ratio was reasonably constant for all waterways.

d. Concentrations of ^{238}Pu in biota samples. - There is very little biota in or near the affected contamination area which is normally consumed directly by man (except for the fish in the waterways). It is assumed that the biota samples taken, however, provide an index for worst case consumable biota contamination considering the nature of the area.

The plutonium-238 concentrations found in grass, algae and fish samples collected in or near the waterways are listed in Table 6.

Table 6

CONCENTRATION OF ^{238}Pu IN BIOTA SAMPLES TAKEN IN OR NEAR THE WATERWAYS

Biota	^{238}Pu Concentration (nCi/g ± 2σ)
Grass taken along banks in areas not subject to frequent flooding	0.000018 ± 0.000020
Grass taken along banks subject to frequent flooding (Areas A and R)	0.000874 to 0.00305
Green algae (samples from North Canal and Pond water)	0.00239 to 0.111
Fish (edible portion)	
Bottom feeders (carp)	0.00000512
Other (blue gill)	0.00000079

Grass samples, taken along the immediate bank where flooding is unlikely for physical reasons averaged 0.000018 $\pm$ 0.000020 nCi/g (dry weight). Some of these values are higher than might be expected from systemic absorption into the grass and might indicate some surface contamination from light tracking. Samples of grass taken in two localized areas, where frequent flooding is observed, were higher and indicated definite external surface contamination from deposited sediment.

Algae samples taken from the surface of the ponds and the North Canal indicated, as expected,[2] these algae tend to concentrate plutonium probably directly from the water. Two species of fish, blue gills and carp, were obtained from the waterways. Although only a few samples could be collected, the bottom feeding carp showed a higher concentration than the blue gills, as would be expected.[2]

2. Plutonium-238 Concentration Potentially Available to Man

Plutonium-238 in soil or sediment that is below the surface is normally not available to enter the "pathway to man" except for an extremely small fraction taken systemically into subsurface biota consumed by man. In order to become significantly available, it must be carried to the surface by physical actions of man or nature. The probability of this occurring decreases with the depth of the deposit.

The ^{238}Pu concentration in the first foot is considered potentially available since it could be brought to the surface without extreme measures. The concentrations which are deposited deeper than 1 ft are much less likely to be exposed and are, therefore, only to be considered as *worst cases*.

The concentrations reported here are taken on 1-ft core sections. Although higher concentrations may exist in thinner stratified layers, the physical action required to bring them to the surface would also tend to mix and dilute the more concentrated zones with the lesser concentrated strata which lie above and below.

The first foot of ^{238}Pu concentrations on the banks of the waterways and the adjacent land areas are lower than the "very surface" values reported previously and so will not cause a future potential increase in the surface contamination.

The maximum ^{238}Pu concentrations in the first foot of sediment in each waterway are listed in Table 7, and vary from 0.0003 to 3.80 nCi/g. The maximum concentrations in the waterways and the depth at which they occur are listed in Table 8.

Table 7

MAXIMUM FIRST-FOOT ^{238}Pu CONCENTRATIONS
OF SEDIMENT IN WATERWAYS

Waterway	Maximum First-Foot Concentration (nCi/g ± 2σ)
Runoff Hollow	0.0314 ± 0.0066
North Pond	0.0062 ± 0.0019
South Pond	
North Basin	0.0309 ± 0.0065
South Basin	0.0096 ± 0.0027
North Canal	1.14 ± 0.10
Drainage Ditch	0.749 ± 0.013
South Canal	3.80 ± 0.25
Overflow Creek	0.0744 ± 0.0126
River	
East Bank Near Canal Outfall	0.0367 ± 0.0074
East Bank Downstream	0.0016 ± 0.0007
Away from East Bank	0.0003 ± 0.0002

Table 8

MAXIMUM ANY-DEPTH ^{238}Pu CONCENTRATIONS
OF SEDIMENT IN WATERWAYS

Waterway	Depth (ft)	Maximum Any-Depth Concentration (nCi/g ± 2σ)
Runoff Hollow	1	0.0314 ± 0.0066
North Pond	0	0.0223 ± 0.0051
South Pond		
North Basin	0	0.0653 ± 0.0114
South Basin	0	0.208 ± 0.028
North Canal	3	4.56 ± 0.20
Drainage Ditch	1	0.749 ± 0.013
South Canal	1	3.80 ± 0.025
Overflow Creek	0	0.270 ±0.034
River		
East Bank Near Canal Outfall	2	0.0415 ± 0.0081
East Bank Downstream	7	0.0037 ± 0.0013
Away from East Bank	4	0.0006 ± 0.0002

Unlike "very surface" concentrations that tended to be reasonably uniform within a given area, the plutonium deposited below the surface tends to be much more localized. These localized deposition patterns result from the nature of the transport and deposition mechanisms, which will be discussed in more detail later in this report.

The first-foot concentrations found in the North and South Canals, which vary greatly as a function of length and width, are shown in Figure 32. The concentration profiles across the canal that are presented are typical of many others measured. The highest concentrations are very localized near the middle of the South Canal.

Figure 33 shows the maximum concentrations (worst case) at any depth along or across the North and South Canals. The maximum levels occur just north of the earthen dam and midway down the South Canal.

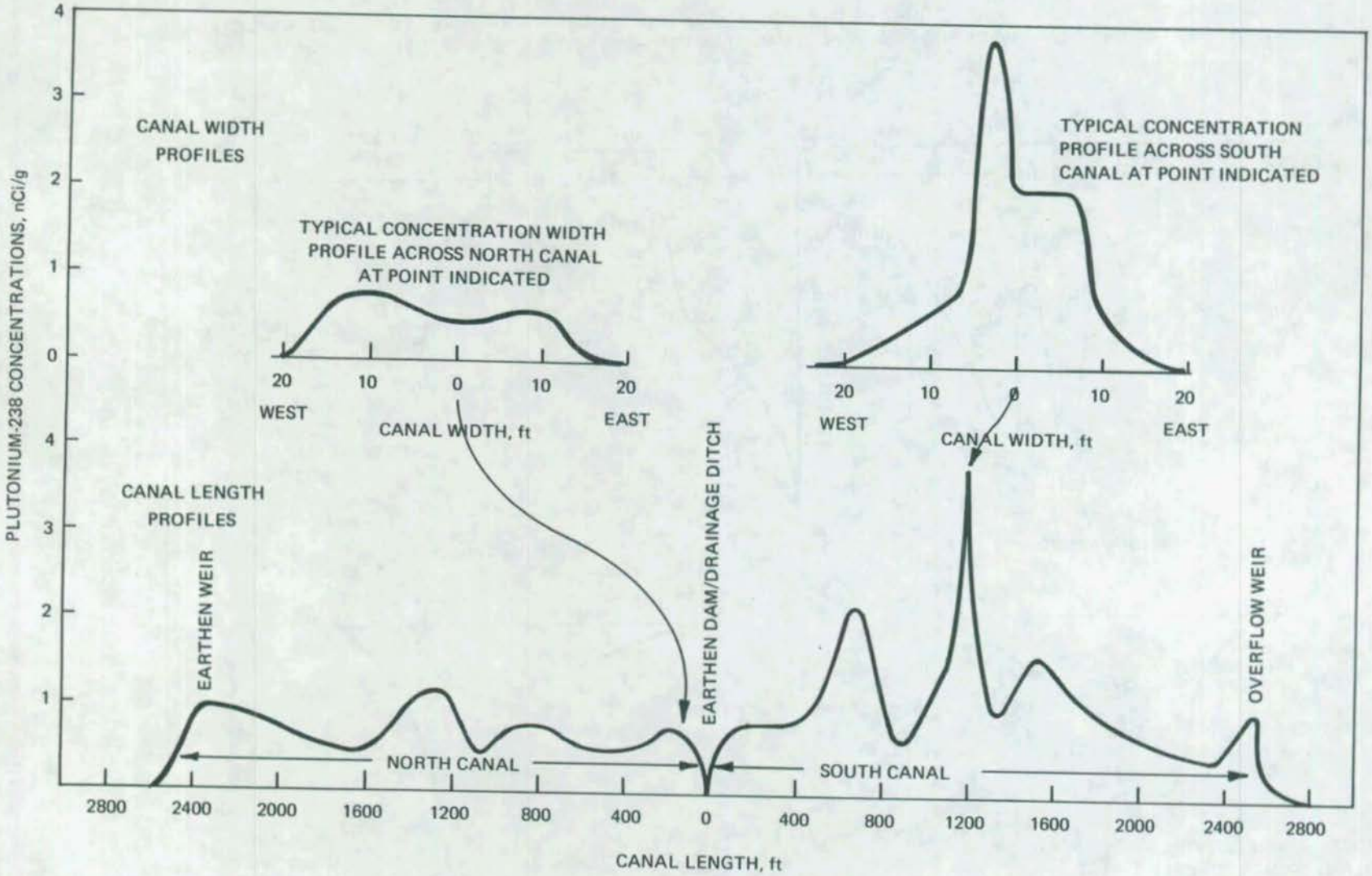

FIGURE 32 - Maximum first-foot ^{238}Pu concentration along the Miami-Erie Canal as a function of canal length and width.

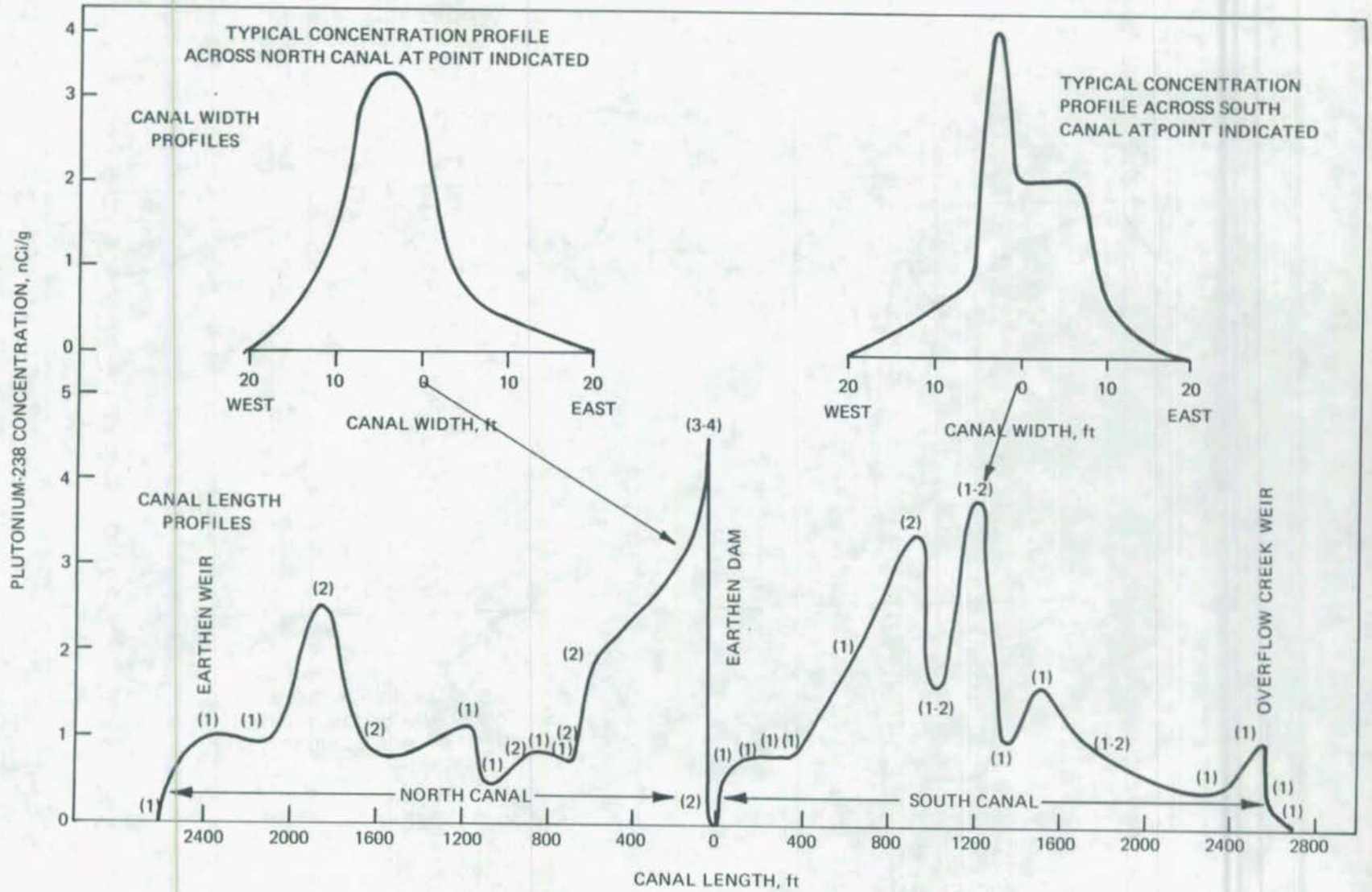

FIGURE 33 - Maximum ^{238}Pu concentration at any depth along the Miami-Erie Canal. (Numbers in parentheses indicate depth at which the concentration occurred.)

The data confirm that the significant contamination is confined to the waterways. The difference between the shape of the width-concentration profiles in the North and South Canals results from the nature of these waterways. The water level in the North Canal remains reasonably constant for nearly all flow conditions while the water level in the South Canal can vary from near zero to more than 4 ft. At low flow conditions, the South Canal maintains a narrow meandering channel, while at high flow, the entire canal channel is utilized. This meandering channel tends to contain less plutonium due to less original deposition or subsequent erosion.

Figure 34 shows typical depth concentration profiles. The solid-lined curves are the depth distribution at the locations of the highest any-depth concentrations in the North and South Canals. The broken-lined curves are depth distribution at the extreme north and south ends of the North and South Canals, respectively.

3. Inventory of Plutonium Deposited in Waterways

The total plutonium-238 inventory deposited in the waterways near Mound Laboratory was estimated by numerical integration of the plutonium concentration and dry sample weight data collected on the 261 cores taken with respect to depth, length, and width. The inventory of plutonium-238 in each waterway is presented in Table 9. The total plutonium-238 inventory in the off-site waterways was found to be 5.2 Ci.

Table 9

INVENTORY OF ^{238}Pu IN THE WATERWAYS

Location	Plutonium Inventory (Ci)
Run-off Hollow	0.0018
Drainage Ditch	0.082
North Canal	1.65
South Pond	0.0058
North Pond	0.0020
South Canal	3.17
Overflow Creek	0.076
Overflow Creek Outfall	0.260
TOTAL	5.2

From the data presented in the previous sections, most of the plutonium inventory is under the surface and, therefore, is not readily available to man.

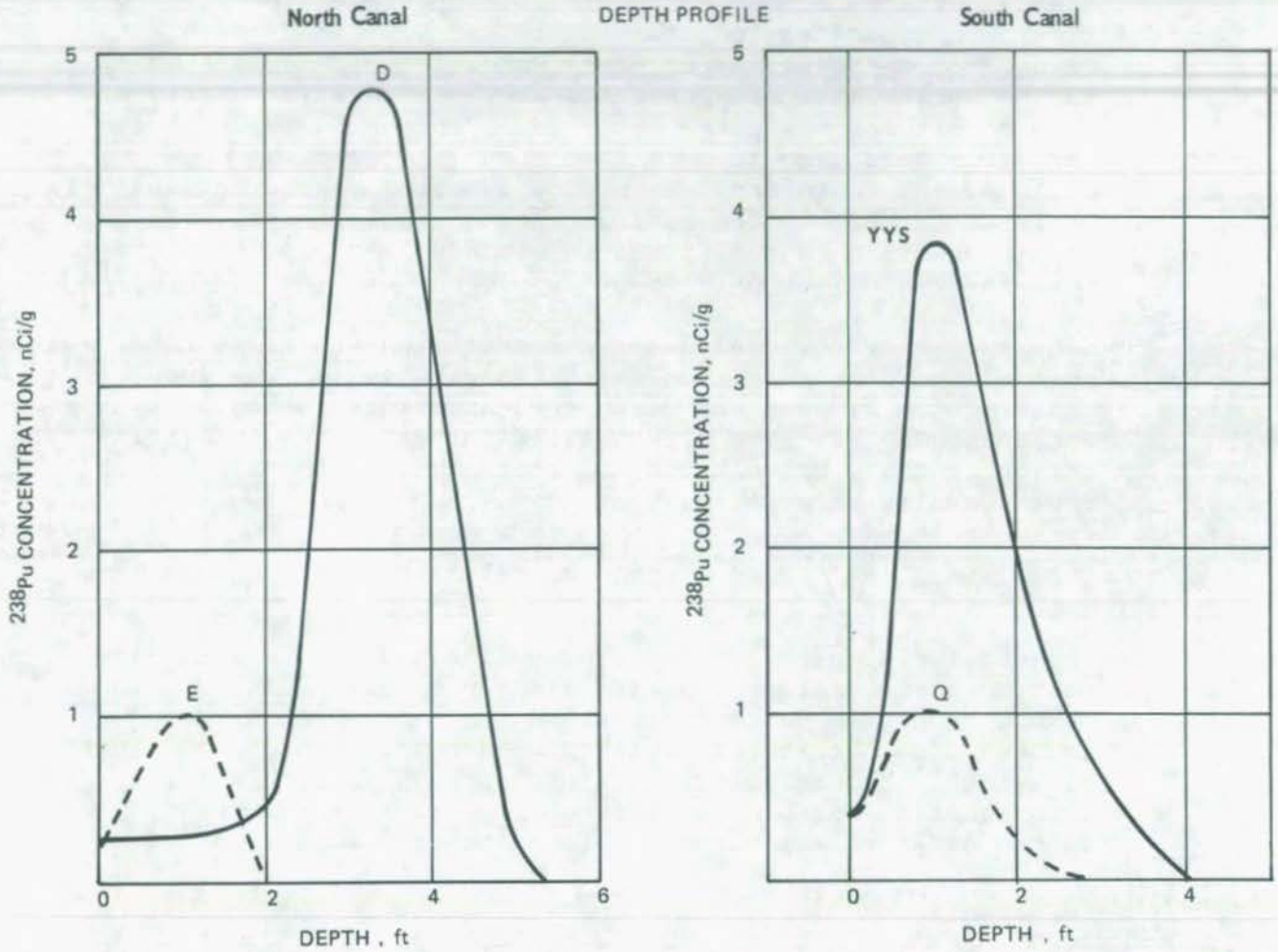

FIGURE 34 - ^{238}Pu concentration as a function of sediment depth at locations indicated (see Figures 18 and 19). Solid curve is distribution at location of maximum concentration; broken line is distribution at the extreme ends of the north and south canals downstream.

IV. CAUSE INVESTIGATIONS AND MECHANISMS

A. Cause Investigations

1. Introduction

Intensive investigations were performed concurrently by a Mound Laboratory Investigation Team* and a United States Energy Research and Development Task Group** to find the source and cause of these plutonium deposits in the waterways.

The Mound Laboratory team pursued their investigation using a systematic-analytical-investigative technique. These activities included:

- A complete and detailed review of all environmental data related to the possible release of plutonium to these waterways.
- A detailed examination of all appropriate laboratory records.
- Private interviews and group discussions with knowledgeable Mound Laboratory personnel.
- Examination of all appropriate engineering drawings, data, and information.

Additional information was collected from field and laboratory investigations. Among these were:

- A radiological surface survey of the Laboratory site using the Field Instrument for Detection of Low-Energy Radiation (FIDLER).
- On-site ^{238}Pu soil sampling and analysis program to supplement available data routinely collected.
- Other field observations, physical inspections and tests.
- Laboratory tests on the waterway sediments and on-site soil found to be contaminated with ^{238}Pu.
- Laboratory studies on the reactions of plutonium solutions with soil and sediment.

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*D, R. Rogers, Mound Laboratory, Chairman
**W. B. Johnston, U.S. ERDA/ALO, Chairman

From this investigation and these scientific studies, the source of the plutonium, the place and nature of the release, the behavior of the plutonium and the transport and deposition mechanisms of the plutonium were identified and confirmed. The U.S. ERDA investigation task group concurred with these findings.

2. Source and Nature of Plutonium Released

During the processing of plutonium-238 at Mound Laboratory, plutonium solutions that are too dilute for recovery operations are solidified and/or immobilized, drummed for burial, and shipped to ERDA-approved burial sites in the United States. Prior to 1967, this drumming operation was performed in the plutonium processing facilities (SM Building).

In 1967, an underground pipeline was installed between the SM Building, located on the southeast hill across the valley, to a waste drumming facility in the Waste Disposal (WD) Building, located on the side of the northwest hill (Figure 35). A pumping station (Building 41), located in the valley just south of the WD Building, was constructed to lift the solution up the final leg of the pipeline to hold tanks in the WD drumming facility.

This specifically-designed 1-1/2 in. pipeline was placed in operation in 1968, and both acidic and caustic solutions containing residual plutonium were thereafter routinely transferred from the plutonium processing hold tanks to hold tanks in the WD drumming facility.

In 1969, during such a routine transfer of an acidic process waste solution, the underground pipeline ruptured between Building 41 (pumping station) and WD Building, releasing the solution to the soil around the pipeline. This plutonium solution and the event were identified as the source of the plutonium found in the waterways off-site.

From the records available and from its observed behavior, the process waste solution was acidic (>1M HNO_3) and contained approximately 4.6 ppm $^{238}Pu(NO_3)_4$.

3. Description of the Rupture Site

The rupture occurred between Building 41 and the WD Building. The ruptured area was located about 4 ft underground on the edge of a roadway running east-west by the WD Facility. Immediately to the south of the rupture site, the land slopes sharply downward (~19° slope) toward Building 41, as shown in Figure 36. An open concrete flume runs downward and across this hillside. This flume carries surface water from

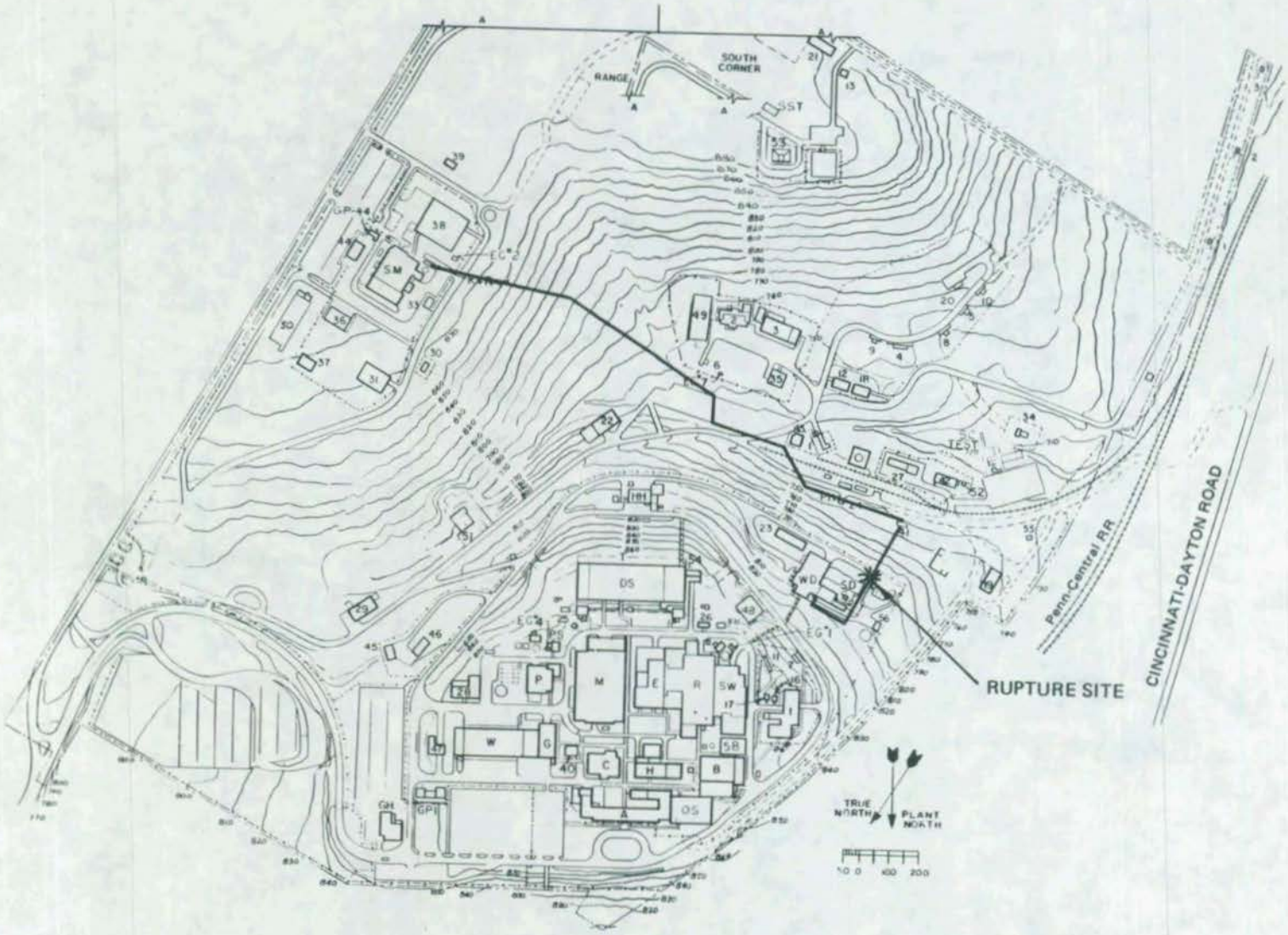


FIGURE 35 - Underground process waste solution transfer pipeline from PP to WD.

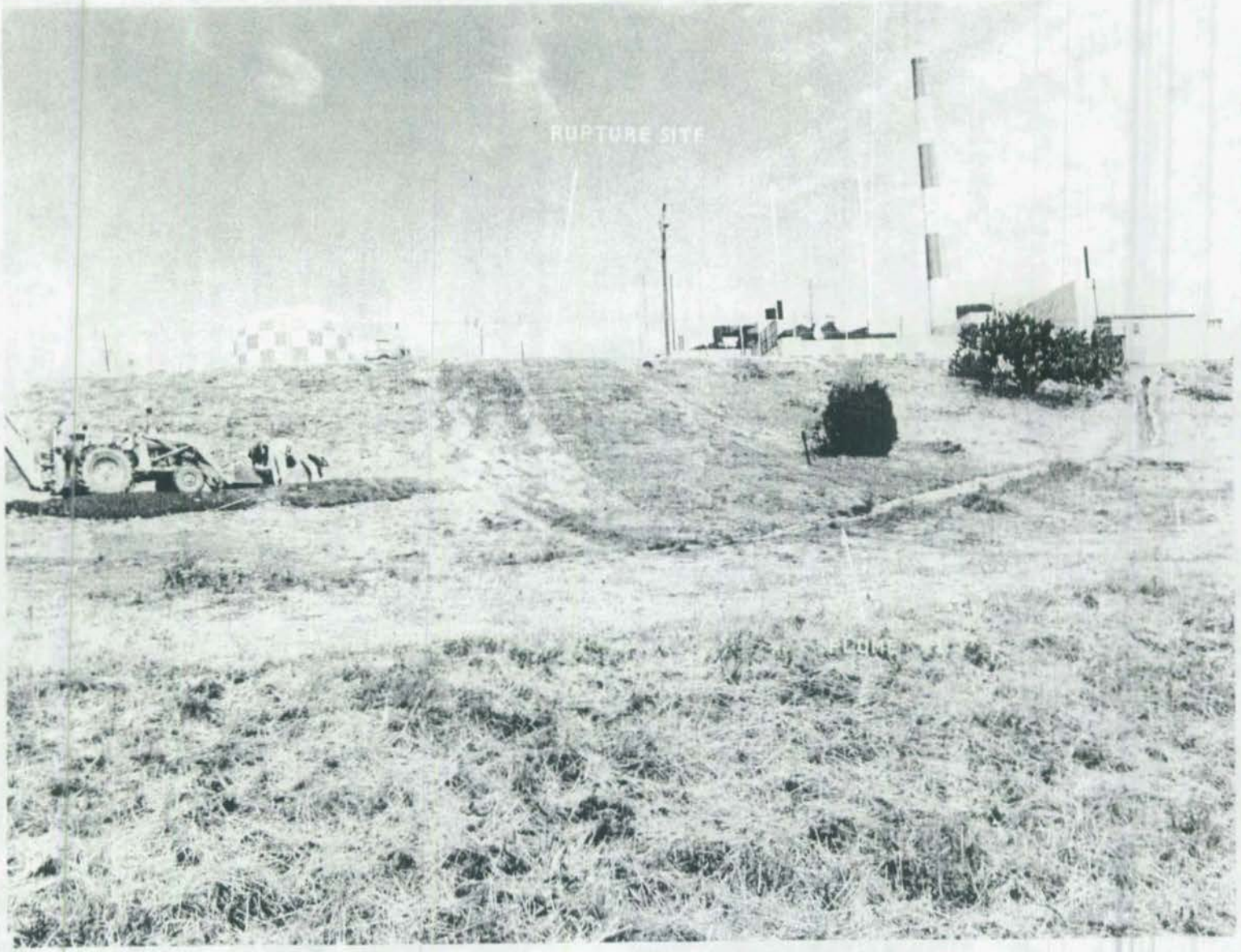


FIGURE 36 - Hillside below pipeline rupture site near the Waste Disposal Building.

the WD street to an underground storm drain at the edge of the Building 41 road, west of Building 41. An underground storm sewer carries the water south under the road and the railroad tracks to an open tributary drainage ditch which flows to the main drainage ditch.

4. Description of the Event

On January 23, 1969, while an acid plutonium(IV) nitrate solution was being transferred to the WD Drumming Facility, the pipeline ruptured at a corroded joint. The plutonium solution was discharged to the soil surrounding the pipeline. A small amount of the solution came to the surface and soaked into the soil at the rupture site and on the hillside.

The operators observed the solution at the surface and shutdown the transfer. They noted that the solution was giving off brown fumes and there was a vigorous effervescent reaction with the soil.

Health Physics survey teams found that the contamination was confined to the immediate area of the rupture site and on a small area of the hillside below. According to their survey data and observations, it appears the plutonium did not reach the drainage flume.

The weather was cold with intermittent light snow flurries when excavation and repair operations were initiated. The contaminated soil removed during excavation was placed in 55-gal drums for subsequent disposal. These drums were stored at the west end of the WD street. Excavation and drumming operations resulted in the contamination of the work area and street, and these areas were restricted by Health Physics.

Before excavation and repair operations were completed, the weather warmed and three days (January 28-30) of intense rainfall occurred.[3] The street became extremely muddy and the excavation filled with water. Water samples, taken in the drainage system below by the Environmental and Health Physics personnel, gave no indication (less than RCG for water) that significant concentrations of plutonium were moving off-site. The size of the contaminated area increased, but the situation appeared to be under control.

Following the rainstorms, repair operations were completed and the area was cleaned up. The area was surveyed with portable alpha counters and the pipeline was tested and placed back in operation.

The drums containing the contaminated soil were cleaned and shipped to an approved burial site.

5. Cause Conclusions

Although it was concluded at the time of this occurrence that no significant amounts of plutonium were released off-site; the 1974 investigations clearly indicated this event as the source of the off-site plutonium. The major evidence is briefly summarized:

- Using more sensitive survey instruments (FIDLER), a trail from the rupture area to the main drainage ditch and the runoff hollow was located in 1974.

- Plutonium-236 dating of the plutonium in the off-site areas were consistent with the time of the rupture. The plutonium-236 content of the plutonium in the waterways matched the plutonium found on the hillside near the rupture site.

- The magnitude of the plutonium release to the soil during the January 23, 1969 pipeline rupture was consistent with the amount of plutonium found off-site considering the mechanisms of transport and deposit.

- The distribution of the plutonium inventory in on-site and off-site areas was consistent with the location of the rupture and the mechanisms of transport and deposit.

- The magnitude of the plutonium concentration in the off-site sediment was consistent with the concentration of plutonium in the soil near the rupture site considering dilution during transport and deposit.

6. Actions Taken to Prevent Reoccurrence

Several actions were planned, initiated or implemented in 1974 to further reduce or eliminate the potential for release of plutonium, especially to off-site areas.

- The surface soil contamination trail, found on the WD hillside, was excavated and removed in 1974 to eliminate the potential for additional erosion of contaminated soil. This soil was drummed and shipped to the ERDA burial site in Idaho.

- A process waste drumming facility was constructed in the plutonium processing facility (SM-PP) and the use of the process waste solution transfer pipeline was discontinued in 1974.

- A comprehensive soil erosion control program has been initiated to reduce the erosion potential of all on-site soil. This program includes the use of ground cover and improvement of the site surface water drainage system.

- Several temporary small dams were installed on the main drainage ditch in early 1974 to settle out the erosion products and reduce the amount of suspended solids leaving the site in the water.

- Construction projects have been initiated or planned to install large permanent settling basins, ponds and a reservoir to reduce the suspended solids in the water flowing to off-site areas to the lowest practical level.

- A routine sediment and suspended solids sampling plan was implemented by the Environmental Monitoring Group.

B. Mechanisms of Release of Plutonium-238 to Off-Site Areas from the Pipeline Rupture Site

1. Mechanistic Model of the Release, Transport, and Deposition

The mechanisms that permitted the plutonium to move from the rupture site to off-site areas is of considerable interest since none of the information taken at the time of the rupture in 1969 indicated a significant release to off-site areas.

From laboratory studies, field tests, and observations, a general mechanistic model has been developed, consistent with all of the available information.

A flow diagram of the release mechanism is summarized in Figure 37. Using this model, the sequence of events leading to the deposition of plutonium in the off-site waterway can be summarized as:

- Acidic plutonium solution released to the soil (mainly underground) on January 23, 1969.

- The acid was neutralized by the calcareous soil.

- The plutonium was strongly sorbed and fixed on the soil and was immobilized.

- The soil onto which plutonium was sorbed was brought to the surface and/or loosened by the excavation and repair operations.

- The heavy rainfall on January 28-30 eroded the soil loosened by excavation and the soil dropped on WD street, into the surface water system.

- The erosion products were carried by stream action to the main drainage ditch, then off-site to the waterways.

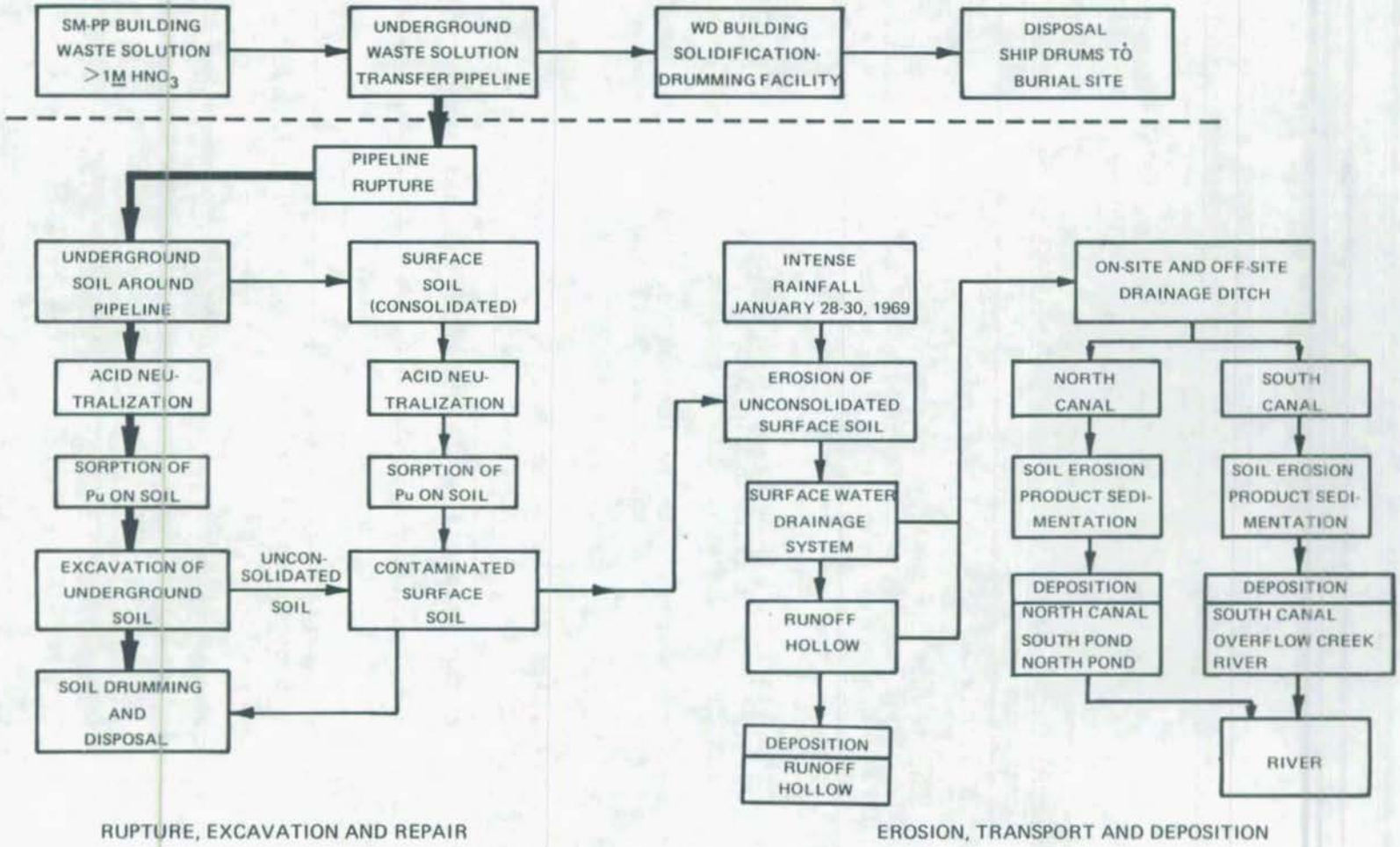


FIGURE 37 - Flow diagram of release transport and deposit of ^{238}Pu in off-site waterways.

- The erosion products settled in the waterway locations most favorable for particle sedimentation.

- Subsequently, erosion of noncontaminated soil tended to result in sedimentation in the same locations and to cover the plutonium-contaminated sediment.

The key elements of this model are:

- Acid neutralization by the soil,

- Plutonium sorption on the soil,

- Erosion of contaminated soil,

- Transport of soil erosion products by stream action, and

- Deposition of the soil erosion products in off-site waterways by sedimentation processes.

Perhaps the most important mechanism examined in this case is the interaction of plutonium solutions and Mound Laboratory soil. From the time the pipe ruptured to the deposit of plutonium in the off-site waterways, the behavior of plutonium was controlled by the plutonium/soil chemical interactions and the physical properties of the Mound Laboratory soil.

2. Interactions of Plutonium Solutions With Soil

a. Soil properties.- The soil indigenous to Mound Laboratory is glacial till, a heterogeneous mixture of gravel, sand, silt, and clay.[4] A typical fully dispersed glacial till particle size distribution is presented in Figure 38.[4] The clay and silt fraction in glacial till which accounts for most of the surface area averages about 40 to 50%. The major chemical constituents of the soil are:

- Clay (hydromicas and other clays)

- Calcite ($CaCO_3$)

- Dolomite [$MgCa(CO_3)_2$]

- Quartz (SiO_2)

- Limestone and Shale Rocks

- Organic Materials

- Water

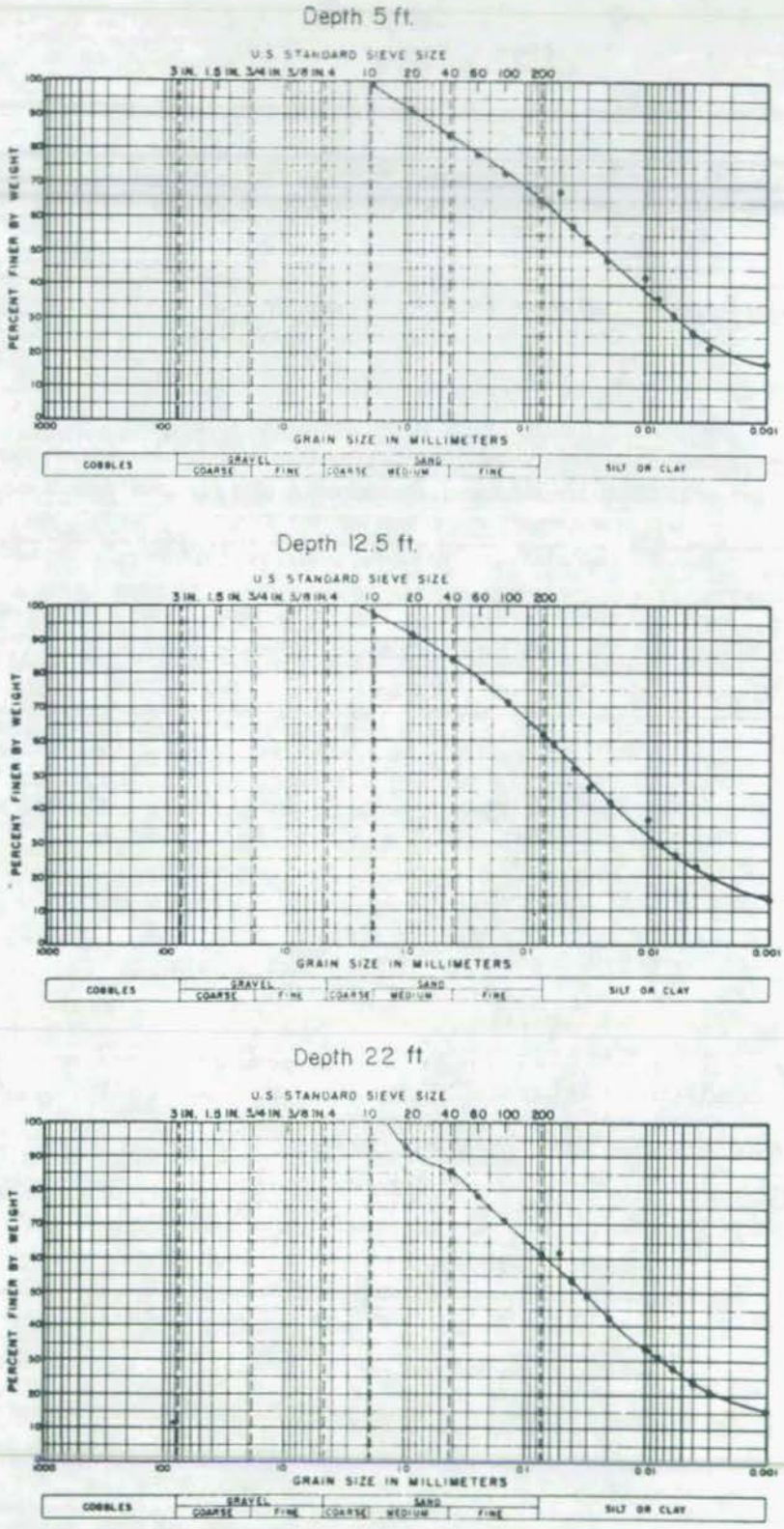


FIGURE 38 - Fully dispersed particle size distribution of glacial till.

The calcite, limestone, and dolomite gives the soil its buffer (acid neutralizing) capacity while the silt and clay fraction is responsible for most of its sorption or ion exchange capacity. Table 10 lists these important chemical properties for both whole soil, and the silt and clay fraction. The pH of water in equilibrium with the soil is typical of calcareous soils.

Table 10

CHEMICAL PROPERTIES OF
GLACIAL TILL AT MOUND LABORATORY

	Whole Soil	<50 μm Fraction
Ion Exchange Capacity*	5-20 meq/100 g	20 meq/100 g
Buffer Capacity (pH = 7)**	400-1100 meq/100 g	400 meq/100 g
pH of Equilibrium Water	7.5-8.3	7.5-8.3

*Varies with particle size distribution
**Varies with amount of soil carbonates (minerals and rocks)

b. Acid neutralization by the soil. - When acidic solutions are placed in contact with the soil, a violent effervescent reaction takes place liberating large amounts of carbon dioxide gas and reducing the acidity of the solution:

$$2HNO_3 + CaCO_3 \rightleftarrows CO_2\uparrow + Ca(NO_3)_2 + H_2O$$

(calcite)

$$4HNO_3 + MgCa(CO_3)_2 \rightleftarrows 2CO_2\uparrow + Ca(NO_3)_2 + Mg(NO_3)_2 + 2H_2O$$

(dolomite)

The rate of acidity neutralization, shown in Figure 39, is rapid. After contact with the soil, the pH increases rapidly to about pH 5 or 6, then more slowly to pH 7.5 to 8.3 as the dissolved CO_2 is expelled from the solution.

The capacity of the soil near the rupture site to neutralize the acidic plutonium solution released is about

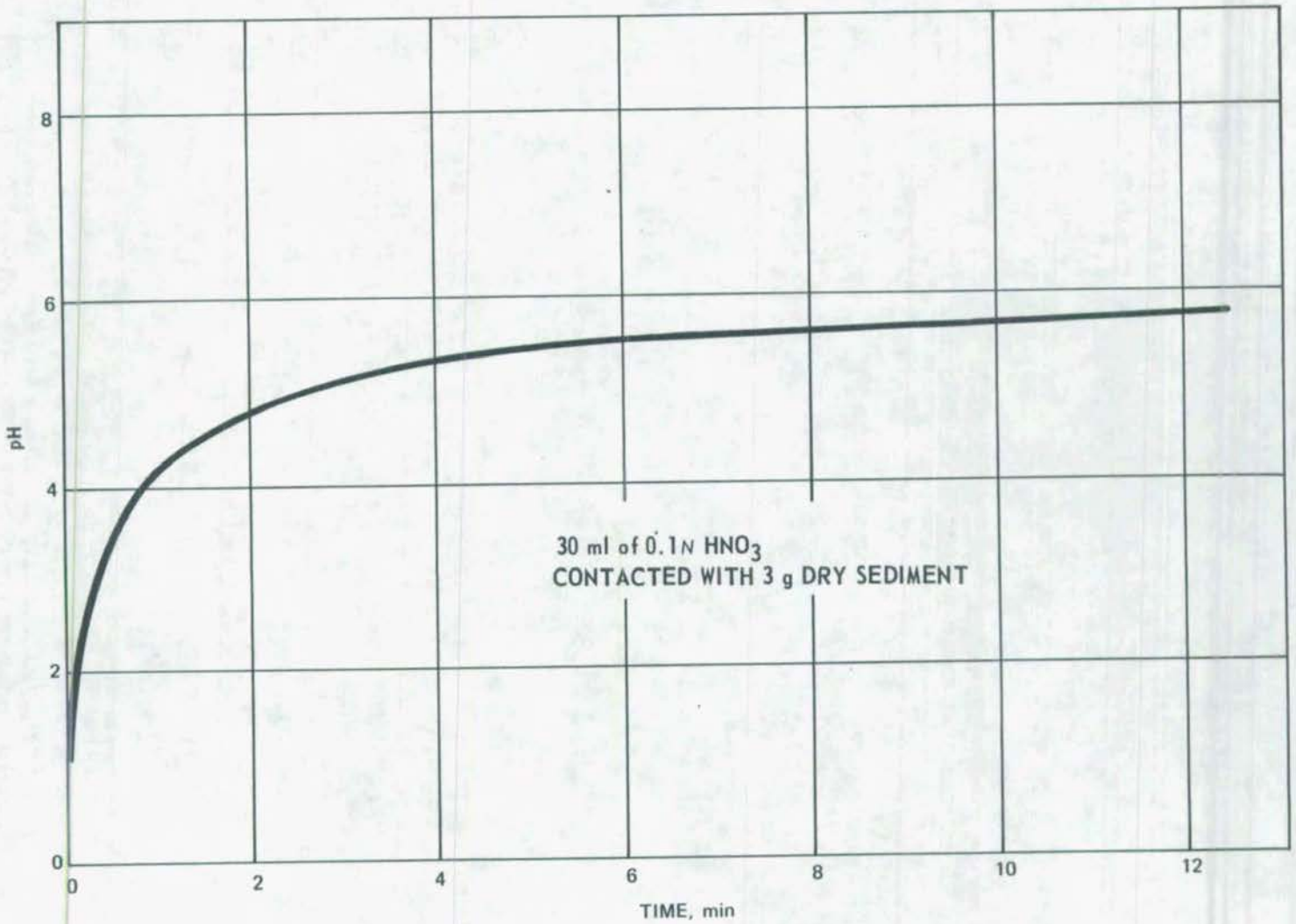


FIGURE 39 - Rate of acid neutralization by glacial till.

4 to 11 milliequivalents of acid per gram of soil depending on the size, distribution, and composition of the soil. This capacity is large enough to have consumed all of the acid released at the rupture site.

c. Sorption of plutonium-238 onto the soil. - The plutonium/soil sorption studies were performed with the clay and silt fraction of the soil and plutonium-238 nitrate [$Pu(NO_3)_4$] solutions prepared and diluted in the laboratory to avoid nonequilibrium hydrolysis or "polymerization". The sorption studies involved contacting 20 ml of the plutonium solution with 2 g of air-dried soil. These samples were equilibrated with agitation at room temperature for 7 to 14 days. Separation of the two phases was performed using centrifugation and filtration.

(1) pH dependence: The equilibrium distribution, $D_{w/s}$ defined as:

$$D_{w/s} = \frac{\text{Concentration of Pu in the water (nCi/g)}}{\text{Concentration of Pu in the soil (nCi/g)}}$$

was found to vary greatly with the equilibrium (final) pH of the solution as shown in Figure 40. (For a perspective of the percent of plutonium sorbed versus pH, see Figure 41.) It is noteworthy that the plutonium is extensively sorbed ($D_{w/s} \approx 2 \times 10^{-2}$) onto the soil even when the final acidity is about 1 M HNO_3. The distribution ratio decreases (sorption increases) as the pH is increased and reaches a minimum value of 6×10^{-6} at about pH 5.5, then appears to increase again.

The increasing distribution ratio with increasing pH in the alkaline region may be due, in part, to the formation of plutonium hydrolytic species and, in part, to clay dispersion. The clay tends to disperse in alkaline solutions and form colloidal suspensions which are difficult to remove by centrifugation and filtration. Some of the measured plutonium in the solution is undoubtedly sorbed onto these colloidal clay particles. The minimum distribution ratio (maximum sorption) found in these laboratory tests (6×10^{-6}) agrees fairly well with the distribution ratio measured on the contaminated sediment off-site (1×10^{-5}). This agreement tends to support the proposed mechanistic model.

(2) Sorption as a function of plutonium solution concentration: At near neutral pH, the sorption of plutonium did not vary significantly using pluton-

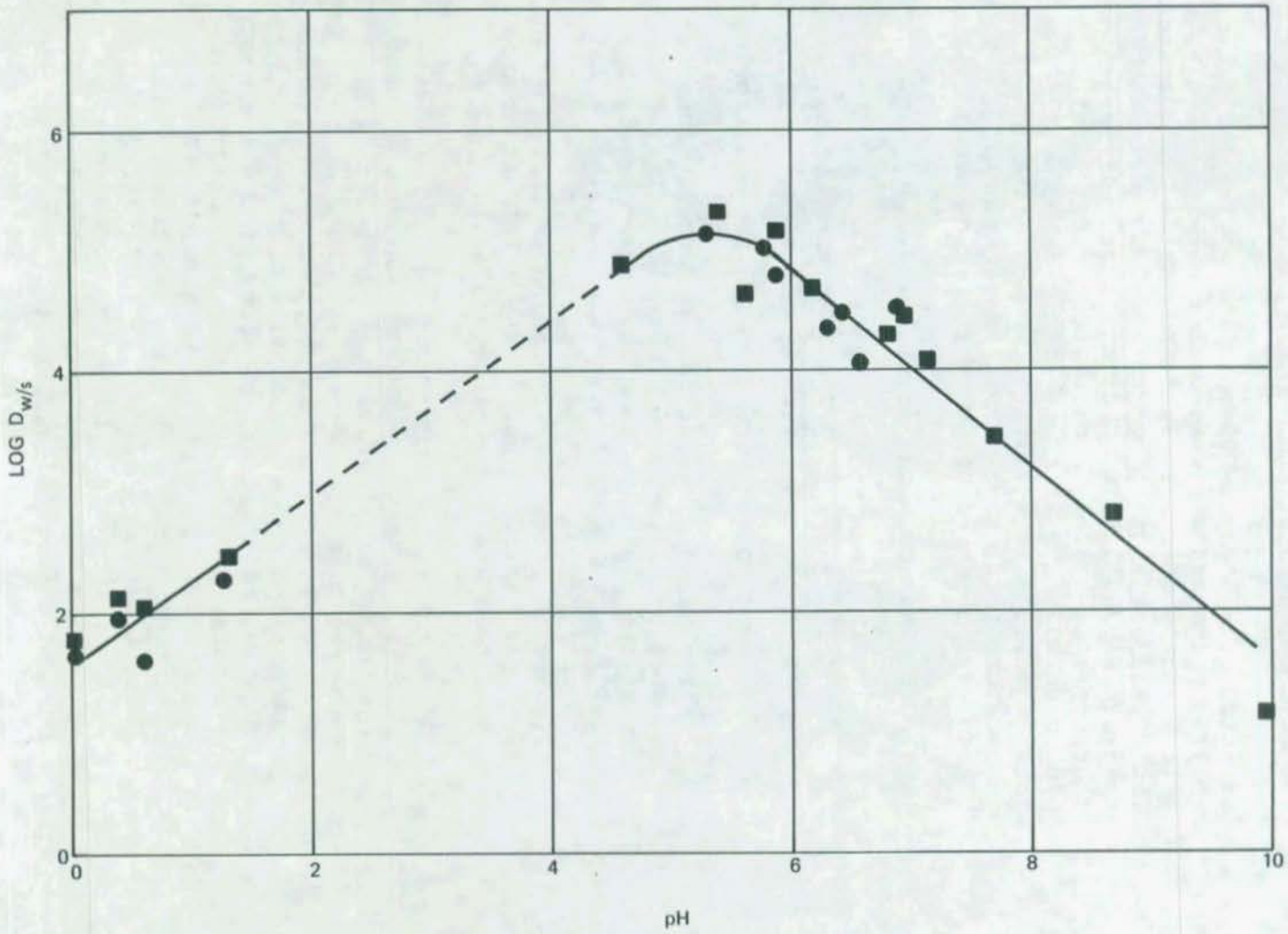


FIGURE 40 - Distribution ratio ($D_{w/s}$) of plutonium-238 between water and soil as a function of pH.

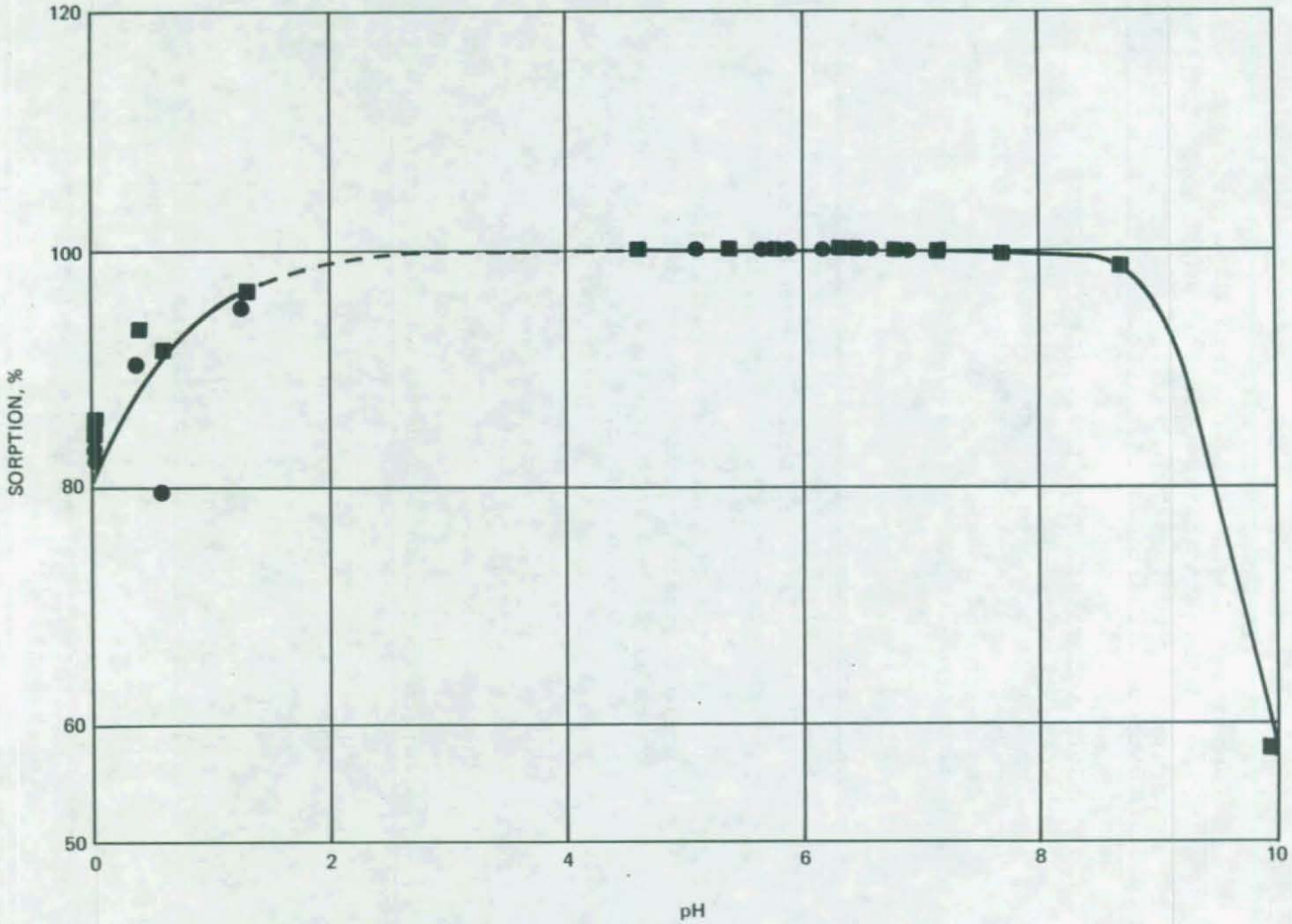


FIGURE 41 - Percent sorption of ^{238}Pu (20 ml solution) onto two grams of soil as a function of pH.

ium solution concentrations from 7×10^{-15} M to 2×10^{-7} M. This is not surprising since the soil exchange capacity (0.2 meq/g) is not challenged significantly ($<1 \times 10^{-2}$ %) using this concentration range. In an additional test, the filtrate solution from the sorption of the 2×10^{-7} M was equilibrated again with fresh soil and yielded approximately the same distribution ratio the second time. This indicates that essentially none of the plutonium would remain in solution after moving an extremely short distance through or on fresh soil.

(3) Sorption rate: Some preliminary rate studies were performed and the reaction was found to be about 98% complete in less than 5 min. As expected, the sorption reaction appears to be a half-time of less than 1 min. The rate of reaction tends to decrease with increasing pH, but does not appear to vary much with solution concentration.

The importance of these findings is that the plutonium would have been almost immediately sorbed onto the soil after being released through the pipe rupture.

(4) Distribution of plutonium as a function of soil particle size: In order to determine how the plutonium is sorbed onto the soil, a joint study was performed by Mound Laboratory and LFE.[5] Samples of soil were treated with plutonium solutions, as described previously, and air dried. Each sample was dispersed and separated by sedimentation techniques into six particle-size ranges. Each of these fractions were then analyzed for plutonium. As is shown in Figure 42, the amount of plutonium sorbed on the soil increases with decreasing mean Stokes diameter. The solid line in this plot is the theoretical slope relationship between ^{238}Pu concentration and sorption capacity. The points shown are the values determined experimentally during this study.

As can be seen, the experimental data are in reasonable agreement with a plutonium concentration dependence on surface area and/or ion exchange capacity. The fact that the small particles tend to provide better general agreement may indicate that there is a change in chemical composition with size.

Also in Figure 42, the plutonium concentration vs particle size data for the synthetic samples are

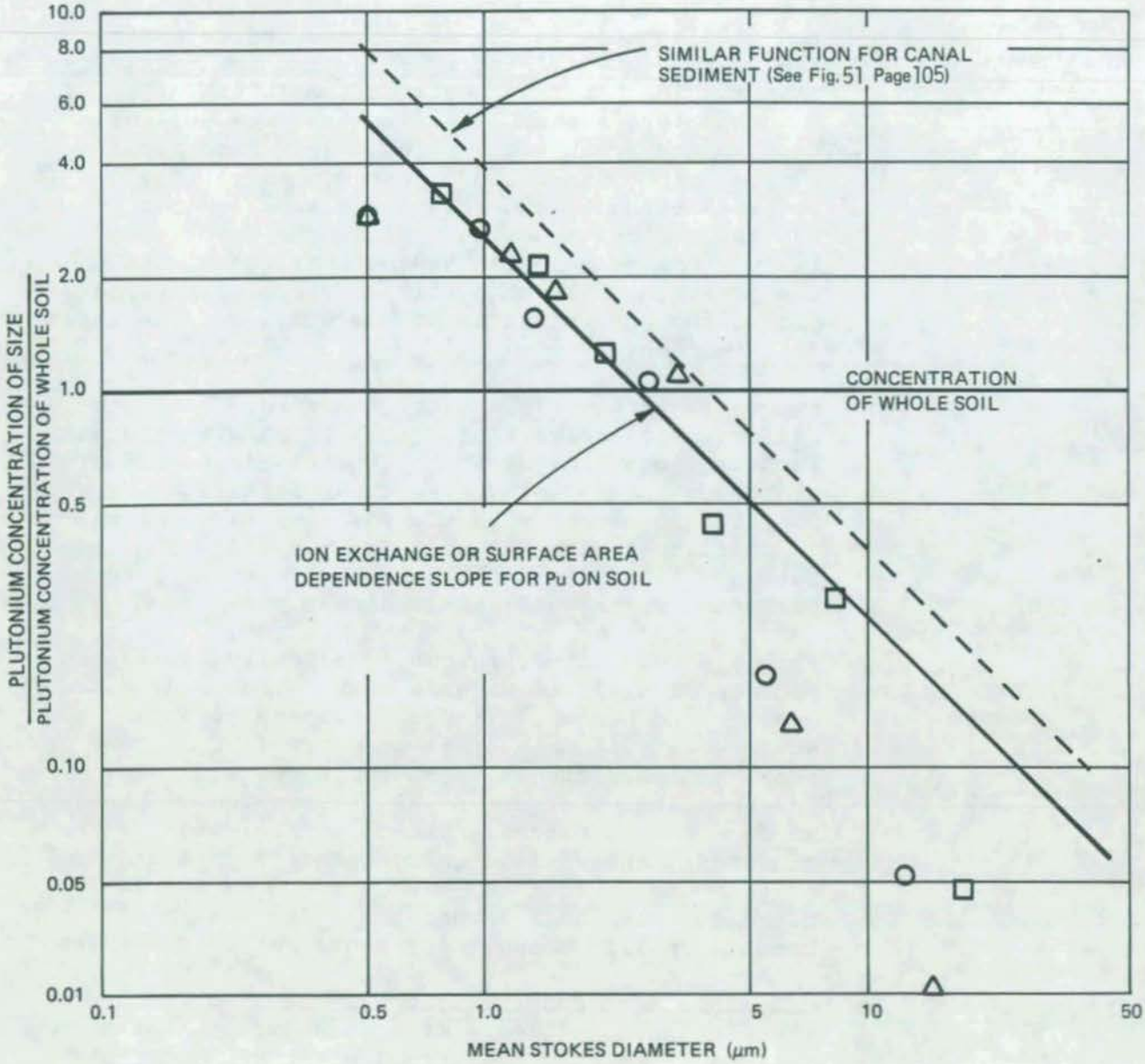


FIGURE 42 - Variation of ^{238}Pu concentrations in soil as a function of fully dispersed particle size.

compared with similar data collected on contaminated sediment from the canal. Except for a displacement in the two functions, there appears to be agreement that the plutonium sorbs as a function of ion exchange capacity or surface area. In both determinations, the smallest fraction exhibited plutonium concentrations from three to eight times the plutonium concentration of the whole sample.

The displacement in the two curves indicates that the average particle size of the two soils is different. The waterway sediment sample exhibits somewhat larger particle sizes than the soil used to make the synthetic samples. This also explains the fact that the ratio of the concentrations of the smallest size to the average size is greater for the sediment samples. Since the majority of the plutonium is sorbed on the small particles, the presence of large particles serve mainly, to dilute the concentration of the whole soil/sediment distribution.

(5) Autoradiographic alpha track analysis:[5] An autoradiographic analysis of soil treated with plutonium solutions indicates the general dispersion of plutonium in the soil rather than formation of PuO_2 particulates mixed with the soil. This observation is consistent with the sorption behavior of plutonium on soil and is in agreement with similar tests performed on the off-site sediment.

(6) Proposed bonding mechanisms: Basically, soils are made up of silicate materials and other minerals. Quartz sand has a continuous silicate structure where each silicon atom is bound to four oxygen atoms and each oxygen is bound to two silicons. This continuous structure is interrupted at the surface and in natural systems (where water is abundant), the surface is composed of unsaturated oxygen bonds. That is, each surface oxygen is bound to only one silicon atom. The remaining bond is usually occupied by other cationic species.

Clays are more complicated, but are also based on the continuous silicate structure except that some silicon atoms have been replaced by Mg^{2+}, Al^{3+}, Fe^{3+}, and Fe^{2+} (mostly Al^{3+}). These substituted atoms in the silicate structure result in variations in long range crystal structure. Rather than forming three dimensional silicate networks as in quartz, many clays form two-dimentional sheets.

which cleve easily to form plates. The surface of these layered sheets that make up the clay particles also exhibit unsaturated oxygen bonds. This, in part, accounts for the higher sorption capacity in clays (relative to the same size silicate particle) since sorption can take place between the silicate layers within the clay particles.

The unsaturated oxygen bonds in natural soils and clays are occupied by cations such as H^+, K^+, Ca^{2+}, Mg^{2+}, or other available cations. The bonding strength order of these cations is:

$$Pu^{4+} > H^+ > Al^{3+} > Ba^{2+} > Ca^{2+} > Mg^{2+} > NH_4^+ > K^+ > Na^+.$$

In order to bond to the silicate, an ion must displace or exchange with the cation already bonded:

$$\text{A-Clay} + B^+ \leftrightarrow \text{B-Clay} + A^+.$$

The extent of the exchange depends on the relative strength of the bonds and the relative solution concentrations of the two cations.

Some cations form silicate bonds that are fairly weak (such as Na^+ and K^+) and may be only electrostricted while other metal cations may form bonds that are much stronger and may even develop covalent character.

Tetravalent plutonium ions are well noted for the formation of strong bonding (complexing) with oxygenated ligands.[6-8] The strength of plutonium oxygen bonds is also indicated by the acidic character of plutonium hydroxide forming hydrous plutonium oxide.

It is not surprising then that plutonium ions can compete with hydrogen ions for the bonding sites on the silicates even when the Pu^{4+}/H^+ concentration ratio is 10^{-11} or less. The very large bonding potential of plutonium[6-8] suggests that sorbed plutonium cannot be significantly displaced from soil by the concentrations of cations existing in nature.

Chemicals that complex the plutonium[6-9] compete with the silicate particles for the plutonium and tend to reduce the extent of sorption of the plutonium on soil. For example, the formation of plutonium hydrolytic species $PuOH^{3+}$, $Pu(OH)_2^{2+}$, $Pu(OH)_3^+$ and $Pu(OH)_4$ (as well as "polymeric" forms) tends to reduce ion exchange sorption.[10]

However, even at pH 10 the distribution of plutonium in the aqueous phase is only 7.2×10^{-2} (Figure 40).

Some of the hydrolytic plutonium "polymeric" forms may adsorb to the surface of the soil particles and the precipitation of $PuO_2 \cdot (H_2O)$ may be nucleated by the colloidal soil particles when the plutonium concentrations are relatively high ($>10^{-6}$ M).[11,12] These physical adsorption mechanisms are not expected to be extensive at the low plutonium concentrations (10^{-9} M) used for this study.[10]

Moderately strong organic complexing agents, such as citric acid, have been shown to reduce the sorption of plutonium on soil. However, relatively high citric acid concentrations (0.1 M) were required to reduce the sorption to $D_{w/s}$ values of 4.2×10^{-2}.

3. Soil Erosion and Transport Mechanisms

The extent of soil erosion by surface water depends on:[13]

- The velocity of the water,
- The soil particle-size distribution,
- The degree of consolidation of the soil, and
- The amount of ground cover or root mats holding the soil in place.

The erosion behavior of soil has been reported and is presented in Figure 43.[13,14] The most erodible soil fractions are sands and unconsolidated silts and clays. The effect of consolidation decreases the erosion potential of silts and clays, but has little effect on the larger size fractions.

The effect of ground cover or root mats is to hold the mass of soil in place and reduce total erosion. The upper parts of the plants also tend to reduce the water velocity near the water/soil interface.[13]

At the time of the pipe rupture, most of the plutonium was sorbed onto the subsurface soil surrounding the pipeline and, therefore, was not subject to erosion. The plutonium solution which came to the surface was quickly sorbed onto soils protected by ground cover and root mats (winter conditions). Since there was no rainfall at the time of the rupture, the erosion potential was nearly zero. Had it rained while these conditions prevailed, the erosion of

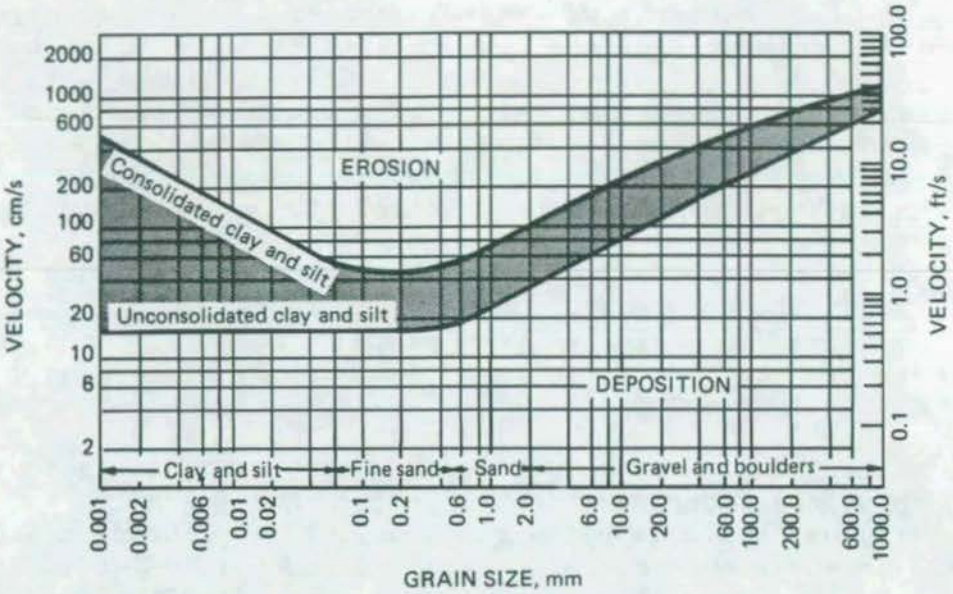


FIGURE 43 - Erosion as a function of particle size and stream velocity.[13,14]

contaminated surface soil would have been minimized by the ground cover and consolidation of the soil. On the other hand, the presence of plutonium contaminated soil on the 19° slope of the hillside would have been greater than at the rupture site because the velocity of the water would have been higher. Erosion from this grassy hillside, however, is not extensive as indicated by the presence of very little sediment in the concrete flume.

During the period between the rupture, January 23, 1969 and the heavy rainfall on January 28-30, 1969, extensive excavation was taking place at the rupture site. The loosened soil removed from around the ruptured pipeline dropped onto the street, increased the total amount of contaminated soil available for erosion and presented unconsolidated soil rich in sand, clay, and silt particle sizes.

When the rains came on January 28-30, the erosion potential was very high. The large amount of water, collected on the paved surfaces, easily eroded this unconsolidated contaminated soil from the street and excavations to street storm drains and over the hillside where it followed the natural drainage pathways. The velocity of water in all of these surface water drainage channels would have precluded extensive sedimentation until the larger off-site channels and lower stream velocities were encountered.

The movement of plutonium from the rupture site to the run-off hollow probably resulted from erosion of contaminated soil

from the outside of drums stored at the end of the street. The flow patterns of water in the area would not have carried the sediment from the rupture site to the run-off hollow. Since only a limited amount of soil was clinging to the outside of these drums, very little plutonium was found in the sedimentation areas of the run-off hollow. The general pathways of erosion of contaminated soil from the rupture site are shown in Figures 44 and 45.

Since the rainfall was very heavy,[3] the suspended and bedloads of uncontaminated erosion products in all of the drainage channels was very large which diluted the concentration of the contaminated sediment by perhaps as much as 1000 times or more.

Water samples taken during this 1969 rainfall indicated plutonium concentrations less than RCG ($<5 \times 10^{-3}$ nCi/ml). The extensive sorption reaction between plutonium and soil was not known to the emergency personnel in 1969; therefore, the absence of plutonium in the water falsely indicated that no significant amounts of plutonium were leaving the site and the situation was under control.

It is believed that the majority of the plutonium/sediment found off-site was eroded and transported off-site during the heavy rainfall of January 28-30, 1969. An additional smaller amount probably was eroded and transported in subsequent 1969 spring rains[3] before ground cover returned. After the area was cleaned up, grass grew on the hillside, and the soil became consolidated, additional erosion of the plutonium/soil which remained on the hillside would have decreased continually.

4. Deposit Mechanism

As in the process of erosion, the important parameters for sedimentation of the erosion products are particle size and water velocity including turbulence.[13]

The particle size distribution of erosion products, in a natural water system, exhibits extensive agglomeration due to incomplete dispersion and the presence of natural flocculants in the water. In order to measure the extent of agglomeration that might be expected in the natural system, two types of laboratory sedimentation measurements were performed.

The first test was performed[15] by methods recommended by the American Society for Testing and Materials (ASTM)[16] to determine the *basic* particle size distribution of the sediment. The second test was performed on an aliquot of the same sediment sample using a modification of the ASTM method to determine the natural particle size distribution of erosion products. In the second test,

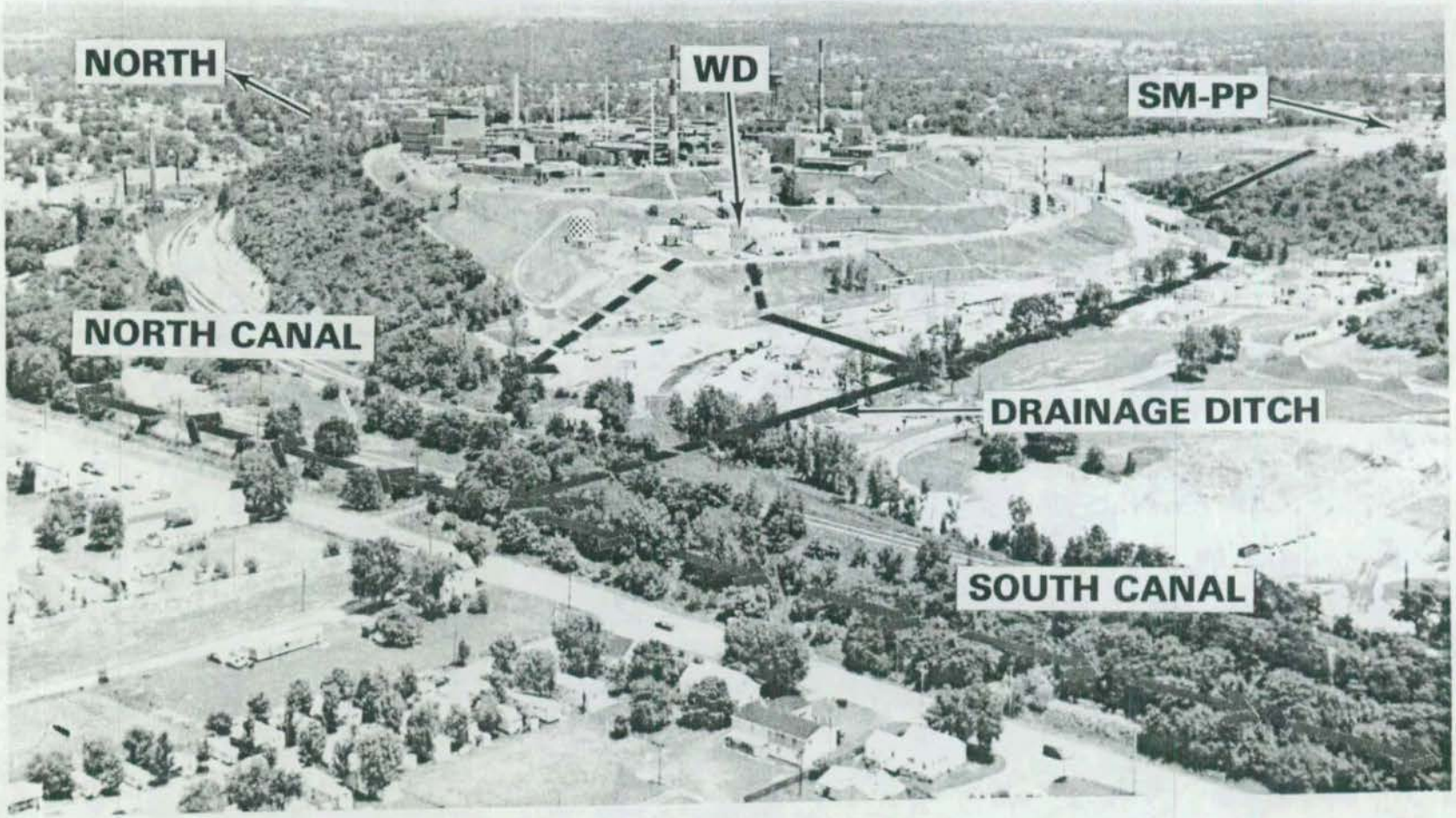


FIGURE 44 - Aerial view of Mound Laboratory from the west, showing pathways contaminated soil erosion in 1969.

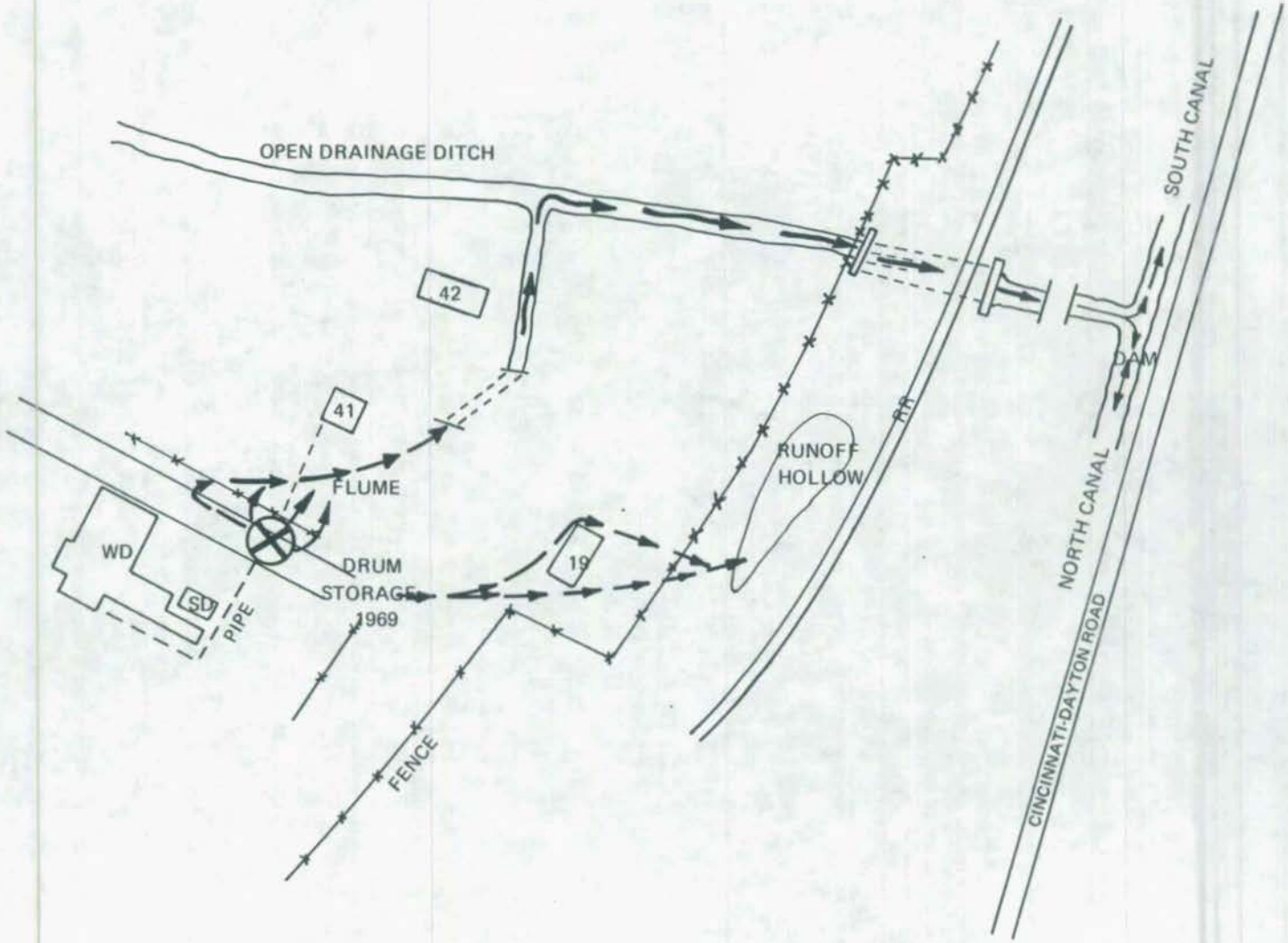


FIGURE 45 - Schematic diagram of contaminated soil erosion pathways.

no dispersant was used and the sedimentation was performed in natural water taken from the sampling site. Everything else was performed[15] exactly as the ASTM method described.

The results of these tests are compared in Figure 46. In the absence of the dispersing agent and in natural water, the smaller particles are extensively agglomerated such that the sedimentation proceeds much faster than would be predicted by the basic particle size of the sediment.

Sedimentation of erosion products in the waterways occurs when the velocity and turbulence of the streams fall below that necessary to maintain the suspension of a particular size range of particles.[13] For example, in very swift sections of the waterways, the bed contains very few deposits, or the deposits are composed mainly of larger gravel and rocks. As the velocity decreases downstream, the deposits indicate successively smaller particle size distributions. This mechanism describes the overall picture of the deposition patterns; however, local variations in the stream velocity such as near an obstruction or on grassy banks of flooded waterways may result in the local deposition of smaller particles.

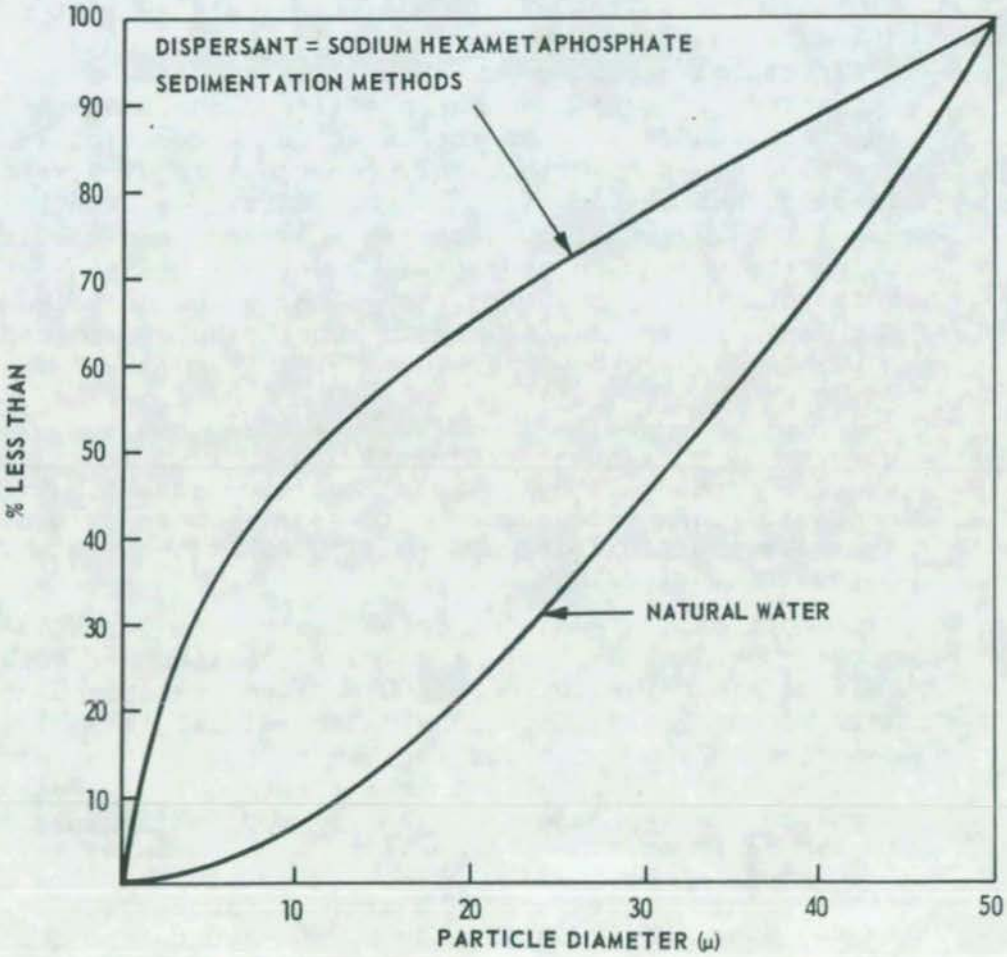


FIGURE 46 - Extent of agglomeration of erosion products in the natural water system.

During the environmental survey sampling program, the size fraction greater than 1000μ was routinely removed from each of the samples and weighed. The percent of this larger size fraction in the first foot is probably indicative of average stream velocity during high velocity waterflows. The percent of the larger size fraction was generally quite large in the sediment of the swift on-site and off-site drainage ditch (central channel). The large size fraction on the grassy banks, which are often flooded, was lower.

In the North and South Canals, an extensive deposition of the >1000 μm size fractions (Figure 47) always preceeded the major deposits of the plutonium bearing sediment. This observation indicates the partial classification of particle sizes in the sediment as a result of flow velocity characteristics of the channel and in preferential sedimentation locations.

There are two main plutonium/sediment deposits in the off-site waterways at locations most favorable for erosion products sedimentation. The deepest sediment (and highest plutonium deposits) was found approximately 55 ft north of the earthen dam in the North Canal. Figure 48 shows the differential plutonium inventory distribution along the length of the canal and the profile of the sediment depth in the North Canal. As the turbulent water flows through the pipes to the North Canal, the canal is wide, the water is calm, and the erosion products settle quickly. In addition, the very shallow water in this area has resulted in an extensive growth of cattail reeds which further enhances sedimentation by reducing the velocity and turbulence of the water flow. The result is a build-up of sediment in this area as indicated by the sediment profile.

The plutonium/sediment was deposited in 1969 and has been covered by subsequent sedimentation of uncontaminated erosion products. By this action, the highest plutonium concentrations were buried 3 to 4 ft. There is evidence that some vertical (upward) mixing has taken place during turbulent periods.

The depth of the sediment decreases to the North due to erosion product depletion; however, smaller particles, bearing plutonium, have been deposited in subsequent favorable sedimentation locations. The initial 1969 plutonium deposits in the locations further north, are therefore thinner, and are usually in the first foot of sediment. There is some evidence of minor secondary transport of plutonium sediment from the sediment area north of the dam to locations further north during high turbulent flow conditions. This general trend continues through the system into the south basin of the South Pond and to the North basin into the North Pond, exhibiting deposits of less and less plutonium bearing sediment.

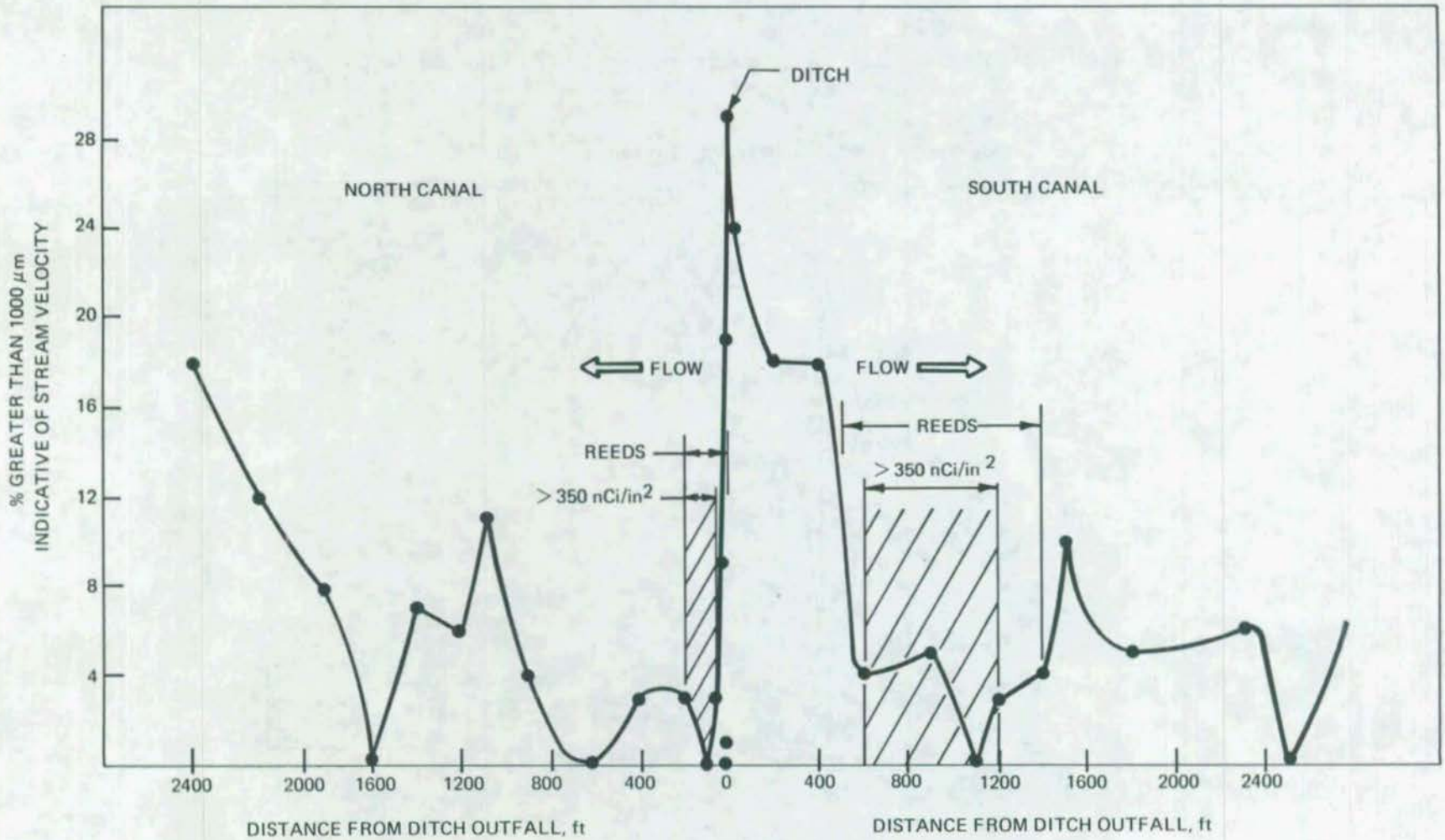


FIGURE 47 - Percent of large particle size fraction in first-foot sediment samples taken along the Miami-Erie Canal.

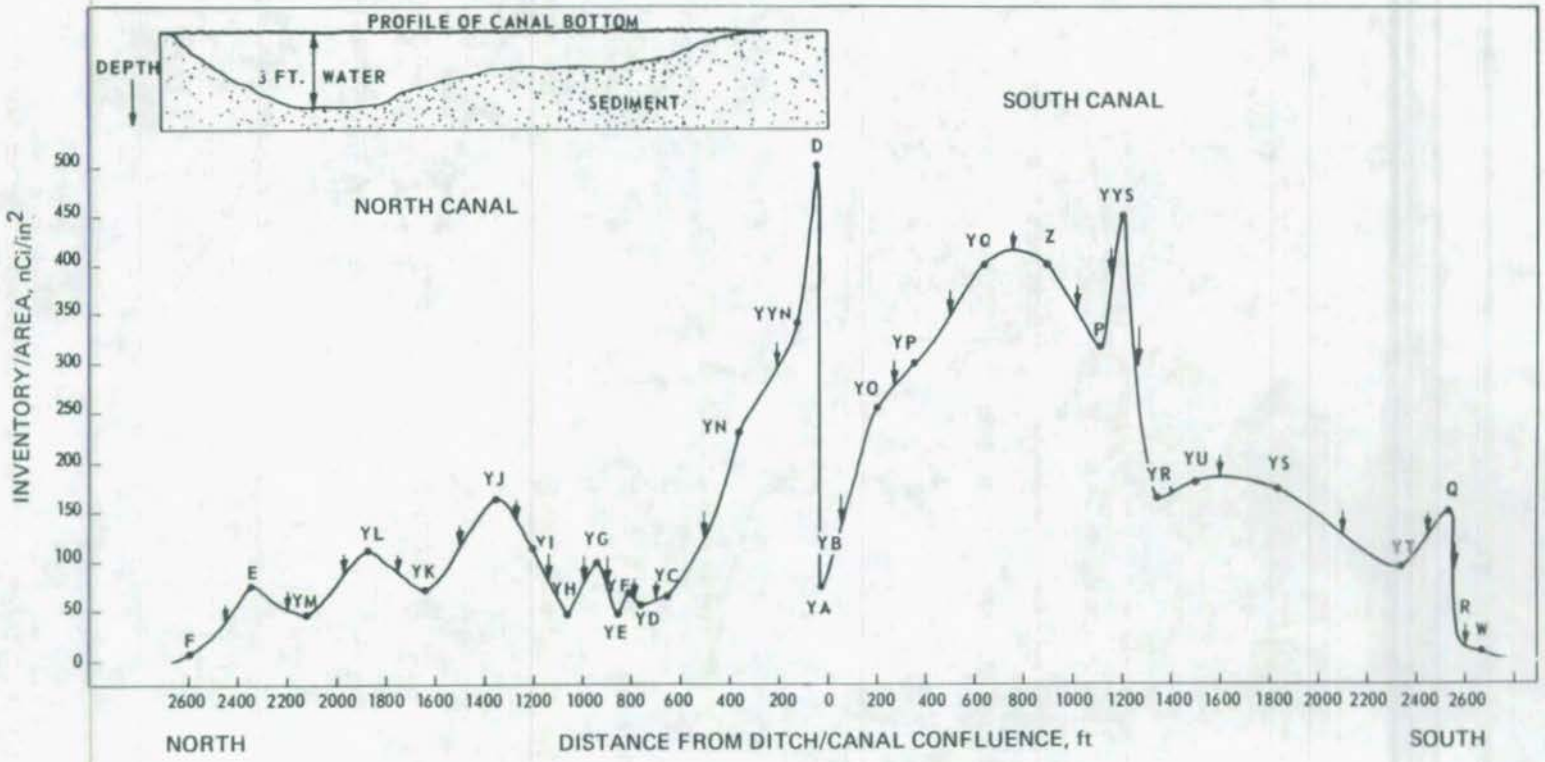


FIGURE 48 - Inventory of ^{238}Pu along the length of the Miami-Erie Canal.

In the South Canal, the initial channel running south of the ditch/canal confluence area is rocky for several hundred feet. Beyond this swift and turbulent zone, another major sedimentation area is evident. At this point, approximately 500 to 600 ft south of the earthen dam, the percentage of >1000 μm particles decreases rapidly, indicating a reduction in the stream velocity. In this area there is also a growth of cattail reeds.

The apparent change in the stream velocity is not as sharp as at the North Canal sedimentation location, therefore, the deposition of plutonium sediment (Figures 47 and 48) is much broader than in the north (~800 ft). In the South Canal sedimentation area, the thickness of plutonium sediment deposit is less, existing mainly in the first 2 ft of sediment. The plutonium inventory trails off in the remaining 1000 ft of the South Canal except for a small additional sedimentation area at the end of the South Canal behind the spillway leading to the overflow creek, where water tends to pool.

The overflow creek is narrow and deep; therefore, the velocity of the water flow is very swift as indicated by very few sediment deposits in this channel. Near the river outfall, sediment containing plutonium tends to collect and deposit. Minor sediment deposits extend downstream in the river along the eastern bank of the river for several hundred feet. This results from the lower velocity of the river near its banks.

The river itself exhibits very little silt and clay deposit. The river bottom was found to be mostly sand and gravel except in selected localized areas such as near the ends of dams.

Due to the minute concentrations of plutonium-238 (<0.0001 nCi/g), found in the downstream river sediment, it is not possible to statistically prove that the downstream river inventory is different from zero, based on the analytical data. It is likely, however, that some of the contaminated erosion products may have passed through the waterways and were carried down the river. These contaminated erosion products would have been severely diluted by the extremely large amount of uncontaminated erosion products carried by the river and are, therefore, very difficult to detect in the river sediment.

It will be shown later in this report that the river sediment concentrations are insignificant from a health and safety viewpoint. However, in order to place a value on the plutonium-238 inventory which might have been discharged to the river by this mechanism, a set of conservative conditions were assumed.

- 40 in. of rain ran off the 200-acre drainfield, flowed down the South Canal, through the overflow creek into the river (8.22×10^8 liters or 2.18×10^8 gal).

- The average suspended erosion product load in the water at the overflow creek outfall was between 500 and 1000 mg (dry weight)/ℓ. The amount of erosion products discharged to the river, therefore, would have been about 450 to 900 tons dry weight.

- The plutonium-238 concentration in these erosion products was equal to the highest concentration foun in sediment (4.6 nCi/g).

Based on these assumptions, it is estimated that about 1.9 to 3.8 Ci of plutonium-238, sorbed onto erosion products, could have been carried down the river as a result of this release.

Summing this estimated amount with the plutonium inventory analytically found deposited in the waterways (5.2 Ci), the total off-site release of plutonium from this incident may have been about 7 to 9 Ci.

5. Anticipated Future Movement of the Plutonium Deposits

a. Future water erosion.- In general, a portion of the surface plutonium/sediment in each of the waterways will continue to be re-entrained during high-flow periods and deposited downstream. The concentration of these surface deposits will decline as they are diluted by uncontaminated erosion products.

In the North Canal, the majority of the plutonium is buried 1 to 4 ft deep in reasonably consolidated deposits. These deposits are not likely to move except under incredibly extreme flow conditions. In the first 5 yr, only 5% of the total North Canal inventory has been transported into the South and North Ponds. The majority of this inventory was probably transported in 1969-70 before the plutonium sediment was consolidated or buried by subsequent sedimentation. The rate of transfer to these ponds is believed to be very small today and should decrease further in the future.

In the South Canal, where waterflow has a considerably higher velocity, more extensive surface erosion could potentially take place. However, this channel is extensively covered with vegetation and the sediment is reasonably well consolidated due to the wet/dry cycling. In addition, most of the plutonium is buried and is therefore less susceptible to erosion. For these reasons, extensive movement of the plutonium deposits

to downstream areas is not likely. A continuous trickle of surface erosion will probably result in very slight inventory increases in the south half of the South Canal in the future. These deposits will be diluted by non-contaminated erosion products carried by the stream reducing the plutonium-238 concentration. The future surface deposits at the overfall creek outfall should provide a measure of the erosion from the South Canal.

b. Surface leaching. - According to the distribution ratio of plutonium between water and sediment ($D_{w/s} = 1 \times 10^{-5}$), the concentration of plutonium in the water flowing through these waterways will be very low and will not provide a significant mechanism for transport of the plutonium from the deposits to the river.

c. Migration through soil. - Due to the sorption of plutonium on soil and the very low water permeability of the clay waterway beds,[4] the movement of plutonium through this medium is expected to be insignificant. The rate of movement of the plutonium by this mechanism can be described by:[17]

$$\frac{d[Pu]}{dz} = \frac{dw/dz}{1/D_{w/s} + B}$$

where d[Pu]/dz is the rate of plutonium migration through the clay; dw/dz is the rate of water flow through the sediment; $D_{w/s}$ is the plutonium distribution ratio between water and sediment; and B is the fraction of free volume in the clay.

Water permeability in clay was measured to be 0.1 to 1.0 m/yr,[4] the distribution ratio is 10^{-5}, and the fraction of free volume in the clay is <0.4; therefore, the migration rate can be estimated at about 1×10^{-6} to 1×10^{-5} m/yr.

V. HEALTH AND SAFETY ANALYSES

A. Introduction

The extensive work summarized in the previous sections of this report was performed by Mound Laboratory in order to make a knowledgeable evaluation of the hazards associated with the plutonium-238 deposited in the waterways adjacent to Mound Laboratory.

It is recognized that the health and safety aspects of plutonium are under public discussion and debate. Most of the debate centers around the setting of national priorities on energy production and fears that *general plutonium levels* in the environment will increase to a level hazardous to man. This is worthwhile scientific controversy; however, it tends to confuse the general public and generate fear, even when it is not justified by the conditions and circumstances accompanied by accidental releases. Whenever such an accidental release occurs, the public has a right to demand and get a timely, systematic, and realistic evaluation of the hazards it presents to them.

Plutonium, like most chemicals, behaves quite differently under different conditions and circumstances. For this reason, the Health and Safety Analyses presented in this report applies *only* to the specific set of conditions and circumstances which are present and operable in and adjacent to the waterways near Mound Laboratory.

There has been an honest attempt to make judgments on the conservative side. Overall, the conclusions reached from these health and safety analyses are considered to be *substantially* on the safe side.

B. Composition and Radiation Properties of Plutonium-238[18]

Plutonium-238 is a manmade element, produced by nuclear reactor irradiation of neptunium-237. Since Mound Laboratory does not have nuclear reactors, the plutonium-238 is prepared elsewhere, and sent to this site in a "pure" condition. That is, it contains no significant fission products, and therefore has very little penetrating radiation. The isotopic composition of this plutonium is listed in Table 11 and its associated radioactive impurities are listed in Table 12. The radioactivity is almost entirely short-ranged alpha radiation with a small amount of weak (20 keV) beta radiation from plutonium-241. Over 99.9% of the alpha radiation results from the decay of plutonium-238 which has specific alpha activity of 17.11 Ci/g. The energies of these alpha particles are listed in Table 13.

The energy and amount of penetrating radiation (x-ray and gamma) from plutonium-238 are listed in Table 14. As can be seen, these dose rates are insignificant at the ^{238}Pu concentrations under discussion.

Table 11

ISOTOPIC COMPOSITION (TYPICAL OF MOUND LABORATORY ^{238}Pu)[18]

Plutonium Isotope	Principal Decay	Half-Life (yr)	Wt %
236	α	2.85	0.0001
238	α	87.78	80.0
239	α	24082	16.5
240	α	6537	2.7
241	β	14.35	0.7
242	α	3.87×10^5	0.1

Table 12

^{238}Pu IMPURITIES[18]

Isotope	Wt %	Remarks
^{234}U	~0.19	Increases with Decay of ^{238}Pu
^{237}Np	0.03	
^{241}Am	0.05	Increases with Decay of ^{241}Pu
^{231}Pa, ^{232}Th, ^{232}U, ^{235}U, ^{236}U, ^{227}Ac	<0.1	

Table 13

ALPHA EMISSION FROM ^{238}Pu[18]

Alpha Energy (MeV)	Emission (Ci/g ^{238}Pu)
5.499	12.15
5.456	4.95
5.358	1.5×10^{-2}
5.214	8.6×10^{-4}
4.70	1.2×10^{-5}
Total	17.11

Table 14

COMPUTED PENETRATING RADIATION DOSE RATE AT ONE METER FROM AN UNSHIELDED ^{238}Pu POINT SOURCE[18]

Approximate Gamma or X-Ray Energy (keV)	Dose Rate (mR/hr/nCi ^{238}Pu)
17	3.8×10^{-8}
44	1.6×10^{-11}
99	4.3×10^{-12}
150	7.8×10^{-13}
760	4.7×10^{-15}
875	2.3×10^{-13}

C. Potential Hazards of Plutonium-238

The potential hazards of radiation to man results from external and internal exposure of vital organs. Since plutonium-238 emits very little penetrating radiation, the potential for external exposure from the amount of ^{238}Pu under consideration is insignificant. The short-ranged alpha and beta radiations cannot penetrate the skin to irradiate vital organs. Therefore, the potential hazard of plutonium-238 is almost entirely associated with internal exposure. Plutonium taken into the body of man deposits in vital organs where the short ranged energetic alpha radiation can irradiate vital cells and may cause harm.

The major entry mechanisms for internal uptake of plutonium in man are:

- Ingestion of plutonium in food, water, or other materials swallowed.
- Absorption of plutonium through the skin or wounds.
- Inhalation of plutonium from the air.

Therefore, to be taken internally, the plutonium must transmigrate from its present location to materials which might be ingested; onto the skin or in wounds where it might be absorbed; or suspended in the air where it might be inhaled. The path that the plutonium follows to finally be taken internally by man can be described as the "pathway to man." The potential pathways to man for the plutonium under consideration will be the subject of detailed discussion later in this report.

Two groups of scientific experts:

- National Council for Radiation Protection (NCRP)

- International Commission on Radiological Protection (ICRP)

have reviewed the large body of biomedical data which has been collected by experiments on animals as well as from the medical histories of occupational workers who have been accidentally exposed. Based on these data, they have established safe occupational plutonium guidelines or standards[19-21] which are listed in Table 15. The most critical points of reference are bones and lungs where the permissible body burden is established as 40 nCi and 15 nCi, respectively.

Table 15

PERMISSIBLE TOTAL BODY AND LUNG BURDENS FOR ^{238}Pu IN MAN[22,23]

Critical Reference Organ	Permissible Total Body or Lung Burden: Occupational Workers (nCi)	Permissible Total Body or Lung Burden: Individual in General Population (nCi)
Total Body	300	30
Bone	40	4
Liver	200	20
Kidney	300	30
Lung	15	1.5

Although the occupational standards include safety factors, these standards are reduced by an order of magnitude before they are applied to individuals in the general population.

From these plutonium standards some important RCG have been derived for plutonium in water and air as are listed in Table 16. If an individual is continuously exposed, during his lifetime, to water or air which has plutonium concentrations at the RCG, he will not exceed a permissible body or lung burden of plutonium.

Table 16

RADIOACTIVITY CONCENTRATION GUIDES FOR ^{238}Pu IN UNCONTROLLED AREAS[22]

Reference Material	Plutonium-238 Radioactivity Concentration Guide (RCG): Soluble Forms	Plutonium-238 Radioactivity Concentration Guide (RCG): Insoluble Forms
Air	7×10^{-14} μCi/cm^3	1×10^{-12} μCi/cm^3
Water	5×10^{-6} μCi/cm^3	--------*
Soil	None	None
Sediment	None	None

*Depends on solubility.

To date there is no officially accepted RCG for plutonium concentrations in soil or sediment. The derivation of safe and reasonable RCG values for plutonium in soil and sediment is more difficult than for water and air. Water and air have nearly a constant composition, behavior and properties everywhere, while soil/sediment composition, behavior, and properties vary greatly from place to place.

In addition, because soil and sediment are not normally considered to be in the direct pathway to man, many other factors must be considered to evaluate man's future potential plutonium uptake from this source. These considerations will be discussed in more detail later in this report.

D. Health and Safety Evaluation Under the Prevailing Conditions

From the time of discovery until the full extent of the contamination was known, a continuous health and safety analysis was performed and re-evaluated as the data became available. This analysis was based on actual concentrations of plutonium in the air or of other materials which were similar to things that might be ingested. The final results of this analysis are summarized in Table 17.

Table 17

EVALUATION OF HEALTH AND SAFETY UNDER PRESENT CONDITIONS

Material Analyzed	RCG	Maximal ^{238}Pu Concentration Found	% RCG	Permissible* Ingestion Amount (lb/yr)
Sediment	None	4.56 nCi/g	-	0.9
Water	5×10^{-6} μCi/ml	1×10^{-8} μCi/ml	0.3	-
Air** (permanent station continuous)	1×10^{-12} μCi/cm^3	7×10^{-17} μCi/cm^3	0.007	-
Air*** (worst-case grab samples)	1×10^{-12} μCi/cm^3	3×10^{-15} μCi/cm^3	0.3	-
Vegetation (grass)	None	3×10^{-3} nCi/g	-	1400
Vegetation (algae)	None	1×10^{-1} nCi/g	-	42
Fish (edible portion)	None	5×10^{-6} nCi/g	-	>800,000

*Ingestion of this amount could potentially result in an uptake of 0.057 nCi/yr (1/70 of a permissible body burden)

**Permanent continuous air sampling station between North and South ponds

***Air samples taken one foot above ground at ditch/canal confluence and east of ponds (average value)

1. Air Concentration

Since inhalation of plutonium is believed to be an important pathway to man for plutonium, measured air concentration was used as one of the major indicators of potential hazards. As a part of the routine environmental survey program, Mound Laboratory has maintained five permanent air sampling stations on site since 1972. The three sampling stations along the western edge of the site are only a few hundred yards east of the North and South Canals. Prior to 1971, portable air samplers were used to periodically sample the air especially along the boundaries of the Laboratory site and at selected off-site areas. In 1972, additional permanent air samplers were installed at the City sewage plant near the overflow creek, across the river from the Laboratory and in Miamisburg (north) as well as many other locations off-site in other directions and at distances up to 28 mi.

A total of 27 such permanent stations is maintained by the laboratory's environmental staff. From this air sampling data, which are routinely published,[24] it is confirmed that plutonium air concentrations have been and are substantially below the RCG for plutonium in air.

In order to obtain more localized air concentration values, portable air samplers were used to collect samples immediately east (downwind) of the canal and ponds. These samples were taken with higher velocity air flow and much closer (1 ft) to the ground, than the permanent station (3-5 ft) to obtain a worst-case air concentration value. As expected, these localized samples indicated higher plutonium concentrations than the permanent station, but they were still substantially below RCG values. A permanent continuous air sampling station installed in 1975 between the North and South ponds is indicating much lower (by a factor of approximately 40) air concentrations than was found with the portable samplers.

From periodic physical inspections of the contaminated waterways, the contaminated sediment is essentially underwater, moist, or covered with vegetation. These conditions tend to severely limit the potential for suspension of the contaminated sediments in the air. Because the plutonium is strongly sorbed and fixed to the sediment, plutonium airborne suspension behavior is that of the sediment itself. Very little, if any, plutonium is present in PuO_2 particulate form which might separate itself from the sediment and be suspended independently in air.

Based on these considerations:

- The air concentrations of plutonium-238 are presently substantially below the RCG standards,

- The physical form of the plutonium in the sediment is known, and

- The sediment is underwater, moist, or covered with vegetation,

it is very unlikely that plutonium concentrations in air will approach the RCG standard under prevailing conditions.

2. Water Concentration

Water samples have been routinely and periodically collected in the canal for years. These samples have consistently indicated concentrations less than the RCG for plutonium in water.[24] Water samples collected in the waterways during this study also confirmed that the water samples are much less than RCG (for water) even when in contact with sediment that may have much higher concentrations. Many samples of contaminated sediment from the various waterways were maintained in contact with the natural water for more than a month to reach "equilibrium". The "equilibrium" distribution ratio (concentration of plutonium in natural water/concentration of plutonium in the sediment) was found to be 10^{-5}. Because of this very low solubility, the plutonium concentration is not expected to approach RCG concentration of plutonium in water.

3. Concentration in "Ingestible Materials"

As far as can be determined, there is very little vegetation in the immediate area of the waterways which can be described as edible by man. The highest land vegetation ^{238}Pu concentrations were found in the grass contaminated externally by sediment in localized flooding areas along the banks of the waterways. Even if this worst-case land vegetation concentration value is taken as indicative of edible plants and none is removed during preparation, then one would have to consume as much as 1400 lb of this vegetation per year to absorb 0.057 nCi/yr of plutonium (1/70 of body burden dose standard) through the gastrointestinal tract.

Again, there is no known aquatic plant life in these waterways which is normally consumed by man. The highest concentrations found in aquatic plant life were in the green algae. One would be required to eat 42 lb/yr of green algae to take up 0.057 nCi/yr of plutonium.

The waterways do contain some edible fish, mainly bluegills and carp. People in the Miamisburg area occasionally fish in the canals, river and ponds, and consume the few fish they catch. Based on the concentration of plutonium in the edible portions of these fish, they would be required to eat over 800,000 lb/yr of fish to approach the 0.057 nCi/yr uptake value.

As an absolute worst case, one would have to eat almost a pound of the highest concentration sediment found in the waterways per year to get 0.057 nCi/yr of plutonium uptake.

While it might not be impossible for these materials to be consumed in the amounts indicated, it seems extremely improbable. Therefore, it has been concluded that the plutonium deposited in these waterways does not and will not present a credible hazard to the public *under the prevailing conditions*.

E. Derivation of Plutonium-238 Sediment Concentration Decision Guides

1. Pathway Analysis of Plutonium in the Environment

While it is clear that the plutonium-238 in the off-site areas near Mound Laboratory does not present a hazard to the public under prevailing conditions, these conditions may be subject to change in the future. To make a definitive determination of these potential future hazards, a rather comprehensive pathway analysis has been performed.

Pathway analysis is an analytical technique for calculating the amount and the concentration of plutonium transmigration from its present state and location, into the materials which might be ingested, absorbed and taken into the body or suspended into the air where it might be inhaled by man and taken into the lungs.

A schematic diagram of the plutonium pathway analyses is presented in Figure 49. In order to perform these analyses,

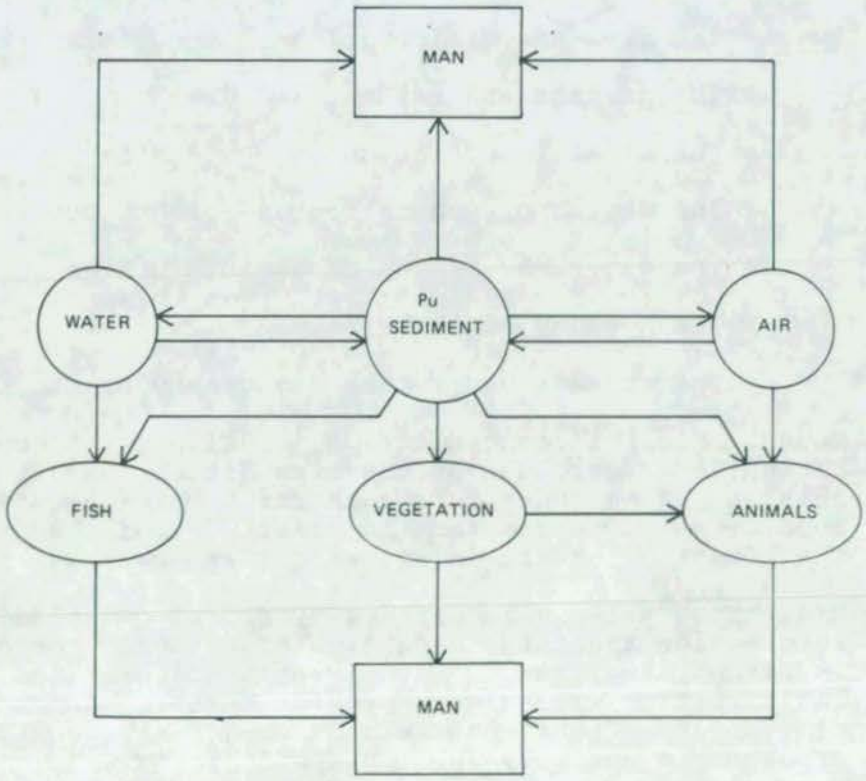


FIGURE 49 - Pathways to man.

it is necessary to 1) determine the present state/behavior and location of the reservoir of plutonium; 2) determine a credible worst-case set of future conditions and circumstances which could affect the transmigration of the plutonium along the pathway to man; and 3) experimentally determine or obtain from scientific literature, translocation transfer factors or coefficients which will adequately describe the magnitude of the transmigration of plutonium along the pathway to man.

In early 1974, J. W. Healy[25] of Los Alamos approached a similar problem in order to determine a general interim soil concentration guide from the available scientific information. The soil concentration guideline he proposed (0.225 nCi/g) is less than some of the concentrations found in the sediment of the waterways near Mound Laboratory. It must be remembered however, that the soil concentration guide he proposed was for general application to widespread contamination in a densely populated area, under all conditions and circumstances, at any location, for an indefinite period. By making his soil concentration guide "for general application," a large number of parameters which are known to vary widely with location, conditions, and circumstances had to be *assumed*. Among these parameters are:

- The chemical and physical properties of the plutonium
- The chemical and physical properties of the *in situ* soil
- The interaction and reactions between the plutonium and the soil
- The area and distribution of the contamination
- The present and future land use
- The climatic conditions
- The degree of vegetation and ground cover
- The geography of the contaminated area
- The geology of the area
- The present and future demography of the area.

Because his soil concentration guideline, if adopted, would be applied regardless of these conditions, Healy was more or less *obligated* under this constraint to assume "worst-case" conditions and parameters in order to *avoid underestimating* the potential hazard in rather extreme cases.

Healy recognized this difficulty and recommended that specific information regarding a particular situation or event be used in pathway analyses. "Such investigations aimed at a particular situation will always provide better answers than a general standard of this nature and such an approach to individual problems is entirely appropriate."[25]

Taking Healy's suggestion, his general approach to pathway analysis has been expanded and adopted but information and assumptions specific to this particular situation and location were factored in.

Examination of the major conditions and circumstances in the contaminated area near Mound Laboratory reveals that there are (and will be in the future) substantial differences from the case Healy assumed. The consideration of these differences is critical to a reasonable evaluation of the hazards in this case.

2. Specific Local Information Available

The major differences between Healy's assumption and conditions near Mound Laboratory are summarized in Table 18.

Table 18

COMPARISON OF THE CONDITIONS USED BY HEALY AND BY MOUND LABORATORY

CONSIDERATIONS	Conditions Assumed by Healy	Conditions at Mound Laboratory
Extent of Contamination	Wide Spread ~39 mi^2	Very Small ~0.01 mi^2
Physical Nature of Contamination	PuO_2 Fallout (Not fixed)	Pu Sorbed on Sediment (Fixed to Sediment)
Nature of Climate and Soil	Arid Western Soil	Moist Fertile Ohio Soil Large Clay Fraction
Nature of the Area	Wide Open Level Plain	Recessed Canal in River Valley - Lined with Trees
Solubility of Pu	Considered Soluble or Nearly Soluble	Solubility Very Low $\frac{\text{Pu Concentration in Water}}{\text{Pu Concentration in Sediment}} = \frac{1}{100,000}$

a. Nature of the area and degree of ground cover.- In Healy's model,[25] he assumes the contaminated area is an open, level, desert plain with little or no vegetation. One can envision this area being raked with high winds to stir up a huge dust cloud from the uncovered unconsolidated desert soil.

The waterways near Mound Laboratory, on the other hand, are located in a fertile river valley in Ohio where vegetation is lush and plentiful. The canal and other waterways are recessed into the ground and are generally lined with trees, grass and other vegetation. A large amount of the bed in the South Canal, which is under water only during

rainfalls, has been overgrown with vegetation. There is every reason to believe that if the entire waterway system were to dry up, vegetation would soon cover the entire area.

This heavy ground cover of thick root mats would almost certainly reduce the airborne suspension characteristics of the sediment even in high winds. The trees and shielding effect of the recessed nature of the waterways also tend to act as windbreaks which would also moderate the effect of high winds.

The low-form vegetation cover also tends to greatly reduce further water erosion of the contaminated sediment, thus reducing the potential for more widespread redistribution of the contamination.

b. Climate. - Healy assumes[25] a climate typical of the southwestern United States. The rainfall is very sparse in this arid or semiarid desert location and annual average windspeed was assumed to be about 11 mph.

The climate near Mound Laboratory[4] is typical of the central midwest. Summers are rather warm and humid, but temperatures rarely exceed 100°F. Winters are moderately cold with an average of about two days of subzero weather. The monthly average temperature and relative humidity are listed in Tables 19 and 20, respectively.

Table 19

TEMPERATURES IN THE MOUND LABORATORY AREA

Month	Average Daily Maximum (°F)	Extreme Maximum (°F)	Average Daily Minimum (°F)	Extreme Minimum (°F)
Jan	39.4	74	23.1	-14
Feb	41.8	73	24.0	-13
Mar	50.5	82	30.7	- 3
Apr	63.7	89	41.1	19
May	73.9	95	51.2	27
Jun	83.2	103	60.9	40
Jul	86.9	107	64.2	48
Aug	85.5	104	62.6	42
Sep	79.1	102	55.1	29
Oct	67.8	91	44.4	19
Nov	52.0	81	33.2	- 1
Dec	40.9	71	24.6	-13
Annual	63.7	107	42.9	-14

Table 20

RELATIVE HUMIDITY IN THE
MOUND LABORATORY AREA

Month	Relative Humidity at Four Times Per Day, Hour (EST)			
	01	07	13	19
Jan	80	81	71	75
Feb	79	80	67	71
Mar	76	79	58	64
Apr	75	77	55	60
May	79	78	54	61
Jun	82	80	55	61
Jul	81	81	52	58
Aug	81	84	52	61
Sep	80	85	50	62
Oct	78	84	52	65
Nov	77	81	61	68
Dec	80	82	69	74
Annual	79	81	58	66

The annual average precipitation is about 40 in./yr and is rather evenly distributed throughout the year as shown in Table 21. Snowfall averages about 27 in./yr.

Table 21

PRECIPITATION IN THE
MOUND LABORATORY AREA

Month	Precipitation	
	Normal (in.)[1]	No. of Days ≥0.5 in.[2]
Jan	3.66	2
Feb	2.62	2
Mar	3.61	2
Apr	3.74	2
May	4.14	3
Jun	4.71	4
Jul	3.40	2
Aug	3.03	1
Sep	3.28	2
Oct	2.53	1
Nov	3.12	2
Dec	2.75	2
Annual	40.59	25

[1]Based on 30 years of record
[2]Based on 10 years of record

The median annual wind velocity is about 8.5 mph predominantly from the south and west. The seasonal median wind velocity is listed in Table 22.

Table 22

WIND VELOCITY IN THE MOUND LABORATORY AREA

	Median Wind Velocity (mph)	Frequency of Wind in Excess of 24.2 mph (%)
Winter	9.9	0.8
Spring	9.5	0.6
Summer	6.9	0.2
Autumn	7.5	0.3
Annual	8.5	0.4

The climate at Mound Laboratory therefore results in higher average soil moisture and less wind erosion than might be expected in the arid setting Healy[25] describes. Most extreme winds observed in the Mound Laboratory area are associated with storms which deliver heavy rainfall. Therefore, under these conditions, the airborne concentration of suspended soil might be expected to decrease rather than increase.

c. Properties of the sediment.- The physical and chemical properties of the soil or sediment are quite important since the transmigration of the plutonium along the pathway to man and behavior in the environment is largely controlled by these properties as has been discussed previously. The sandy desert soil assumed by Healy would be expected to behave entirely differently from the sediment found in the waterways in Ohio.

The sediment properties expected to affect the behavior of the plutonium in the environment are:

- Large clay/silt fraction
- Low water permeability
- Drys to very hard, difficult to grind material
- Large ion exchange capacity

- Calcareous ($CaCO_3$)
- High acid neutralization capacity
- Agglomerates in natural water
- Remains agglomerated in dry state

The sediment in the waterways results from the water erosion of the glacial till and natural sedimentation processes. One of the most important features of the sediment is the very large clay and silt fraction which varies from 49 to 92% of the sediment as shown in Table 23. As a result, the sediment exhibits low water vertical permeability (0.1 to 1.0 m/yr)[4] and dries to a very hard mass similar to pottery clay. When this drying occurs, the surface cracks from shrinkage; however, very few fines are formed unless the mass is mechanically broken up. This very hard dry mass is very difficult to grind into small particles. A sample was air dried, broken, and ground in a mortar and pestle until it would pass through a 50-μm screen. The particle size distribution of this powder is shown in Figure 50. It should be noted that only about 2 to 3% of the powder is less than 5 or 6 μm (aerodynamic diameter = 10) which is considered respirable.

The major chemical constituents are clay (mostly hydromicas), $CaCO_3$ (calcite), $CaMg(CO_3)_2$ (dolomite), Fe_3O_4 (magnetite), SiO_2 (sand), organic materials, and water.

The large clay fraction is composed of mostly hydromicas with some kaolinite and montmorillonite. The sediment is very calcareous containing a large amount of calcite ($CaCO_3$) and dolomite ($CaMg[CO_3]_2$).

Table 23

PHYSICAL COMPOSITION OF SEDIMENT

Components	Composition (%) A	Composition (%) B
Clay and Silt	92	49
Fine sand	8	28
Medium sand	-	10
Coarse sand	-	5
Fine gravel	-	6
Coarse gravel	-	2

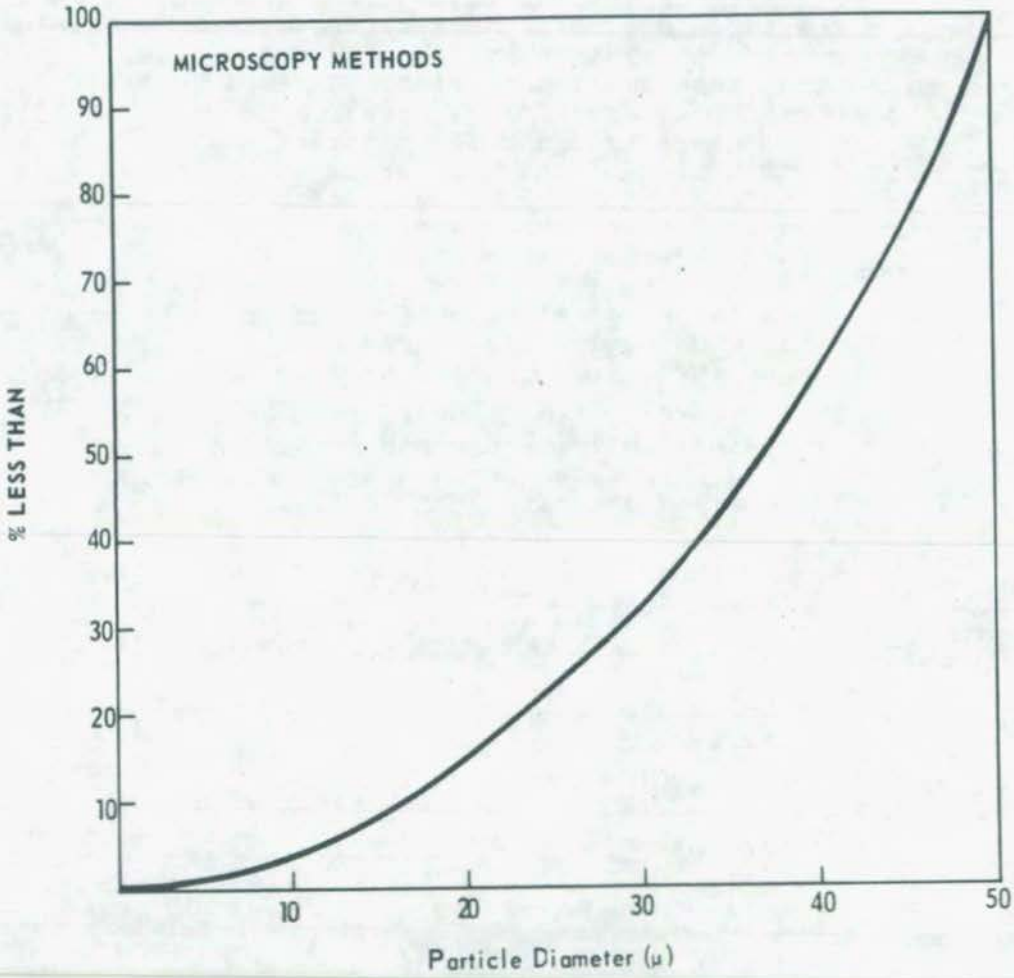


FIGURE 50 - Agglomerated particle size distribution of soil which was dried and ground to less than 50 μm.

The clays account for most of the ion exchange capacity and surface area while the calcareous constituents account for the buffer capacity (acid neutralization) and the pH of the natural surface water. These soil properties and their values are listed in Table 24.

Table 24

CHEMICAL PROPERTIES OF SEDIMENT

Property	Value
pH of water in equilibrium with sediment	7.5 - 8.3
Acid neutralization capacity	4 meq/g
Ion exchange capacity	20 meq/100 g

From the time of the release of the plutonium solution to the on-site soil, the properties of the soil and erosion products controlled the behavior of the plutonium. It is expected that the properties of the contaminated sediment will continue to be one of the major controlling factors in the transmigration of the plutonium along the pathway to man.

d. Physical nature and characteristics of the contamination. - The physical form of the contamination in Healy's analysis[25] is assumed to be fine PuO_2 particulate material such as would be present in nuclear fallout or from the release of fine PuO_2 powder. When such fine particulate PuO_2 powder is freshly deposited on soil, it tends to exhibit behavior independent of the soil properties. With time, however, these tiny PuO_2 particles become mixed with the soil and/or fixed onto soil particles and the airborne resuspension properties of the PuO_2 decreases.[26-29]

This process proceeds (depending on the nature of the soil and the climate) quite rapidly at first exhibiting half times of 35 to 70 days[26-27] over the first six to eight months.

This reduction in resuspension behavior apparently approaches a lower limit in a few years since 20-yr deposits have been reduced by only a factor of 1000.[30] It seems reasonable that a lower limit might represent the resuspension characteristics of the soil itself.

Samples of sediment in the waterways at Mound Laboratory have been examined by an independent testing laboratory, LFE,[5] using autoradiographic techniques to detect particulate PuO_2 content, by examination of the alpha tracks. Very few observable PuO_2 particles were detected in these samples. The radioactivity was generally quite

diffuse throughout the sample. This observation is consistent with the information presented in the cause analyses regarding the nature of the release. An acidic plutonium solution was released to on-site soil. The acidity was neutralized by the calcareous soil and the plutonium was strongly sorbed to the silt and clay fractions of the soil.

Since the plutonium is chemically sorbed onto the soil, it can be considered fixed in that the plutonium is not free to act physically independent of the soil. Under these conditions, one would expect the resuspension properties of contaminated sediment (from the beginning) to be similar to the behavior of well-aged fallout deposits.

Suspension characteristics of contaminated soil at Mound Laboratory were estimated by measuring the air concentration 2 ft above the ground under prevailing conditions. The ground contamination levels were measured by surface FIDLER Survey using the 17 keV x-rays emitted as a result of plutonium-238 decay. This technique is responsive predominantly to soil plutonium-238 concentration in about the top 0.1 cm of soil[31] ("very surface" of the soil). The measured suspension factors, K m^{-1} (defined as the air concentration in $\mu Ci/m^3$/surface contamination ($\mu Ci/m^2$), are listed in Table 25.

Table 25

RESUSPENSION FACTORS ESTIMATED ON MOUND LABORATORY SOIL

Geometric median of maximum values	3×10^{-9} m^{-1}
Geometric median of minimum values	5×10^{-10} m^{-1}
Geometric median value	1×10^{-9} m^{-1}
Maximum value on downwind edge	1.6×10^{-8} m^{-1}
Minimum value on downwind edge	9.0×10^{-10} m^{-1}
Maximum value on upwind edge	1.8×10^{-9} m^{-1}
Minimum value on upwind edge	3.2×10^{-10} m^{-1}

These resuspension factors are comparable (perhaps lower) to those measured on the 20-yr-old contamination in the GMX area at NTS which are in the 10^{-8} to 10^{-9} m^{-1} range.[30] These values indicate that the sediment even if allowed to dry would exhibit a factor of about 1000 fold less contaminated airborne suspension than was assumed by Healy.[25]

e. Chemical behavior of the contamination. - Although Healy[25] assumes the contamination is in the form of PuO_2 particles which are generally difficultly soluble, he considers the contamination soluble or partially soluble in most cases. For example, he assumes complete solubility in the ingestion and absorption intake mechanisms and partial solubility for inhalation. He makes these assumptions because the chemical behavior of the contamination in his case is truly unknown. The solubility of the plutonium in the contaminated sediment under the action of various reagents has been measured and these values are listed in Table 26.

Table 26

DISTRIBUTION RATIOS (WATER/SEDIMENT) AND SOLUBILITY OF PLUTONIUM FROM SEDIMENT

Solution	Distribution Ratio Water/Sediment $D_{w/s}$	Solubility(%) (20 ml Solution/1 g Sediment) S
Canal Water	1×10^{-5}	2×10^{-2}
Dilute Acid Initial pH = 1 Final pH = 5.5	1×10^{-5}	2×10^{-2}
Moderate Acid Final pH = 1	1.6×10^{-3}	3.1
High Acid (8M HNO_3)*	2.5×10^{-1}	83
Moderate Base pH = 10	Clay disperses	(61)**
Complexing Agent (0.1 M citrate)*	4.2×10^{-2}	46

*Tamura (HNL)

**Pu believed to be sorbed to colloidal clay suspension

When the sediment is placed in the natural water and permitted to reach equilibrium, the plutonium distribution ratio between the water and sediment was found to be 10^{-5} which demonstrates the extent of the sorption reaction. When placed in mild acid solutions (pH 1), the calcareous soil neutralizes the acid and the distribution ratio remains unchanged. When sufficient acid is introduced to adjust the solution to a final pH of 1, then the distribution ratio is slightly increased (more in solution) to 1.6×10^{-3}. Even when placed in strong acid (8 M HNO_3),

the plutonium is not completely removed from the sediment. The 8 M HNO_3 is also a complexing agent for plutonium, in that the high nitrate ion concentration would tend to assist the plutonium into solution forming $Pu(NO_3)_6^{2-}$.

The distribution ratio of plutonium between 0.1 M citric acid is 4.2×10^{-2}, indicating again that moderately strong complexing agents in reasonably high concentrations do not completely remove the plutonium from the sediment.

The behavior of the sediment in strong alkaline solutions is probably partially related to a physical reaction rather than being completely due to chemical formation of plutonium hydrolytic species and desorption of the plutonium. In strong bases, the clay fraction tends to become dispersed in the solution and is difficult to remove from solution. The apparent increase in solubility may be due, in part, to plutonium sorbed onto colloidal clay suspended in the solution.

The exact nature of the sorption reaction and the plutonium species sorbed on the soil is not unambiguously known at this time; however, from the solubility data, the bonding is quite strong and silicate bonding is believed most likely as discussed previously.

The plutonium is sorbed mostly on the clay and silt fraction of the sediment which has a very high ion exchange capacity and specific surface area. It has been demonstrated that the extent of sorption increases with decreasing surface median particle size indicating a surface area or ion exchange capacity dependence as shown in Figure 51. The smallest fully dispersed size fraction, however, exhibits no more than four to eight times the plutonium concentration measured on the full range of particle sizes in the sediment. Under normal circumstances, however, the sediment remains highly agglomerated and the particle size dependence of the plutonium concentration should be much less dramatic. These large agglomerated particles exhibit the surface area and ion exchange capacity and, therefore, plutonium concentration of the smaller particles from which they are constituted.

f. Distribution of the contamination.- The distribution of the contamination assumed by Healy is quite different than is observed in this case. The contamination he assumes is idealized to be on the "very surface" of the desert soil and evenly distributed throughout the area.

In this case, the contaminated sediment is very unevenly distributed with regard to both area and depth profile as has been described previously. The deposition of the sediment was controlled by sedimentation conditions which vary greatly within the waterways. No attempt will be made to use an area-wide average concentration value for the 0.01 mi^2 area even though the concentration varies over greater than four orders of magnitude. Instead the maximum values will be used in all cases as though it were

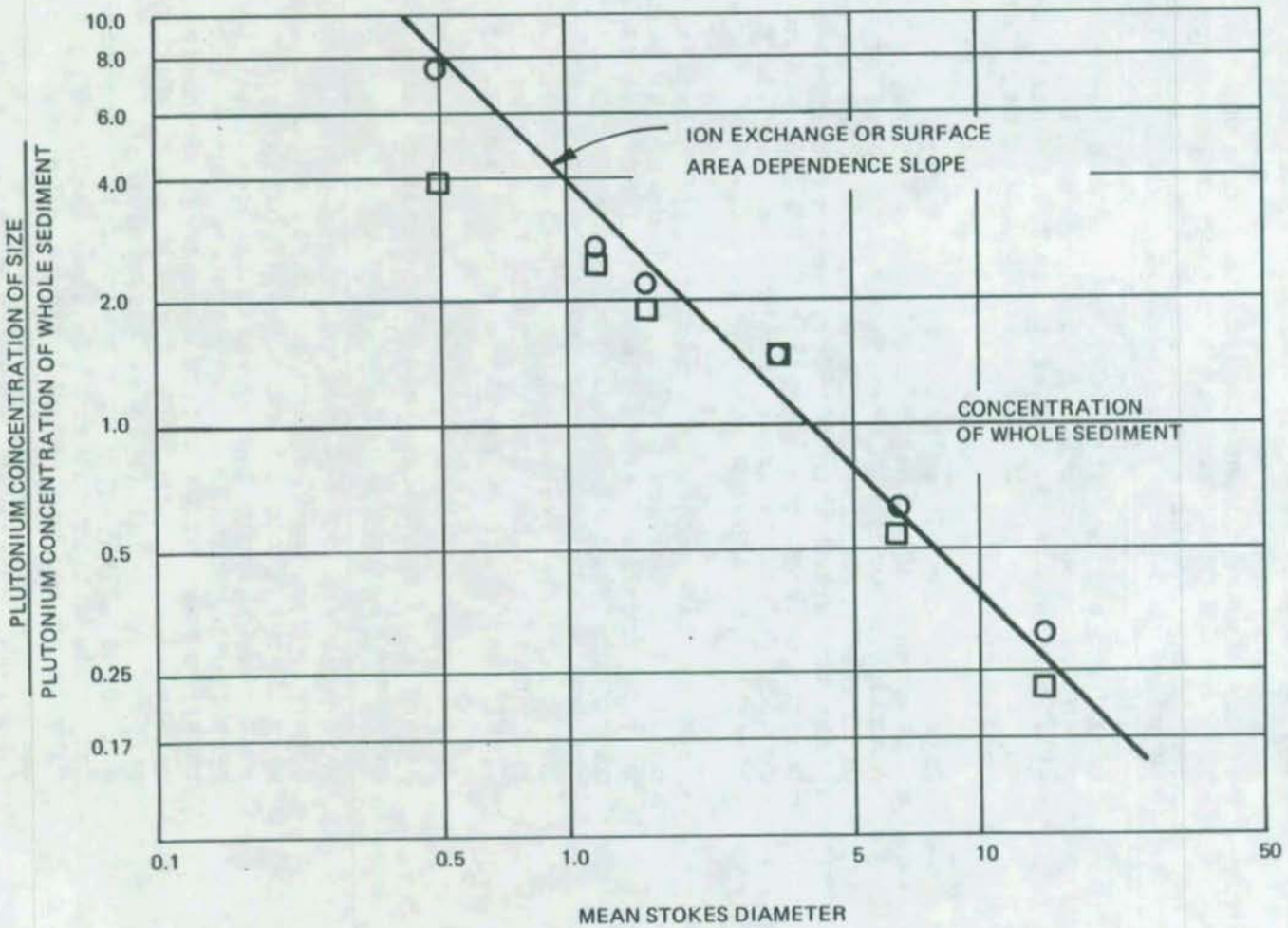


FIGURE 51 - Variation of ^{238}Pu concentration in sediment as a function of fully dispersed particle size.

evenly distributed throughout the 0.01 mi^2 area. The localized nature of these maximum values, however, is clearly less hazardous than the case Healy describes.

Because events which resulted in these contaminated sediment deposits occurred in 1969, the major part of this contamination has been buried beneath the surface by continued sedimentation of uncontaminated sediment. This buried plutonium presents a less hazardous condition than the surface deposited contamination assumed by Healy because the plutonium under the surface is not available to man. In the future, some of the more concentrated buried plutonium contaminated sediment could potentially be brought to the surface and, therefore, will be considered. However, it is expected that such occurrences would be localized and that dilution with less contaminated sediment would occur.

g. Extent of the contamination.- Healy assumes a very widespread contaminated area in his pathway analysis. Although he does not state the exact size, it has been estimated from his wind resuspension model that the area might be as large as 39 mi^2. He generally assumes the uniform surface contamination to be omnipresent in a densely populated area, even to the very doors of the homes and businesses.

At Mound Laboratory, the significant contamination is confined to the sediment in the beds of the waterways or to very localized areas along the immediate banks where minor but frequent flooding and sediment deposition has occurred. The total area (even the area under water) with contamination levels greater than 0.001 nCi/g including the runoff hollow, ditch, North Canal, two ponds, South Canal, and overflow creek is about 277,000 ft^2 or 0.01 mi^2.

This 0.01 mi^2 area will most certainly present a lesser hazard than the widespread contamination area Healy assumes.

This area is not expected to expand significantly in the future. The overwhelming source of water to these waterways is the Mound Laboratory site drainfield. This water must pass through the culverts under the railroad before reaching the canal waterway system. These culverts tend to set a maximum waterflow rate to those contaminated waterways. In June 1974, the area observed a very intense 6-in. rainfall in 24 hr. During this storm (which has about a 100 yr return frequency), the Mound Laboratory side of the culvert was completely submerged and the culvert system was transmitting the water at about the maximum rate. The system did not flood beyond the localized areas described as flooding areas.

3. Major Assumptions Regarding Future Conditions

The major assumptions about future conditions are guided by the philosophy of selecting *most credible worst-case conditions* with regard to health and safety for each pathway considered. This philosophy was applied in order to avoid underestimating the potential hazard to the public. It is believed that these assumptions are truly worst case and add substantial safety to the analyses.

- It is assumed that the plutonium contaminated sediment will remain in the waterways for at least 70 years.
- The maximum values of the ^{238}Pu concentration at the "very surface," "first foot," and "any depth" have been assumed to be evenly distributed over the entire 0.01 mi^2 area of the waterways.
- The public is assumed to consume 10% of their intake of sediment, food, and water from the contaminated area.
- As much as 10% of the mud or dust which the public normally has on their skin,in wounds, on their clothing, and in their homes continuously is assumed to come from the contaminated waterways.
- The entire waterway system is assumed to be *permanently dry*.
- The public will visit the contaminated area daily (8 to 24 hr) for the entire 70 yr of life.

4. Sediment Concentration Decision Guides Estimated by Pathway Analyses

Using the information specific to this situation, the assumption regarding credible worst-case future conditions and "best value" pathway parameters from the scientific literature, plutonium concentration decision guides for the sediment were calculated using pathway analytical techniques.

These decision guides are defined as the critical plutonium concentration in the sediment of the waterways which could, under credible worst-case conditions, result in a plutonium uptake in man approaching maximum permissible dose levels with a continuous lifetime exposure of 70 yr.

The decision guides include an additional safety factor of 10 which was applied to account for uncertainties in the pathway parameters and changes that are anticipated in the plutonium dose standards. The decision guides were then compared to the plutonium concentration found in the sediment of the waterways in order to determine if a significant hazard will exist even under these maximum credible worst-case conditions.

a. Ingestion pathways.- Materials ingested or swallowed by man pass through the gastrointestinal tract (GIT) in about 24 hr. The GIT has been found to absorb only a small fraction of soluble plutonium forms ingested. The ICRP reports[20,23] that only 3×10^{-3} % of the soluble plutonium passing through the GIT is absorbed by the body, the remainder passing in the stool, leaving no reservoir for future absorption. Tests have also shown that insoluble forms of plutonium are absorbed to even a lesser degree by the GIT.[32] The presence of complexing agents, such as citrate ions, have been shown to increase absorption probably by increasing the solubility of the plutonium in the GIT.[25]

Before plutonium can be absorbed into man, however, it must first transmigrate from the sediment deposits to the GIT. The pathways describing this transmigration, considering soil or sediment directly, water, vegetables, meat from both wild and domestic animals and fish, are summarized in Figure 52.

(1) Accidental or deliberate ingestion of sediment or soil: Sediment and soil are not normally a part of the human diet and usually are ingested only accidentally. The pathways considered for this accidental ingestion are shown in Figure 53. The sediment can be transferred to hands and then to the mouth.

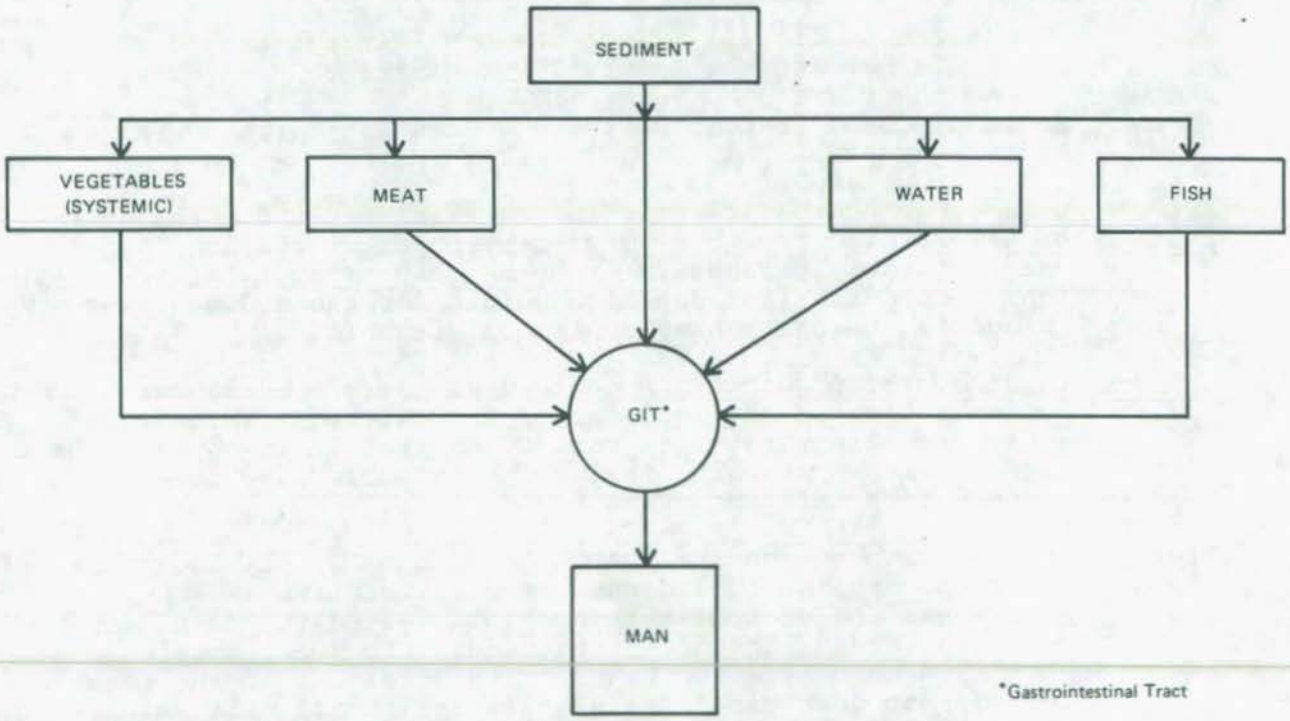


FIGURE 52 - Ingestion pathways.

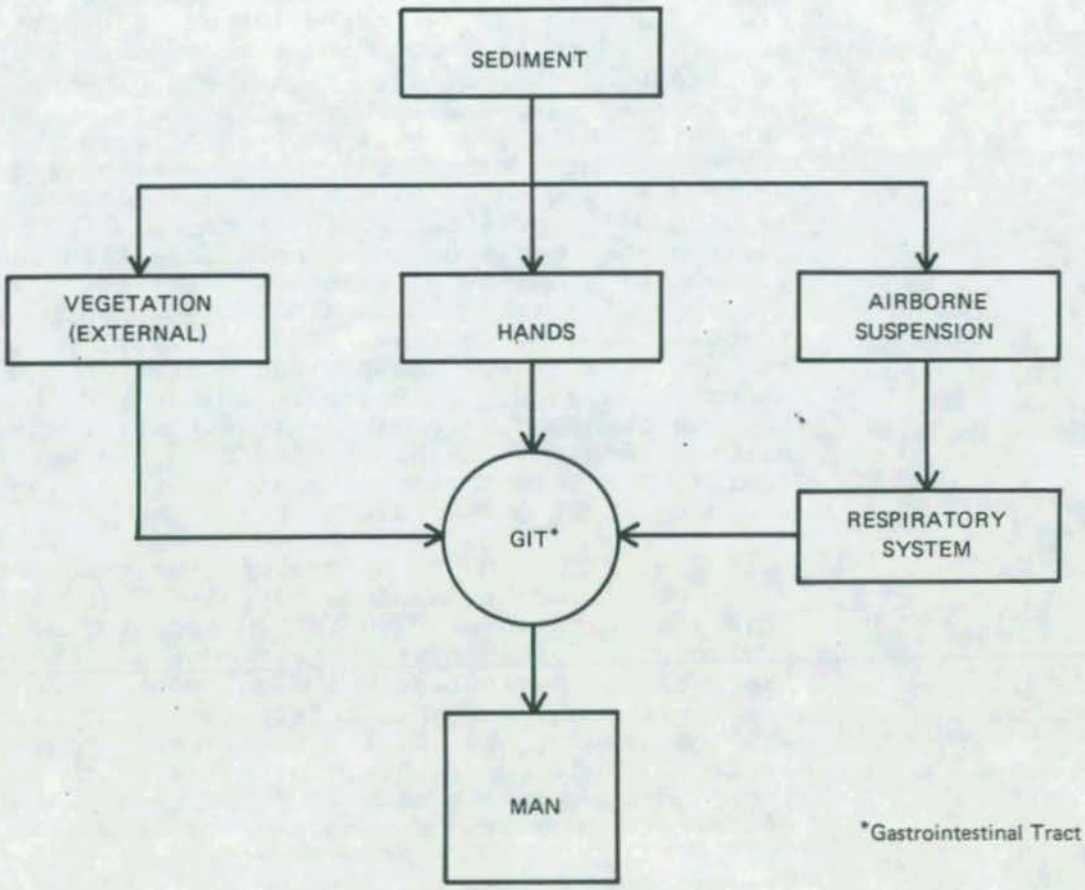


FIGURE 53 - Pathways for ingestion of sediment.

Sediment can be transferred mostly by water action to the surface of food plants which are ingested. Part of the sediment which is suspended in air and taken into the respiratory system is transferred into the GIT.

The amount of soil or sediment that might be transferred from dirty hands into the mouth has not been experimentally determined and probably occurs only sporadically. A worst-case upper limit of this value might be developed by the following logic. The surface area of the hands, fingers and about one-third of the forearm is assumed to be of the order of about 1000 cm^2. Healy[31] assumes that a person might have as much as 0.1 mg/cm^2 of dust on the skin constantly all his life. This would mean that an individual might have as much as 0.1 g on his hands. Assuming that this much dirt is constantly present and as a worst case, 10% of this dirt was transferred to the mouth and swallowed per day, up to 3 to 4 g of total dirt might be accidentally ingested per year. This value compares favorably with the 2 g/yr soil accidentally ingested by a person living continuously in a desert environment which was assumed by Martin.[33] Healy assumes as much as 36.5 g/yr for a desert environment which seems quite high. For accidental soil ingestion in the case under study here, in a nondesert environment 3 to 4 g of total soil intake per year seem quite adequate as a worst case. Additionally one need not assume that all the dirt deposited on the hands and arms would originate in the beds of the contaminated waterways. If 10% of all the dirt assumed to be on the hands come from the waterway sediment, the contaminated sediment ingestion rate would be about 0.3 to 0.4 g/yr.

Healy estimates as an upper limit that young children might be subject to a higher uptake potential from this source because of their tendency to get dirtier, and to put their hands in their mouths. He believed the rate may be an order of magnitude higher at least for 1 yr. Applying this safety factor of 10, it will be assumed that as a worst case, 3 to 4 g of sediment might be transferred to the mouth and ingested per year to account for more or less deliberate ingestion during 1 yr of life.

The fraction of the sediment which is transferred from the respiratory system to the GIT is assumed by the ICRP[20,23] to be 0.625 of the total amount inhaled. The amount of dust in the air in this area ranges between 40 to 100 $\mu g/m^3$ of air. It will be shown later that only about 1% of this dust can be assumed to come from the contaminated area.

A person breathes air at the rate of about 20 m^3/day and thus would inhale from 0.2 to 0.5 g of dust

per year, but only 0.002 to 0.005 g would be from the contaminated area. This is insignificant relative to the transfer from hand to mouth.

The amount of sediment deposited on vegetation might be as much as 10 $\mu g/cm^2$. One would expect that these would be washed prior to ingestion and only 10% of this sediment would be actually ingested. It has been estimated the specific surface area of leafy food plants is 6.45 cm^2/g.[34] The average amount of vegetables consumed by people in North America is 73,000 g/yr,[34] and as before, one need only assume 10% (as an upper limit) or 7300 g would come from the contaminated area. Combining these values would result in the ingestion of about 0.05 g of sediment from this source which is well within the uncertainty of the amount transferred from hand to mouth.

The worst-case amount of soil ingested as a lifetime average is therefore assumed to be about 0.3 to 0.4 g. Children, during 1 yr, might ingest as much as 3 or 4 g deliberately.

The "sediment concentration decision guide" was calculated for ingestion of sediment as shown in Table 27 and found to be about 52 nCi/g for deliberate ingestion for one year and 520 nCi/g for accidental ingestion.

Table 27

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON INGESTION OF SEDIMENT/SOIL

Decision Guide Equation for Ingestion of Sediment:

$$DG_{(sediment)} = \left(\frac{B/T_e}{G_s F_{m/gi}}\right)\left(\frac{1}{S}\right) = 52 \text{ nCi/g (deliberate/one year)}$$

$$= 520 \text{ nCi/g (accidental)}$$

Pathway Parameters:

B	= permissible body burden	= 4 nCi
T_e	= years of exposure	= 70 years
G_s	= amount of sediment ingested	= 3 to 4 g/yr (deliberate) 0.3 to 0.4 g/yr (accidental)
$F_{m/gi}$	= fraction absorbed from GI tract	= 3×10^{-5} g/yr
S	= overall safety factor	10

(2) Ingestion of natural water in equilibrium with sediment: It is not expected that anyone would deliberately consume water from this source since the water is quite dirty. However, a small amount might be swallowed if swimming is permitted or someone falls in the waterways. The pathway considered for ingestion of water is shown in Figure 54.

Previously, the distribution ratio ($D_{w/s}$) of plutonium between water and sediment has been discussed. The concentration of plutonium in water is only 10^{-5} fraction of the concentration in sediment under "equilibrium" condition.

Since the consumption rate of water (as water) is about 1 liter/day or 365,000 g/yr, an uptake of 36,500 g/yr (10% of total intake) is assumed as a worst case upper limit. Safe water ingestion from this area might limit the sediment concentration to about 522 nCi/g as shown in Table 28.

*Gastrointestinal Tract

FIGURE 54 - Pathway for ingestion of water.

Table 28

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON INGESTION OF WATER

Decision Guide Equation for Ingestion of Water:

$$DG_{(sediment)} = \left(\frac{B/T_e}{G_w D_{w/s} F_{m/gi}}\right)\left(\frac{1}{S}\right) = 522 \text{ nCi/g}$$

Pathway Parameters:

B	= Permissible body burden	= 4 nCi
T_e	= years of exposure	= 70 yr
G_w	= amount of water ingested	= 36,500 g/yr
$D_{w/s}$	= ratio water/sediment concentration	= 10^{-5}
$F_{m/gi}$	= fraction Pu absorbed by GI tract	= 3×10^{-5}
S	= overall safety factor	= 10

Sediment, which might be swallowed with the small amount of water during swimming, can be assumed to be part of the sediment ingestion amount previously considered.

(3) Ingestion of vegetables grown in sediment: At the present time there is little, if any, edible vegetation growing in or near the contaminated waterways. In this pathway, shown in Figure 55, the plutonium in the sediment must first be taken up systemically into the plant and then consumed by man. Previously (sediment ingestion) the external deposit of sediment on the plant has been considered.

Plants show very little tendency to extract plutonium from soils. In 1970, Romney[35] grew clover in soils contaminated with plutonium solutions for several years. Each year he harvested the top parts of the clover and analyzed for plutonium. He found that the concentration in the clover tops tended to increase from year to year probably as the root structure developed and maintained more contact with the soil. The ratio ($F_{p/s}$) of plutonium concentration taken into the plant and the plutonium concentration in the soil seemed to follow the empirical equation:

$$F_{p/s} \simeq 1.91 \times 10^{-5} \exp\,[0.4971(t-1)].$$

After one year $F_{p/s}$ reached a value of about 2×10^{-5} and then after 5 yr increased to 1.40×10^{-4}.

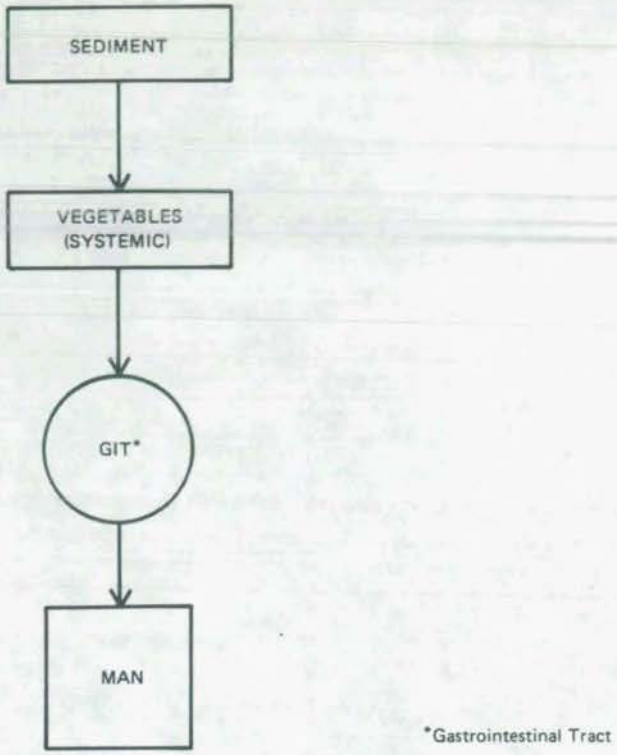


FIGURE 55 - Pathway for ingestion of vegetables.

Because almost all vegetable crops consumed by man are annual plants, it seems safe enough to use Romney's first year uptake value ($\sim 2 \times 10^{-5}$).

Wildung and Garland[36] recently have suggested that the plutonium uptake in root crops may be substantially higher than in leafy vegetation. On the basis of tests they performed on barley plants, the root uptake was found to be up to eight times higher than for the barley shoots. It is somewhat understandable that the small feeder roots of barley might exhibit a large uptake; however, the cellular structure and function of the massive roots that are consumed by man might be expected to take up much less plutonium than the tiny barley roots.

The potential for a larger plutonium uptake in man from the root crop portion of the diet is believed to be adequately offset by the "worst case" assumptions regarding the dietary intake from this area.

Coleman[34] estimated the average vegetable consumption in North America to be 73,000 g/yr. Most vegetables consumed on a yearly basis are grown on commercial farms and purchased fresh or canned. Since the contamination is confined to the waterways and the immediate banks, the vegetation to be considered in this case must be grown in these confined areas.

As a worst case, it is assumed that no more than ten percent of the total dietary vegetable intake would be grown in this contaminated sediment. The sediment concentration necessary to result in an uptake of 0.057 nCi/yr was estimated to be 1300 nCi/g as shown in Table 29.

Table 29

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON INGESTION OF VEGETABLES

Decision Guide Equation for Ingestion of Vegetables:

$$DG_{(sediment)} = \left(\frac{B/T_e}{G_s F_{v/s} F_{m/gi}}\right)\left(\frac{1}{S}\right) = 1300 \text{ nCi/g}$$

Pathway Parameters:

B	= permissible body burden	= 4 nCi
T_e	= years of exposure	= 70 yr
G_s	= amount of vegetables ingested (10% of total dietary intake)	= 7300 g/yr
$F_{v/s}$	= ratio plant/sediment concentration	= 2×10^{-5}
$F_{m/gi}$	= fraction Pu absorbed from GI tract	= 3×10^{-5}
S	= overall safety factor	= 10

(4) Ingestion of meat from animals grazing or living in contaminated waterways: Presently there is no known consumption of animals from this waterway area since it is undeveloped land inside the City of Miamisburg. However, as a worst case, the ingestion of the muscle portion of both small and large animals will be considered. The transmigration pathway of plutonium from the sediment through animals to man is shown in Figure 56.

Martin[33] has considered this pathway in a desert environment for ingestion of large animals (beef). From his pathway analysis, he estimates that the ratio of plutonium concentration in the muscle to the concentration in the soil to be about 2×10^{-5}. To account for smaller animals which may burrow in the banks of the waterways, this value was increased by a factor of 10 in this analysis

If ten percent of the 74,000 g of meat[34] or 7400 g were consumed from this area, the sediment concentration guide would be safely between 129 and 1290 nCi/g as summarized in Table 30.

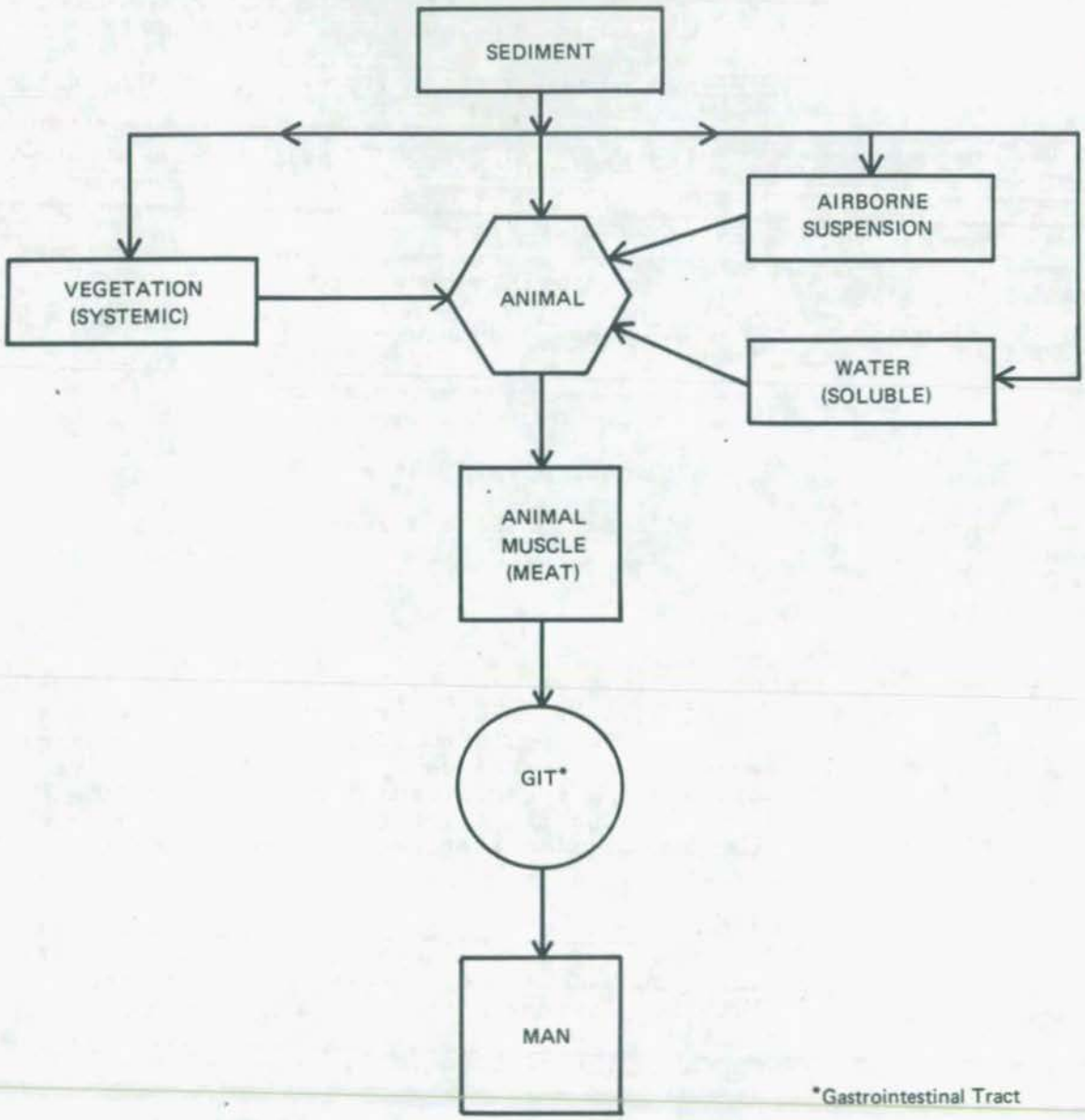


FIGURE 56 - Pathway for ingestion of meat.

Table 30

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON INGESTION OF MEAT

Decision Guide Equation for Ingestion of Animals:

$$DG_{(sediment)} = \left(\frac{B/T_e}{G_a F_{a/s} F_{m/gi}}\right)\left(\frac{1}{S}\right) = 129 - 1290 \text{ nCi/g}$$

Pathway Parameters:

B	= permissible body burden	= 4 nCi
T_e	= years of exposure	= 70 yr
G_a	= amount of muscle ingested	= 7400 g/yr
$F_{a/s}$	= ratio muscle to sediment	= 2×10^{-5} (large animals) 2×10^{-4} (small animals)
$F_{m/gi}$	= fraction Pu absorbed from GI tract	= 3×10^{-5}
S	= overall safety factor	= 10

(5) Ingestion of fish from the contaminated waterways: The fish in the waterways are mostly bluegill and carp. The public occasionally fish in the canals and ponds and eat the bluegills. The carp are not usually consumed, but are edible.

The pathway from the sediment to man through ingestion of fish is shown in Figure 57. The pathway from sediment to fish might include ingestion of water, biota, and sediment or absorption of plutonium from the water. The ^{238}Pu concentration in each of the components is assumed to be in equilibrium with the other components and all are related to the water concentration.

The concentration of plutonium in the water is controlled by the sediment/water equilibrium which has a water to sediment distribution ratio of 10^{-5} as discussed previously. Noskin[37] has reported that the edible portion of the fish might have a ^{238}Pu concentration four or five times higher than the water concentration. The highest concentration factor of plutonium-238 in the fish (carp) taken from the waterways indicates this fish to water ratio might be considerably less (0.5 to 1.0) than Noskin's value; however, the age of these fish is not known and Noskin's value will be assumed.

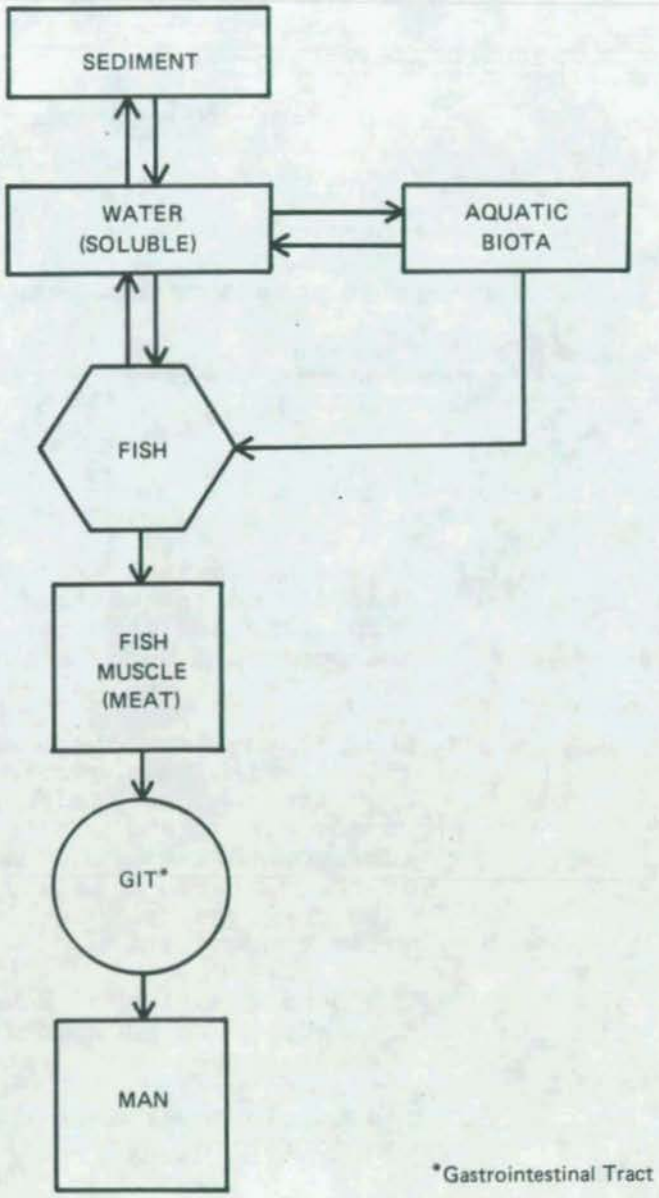


FIGURE 57 - Pathway for ingestion of fish.

Coleman[34] reports the average fish consumption by man is about 9000 g/yr. Since the fish population in these waterways (except the river) is quite limited, it is assumed that 10% of the total fish consumed (or 900 g/yr) were taken from the contaminated waterways.

Using the parameters and assumptions described above, the sediment concentration decision guide for the fish ingestion pathways was estimated to be 4230 nCi/g as summarized in Table 31.

Table 31

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON INGESTION OF FISH

Decision Guide Equation for Ingestion of Fish:

$$DG_{(sediment)} = \left(\frac{B/T_e}{G_f D_{w/s} F_{f/w} F_{m/gi}}\right) \cdot \left(\frac{1}{S}\right) = 4230 \text{ nCi/g}$$

Pathway Parameters:

B	= permissible body burden	= 4 nCi
T_e	= years of exposure	= 70 years
G_f	= amount of fish ingested	= 900 g/yr
$D_{w/s}$	= ratio of water to sediment concentration	= 1×10^{-5}
$F_{f/w}$	= ratio of fish to water concentration	= 5
$F_{m/gi}$	= fraction plutonium-238 absorbed through GI tract	= 3×10^{-5}
S	= overall safety factor	= 10

b. Absorption pathways. - Contaminated sediment deposited on the skin and in wounds might be absorbed into the body. The intact skin provides an excellent barrier against this pathway to man. The entry of solid or insoluble material by this mechanism is negligible and even the rate of absorption of soluble plutonium is very small. Langham[38] measured the rate of absorption of a 0.4 M HNO_3 solution of plutonium through the skin of a human subject and found the rate to be less than 1×10^{-2} % per day. (This acidity might be high enough to cause some skin change and increase absorption.) After reviewing the animal and human absorption studies performed with acidic solution of plutonium, Healy[25,39] recommends an upper limit for absorption rate to be about 1.4×10^{-2} % per day for soluble plutonium.

Data on the rate of absorption through open wounds indicate that this rate may be 10 to 100 times faster than through intact skin.[25]

In order for plutonium to transmigrate from the contaminated sediment to man, it must be deposited on the skin or in wounds, become solubilized, and then absorbed into the body. The pathways for these mechanisms are shown in Figure 58.

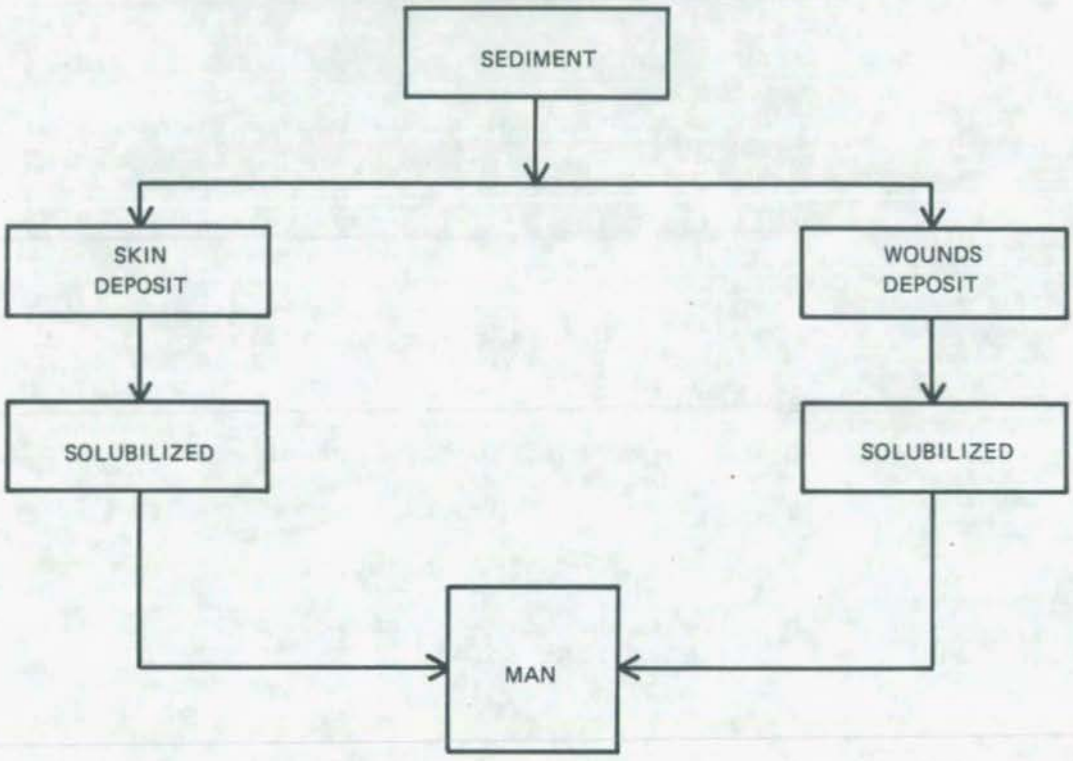


FIGURE 58 - Absorption pathways through skin and wounds.

(1) Absorption through intact skin: The amount of total dirt which may be deposited on the skin was estimated by Healy[25] to be about 1.0 g/m^2 of skin area. Since the skin area of a "standard man" is estimated at 1.85 m^2,[39] this amounts to about 1.85 g of dirt deposited continuously on the skin 24 hr/day for a lifetime.

If a person is clothed, and this part (90%) of the skin is protected,[39] the amount of dirt on the exposed areas might be as high as 10 g/m^2. Tests performed with the silty-clay sediments indicate the appearance of this amount of deposit is very muddy and might be unreasonable for a continuous lifetime deposit especially with periodic bathing.

But, even if this amount of dirt were deposited, one would not expect all of it to come from the contaminated waterways. For these reasons, a deposition of 0.1 g/m^2 on the 1.85 m^2 individual is believed to safely represent a worst-case upper limit for skin deposits of sediment from the contaminated waterways. The solubility of the plutonium in the sediment deposited on the skin is assumed to be of the order of 10^{-2} % if there is about 2 ml (0.001 ml/cm^2 x 1850 cm^2 of skin area exposed) of perspiration continuously in the skin areas where the sediment is deposited.

The sediment concentration guide considering absorption through intact skin was estimated to be greater than 6000 nCi/g as shown in Table 32.

Table 32

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON ABSORPTION THROUGH SKIN

Decision Guide Equation for Absorption Through Skin:

$$DG_{(sediment)} = \left(\frac{B/T_e}{(G/a)_s A_m R_{abs} d/y S_{w/s}}\right)\left(\frac{1}{S}\right) \simeq 6000 \text{ nCi/g}$$

Pathway Parameters:

B	= permissible body burden	= 4 nCi
T_e	= years of exposure	= 70 yr
$(G/a)_w$	= Amount of sediment deposited per unit area (10% of total dirt)	= 0.1 g/m^2
A_m	= skin area, standard man	= 1.85 m^2
R_{abs}	= absorption rate through skin	= 1.4×10^{-4}/day
d/y	= days per year of absorption	= 365 days
$S_{w/s}$	= fraction of plutonium soluble	= 1×10^{-4}
S	= overall safety factor	= 10

(2) Absorption through wounds: If no more than 1% (0.185 m^2) of the skin area is continuously abraided, the uptake by absorption through wounds would lead to about the same decision guide as for intact skin as shown in Table 33.

Table 33

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON ABSORPTION THROUGH WOUNDS

Decision Guide Equation for Absorption Through Wounds:

$$DG_{(sediment)} = \left(\frac{B/T_e}{(G/a)_s A_w R_{abs} d/y S_{w/s}}\right)\left(\frac{1}{S}\right) \simeq 6000 \text{ nCi/g}$$

Pathway Parameters:

B	= permissible body burden	= 4 nCi
T_e	= years of exposure	= 70 yr
$(G/a)_s$	= amount of sediment/unit area of wounds (10% of total dirt)	= 0.1 g/m
A_w	= area continuously abraided (1% of total skin area).	= $1.85 \times 10^{-2} m^2$
R_{abs}	= absorption rate through wounds	= 1.4×10^{-2} /day
d/y	= days/year of absorption	= 365 days
$S_{w/s}$	= fraction of plutonium soluble	= 1×10^{-4}
S	= overall safety factor	= 10

c. Inhalation pathways

(1) Introduction: Before the contaminated sediment can enter the inhalation pathway to man it must first be suspended in the air and exhibit a particle size range which can be breathed in and deposited in the lungs. These pathways are shown in Figure 59.

At the present time, the sediment is showing very little tendency to become airborne because it is underwater, moist, or covered with vegetation. These conditions tend to suppress or eliminate the air suspension potential of the sediment. If, however, in the future, the surface hydrology is altered to reduce or eliminate the water flow in these waterways, this would potentially affect the suspension characteristics of the sediment.

Even without the extensive surface waterflow now observed, these waterways cannot be assumed to take on desert-like conditions. More likely, it can be safely assumed that the beds of the waterways will revert to the conditions in the area nearby which are not subject to extreme flooding. Therefore, if

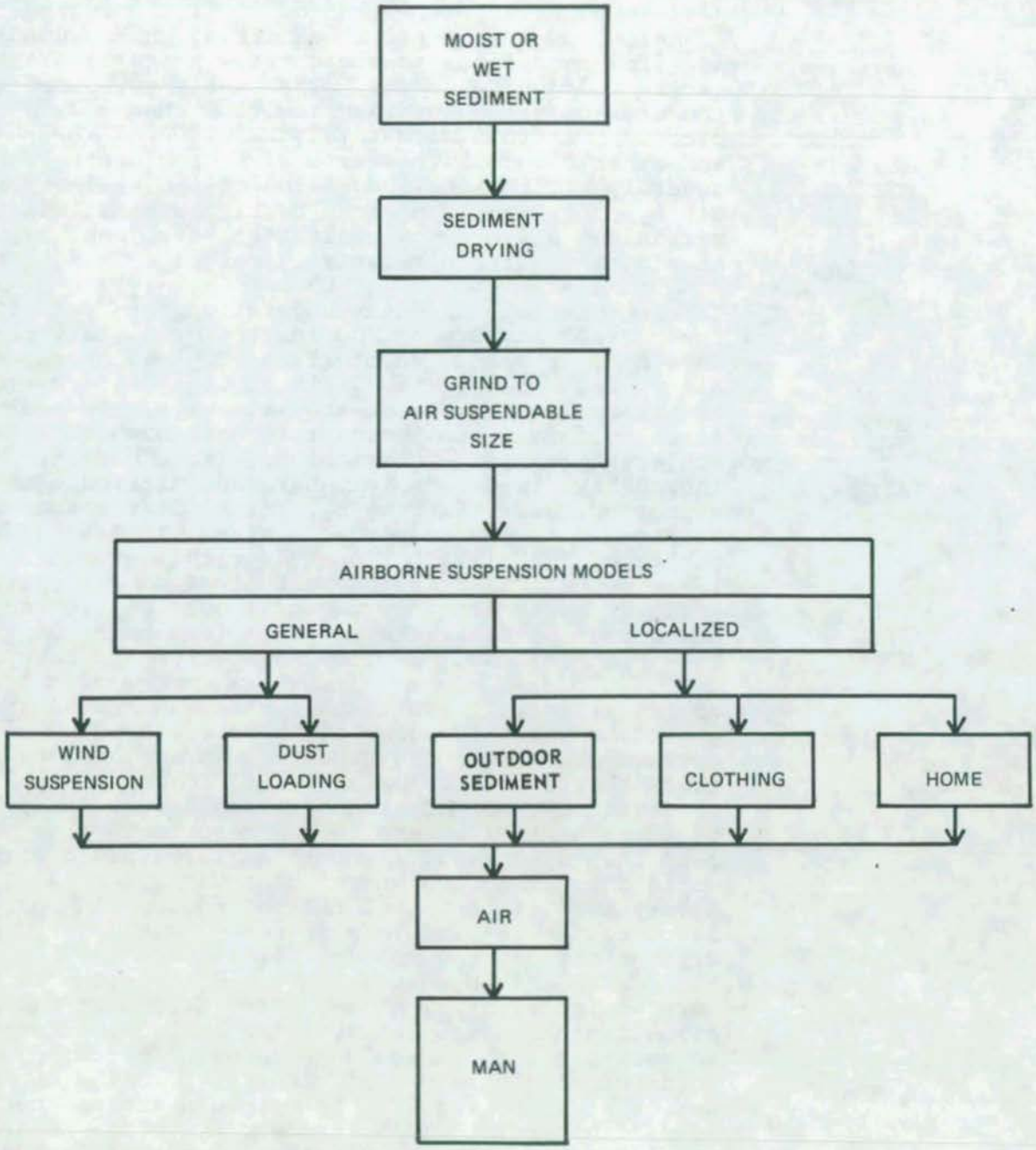


FIGURE 59 - Pathways for inhalation.

the waterflow was somewhat permanently decreased or eliminated, the beds of the waterways would be expected to become overgrown with thick vegetation in 1 or 2 yr. That this assumption is valid is indicated by the conditions prevalent in the South Canal. This portion of the waterway is subject to considerable variation of flow, ranging from a small flow in a central meandering channel under low flow conditions to a high flow that fills the entire bed during storms. The canal bed, other than the area of the meandering low flow channel, is completely overgrown with thick grass, weeds, brushes, and other plants. This extensive ground cover severely reduces the suspension of the sediment in air by providing a physical barrier against wind, mechanical disturbance, saltation, etc., and by keeping the surface relatively moist.

In the interim, while the vegetation gets established, the surface may dry out during one or two dry periods in summer or fall. When extreme drying occurs, the silty clay sediment drys, almost like pottery clay, to a very hard cake. In the waterways, the sediment tends to form large agglomerate masses which are not subject to airborne suspension. These masses, thoroughly dried, are very hard and difficult to grind, but under extreme mechanical disturbance might be reduced to air suspendable size. To determine the particle size distribution that might result from extreme mechanical grinding, a typical sample of the sediment taken from the top of a core sample was air and vacuum dried at temperatures less than 40°C (104°F). This action produced a very hard cake, about 2 in. in diameter and 1 in. thick, and almost no fine material. Next, this cake was broken and ground to pass through a 50 μm screen simulating an extreme mechanical disturbance. An agglomerate particle size distribution analysis (Figure 51) performed by microscopy methods indicated that only about 2.1% of these particles were in the respirable size range, less the ~6 μm (equivalent to an aerodynamic diameter of 10 μm). The mass median diameter was about 35 μm. A very small fraction of the sediment would be reduced to the respirable size range even under dry conditions and extreme mechanical disturbance. In addition, when this powder is rewetted, it seems to return to its original state. After the next rainfall, all sediment in the waterways which had been subjected to mechanical abuse would be reconsolidated. Therefore, continuous dry conditions or repeated mechanical action would be required to maintain the sediment in suspendible form. This is contrary to the nature of local weather conditions. In all probability, if these waterways were no longer needed to carry surface water and were allowed to dry, the city would fill the waterway beds to provide more level land. This action would bury the plutonium sediment deposits and virtually eliminate the air suspension potential.

Very few of these moderating conditions and circumstances were used in the pathway analysis performed because they are very difficult to define quantitatively.

Several methods have been used to estimate air suspension characteristics. General or widespread airborne contamination behavior is examined using dust loading and wind plume suspension models. Localized air concentrations near the waterways or resulting from sediment contamination of personal clothing and homes are estimated from airborne resuspension factors.

Using these models to define the pathway to man, the sediment concentration which would potentially result in air concentration approaching the levels permitted by the RCG for air were calculated.

The ICRP recommends two RCG values reflecting the solubility of the plutonium inhaled. The "insoluble" air RCG (1×10^{-12} $\mu Ci/cm^3$) was established to prevent the accumulation of a permissible lung burden (1.5 nCi) with a lifetime continuous exposure. The "soluble" RCG (7×10^{-14} $\mu Ci/cm^3$) was established to prevent the accumulation of a permissible body burden with a continuous lifetime exposure.

The "insoluble" RCG is usually applied to PuO_2 forms which are relatively insoluble and tend to remain in the lungs for an extended period of time. The "soluble" RCG relates to inhalation of very soluble forms like acidic $Pu(NO_3)_4$ solutions.

While the plutonium sorbed on the sediment is probably more soluble in lung fluids than the refractory forms of PuO_2, it is almost insoluble when compared to acidic plutonium nitrate solutions. Therefore, it was assumed, due to the relative insolubility of the plutonium/sediment in reasonably harsh conditions and the worst-case nature of the suspension mechanisms, that the insoluble RCG for air (1×10^{-12} $\mu Ci/cm^3$) is severe enough to adequately define the hazardous levels of plutonium in sediment.

(2) Inhalation estimated by a dust-loading model: Under normal circumstances, there is a sizable concentration of dust in the air. The source of these airborne particulates varies from location to location, but usually results from soil suspension in primarily agricultural areas, from automobiles and industry in urban areas. In any case, the airborne dust loading at any location originates from very wide areas.[25] Localized air concentrations can be enriched temporarily from a single ground source or small area where mechanical disturbance is occurring, but this affect is transient and averaged out over a period of time will be insignificant.

The dust loading in this area varies from about 40 $\mu g/m^3$ to 100 $\mu g/m^3$. How much of this dust is suspended soil is not known, but the fossil fuel electrical generation station to the southwest is known to greatly affect the dust loading.

If it is assumed, as a worst case, that the dust loading values reflect only airborne *soil* suspension and that the area supplying dust (a reference location) is much greater than 1 mi^2, the sediment concentration in the air near the waterways (area = 0.01 mi^2) that are assumed to be dry would contribute less than 1% of the total dust.

Since the dust load model implies a general average air concentration, it has been assumed as a worst case that the public in this area would be exposed to this air, 24 hr/day for 70 yr.

The sediment concentration which could potentially result in exposure of the public to air concentrations approaching the RCG value for air has been estimated as shown in Table 34. This pathway model results in a sediment concentration decision guide of about 100 to 250 $\mu Ci/g$.

Table 34

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON INHALATION: DUST LOADING MODEL

Decision Guide Equation for Inhalation (Dust Loading)

$$DG_{(sediment)} = \left(\frac{RCG(air)}{C_p A_s / A_p}\right) \cdot \left(\frac{1}{S}\right) = 100\text{-}250 \text{ nCi/g}$$

Pathway Parameters:

RCG(air)	= permissible air concentration (plutonium-238)	= 1×10^{-9} nCi/cm^3
C_p	= total dust loading this area	= 40×10^{-12} to 100×10^{-12} g/cm^3
A_s	= area of contaminated sediment	= 0.01 mi^2
A_p	= total area supplying particulate matter to this area	= >1 mi^2
S	= overall safety factor	= 10

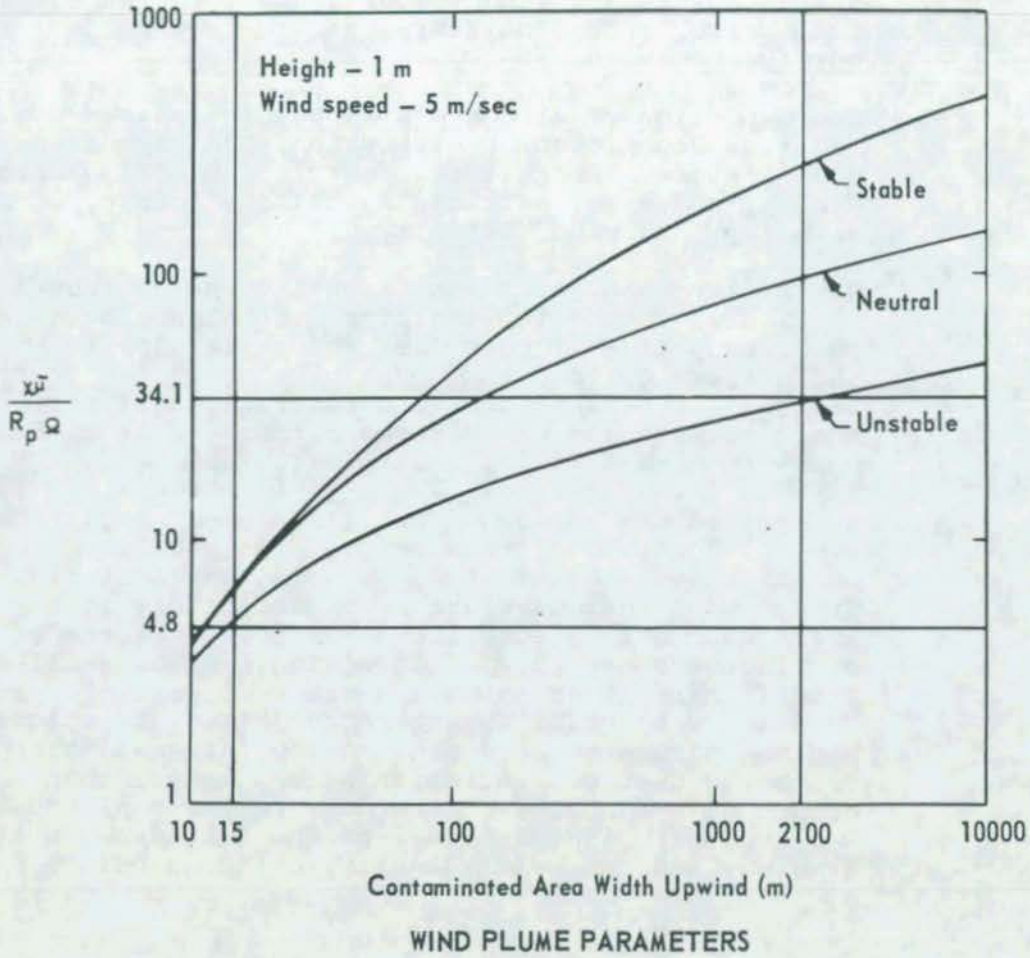


FIGURE 60 - Calculated concentration integrals (plume parameters).

(3) Inhalation estimated by a wind suspension model: When the wind blows over an area, it tends to pick up loose soil particles from the surface and form a dust cloud or plume. The airborne particles rise and are subsequently dispersed in the air or deposited downwind.

Many factors determine the amount (and rate) of soil particles which will be picked up by the wind. Among these factors are wind velocity and turbulence, particle size, ground moisture, ground cover and surface roughness.

The depletion of airborne soil concentrations resulting from deposition or dispersion also depends on many factors. Among these factors are soil particle density and size, particle settling velocity, wind velocity, and wind stability.

The maximum concentration of dust downwind from a particular source is, therefore, a balance between the pickup of soil particles and their deposition and dispersion downwind. Healy[25] considered these factors and presented a wind generated dust plume model to estimate the maximum airborne contamination downwind of a strip of contaminated soil.

A detailed discussion of Healy's plume calculation is beyond the scope of this report. He performed a calculational parameter study assuming the contaminated strip is of various widths, but infinitely long. The infinite length assumption tends to reduce the effect of minor changes in wind direction. Using an 11 mph wind (5 m/sec), he assumed a range of particle properties and wind stability characteristics and calculated a plume parameter or concentration integral which defines the dust concentration at one meter above the ground as a function of the dust particle pickup rate, R_p, the contamination level on the ground per unit area, Ω, and the wind velocity, as shown below:

$$I = \frac{x\bar{u}}{R_p \Omega}$$

These concentration integral values are presented as a function of contaminated strip width and wind stability in Figure 60.

As was discussed previously, the wind at Mound Laboratory blows mostly from the south and west. These extremes correspond to a wind direction essentially perpendicular or across the waterways and parallel along the length of the waterways. The dimensions of the contaminated waterway beds presented to a west or southwest wind is assumed to be an average of about 49 ft wide (15 m) and infinitely long. In this case,

the assumption of infinite length is probably justified since the waterways are very long (6890 ft) relative to the width.

For a south or southwest wind, a contaminated strip is assumed to have a width of 6890 ft (2100 m) and an infinite length. This assumption is clearly worst case since this would define a contaminated area very much larger than is observed. However, using these width assumptions the extreme concentration integrals were taken from Figure 62 to be 4.8 for the 15 m strip (west wind) and 34.1 for the 2100 m strip (south wind). Healy's assumption of an unstable 11 mph wind is not substantially different than is observed in this area and was adapted in the pathway calculation.

Using ZnS particles to simulate fresh fallout, Healy[25] determined a pickup factor, R_p to be 2×10^{-8}. Therefore, this value applies to fresh fallout and is not applicable to this case where the plutonium is fixed to the soil. As has been stated previously, the suspension properties of the contaminated sediment is controlled by the soil properties and should be similar to fallout contamination on soil which has been well aged and permitted to become fixed onto the soil.

The wind pickup factor, R_p, has been estimated to be 2×10^{-12} sec/m^2 for the 20-year GMX area at NTS.[25] This value was selected to represent the wind pickup factor in this area.

Healy[23] assumes that only the top 0.1 cm (or the very surface) of the soil is vulnerable to wind suspension. Using a density of about 1.6 g/cm^3, then there are about 1600 g/m^2 of particles available for suspension.

As in the case of dust loading, the wind suspension of dust must be considered generally distributed and as a worst case the public might be exposed to this air 24 hr/day for 70 yr.

With these assumptions and parameters, the sediment concentration which potentially could result in the exposure of the public to air concentration approaching the air RCG is calculated as shown in Table 35. These sediment concentration decision guides based on wind suspension for the *dry* waterway were then estimated to be between 183 to 13,000 nCi/g.

Table 35

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON INHALATION: WIND SUSPENSION MODEL

Decision Guide Equation for Inhalation (Wind Suspension)

$$DG_{(sediment)} = \left(\frac{RCG(air)}{\frac{\chi \bar{u}}{R_p \Omega_w} \frac{R_p}{\bar{u}^2} \bar{u} (G/a)_w} \right) \cdot \left(\frac{1}{S} \right) = 183\text{-}1300 \text{ nCi/g}$$

Pathway Parameters:

RCG(air)	= permissible air concentration (plutonium-238)	= 1×10^{-3} nCi/m^3
$\frac{\chi \bar{u}}{R_p \Omega_w}$	= dust concentration integral (plume parameter)	= N/S 34.1 = E/W 4.8
$R_p/\bar{u}^2$	= wind pickup rate constant	= 2×10^{-12} sec/m^2
$\bar{u}$	= wind velocity	= 5 m/sec
$(G/a)_w$	= sediment available for suspension per unit area	= 1600 g/m^2
S	= overall safety factor	= 10

(4) Inhalation estimated by resuspension factors: A useful technique for estimating the air concentrations, which might be present in the immediate area of the contamination, is defined by the resuspension factors.

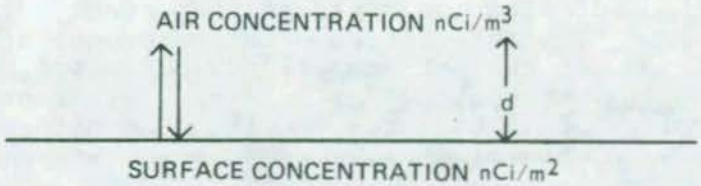
The resuspension factor, Km^{-1}, is defined as the ratio of the air concentration per unit volume to the ground concentration per unit area as shown in Figure 61. This parameter does not relate to any particular mechanism for air resuspension of ground contamination but is simply a functional measure of the air concentration which would be expected in the immediate area of the ground contamination under prevailing conditions.

Resuspension factors have been reported for a variety of conditions and circumstances and (10^{-2} to 10^{-11})[27,28,40] vary over about nine orders of magnitude. When these values are sorted according to the condition prevailing at the time:

- Nature of the contamination
- Amount of ventilation (indoors or outdoors)

- The nature of the surface (hardness, smoothness)
- The amount and nature of the mechanical disturbance

The range of values fall into a pattern as shown in Table 36.



$$K = \left(\frac{\text{AIRBORNE CONCENTRATION nCi/m}^3}{\text{SURFACE CONCENTRATION nCi/m}^2} \right) m^{-1}$$

FIGURE 61 - Resuspension factors.

Table 36

SUMMARY OF RESUSPENSION TESTS ON OUTDOOR SOIL

Nature of Contamination	Disturbance	Km^{-1}
Fresh Fallout	Low	10^{-7} to 10^{-6}
Fresh Fallout	Auto Traffic	10^{-6} to 10^{-5}
Aged Fallout (20 yr)	Low	10^{-9}
^{91}Y sorbed on soil	Low	10^{-9}
^{238}Pu sorbed on soil at Mound Laboratory	Low	10^{-9}
	High	10^{-8}

The range of resuspension factors measured for fresh PuO_2 fallout particulates deposited on outdoor soil during nuclear testing range from 10^{-7} to 10^{-6} under low mechanical disturbance conditions.[28] (In areas where the surface is rocky or paved, the resuspension factors may range up to 10^{-3} due to these smoother and harder surfaces and because little mixing with noncontaminated surfaces can occur.)

Mechanical disturbances (even automobile traffic) usually increase the resuspension factor by a factor of 10 to 100.[39] Steward[28] and others[26] recommend a resuspension factor of 10^{-6} m^{-1} for fresh fallout under quiescent conditions increasing this value to 10^{-5} m^{-1} for moderate activity. As the fresh fallout becomes aged, fixed to the soil or mixed with the soil, the resuspension characteristics of the contamination apparently approaches the soil suspension characteristics. The resuspension factors measured at the GMX area (20-yr-old deposit) is estimated at about 10^{-9} m^{-1}.[30,33] This represents a reduction of about three orders of magnitude from the time it was originally deposited.

Interestingly, the suspension of radioactivity, which is sorbed onto soil is apparently controlled by soil suspension characteristics without aging. A test performed with $^{91}YCl_3$ solution sorbed onto soil indicated a 10^{-9} m^{-1} resuspension factor.[28] Plutonium-238 sorbed onto Mound Laboratory soil also indicated a resuspension factor of about 10^{-9} m^{-1} as has been discussed previously.

On this basis, sediment would exhibit a resuspension factor of about 10^{-9} m^{-1} from the time of sorption.

Using Healy's "very surface" criteria,[25] the amount of sediment available for resuspension is assumed to be 1600 g/m^2.

In this case, since the resuspension factor defines the air concentration only in the immediate area of the waterway beds, it has been assumed that the public would be exposed no more than 8 hr a day, every day, for 70 yr.

The sediment concentration in the dry waterway beds which could potentially result in exposure of the public to air concentrations approaching the RCG for air was estimated, as shown in Table 37.

The sediment concentration decision guide based on resuspension factors was established at 188 nCi/g. During periods of high mechanical disturbance, the RCG might be exceeded temporarily; however, these short term exposures will not affect the overall evaluation significantly.

Table 37

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON INHALATION: RESUSPENSION FACTORS

Decision Guide Equation for Inhalation
(Resuspension Factors)

$$DG_{(sediment)} = \left(\frac{RCG\ (air)}{(Km^{-1})\ (G/a)_r F_t}\right) \cdot \left(\frac{1}{S}\right) = 188\ nCi/g$$

Pathway Parameters:

RCG(air)	= permissible air concentration (plutonium-238)	= $1 \times 10^{-3} nCi/m^3$
Km^{-1}	= resuspension factor	= $1 \times 10^{-9}/m$
$(G/a)_r$	= resuspendable sediment per unit area	= $1600\ g/m^2$
F_t	= fraction of time exposed/day (8 hours/day)	= <0.33
S	= overall safety factor	= 10

(5) Inhalation estimated from clothing contamination: Should a person contaminate his clothing with waterway sediment and perform a vigorous activity, one might expect an increased inhalation exposure. This is assumed to be due to the more active mechanical disturbance, the drier nature of the dust deposit, and the proximity of the clothing to the mouth and nose. As a measure of assurance that this mechanism is not underestimated, the normal sediment resuspension factor has been increased by three orders of magnitude to about $10^{-6}\ m^{-1}$.

Healy[25] assumes that 10 g/m^2 of dust might be present on clothing in a desert area. While it would certainly be less in this area and all of it need not be assumed to be contaminated sediment, a value of 1 g/m^2 of sediment dust on the clothing seems to be safe.

One can logically assume that the clothing, removed for sleeping, would not be subject to mechanical agitation. Therefore, it is assumed that a person would, in the worst case, only be exposed to this source 16 hr a day, every day, for 70 yr.

Using these parameters and assumptions, the calculation of sediment dust concentration which might result in an exposure of the public to air concentrations approaching the RCG for air is summarized in Table 38.

Table 38

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON INHALATION:
PERSONAL CLOTHING CONTAMINATION

Decision Guide Equation for Inhalation (Clothing)

$$DG_{(sediment)} = \left(\frac{RCG(air)}{(Km^{-1})_c (G/a)_c F_t}\right) \cdot \left(\frac{1}{S}\right) = 150 \text{ nCi/g}$$

Pathway Parameters:

RCG(air)	= permissible air concentration (plutonium-238)	= 1×10^{-3} nCi/m^3
Km^{-1}	= clothing resuspension factor	= 10^{-6}/m
$(G/a)_c$	= grams sediment per unit area of clothing	= 1 g/m^2
F_t	= fraction of time exposed/day (16 hr/day)	= 0.67
S	= overall safety factor	= 10

The sediment concentration decision guide is estimated at 150 nCi/g based on clothing contamination.

(6) Inhalation from home contamination: Sediment from the waterways might be tracked into nearby homes or carried in on clothing. The total amount of soil that might be considered to be continuously in the home is not known. However, one can safely assume, with normal routine cleaning, that the soil in the home will not exceed 10 g/m^2. This amounts to about three pounds of dirt in a modest 1500 ft^2 house.

For the purpose of this pathway one need not assume that all dirt in the house originated in the contaminated waterways. Therefore, as with personal clothing, the sediment deposition in the home will be assumed to be 1 g/m^2 of floor space.

Because some of the floors are harder and have a smoother surface than outside soil and because the ventilation may be low and cannot disperse the dust, the outside resuspension factor has been increased, by a factor of 1000, to 10^{-6} m^{-1} as a worst case. Even though this is a localized concentration, the daily exposure time has been assumed to be 24 hr a day, every day, for 70 yr to account for persons who spend a great deal of time working in the house.

The worst-case sediment concentration, to potentially result in RCG air concentrations, in nearby homes has been estimated as shown in Table 39. The sediment concentration decision guide based on this pathway is estimated to be of the order of 100 nCi/g.

Table 39

SEDIMENT CONCENTRATION DECISION GUIDE
BASED ON INHALATION: HOME CONTAMINATION

Decision Guide Equation for Inhalation (Home)

$$DG_{(sediment)} = \left(\frac{RCG(air)}{(Km^{-1})_H (G/a)_h F_t}\right) \cdot \left(\frac{1}{S}\right) = 100 \text{ nCi/g}$$

Pathway Parameters:

RCG(air)	= permissible air concentration (plutonium-238)	= 1×10^{-3} nCi/m^3
$(Km^{-1})_H$	= resuspension factor (indoors)	= 10^{-6}/m
$(G/a)_h$	= grams of sediment per unit area of home (10% of total dirt)	= 1 g/m^2
F_t	= fraction of time/day exposed 24 hr/day	= 1.0
S	= overall safety factor	= 10

F. Applied and Implied Safety Factors in Sediment Concentration Decision Guides

In the previous section, the sediment concentration decision guides were estimated for a number of worst-case pathways. In effect, these decision guides attempt to answer only the question: "What sediment concentration in or around these waterways should cause *the slightest concern* about allowing the plutonium sediment deposits to remain in these locations based only on health and safety considerations?"

In order to avoid *underestimating the potential hazard,* a number of directly applied and implied safety factors have been built into the analyses.

- The overall dose standards recommended for *continuous* lifetime exposure to occupational plutonium workers were reduced by a factor of 10 before they were applied to the general public in this analysis. The occupational internal dose standards also contain safety factors for they have protected workers

and scientists working with plutonium for many years. It is a high recommendation, that no one is known to have been killed or significantly harmed[41] by internal exposure to plutonium at much higher levels than the occupational standard. Few other safety standards have been this successful.

- An honest attempt was made in this analysis to develop as many pathways to man as possible, even to the point of forcing the issue on a few.

- An attempt was made to develop not only an overall worst case, but the maximum credible worst case for each pathway.

- Pathway parameters which have not been experimentally evaluated were estimated with great caution. An attempt was made to deliberately choose a value many times the value considered reasonable just to remain on the safe side.

- The best values for measured parameters reported in the scientific literature were used in the pathway analyses.

- A continuous exposure, 8 to 24 hr/day, every day, for a 70-yr lifetime was assumed in all pathways.

- An overall additional safety factor of 10 was applied to each pathway decision guide to account for anticipated future reduction of the plutonium dose standards and uncertainties in the pathway parameters.

Now as the decision guides are compared to the actual plutonium concentration found in the waterways, some additional safety factors are implied.

- These decision guides will be compared with the maximum values (rather than an average or mean value) found on the surface in or near these waterways as though they existed evenly throughout the entire 0.01 mi^2 area.

- Next, the decision guides will be compared with the *maximum* first-foot value and the maximum worst-case value found at any depth as though these concentrations existed at the surface and were evenly distributed over the 0.01 mi^2 area. The fact is that these maximum subsurface concentrations are found in a very small fraction of the total contamination area. tamination area.

Overall then the sediment concentration decision guides and their application to the evaluation of hazard to the general public in this particular situation is considered substantially on the safe side from a health and safety standpoint.

G. Overall Health and Safety Evaluation

The plutonium sediment concentration guides developed in the previous section are summarized in Table 40. These guides indicate the plutonium concentrations in sediment, which under credible worst-case conditions, could potentially result in a plutonium uptake approaching the dose standard in a lifetime (70 yr) of continuous exposure. Plutonium concentrations in sediment which are less than the concentration decision guides are considered to be safe.

In order to make the long-term health and safety evaluation, the *maximum* plutonium concentrations found in or near these waterways are compared with the decision guides. The maximum concentrations found are subdivided into the concentrations at the "very surface," in the first-foot, and at any depth as listed in Table 41.

The "very surface" concentration represents the plutonium contaminated sediment or soil that would be available to man. The maximum value found was 0.45 nCi/g, which is at least two orders of magnitude less than the worst decision guide pathway.

The first-foot concentrations represent the plutonium-contaminated sediment which, if it were brought to the surface by some action, could potentially be available to man. The only locations where exposing the first foot concentration would lead to increased surface concentrations of plutonium is in the actual beds of the North and South Canals. The highest first-foot concentration, 3.8 nCi/g, is found near the middle of the South Canal, which is, at least, an order of magnitude less than the worst-case pathway decision guide.

The high concentration found at any depth represents the worst-case potential available plutonium because the probability of being brought to the surface decreases with depth. The highest plutonium concentration (4.6 nCi/g) was found buried 3 to 4 ft in sediment in the North Canal. This too is substantially below the decision guides for all the worst-case pathways.

Table 40

SEDIMENT CONCENTRATION DECISION GUIDES
FOR WATERWAYS NEAR MOUND LABORATORY

Ingestion	Sediment Concentration (nCi/g)
Sediment	52-520
Water	520
Vegetables	>1000
Animals, large	>1000
Animals, small	129
Fish	>1000
Absorption	
Skin	>1000
Wounds	>1000
Inhalation	
Dust Loading	100-250
Winds Suspension	183-1300
Resuspension Factor	188
Clothing Contamination	100
Home Contamination	150

Table 41

MAXIMUM SEDIMENT/SOIL PLUTONIUM-238
CONCENTRATION FOUND IN WATERWAYS NEAR
MOUND LABORATORY

Very Surface (Available)	Maximum Concentration Found (nCi/g)
Sediment Surface in Waterways	0.450
Immediate Waterway Banks	0.060
Contiguous Land Areas	0.001
First Foot (Potentially Available in Future)	
Sediment in Waterways	3.8
Banks	0.010
Contiguous Land Areas	0.0004
Any Depth (Worst-Case Available in Future)	
Sediment in Waterway	4.6
Banks	0.060
Contiguous Land	0.001

Based on worst-case pathway analyses, the overall conclusions about the health and safety aspects of the plutonium deposited in waterways near Mound Laboratory are:

- The plutonium-238 does not and will not, in the future, present a hazard to people living in this area or the public at large.
- There is no apparent reason to restrict the use of the area in or near the waterways because of the plutonium deposits.

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