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PREVIOUS MANAGEMENT PRACTICES FOR NATURALLY OCCURRING RADIONUCLIDE WASTES: CURRENT RADIOLOGICAL STATUS

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Abstract

Many installations used during the early days of the United States' atomic energy program have been released in recent years for unrestricted private uses. These installations include lands and buildings used for the storage of radioactive wastes resulting from refining and processing of uranium and thorium. Waste management practices at these sites in the 1940's and 1950's were not conducted with today's emphasis on as-low-as-reasonably-achievable (ALARA) principles. Consequently, many of these older waste storage areas are contaminated with naturally occurring radionuclides in concentrations which are orders of magnitude greater than those found ordinarily in the earth's crust. Current and potential elevated human exposures at fifteen of these sites are due primarily to radon daughters and external-gamma radiation. A wide variety of exposure conditions may be found at these sites — ranging from slightly above background to more than thirty times the guidelines recommended for the public. Remedial actions are contemplated for a number of these sites where contamination levels or radiation exposures exceed current guidelines.

Introduction

Early in this nation's development of atomic energy, extensive efforts were made to utilize all available resources for a program to demonstrate controlled nuclear fission. Initially, this program was administered by the Department of the Army under the Manhattan Engineer District (MED). Although conducted in wartime secrecy, the MED program encompassed a wide variety of materials research and development activities as well as various commercial source material handling operations. Contracts for needed services were entered and terminated as required.

Administration of the MED program was taken over by the Atomic Energy Commission (AEC) after the conclusion of World War II. Although the initial MED/AEC contractor facilities processed uranium, increased interest in the thorium fuel cycle resulted in a corresponding increase in the number of facilities involved in thorium processing. Services provided by private commercial firms under MED/AEC contracts covered a

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wide variety of activities such as ore transport and storage; dissolution and leaching of ores; production of mill concentrate (yellow-cake); refining of mill concentrate; conversion of this refined product to other compounds and/or metal; smelting, rolling, extrusion, cutting, and packaging uranium and thorium metal products for distribution to other institutions such as national laboratories; and the recovery of uranium from scrap and salvaged material.

Disposal of radioactive residues frequently consisted of shallow-land burial on-site or at some federally owned or leased land in the vicinity of the site. At the termination of contract operations, efforts were made to decontaminate buildings, land, and equipment to levels consistent with guidelines which existed at that time. However, no consideration was given to cleanup in accordance with ALARA objectives.

In many cases, present records are insufficient to document the radiological condition of these sites, most of which are now in the public domain. The overall program to determine the current status of these sites has been described elsewhere (e.g., Ha78, DOE78). This paper will present results found at three sites involved with waste disposal during MED/AEC activities; these results represent the range of findings obtained during the current resurvey program.

Early Solid Waste Management

The handling and processing of ores and ore concentrates produced large volumes of low-level solid residues at MED/AEC contractor facilities. Furthermore, solid wastes were generated when equipment and building surfaces were decontaminated or discarded. These wastes, particularly the residues, contained most of the radioactive material present in the original ore or ore concentrate. Consequently, these wastes are the source of present day radiation exposures at MED/AEC sites.

The highest priority items at MED/AEC sites, particularly during World War II, were the development of processing technology and the production of source material. Typically, a portion of the contractor's land would be dedicated to surface storage or shallow burial of process residues. If this proved to be impractical, a contract for these purposes would be established with a nearby property owner. Residues were generally regarded as wastes when the source material content was no longer recoverable by the contractor's processes. Thus, qualititative and quantitative aspects of the radionuclide inventory of residues were quite dependent on the processing history of the material.

Previous waste management practices are intimately associated with the present radiological status of fifteen former MED/AEC sites surveyed to date by the Oak Ridge National Laboratory (ORNL). The contamination of these sites by radionuclides such as ^{230}Th, ^{232}Th, ^{227}Ac, ^{228}Ra, ^{226}Ra, and ^{210}Pb indicate that problems could be associated with inhalation and ingestion of these long-lived materials. On-site measurements indicate that external gamma radiation exposures and, where

structures are present, indoor radon daughter concentrations constitute the principal radiation exposure problems at these formerly utilized sites (e.g., Le78a).

Radiological survey results indicate that present exposures range from those which cannot be distinguished from background to more than thirty times the guideline values recommended for the general public (Cr78, Le78a). These results are due to a wide range both in exposure rates and in site occupancy. Radiological survey results from three sites will be used to demonstrate the relationship between previous solid waste management practices and present radiation exposures at formerly utilized sites.

Former Vitro Rare Metals Plant, Canonsburg, Pennsylvania

The 8-ha (18-acre) site at Canonsburg, Pennsylvania, was used for the commercial extraction of ^{226}Ra from 1911 to 1922. From 1930 to 1942, radium and uranium salts were extracted for commercial purposes. From 1942 to 1957, uranium was recovered from ores, concentrates, and scrap materials under MED/AEC contracts. The site remained under the control of various AEC licenses until 1966. Since 1967, the property has been developed by the present owner. Various light industries currently occupy the twelve buildings in what is now an industrial park. Approximately 125 persons are employed at this site. None of these employees is performing any work which is related to the nuclear industry.

The site is divided into three separate parcels, designated A, B, and C as shown in Fig. 1. Extraction of radium began on the western side of parcel A. Residues were dumped on parcels A and B. As the plant expanded toward the east of parcel A, new buildings were constructed over the residues. Liquid slurry wastes were impounded in a swampy area located in parcel C; this area was later filled with site residues and covered with uncontaminated dirt.

Results of the radiological survey (Le78a) indicate that a layer of contaminated soil can be found within 1 m of the surface at almost any point on the site. Apparently, all buildings on the site are built over or directly adjacent to contaminated soils. A "typical" boring on parcel A would indicate contamination to a depth of almost 3 m; the average ^{226}Ra content of the core would be about 200 pCi/g. In parcel B, about the same results could be expected except for an area in the center which has up to 2 m of fill material over the contamination. A 150-cm layer of highly contaminated muck (up to 17,000 pCi/g of ^{226}Ra) would be encountered within 1 m of the surface of parcel C. The ratio of ^{226}Ra activity to ^{238}U and ^{227}Ac activities varied widely from sample to sample because of the wide variations in processes and in feed material used to generate the residues.

A summary of radiation exposures being received by the industrial park employees is given in Table 1. These elevated exposures can be attributed directly to the contaminated residues which cover and underlie practically the entire site. Variations in external gamma radiation

levels can be correlated fairly well with variations in subsurface contamination (Le78a). Radon and radon daughter concentrations in buildings can be attributed to ^{226}Ra contamination in surface and subsurface soils, in floor and former process drains, and on interior surfaces of the buildings. Daughters of ^{219}Rn, attributable to ^{227}Ac contamination in surface soils, were also detected inside structures (Le78a).

The ranges of average exposures inside structures shown in Table 1 represent the lowest and highest average obtained in any building on site. Each of these individual building averages represents numerous individual measurements. However, the range for airborne ^{230}Th represents the range observed in individual spot samples; these should not be construed as average values.

A summary of current exposure guidelines for an individual in the general public is given in Table 2. These guidelines, except for radon daughters in commercial structures, assume that the exposure is continuous. Comparisons between Table 2 and Table 1 show that employees at the Canonsburg site are exposed to average radon and radon daughter concentrations which are far in excess of the guidelines. In fact, only a portion of one of the twelve buildings had average radon and radon daughter concentrations below the guidelines. Remedial measures are obviously required to reduce on-site exposures to radon and its daughters. Results of spot samples and the presence of alpha contamination on practically all building surfaces indicate that occupants of some of the buildings on this site may be exposed to average concentrations of long-lived airborne radionuclides (particularly ^{230}Th) which exceed guideline values.

Exposures at the Canonsburg site are far higher than those associated with the other MED/AEC sites surveyed by ORNL. The Canonsburg site is the only MED/AEC site which was specifically included in the "Uranium Mill Tailings Radiation Control Act of 1978" (H.R. 13650, 95th Congress). Under the provisions of this act, the Secretary of Energy is authorized to conduct remedial measures at this site.

Pennsylvania Railroad Landfill Site, Burrell Township, Pennsylvania

The Pennsylvania Railroad Landfill Site is located approximately one mile southeast of Blairsville, Pennsylvania, in Burrell Township. This property, owned by the Properties Division of the Penn Central Transportation Company, lies between the Conemaugh River and the mainline tracks of ConRail (see Fig. 2) and consists of approximately 25 ha (60 acres).

During the period of October 1956 through January 1957, an estimated 10,500 metric tons of radioactive material were shipped by rail from the uranium processing plant in Canonsburg, Pennsylvania, and was dumped on the site. The material contained approximately 5000 metric tons (dry weight) of waste residues containing an average 0.097% U_3O_8 by weight. The uranium (approximately 5 metric tons of U_3O_8) was classified as "unrecoverable material-measured." The wet weight of the residues was

estimated to be 9000 metric tons, and since the material was shipped wet, it appears that approximately 1500 metric tons of possibly nonradioactive materials were mixed with the radioactive materials during loading. The waste residues were generated under an AEC contract at the Canonsburg plant.

The area where the residues were dumped was previously the river bed of the Conemaugh River, which had been diverted approximately 150 m to the south several years earlier. Apparently, this site was chosen, in part, because the dumping site and scattering technique used in unloading from the railroad cars would cause the material to be widely dispersed and intermixed with large volumes of nonradioactive wastes. Furthermore, it was thought that the material would be confined in a large chasm approximately 10 m deep. There were, and still are, no public thoroughfare passes through the site or in its immediate vicinity, other than the railroad. The nearest dwelling is approximately 150 m from the site. There are probably a few persons, for example, railroad workers or hunters, who may occasionally be on the site.

Results of the radiological survey (Le78b) indicated more than 75% of the residues lie at least 3 m beneath the surface. It was estimated on the basis of historic records that about 1.5 Ci ^{238}U was dumped on this site. Auger and core hole analyses performed during the recent survey could account for approximately 1.3 Ci (Le78b). Most of this activity was in an area of less than 2 ha (4 acres). Thus, it appears that most of the dumped residue has been accounted for by the recent survey.

Areas of surface contamination appear to coincide to a large extent with those of subsurface contamination shown in Fig. 2. Beta-gamma dose rates as high as 5.4 mrad/hr were measured at 1 cm above the surface; however, most values were below 0.1 mrad/hr. The average ^{226}Ra content of numerous surface soil samples taken from the shaded area of Fig. 2 was 10 pCi/g; ^{238}U average was 3.9 pCi/g. Background concentrations in the Burrell Township core soils are 1.9 and 0.9 pCi/g, respectively.

A summary of current exposure conditions at the Burrell Township site is given in Table 3. The average outdoor ^{222}Rn concentration on the site was 0.52 pCi/ℓ. However, instantaneous measurements as high as 9.7 pCi/ℓ were observed. Radon daughter concentrations measured on the site were reasonably typical of outdoor radon daughter measurements in that area of Pennsylvania. Average background gamma radiation in the Burrell Township area was found to be 8 μR/hr. Thus, the general average of external gamma radiation on site is slightly above background. Furthermore, the concentrations of radionuclides in all water samples were below the concentration guide for water in 10 CFR 20.

In summary, the Pennsylvania Railroad Landfill Site is contaminated by about 4 Ci of ^{226}Ra and 1.5 Ci of ^{238}U spread over an area of about 2 ha (4 acres). Although most of the contamination is presently a few meters below the surface, the random dumping of materials has resulted in some areas of significant surface contamination. Radiation

exposures on this site are slightly above regional background. Occupancy of the site is very infrequent. Scenarios can be offered, such as building structures over contaminated soils on the site, which could result in overexposures to site occupants.

This site is much more typical of the MED/AEC sites surveyed by ORNL than is the Canonsburg site. The Burrell Township site has a relatively small amount of measurable contamination. However, some type of remedial action, consistent with ALARA principles, could be taken to reduce potential radiation exposures to potential site occupants.

Middlesex Landfill Site, Middlesex, New Jersey

In 1948, about 6000 m^3 of dirt contaminated with pitchblende ores were brought to the 10-ha (23-acre) landfill site from the former Middlesex Sampling Plant. In 1960, elevated gamma radiation levels were detected on this site by civil defense monitors during a local civil defense exercise. A radiological survey of the site was made at that time by the AEC, and it was found that external gamma radiation levels over an area of approximately 2000 m^2 were 20 to 50 times background levels for the surrounding area. The AEC subsequently removed approximately 600 m^3 of the contaminated material nearest the surface and covered the area with about 1 m of uncontaminated dirt. This action reportedly lowered the external gamma radiation levels to no more than 50 μR/hr.

In 1974, a second survey of the site was performed to reevaluate the radiological conditions. During the time between the 1960 and 1974 surveys, an approximately 2-ha parcel of the landfill site (originally owned by the Borough of Middlesex) had been sold to the Middlesex Presbyterian Church, and a church had been constructed on the parcel. During weekdays, part of the church building and grounds is used as a day care center for local children. The church and the Middlesex Municipal Building are located on the western edge of the site. It appears from discussions with local people that both the church and the Middlesex Municipal Building were constructed on "non-fill" or solid ground. The landfill site is surrounded by residences which approach to within 0.5 km of the south and west and to Bound Brook on the eastern and northern edges. Results of the 1974 AEC survey indicate that the remaining contamination on the property was in an area (see Fig. 3) bounded by the baseline and by the lines designated as 300R, 2+0, and 6+0 (Cr78).

Radiological survey results showed that average surface contamination of ^{226}Ra and ^{238}U in soil were indistinguishable from background activity of about 1 pCi/g of each. A few subsurface samples contained detectable activity caused by small pieces of material, presumed to be pitchblende ore. These contaminated samples were found in the general area referred to in the previous paragraph, generally at depths of less than 4 m.

A summary of present radiation exposures is shown in Table 4. Background gamma radiation measurements in this area ranged from 5 to 10 μR/hr, with an average of 8 μR/hr. Hence, the average external gamma

radiation at this site is within the range of area background. The maximum external gamma radiation was associated with surface contamination in an area of about 50 m^2. Construction of a building on this contaminated area, particularly a building with a basement, could lead to elevated human exposures. Furthermore, the underground contamination poses the potential for producing elevated exposures if future activity at the site were to uncover pieces of uranium ore at or near the surface.

This site represents the radiological condition at those MED/AEC sites where present radiation exposures cannot be distinguished from background over almost the entire site. Minor remedial measures at these sites are expected to involve minimal expenditures.

Summary and conclusions

The three sites summarized in this paper represent the range of results found in ORNL surveys of waste storage areas at former MED/AEC sites. Most of the sites would be typified by the Burrell Township site—current radiation exposures averaged over the site are slightly, but demonstrably, greater than background; small portions of the site contain highly contaminated material in close proximity to the surface of the ground. Exposures to ^{222}Rn and its daughters in structures built on these contaminated areas would probably exceed guidelines. Exposures to external gamma radiation will probably exceed guidelines at several points within the contaminated areas. However, the Burrell Township site does not demonstrate appreciable radionuclide migration due to surface runoff. This is an appreciable problem at three other MED/AEC sites. Remedial actions are required at sites such as Burrell Township.

The Middlesex Landfill Site represents the lowest end of the spectrum of present exposures found at MED/AEC sites. Exposures can be differentiated from background only at small portions of these sites. Minor remedial action would be required at these sites.

The Canonsburg site represents the upper end of the exposure spectrum found at MED/AEC sites surveyed by ORNL. Practically all of the present exposures at Canonsburg can be traced to previous waste management practices at that site. Extensive remedial measures are required to reduce radon and daughter concentrations in buildings as soon as possible at Canonsburg.

Waste management practices employed during the time that MED/AEC contracted activities were performed have a major bearing on the extent of current radiation exposures at these sites. The magnitude of these exposures is directly related to the amount of contamination still present at a site. Hence, sites with the best planned waste management practices generally have the lowest present day exposures.

Table 1. Summary of current on-site exposures at the Canonsburg, Pennsylvania, site

Exposure source	Range of average values observed	Maximum value observed
Radon in air inside structures	2.6 to 107 pCi/ℓ average daytime concentration	227 pCi/ℓ
Radon daughters in air inside structures	0.01 to 0.43 WL* average daytime concentrations	0.5 WL†
External gamma radiation inside structures	20 to 80 μR/hr averaged over buildings	310 μR/hr
Airborne ^{230}Th inside structures	0.003 to 0.2 pCi/m^3 range of spot samples	0.2 pCi/m^3
Radon in air outside structures	2.5 to 17 pCi/ℓ average 24-hr concentration	69 pCi/ℓ
External gamma radiation outside structures	110 to 210 μR/hr averaged over parcels A, B, and C	1600 μR/hr

*The WL (working level) is defined as any combination of short-lived radon progeny per liter of air which will result in the ultimate emission of 1.3×10^5 MeV of alpha energy by decay to ^{210}Pb.

†Measured during good ventilation conditions. Under poor ventilation conditions (cold weather) maximum is estimated to be 1.9 WL.

Table 2. Summary of current guidelines for exposure to a member of the general public

Exposure source	Guideline value	Documentation
Radon in air	3 pCi/ℓ	10 CFR 20
Radon daughters in air	0.03 WL	10 CFR 20
commercial structure	0.03 WL	10 CFR 712
residential structure	0.01 WL	10 CFR 712
Airborne ^{230}Th (insoluble)	0.08 pCi/m^3	10 CFR 20
External gamma radiation (whole body)	500 mrem/yr	10 CFR 20

Table 3. Summary of current on-site exposures at the Burrell Township, Pennsylvania, site*

Exposure source	Average values observed	Maximum values observed
Radon in air	0.52 pCi/ℓ	9.7 pCi/ℓ
Radon daughters in air	0.0009 WL	0.001 WL
External gamma radiation (at 1 m)	11 μR/hr	630 μR/hr
Beta-gamma radiation (at 1 cm)	<0.1 mrad/hr	5.4 mrad/hr

*Includes component of exposure due to background.

Table 4. Summary of current on-site exposures at the Middlesex, New Jersey, landfill site

Exposure source	Average values observed*	Maximum observed value
Radon in air	0.04 pCi/ℓ	Calculated to be 0.01 pCi/ℓ above background
External gamma radiation	5 μR/hr	32 μR/hr

*Includes component of exposure due to background.

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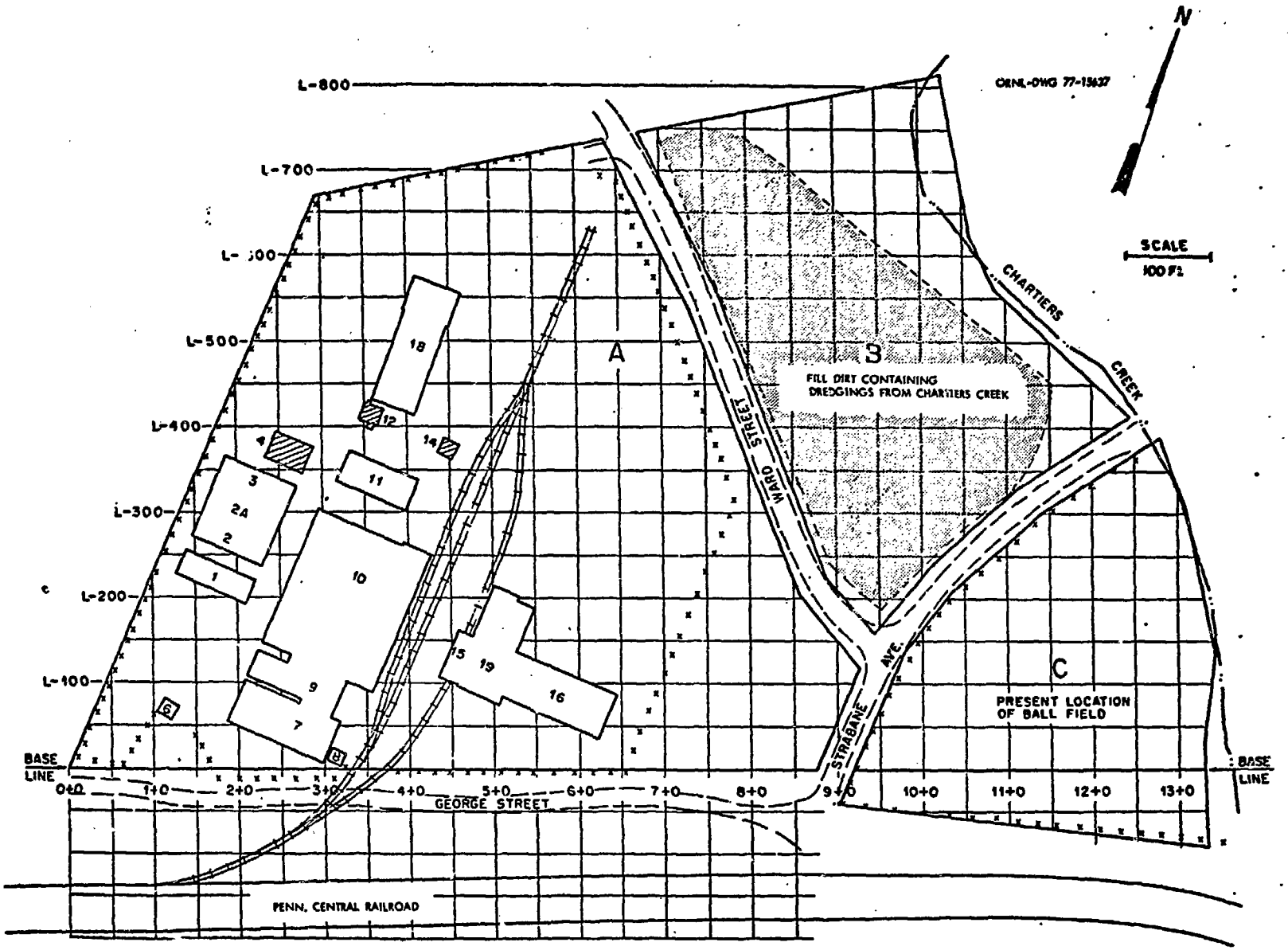

Fig. 1. Layout of the present setting of the site at Canonsburg, Pennsylvania.

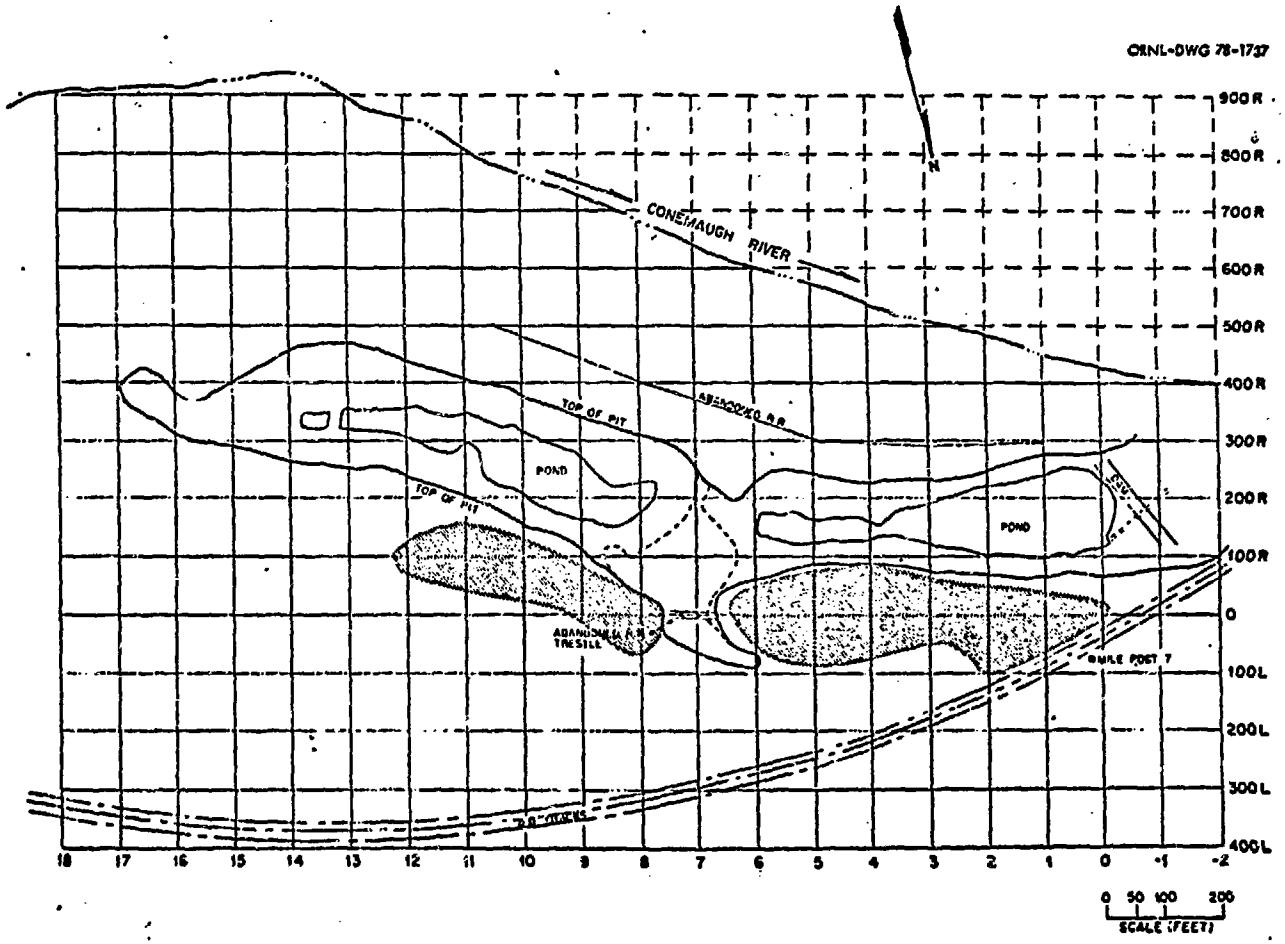

Fig. 2. Layout of landfill site at Burrell Township, Pennsylvania. Shaded areas are those where subsurface contamination has been found.

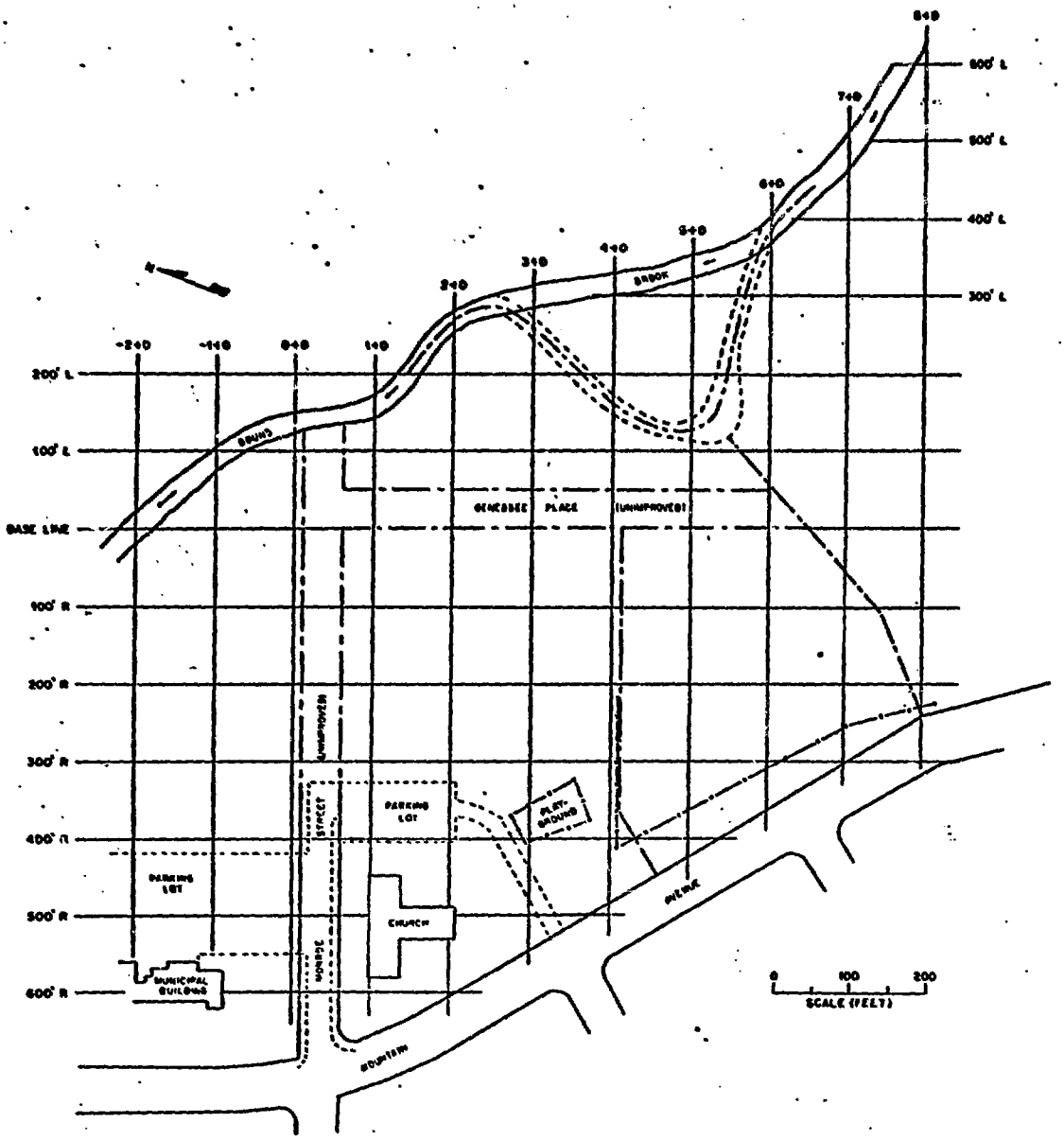

Fig. 3. Present setting of the Middlesex, New Jersey, landfill site.