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RESIDUAL RADIOACTIVITY IN THE VICINITY OF FORMERLY UTILIZED MED/AEC SITES*

F. F. Haywood and W. A. Goldsmith
Health and Safety Research Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

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DISCLAIMER

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ABSTRACT

As demand for uranium and thorium was accelerated during the 1940s, services of chemical and metallurgical firms and major research facilities were contracted as needed by the Manhattan Engineer District (MED). A lack of documentation of the radiological status at the time contracts were terminated at these facilities led the Department of Energy (DOE), and its predecessor the Energy Research and Development Administration (ERDA), to develop a major radiological resurvey program to fill this information void.

Radioactivity may not be confined to the boundaries of property on which operations were carried out. Radioactivity can be relocated to areas outside site boundaries through wind and water erosion, processing effluents transport through groundwater, spillage of incoming feed material along roads and railroads, salvage of excess equipment, and removal of contaminated material for private purposes.

A combination of aerial and ground-level radiological monitoring teams are utilized (to identify and assess off-site radioactivity). Results from comprehensive aerial surveys conducted by EG&G, Inc., a DOE contractor, provide the approximate areal extent of elevated radiation levels on the ground. These aerial survey results lead to two types of ground-level surveys: (1) gamma-ray scanning on foot or from a motorized vehicle (mobile lab based system) to pinpoint the location of residual radioactivity, and (2) comprehensive radiological surveys to determine the amount and type of materials present on specific parcels of private and public property identified during the scanning. This type of investigation was initiated in 1978 and has been successful in identifying and assessing the potential radiation hazard from property on which materials bearing natural radioactivity have been found.

This paper contains a description of the techniques used to find and evaluate radioactive material displaced outside the boundaries of a formerly utilized site. An example is given to illustrate mobile gamma-ray scanning and other radiological survey techniques used to pinpoint the location of elevated radiation levels as suggested by aerial survey results. Numerous properties in the Canonsburg, Pennsylvania, area have been found to contain materials similar to those at the nearby inactive uranium processing site. Surveys have shown that in many instances these off-site anomalies were probably a result of voluntary action by

humans, aided by a lack of security at the site after contractor operations ceased. Another example will be given, where ground-level surveys of the Niagara Falls, New York, area have established that numerous off-site anomalies (suggested from aerial survey results) were caused by the use of a radium- and uranium-bearing slag which was not associated in any manner with MED/AEC activities. In this case, uranium and thorium appeared as trace contaminants in feed material from which nonradioactive minerals (e.g., elemental phosphorus) were extracted.

INTRODUCTION

Large quantities of uranium were needed for development of the nuclear weapon beginning in 1942, and later for development of nuclear power. Once the uranium was obtained, diverse activities such as research and development, processing, production, handling, etc., were carried out by private industry, universities, etc., under contract with the Manhattan Engineer District (MED) and later the Atomic Energy Commission (AEC). Virtually all of this early work involved natural radioactivity; in some cases a purified product (uranium or thorium) was involved, and in others, the radioactivity was contained in an ore matrix.

Some degree of government control was applied to these contract activities for the purpose of protecting the health and safety of workers and the public. The specific nature of health and safety procedures in effect at that time is unknown. However, it is known that efforts were made to control the spread of contamination and procedures existed for the storage or disposition of scrap and residues. Documentation of the radiological status of formerly utilized facilities upon the termination of contracts or upon release of government controls of site activities is limited, and in many cases does not exist. Because of this, the Department of Energy (DOE, then ERDA) initiated a resurvey program designed to: (1) characterize the current radiological status of these facilities, (2) carry out remedial measures (if required) to minimize potential hazards, and (3) certify those sites which are suitable for unrestricted use. Under this program, known now as the Formerly Utilized Sites--Remedial Action Program (FUSRAP),[1,2] most of the site-specific radiological characterizations have been completed. Radiological assessment activities have been carried out by teams from three DOE laboratories: Argonne, Los Alamos, and Oak Ridge. Documentation is in the form of technical reports published by DOE in its DOE/EV-0005 series. This presents techniques used to identify and evaluate the significance of residual radioactivity in the vicinity of formerly utilized MED/AEC sites.

Migration and Movement of Radioactivity

The extraction of uranium or thorium from ores can create the potential for spread of radioactive materials to surrounding areas. Approximately 85% of the original radioactivity in uranium ore can be found in residues (tailings) remaining after extraction of the product.[3] When residues of this type were generated during MED/AEC operations, they were isolated and placed in interim and long-term storage in a few documented locations. It is during this storage period that migration or movement of residues is most likely to occur. The principal mechanisms for movement are:

1. Wind erosion - Wind may cause transport by one of three modes: surface creep, saltation, and suspension. Surface creep is illustrated through the movement of large particles along the surface due to wind or by the impaction of other particles. Intermediate-sized particles move by saltation, wherein the particles are lifted a few feet in the air by wind and quickly return to the surface. The smaller particles, including those in the respirable range, become suspended and remain suspended due to vertical turbulence. The theoretical and experimental studies of these mechanisms are well summarized by Healy[4] and Anspaugh et al.[5] The quantity of material suspended is affected by variables such as the topography and physical properties of the surface, the amount and type of vegetative cover, the extent of surface contamination, local micro-meteorology, and the time elapsed since the surface was contaminated.

2. Water erosion - Flowing surface water can cause the movement of residues from a storage pile to adjacent areas. Unless material is moved to a body of water (river, stream, etc.) for further transport, the quantity of residue along drainage paths decreases rapidly with distance. Material deposited in this manner is subject to movement by other means.

3. Groundwater transport - Radionuclides may be transported in mining groundwater. The magnitude of this type movement is determined by the proximity of residues to groundwater supplies, presence of "leaky" aquifers, differences in permeability in heterogeneous aquifer materials, concentration of cations, and the presence or absence of coprecipitating chemicals in the water. The adsorptive capacity of the subsoil will usually inhibit the movement of all tailings radionuclides except uranium migrating as an anionic uranyl complex. Most of the radioactivity which migrates in groundwater remains below the surface (unless contamination of a "leaky" aquifer containing potable water occurs); therefore, this pathway is not important to the investigations considered here except for nearby private wells.

4. Spillage of transported material - The exact means of packaging (in 1940s) of feed material such as uranium ore is not known in all cases. However, spillage along rail sidings[6] and storage areas[7] has been observed.

5. Equipment salvage - excess equipment which has been removed from the site and salvaged without adequate decontamination and radiation survey represents a potential source of off-site contamination.

6. Removal of material for private purposes - There have been many instances where residues and material bearing radioactivity were removed from a site for use on private property. This type removal has occurred on a small scale during periods of inactivity at some sites, and on a larger scale at tailings piles in western states. The best known case is the tailings removal at Grand Junction, Colorado.[8]

7. Process effluents - discharge of material from stacks, and into streams and rivers from drain lines. Once material containing radioactivity has been moved from the confines of a storage area, it is subject to further movement or distribution as a result of the action of humans (e.g., street and road repair or construction, building, etc.).

Approach Used to Locate Off-Site Radioactivity

The extent of spread of radioactivity in the immediate vicinity of a processing site or storage area may be determined in a relatively short period of time through a systematic search by monitoring personnel using portable instruments.[9] In general, this is not the case for contaminated material which has been removed from the site to private property. Distances from the original storage site may extend to a few miles, thereby involving large areas.

Based on recent experience,[10] it has been determined that an effective approach in identifying off-site residual radioactivity is the use of combined teams of aerial and ground-level radiation survey specialists. Large land areas (up to several square miles) are surveyed by EG&G, Inc., a DOE contractor using fixed wing aircraft for high-altitude surveys and helicopters for low-altitude surveys.[11] Banks of sensitive gamma-ray detectors are used to identify the location (with a high degree of spatial resolution) and approximate magnitude of radiation levels which differ from the normal terrestrial background for that area.

Information derived from an aerial survey may be used as input for ground-level investigations. Areas where elevated radiation levels are indicated by the aerial survey are located on a map. One of two survey techniques may then be employed. Discrete gridpoint property surveys are conducted using portable instruments in cases where a single parcel of property is involved, and a mobile gamma-ray scanning system is used to further identify parcels in cases involving large areas (several city blocks). After needed mobile gamma-ray scanning operations are complete, surveys are then conducted on property which was identified during that exercise.

Other ground-level property surveys have been conducted in Niagara Falls, Tonawanda, and Lewiston, New York, and Middlesex, New Jersey. In the New York areas, ground-level surveys revealed that only one newly identified site showing elevated gamma radiation levels was associated with former Manhattan Project operations. This one site among about thirty identified from the aerial survey, was found to contain ^{226}Ra-bearing soil which had been relocated by the current owner of property in North Tonawanda upon which residues from uranium refinery operations had been placed for interim storage by the U.S. Government in the 1940s. The remaining areas containing elevated gamma radiation fell into two categories: areas known (4) to contain radioactivity from former MED activities, and areas containing radioactivity as a result of non-MED industrial operations where concentrations of natural radioactivity in process residues are enhanced. In the latter category (approximately 25 areas), elevated radiation levels resulted from the presence of a slag material, possibly from phosphate rock operations. This slag, used for various purposes such as fill under roads, driveways, parking lots, etc., was found to contain uranium and radium in concentrations ranging from 30 to 50 times natural terrestrial concentrations for that area of New York; and in samples from one area, thorium was found in concentrations up to 400 times the level in natural soil.

The mobile gamma-ray scanning system now in use (this system was not used in Canonsburg, Pennsylvania) by the Oak Ridge National Laboratory (ORNL) consists of a motorized cargo van (Fig. 1) which has been equipped with specialized equipment. Two 10 cm x 10 cm sodium iodide (NaI) detectors, housed in separate moveable lead shields each with a collimated opening, are used for scanning purposes. The shields can be adjusted to various angles to provide detector coverage over a wide angle of view. Normally, one is positioned in a verticle position and one pointed toward the horizon with slightly overlapping fields of view. A pressurized ionization chamber (PIC) for measuring gamma-ray exposure rates at points of interest is provided on board. The data-gathering system is automated and consists of two multichannel pulse height analyzers, one for each NaI crystal, an electrometer for PIC readout, and a photo-optical distance measuring device mounted on the vehicle's drive train. Signals from all of these devices are transferred by an ORNL Comp-8 microcomputer to a Commodore Pet computer used for central processing. Peripheral equipment for the Pet computer includes a dual-floppy-disk for mass data storage and a highspeed printer. The system is operator controlled through keyboard instructions to the computer. Electrical power is supplied by a gasoline-powered 4 kW generator mounted near the rear of the van. A mobile radio-telephone system is used for communications. An interior view of the scanning van is shown in Fig. 2.

Specific Case Study

In April 1978, an aerial survey was conducted of the former Vitro Rare Metals Plant and surrounding area in Canonsburg, Pennsylvania. Results of this survey are included on the aerial photo in Fig. 3. In this photo, the line which defines the outer boundary of region 1 (approximately 9-50 μR/hr 1 m above the ground) is seen to be approximately symmetrical on three sides of the plant site (most of the site is covered by regions 2-4), but on the other side, this line wanders through the village of Strobane.

In order to determine if off-site radioactivity was present in this village, a study was designed and carried out during the summer of 1978. Case study objectives were:

1. Verify aerial survey results,
2. Determine specific areas where materials bearing radioactivity existed,
3. Outline the magnitude of the problem.

The approach used for radiological assessment in this area included mobile gamma-ray scanning followed by discrete gridpoint property surveys.

Scanning the village was accomplished by driving a mobile laboratory van similar to, but somewhat larger than, the one shown in Fig. 1 along every accessible street and alley in the village. The van was stopped for a short counting period in the front and rear of each parcel of property (a third side of some properties was surveyed when scanning cross streets). If, after a 30-sec count, the gross count equaled or exceeded the 60-sec count rate in areas considered to represent the area background radiation level, a 5-min gamma-ray spectrum was accumulated. Whether or not elevated concentrations of ^{226}Ra existed on the property was estimated by comparing the ratio of observed photo-peak intensities from ^{226}Ra and ^{40}K at background concentrations, to the ratio observed adjacent to individual parcels of private property.

Using the above technique, 54 (confirmed and borderline) of the approximately 500 properties scanned were found to contain ^{226}Ra-bearing material. Shortly after this task was completed, and at the request of DOE, a pilot study involving 33 properties was carried out. In this study, 29 of the properties believed to contain radioactivity and four which appeared to be uncontaminated were included. Comprehensive ground surveys were performed at each address. The survey included measurements (surface α and β-γ, gamma-ray exposure rate, etc.) in every room of each house, measurements of beta and gamma radiation levels at the ground surface and 1 m above the ground outdoors on the property, collection of sediment from basement floor drains, collection of composite soil samples, and collection of samples of contaminated soil, bricks, concrete, and other material. Most contaminated materials were found toward the rear of the property. In some cases, bricks, timbers, and various objects (all containing radioactivity) were found in storage for later use. In other cases, sand and dirt had been used to fill low areas and some miscellaneous materials had been incorporated as building material in garages, sheds, sidewalks, and basement additions. Only minor contamination was found inside residences.

Results of the pilot study were delivered to DOE in the form of brief letter reports stating the findings at each address. Included in each of these reports was a plan view of the property involved in the survey. On this plan view, contaminated areas were depicted as shaded regions of the property. Two properties typical of those containing radioactivity are shown in Figs. 4 and 5.

The significance of contamination found either inside a residence or outside on the grounds was evaluated in accordance with typical radioactivity-to-man pathways analyses. This information, along with recommendations to the property owner, was included in the letter report. An example of some of the information included in a typical letter report is given in the following excerpt:

"<u>Significance of Findings</u>:

Elevated gamma radiation levels found in the house are of no consequence. Elevated gamma radiation levels and radium concentrations in the yard are not a present hazard to occupants of this property under conditions of present use. These levels constitute only a nuisance to occupants of this property.

<u>Recommendations to Property Owner</u>:

It is recommended that no new structures be built on the property without the advice of the Department of Energy (DOE). Radium contaminated soils in shaded areas of accompanying drawing should not be used for vegetable gardens. Until contaminated soil is removed, direct contact with this material should be minimized.

Plans are being drawn up for the removal of contaminated material by authorized DOE contractors. This will be done only with consent of the property owner pursuant to a mutual agreement providing for cleanup and restoration."

The 33 properties included in this pilot study may be classified as follows:

Uncontaminated - 7
Minimal nuisance - 9
Practical concern - 17

Plans have been made to complete the assessment of properties in the Canonsburg, Pennsylvania, area during the spring and summer, 1980. It will be necessary to extend mobile gamma-ray scanning activities to areas not yet surveyed. Once all properties suspected of containing elevated concentrations of radioactivity have been identified, more extensive radiological surveys will be conducted at suspect properties. Some additional properties will be included in the survey in order to demonstrate the reliability of the mobile gamma-ray scanning technique.

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ORNL-Photo 5414-79

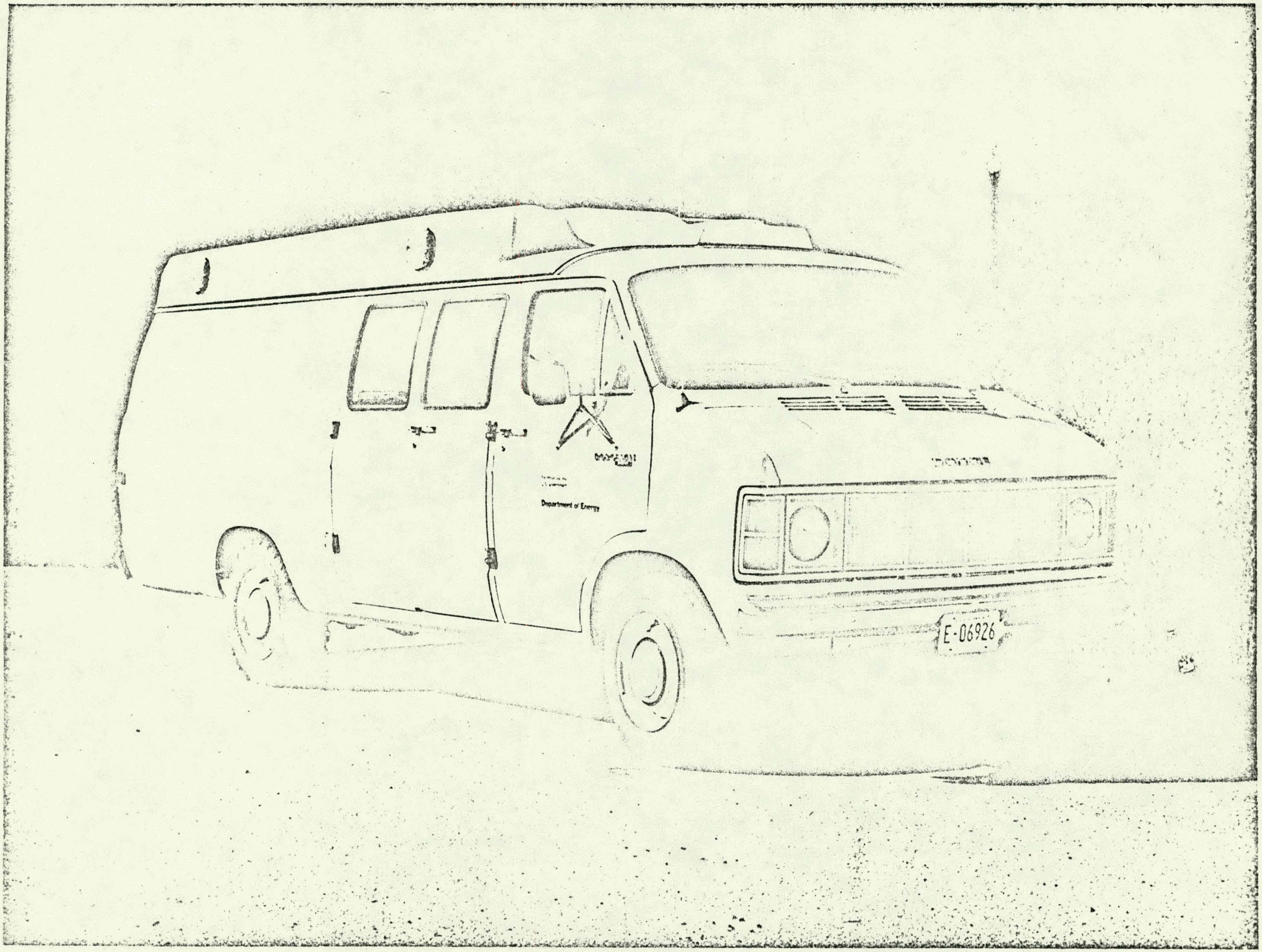

Fig. 1. Mobile gamma-ray scanning van.

ORNL-Photo 5411-79

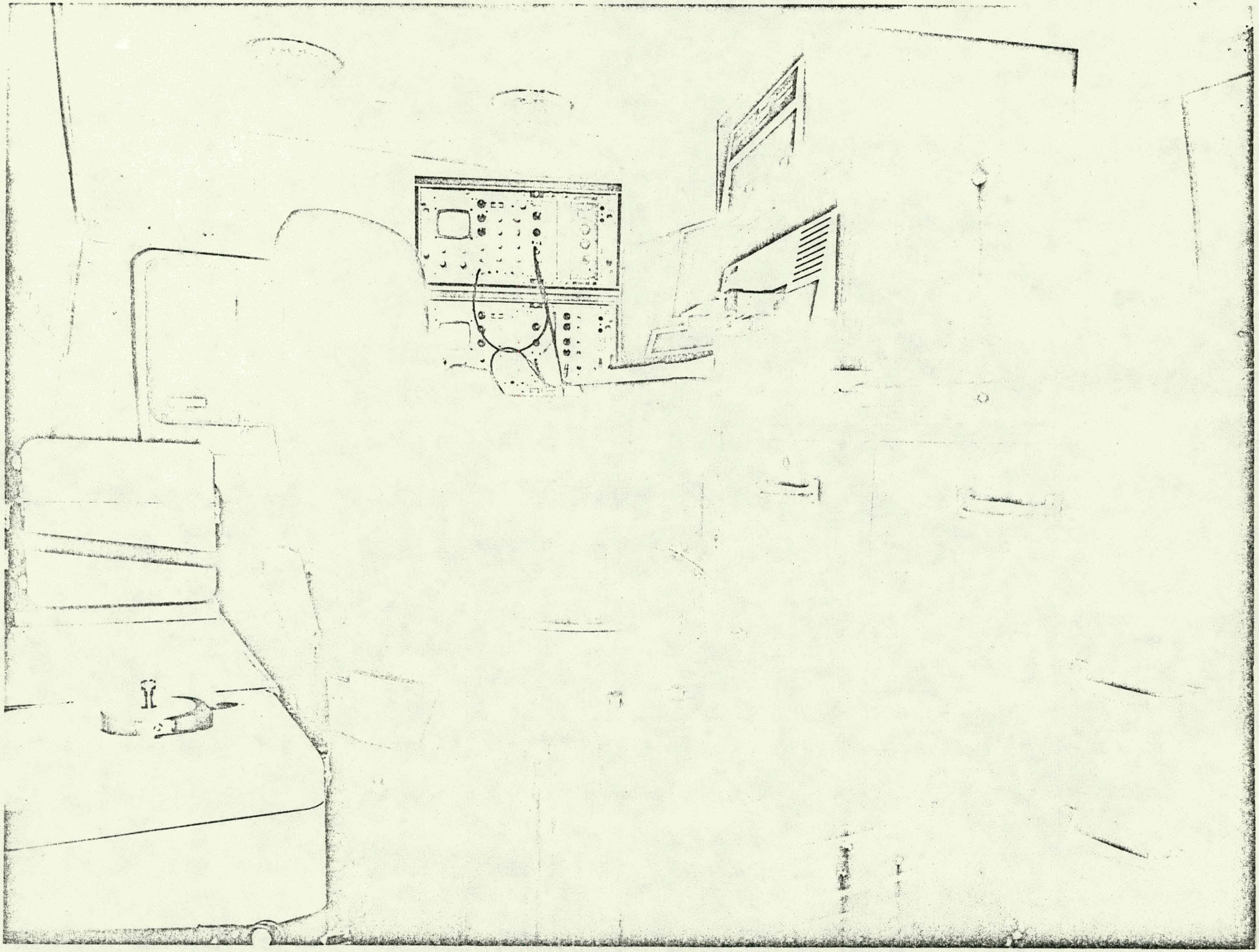

Fig. 2. Interior view of gamma-ray scanning van.

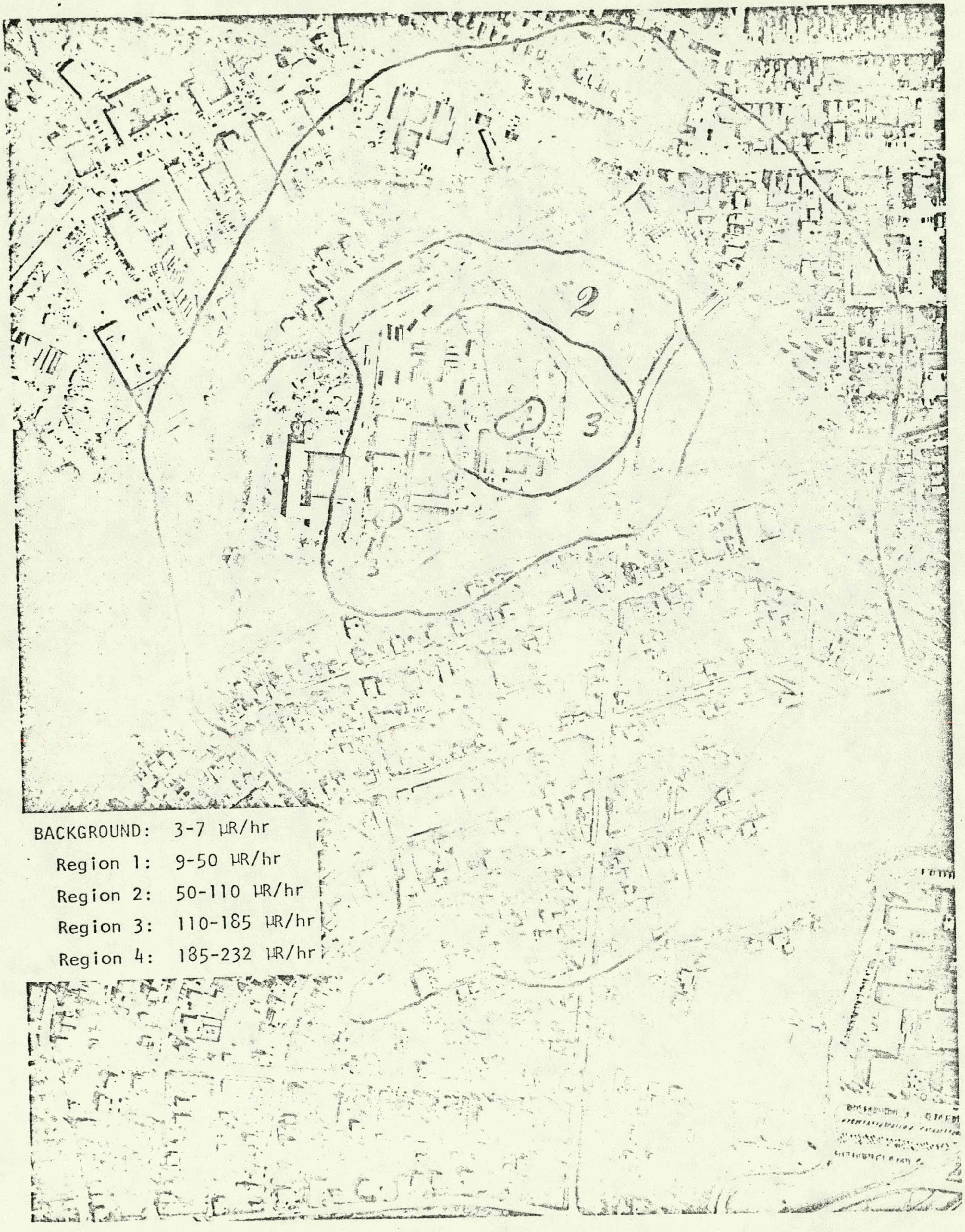

Fig. 3. Aerial view of the former Vitro Rare Metals Plant, Canonsburg, Pennsylvania (regions 2-4), including results of aerial survey (courtesy EG&G, Inc.).

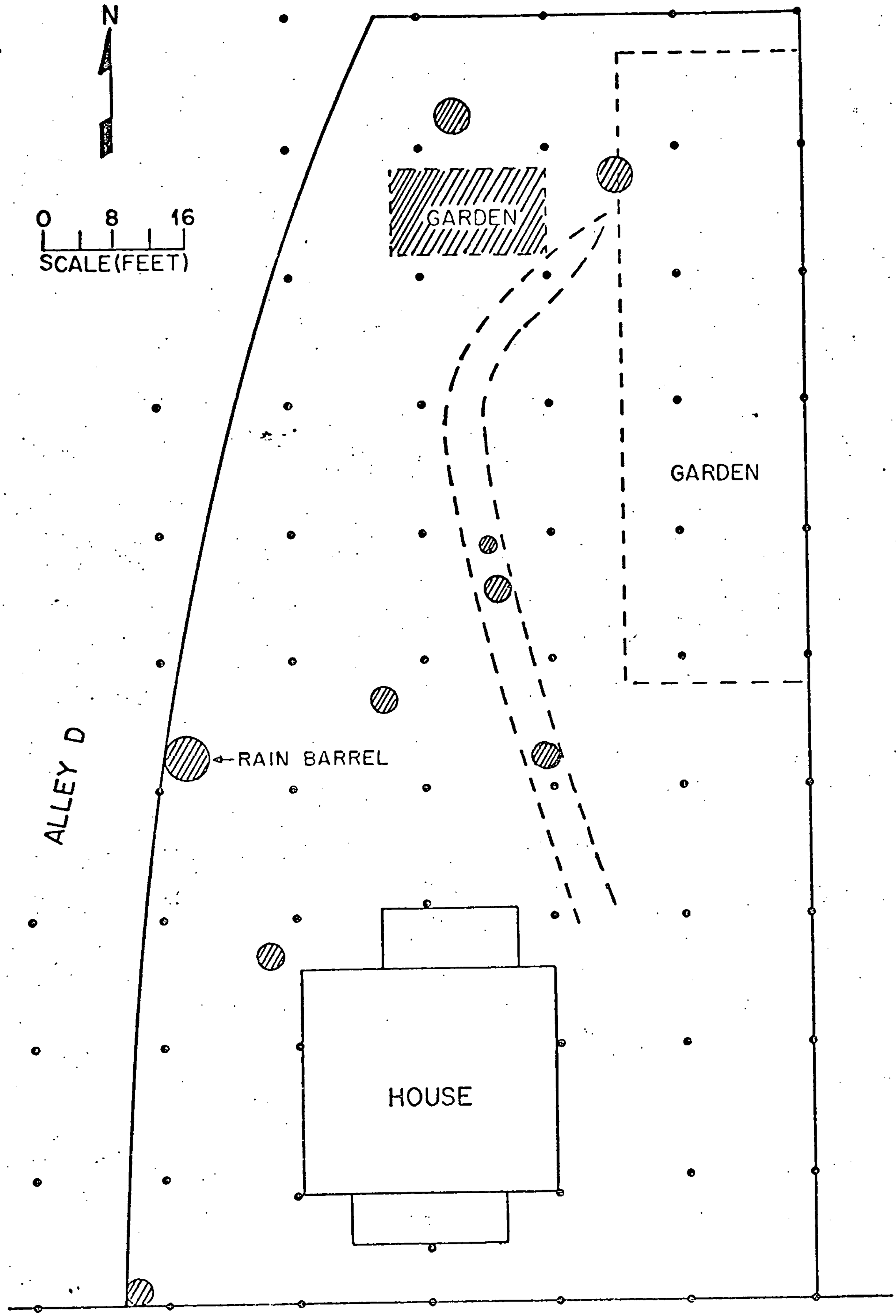

Fig. 4. Plan view of one parcel include in the pilot study of individual properties showing numerous areas (shading) of the property containing elevated concentrations of ^{226}Ra.

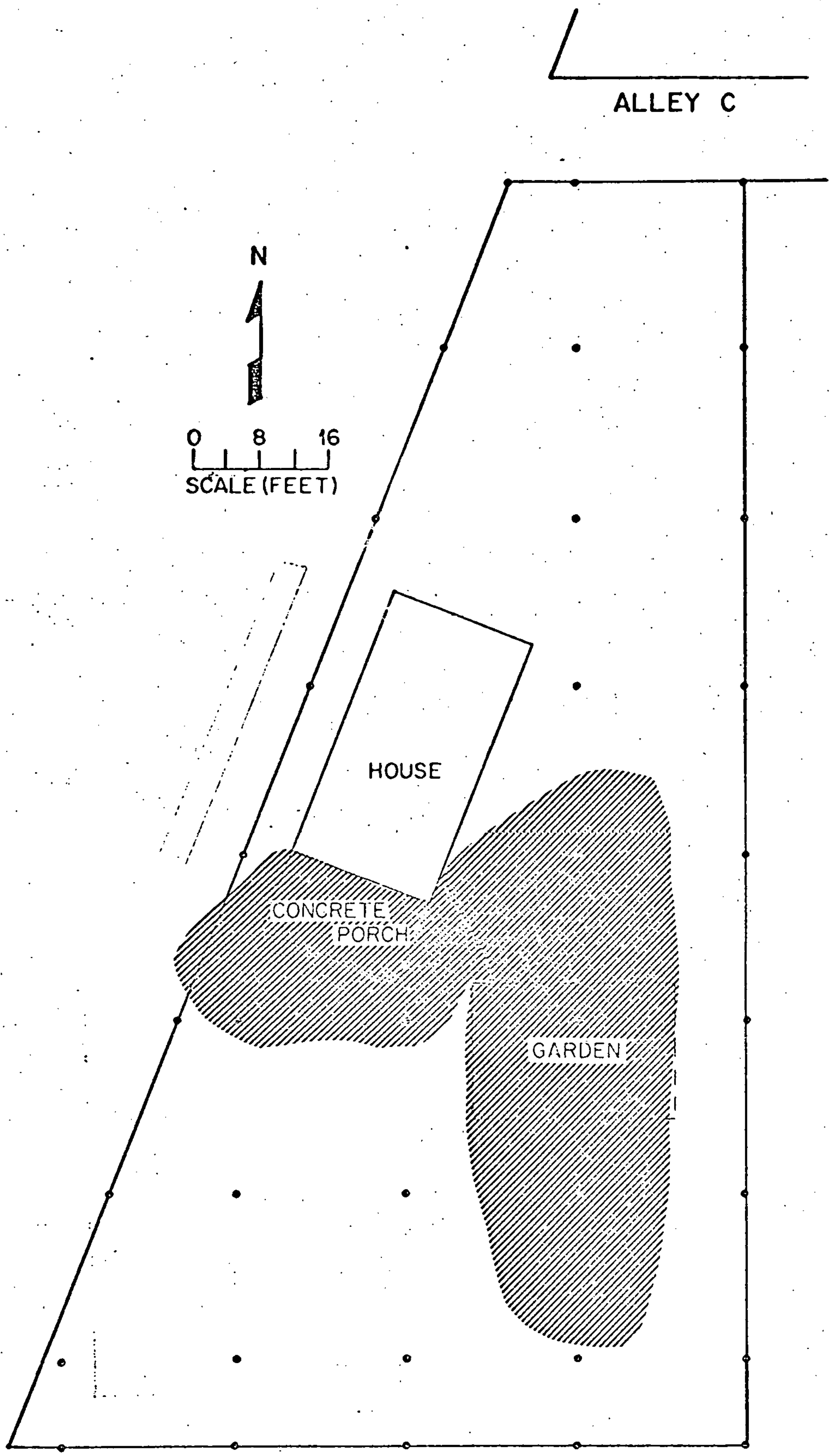

Fig. 5. Plan view of one parcel included in the pilot study of individual properties showing a large area of ^{226}Ra-bearing soil on the property.