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FINAL REPORT

RADIOLOGICAL SURVEY OF THE FORMER AEC-ST. LOUIS AIRPORT STORAGE SITE, ST. LOUIS, MISSOURI

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Work performed by the
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152-C
S 145

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CONTENTS

LIST OF FIGURES

LIST OF TABLES

RADIOLOGICAL SURVEY OF THE FORMER AEC-ST. LOUIS AIRPORT STORAGE SITE, ST. LOUIS, MISSOURI*

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ABSTRACT

The results of a radiological survey of the St. Louis-Lambert Airport property, formerly known as the Atomic Energy Commission (AEC) Airport Storage Site, St. Louis, Missouri, are presented in this report. The survey was conducted over the 21.7-acre area in which uranium and radium-bearing waste materials were stored from the late 1950's to early 1960's. The survey included direct measurements of beta-gamma radiation at the ground surface and external gamma radiation at 1 m above the ground throughout the site, measurements of radionuclide concentrations in sediments and water from Coldwater Creek and external gamma radiation levels along the creek bank which borders the west end of the property, determination of uranium and radium concentrations in samples of soil from the surface and from core holes bored at several locations on the site, and determination of radionuclide concentrations in surface soil and groundwater samples.

INTRODUCTION

At the request of the Energy Research and Development Administration (ERDA), Oak Ridge Operations, a radiological survey was conducted at the

*Research sponsored by the Energy Research and Development Administration under contract with Union Carbide Corporation.

St. Louis-Lambert Airport property, St. Louis, Missouri. This 21.7-acre tract of land is bordered on the north and east by Brown Road, on the south by the Norfolk and Western Railroad, and on the west by Coldwater Creek (see Fig. 1). The site was used as a storage area for wastes generated by the Mallinckrodt Chemical Corporation during their uranium-processing operations from 1946 to 1953. An inventory of the materials and their approximate uranium content is given in Table 1. Also given in this table is a list of the original structures and other facilities on site. This inventory was made as a part of a radiological survey conducted by the U. S. Atomic Energy Commission (AEC) in November of 1965 prior to the removal of the residue piles and disposal of structures. Since that time, access restrictions have been posted at the site, thus barring casual entry.

During 1966 and 1967 the piles were sold and removed from the site. Except for the area where barium sulfate residues (referred to as "airport cake" or AJ-4 residues) were located, the removal of residue piles restored all areas to a condition where the radiation level at the ground surface was less than 1 mrad/hr. In the AJ-4 area, the surface beta-gamma dose rate was about 3 mrad/hr due to residual contamination.[1] As stated in the acquisition permit of November 10, 1969, the St. Louis-Lambert Airport Authority agreed to decontaminate this property. In an agreement with the U. S. Government, it was required that the barium sulfate residue be removed to a federal waste repository at Weldon Springs, Missouri, and that all structures on site except the fence be razed. Building rubble, including a storage shed, truck wash pad, and a

152-I
5 154

concrete storage pit, was to be buried. Also, a minimum of one foot of clean fill was to be placed over the entire site. This work was performed during the period January through December 1969. In January 1970 a radiation survey[2] was made according to the criteria stated in Appendix II of the acquisition permit, and elevated radiation levels were found at eleven points. At these survey points, the approximate areas where radiation levels exceeded 1 mR/hr ranged from 10 ft^2 to 50,000 ft^2. Additional fill (2 to 3 ft) was placed over these areas to achieve acceptable radiation levels. Clean fill elevations were then described by a topographical survey conducted in October 1971. A radiation survey was conducted in November 1971 to document radiation levels over the entire site. Ground surface dose rates were generally less than 0.05 mrad/hr. Certain isolated areas were found exceeding 0.2 mrad/hr and were documented. No readings exceeded 1.0 mrad/hr.

RADIOLOGICAL SURVEY PLAN

The present survey was performed to characterize the existing radiological status of the property. It was conducted by four members of the Health and Safety Research Division of the Oak Ridge National Laboratory during the week of November 14, 1976. The survey included:

1. measurement of external beta and gamma radiation levels at the ground level and gamma levels at one meter. These measurements were performed on a 50-ft grid in the west end of the property (see Fig. 2 between R 15+00 and R 22+74 to the west and between S 0+00 and S 4+00 to the north); and for all areas east of R 15+00, the survey was conducted on a grid with 100-ft sides.

2. measurement of external beta and gamma radiation levels at the ground and gamma levels at a height of one meter within a fine grid in the west end (~ 47,500 ft^2), where buried material is known to exist. This area lies between R 18+50 and R 21+00, and S 0+50 and S 3+00 (see Fig. 3).
3. measurement of external beta and gamma radiation levels at ground level and gamma radiation levels at a height of one meter along the fence line, including selected points both inside and outside the fence bordering the property.
4. measurement of radiation intensities as a function of depth below the ground at bore hole sites and collection of both surface and subsurface soil samples at selected locations within the fenced area (included were some locations from the 1969 AEC survey). (Holes were logged using gamma scintillation and/or collimated glass-walled GM detectors.)
5. measurement of radionuclide concentrations in soil and water at offsite locations including Coldwater Creek and the drainage pathways north and south of the property and leading away from the property.
6. measurement of gamma-ray background and collection of surface soil samples at locations in Missouri far removed from the former AEC storage site.

RADIOLOGICAL SURVEY TECHNIQUES

Measurement of Beta-Gamma Dose Rates and Gamma Radiation Levels

Beta-gamma dose rates were measured 1 cm above the surface at each of the points mentioned in sections 1 through 3 of the Radiological Survey Plan. Within the 50-ft grid area mentioned in section 2 of the Plan (Fig. 3), each 50x50-ft area was scanned using a portable gamma scintillation (NaI crystal) survey meter. At the maximum reading, a beta-gamma measurement was made 1 cm above the ground. These beta-gamma measurements include natural background and were made with Geiger-Muller survey meters. Gamma radiation levels were measured at 1 m above the surface at the same points at which beta-gamma dose rates were measured. These measurements which also include background were made using the above gamma survey meter. Both instruments are described in Appendix I.

Surface Soil Sampling

Surface soil samples from a depth of 0 to 1 inch were taken at points determined by a 100-ft grid west of R 15+00, at points determined by a 200-ft grid east of R 15+00, and at five points where insectivore activity was noted. In addition, 15 samples were taken along the property line bordering Brown Road. Each sample was packaged in plastic bags for transport to Oak Ridge where they were dried for 24 hours at 110°C and pulverized to a particle size of 35 mesh (500 μm). Aliquots from each sample were transferred to plastic bottles (25 ml), weighed, and counted using a Ge(Li) detector. The spectra obtained were analyzed by computer techniques. A description of the Ge(Li) detector and the soil counting

techniques is given in Appendix II. Concentrations of ^{226}Ra, ^{238}U, and ^{227}Ac were determined for all samples.

Subsurface Soil Sampling

Holes were drilled to depths of 8 to 15 ft at 16 grid locations using a motorized drilling rig. An auger with a 5-in. diameter hollow core was used for drilling these holes. Gamma radiation profiles were measured at various depths in the core holes by lowering a scintillation probe inside the open hole. This "logging" of core holes was done as a first step in determining the depth of contamination in the soil.

At 8 of the 16 core hole locations, soil samples were collected using Shelby tube samplers. These loaded tubes were shipped to Oak Ridge where the soil was then removed and analyzed using the techniques described in Appendix II. Concentrations of ^{226}Ra, ^{238}U, and ^{227}Ac were determined for all samples.

Groundwater Sampling

Corings were made at 6 grid locations to a depth where groundwater was reached. At each location a 2-liter water sample was collected. These samples were analyzed at ORNL using sequential separation techniques to determine ^{238}U, ^{226}Ra, and ^{230}Th concentrations.

Offsite Sampling and Analysis

Four water samples were taken from Coldwater Creek, which borders the west side of the site. A sample of sediment was taken from the bed of Coldwater Creek at each of the locations used for sampling water.

Each sediment sample was prepared and analyzed using the soil sample analysis techniques described before. The creek water samples were analyzed using the same sequential separation techniques as for the groundwater samples. Gamma radiation levels were measured at 1 m above the creek bed at each sampling point.

Along each side of Brown Road are drainage ditches which carry run-off water westward into Coldwater Creek. The ditch adjacent to the south side of Brown Road serves as a drainage path for the former AEC storage site. This ditch is connected to the drainage ditch on the north side of the road by two culverts. The south side of the site is drained by a ditch which borders the Norfolk & Western Railroad track and which also drains into Coldwater Creek.

Gamma radiation levels at 1 m and beta-gamma dose rates at 1 cm were averaged over areas of 1 m^2 centered at selected points along these drainage pathways. Also, surface soil samples were taken at five locations in the ditch north of Brown Road, and two Shelby-tube samples were taken in the ditch south of Brown Road.

SURVEY RESULTS

External Beta-Gamma Dose Rates

The average beta-gamma dose rate a 1 cm above the surface was 0.05 mrad/hr with a range of 0.02 to 0.34 mrad/hr at grid points in the 100-ft grid area east of grid line R 15+00; 0.05 mrad/hr with a range of 0.02 to 0.23 mrad/hr at grid points in the 50-ft grid area west of grid line R 15+00; and 1.51 mrad/hr with a range of 0.24 to 4.57 mrad/hr within the

fine grid blocks shown in Fig. 3. Each beta-gamma dose rate reported for the 50 and 100-ft grid points represents the average of several readings taken over an area of 1 m^2 centered at the grid point. The beta-gamma dose rates reported for the fine grid blocks represent readings taken within each grid block in Fig. 3. Individual beta-gamma measurements at grid points in Figs. 2 and 3 are given in Tables 2 and 3.

External Gamma Radiation Levels

The average external gamma radiation level at 1 m above the surface was 16 μR/hr with a range of 4 to 71 μR/hr at grid points in the 100-ft grid area; 14 μR/hr with a range of 5 to 43 μR/hr at grid points in the 50-ft grid area; and 113 μR/hr with a range of 23 to 297 μR/hr within the fine grid area. At grid points S 3+00/R 4+00 and S 3+00/R 22+00, it was observed that the gamma radiation levels at 1 m were higher than the corresponding beta-gamma dose rates at 1 cm above the surface. This is attributed to a nonuniform distribution of radioactive materials on the ground and to abrupt variations in local topography, factors which govern the radiation level as a function of height above the ground. The approximate average background radiation level for the St. Louis area is 8 μR/hr.

At the 11 grid points (marked with an asterisk in Table 2) outside the fence along the north property line, the average external gamma radiation level was 124 μR/hr, with a range of 26 to 238 μR/hr. Detailed gamma measurements for the grid points in Figs. 2 and 3 are given in

S 157

Tables 2 and 3. Erosion of fill earth has occurred along the fence line, exposing some of the contaminated soil. Moles or other insectivores have tunneled extensively in this area, and some of the excavated soil has been brought to the surface. The insectivore activity does not extend more than ten feet from any point along the fence.

Background Concentration of Radionuclides in Soil

Samples of surface soil were collected at ten locations throughout Missouri as shown in Fig. 4. This material was returned to ORNL for analysis using gamma-ray-spectrometry techniques. Results of these analyses are given in Table 4. It was observed that the concentration of ^{226}Ra ranged from 0.31 to 1.33 pCi/g. The average ^{226}Ra concentration ($\bar{X}$) was 1.05 pCi/g with a standard deviation (σ) of 0.3 pCi/g. The range in values for ^{232}Th was 0.76 to 1.19 pCi/g; and for ^{234}Th, which is assumed to be in equilibrium with its parent ^{238}U, the range was 2.14 to 9.97 pCi/g.

Surface Soil Analysis

Locations at which onsite surface soil samples were collected and the results of gamma-ray-spectrometry analyses of these samples are listed in Table 5. These surface soil samples have been divided into three groups--those taken at grid points used for beta and gamma-ray measurements (samples 1 through 49), those taken along a fence bounding the north side of the property (samples F1 through F15), and those collected in areas excavated by insectivores (V1 through V5). Concentrations of ^{226}Ra, ^{238}U, and ^{227}Ac in these samples are listed in Table 5. Much

of the surface soil at grid points inside the fence boundary was found to contain normal terrestrial concentrations of ^{226}Ra and ^{238}U. However, there were 26 sampled grid locations where the concentration of ^{226}Ra exceeded the maximum ^{226}Ra concentration observed in Missouri background samples. The range of elevated values was from approximately 1.35 to 77.9 pCi/g. The range of natural uranium concentrations in background samples was 2.14 to 9.97 pCi/g. There were 11 grid points where uranium was found in excess of the observed maximum background value. The range at these grid points was 11.2 to 757 pCi/g. None of the background samples contained measurable quantities of ^{227}Ac, daughter of ^{231}Pa. However, ^{227}Ac was found at 18 of the grid locations with a range of 0.46 to 77.3 pCi/g. The source of the ^{227}Ac is linked to a precipitate formed in a column where uranium was stripped from diethyl ether using dilute nitric acid. This precipitate was, on occasion, removed from the column by a Sperry Filterpress.[1] This "Sperry cake" was found to be a good source of ^{231}Pa and, hence, of its daughter ^{227}Ac. The largest concentration of this radionuclide, 1084 pCi/g, was found near the area where barium sulfate cake ("airport cake") had been stored. Also, a ^{227}Ac concentration of 77.3 pCi/g was observed in an area where pitchblende raffinate (AM-7) had been stored.

There were 5 locations on the site where there was obvious insectivore activity. Surface samples were collected at each location. Sample V5, just outside the fence (grid location S 5+25/R 10+50), contained 1260, 1076, and 1084 pCi/g, respectively, of ^{226}Ra, ^{238}U, and ^{227}Ac.

S 159

At points just outside the fence and along the north property boundary, 15 surface soil samples were collected. The range of ^{226}Ra activity was 2.9 to 452 pCi/g. Likewise, the range in ^{238}U and ^{227}Ac activity was 2.0 to 1510 pCi/g and 3.55 to 287 pCi/g, respectively.

Offsite Measurements

A summary of the environmental radiation and radioactivity measurements in the vicinity of Coldwater Creek is given in Table 6. External gamma radiation readings at 1 m were highest at the run-off water outfalls where significant erosion of the fill dirt had occurred. At the south outfall, where the highest external gamma radiation level (18 μR/hr) was measured, soil erosion has resulted in a ditch which is 6 to 8 ft deep at the fence. This ditch extends eastward, becomes progressively shallow, and terminates 200 ft due east of the south outfall. The external gamma radiation level at 1 m above the creek bed at the north outfall was 12 μR/hr. Less erosion has occurred at the north outfall, but some fill soil has eroded into Coldwater Creek. The ^{226}Ra concentrations in creek sediment samples were 11.5 and 1.59 pCi/g in the samples taken adjacent to the south and north outfalls, respectively; while the ^{238}U concentrations at the south and north outfall were 26.2 and 12.3 pCi/g, respectively. Analysis of the creek water showed that neither ^{238}U nor ^{226}Ra concentrations varied significantly with location; the average values were 1.04×10^{-3} pCi/ml ± 24% and 1.69×10^{-4} pCi/ml ± 44%, respectively. It should be noted that at a point 50 meters upstream from the south outfall, the concentration of ^{238}U and ^{226}Ra in creek

Surface?

S 160

bed sediments was in the range of normal terrestrial concentrations observed in other parts of the state.

A summary of beta-gamma dose rates at 1 cm above the surface taken in the drainage ditch that runs west along the south property line is given in Table 7. No reading exceeded 0.06 mrad/hr. At grid line R 11+00, the ditch turns due northwest and becomes the south outfall drainage path. The low readings from grid line R 0+00 through R 11+00 suggest that the radioactive material which was transported by erosion to the south outfall originated west of the grid line R 11+00.

Results of the measurements taken in the drainage ditches on either side of Brown Road are given in Tables 8 and 9.

In the drainage path south of Brown Road, the average external gamma radiation level at 1 m above the surface was 37 μR/hr, with a range of 3 to 83 μR/hr; and the average beta-gamma dose rate at 1 cm above the surface averaged over 1 m^2 at each grid point was 0.15 mrad/hr, with a range of 0.02 to 0.57 mrad/hr (Table 8). Two core samples were taken at grid lines R 5+75/S 4+00 (hole No. 16) and R 9+20/S 5+60 (hole No. 15) in this drainage path, and the concentrations of ^{226}Ra and ^{238}U as a function of depth are given in Table 10.

The results of the measurements taken in the drainage ditch north of Brown Road are given in Table 9. The external gamma radiation level at 1 m above the surface averaged 58 μR/hr with a range of 15 to 89 μR/hr. The beta-gamma dose rates at 1 cm from the surface ranged from 0.05 to 0.34 mrad/hr, averaging the readings over an area of 1 m^2 centered about the grid point. Surface soil samples were taken at each of the five

points in the north ditch and were analyzed for ^{238}U, ^{226}Ra, and ^{227}Ac. These results are also given in Table 9.

Subsurface Soil Analysis

Estimates of radium concentrations as a function of depth and the location of sampling points are given in Table 11. A combination of soil sample analyses and scintillation probe "loggings" was used to estimate these concentrations. Graphs of the count rates as measured using the unshielded scintillation probe vs. depth permit an accurate estimation of the depth at which the maximum ^{226}Ra concentration occurs and the thickness of the contaminated layer. Estimates of the depth and extent of the maximum radium concentration are also given in Table 11. The gamma-ray logging technique used during this survey is not specific for a given nuclide. However, some comparisons have been made between observed response of the gamma-ray logging probe and measured ^{226}Ra concentrations in soil taken from points corresponding to gamma-ray measurements. It is thus possible to make an estimate of the thickness of contaminated layers underground. The techniques used in estimating the depth and extent of contamination are explained in Appendix III. At 7 of the 16 core holes, soil samples were taken to a depth ranging from 2 to 4 ft. The concentrations of ^{238}U, ^{226}Ra, and ^{227}Ac in these samples are presented in Table 12. An examination of the data in Table 12 shows that in the majority of cases where ^{226}Ra and ^{238}U were found in excess of observed background values, the average of the ratio ^{238}U:^{226}Ra was 4.6. Therefore, one can, to a first approximation, estimate the percent uranium in the contaminated layers beneath the surface. For example, consider Hole No. 4, Table 11. The average ^{226}Ra concentration from the

surface to a depth of 7 ft was 1115 pCi/g, suggesting an average ^{238}U concentration of 5129 pCi/g. A uranium concentration of 172 pCi/g is equivalent to 0.05 percent uranium by weight. Therefore, the soil in the first 7 ft at Hole No. 4 contains approximately 1.5% uranium. The range for all 16 bore hole locations was 0.01 to 1.9%.

Groundwater Analysis

At 6 of the 16 core hole locations, drilling was continued to the depth at which groundwater was encountered. A 2-liter water sample was collected at each location and analyzed to determine the concentrations of ^{238}U, ^{230}Th, ^{226}Ra and ^{210}Pb. The results of these analyses are presented in Table 13. The point at which water was reached is also given; this point ranged from 17 to 35 ft below the surface, the mean sea level elevation (msl) of which varied from 521 to 528 ft. It is not possible to describe the migration of radionuclides in groundwater on the basis of the limited number of samples collected. However, the total radionuclide concentration in groundwater (^{238}U, ^{230}Th, ^{226}Ra and ^{210}Pb) as a function of depth and the concentration of ^{238}U as a function of depth are presented in Fig. 5 (average values given for 25 and 35 ft). It is seen that to a depth of approximately 23 ft most of the activity is due to uranium. Below that point, ^{210}Pb is the predominant nuclide. Even though the concentrations are significantly above background, the RCG[3] is exceeded only for ^{210}Pb (100 pCi/liter) at 25 and 35 ft.

SITE TOPOGRAPHY

As an adjunct to the radiation survey and at the request of ERDA, a topographical survey was made on January 4, 1977. The purpose of obtaining

elevations on the site was to determine whether there had been a change in surface contour since the previous survey in 1971. Elevations were determined with a resolution of six inches. Results of the survey are presented as contour lines in Fig. 6. A close comparison was made between the latest topographical survey and the one made in 1971. There were numerous points where the present elevation was below the 1971 value. In order to appreciate fully some of the surface changes which have occurred during the past six years, several graphs have been prepared. These depict the surface elevation in 1971 and in 1977 along grid lines and are given in Figs. 7 through 15. These particular grid lines were chosen because the new elevation was six inches or more lower than when last surveyed. In some cases, the surface had sunk from two to three feet (see Figs. 10, 12 and 14).

SUMMARY

At the request of ERDA, a comprehensive radiological survey was conducted at the former Atomic Energy Commission Airport Storage Site in St. Louis, Missouri. This survey included radiation measurements at 1 cm above the surface of the ground and at a height of one meter above the ground both inside the fenced boundary of the storage site and at points outside the fence and property boundaries. The storage site was divided into two distinct sections. In the east section, measurements were made at the intersection of grid lines whose spacing was 100 ft. Beta-gamma radiation levels at 1 cm above the ground in this area ranged from 0.02 to 0.34 mrad/hr (the average was 0.05 mrad/hr), and the average gamma-ray exposure rate was 16 μR/hr with a range of 4 to 71 μR/hr.

In the western section of the site, measurements were made on a grid whose spacing was 50 ft. The average beta-gamma reading in this area was 0.05 mrad/hr with a range of 0.02 to 0.23 mrad/hr. Gamma-ray exposure rates in the western section ranged from 5 to 43 μR/hr with an average of 14 μR/hr. One small area in the western section was found where the readings were elevated in numerous places. The beta-gamma readings in this area ranged from 0.24 to 4.57 mrad/hr, and the gamma-ray exposure rates at one meter above the ground ranged from 23 to 297 μR/hr. Some gamma-ray measurements were made along the outside of the fenced boundary and were found to range from 26 to 238 μR/hr at one meter above the surface. All of these points are accessible to the public. Although no one works near the point of maximum exposure, the potential annual exposure is 0.5 rem based on a 40-hr week.

Samples of soil were collected from the surface at many of the survey points mentioned above. There were 26 points where the ^{226}Ra concentration was found in excess of the maximum level for background concentration observed in Missouri. The range in these data was 1.35 to 77.9 pCi/g. In the same area, ^{238}U and ^{227}Ac concentrations were also found to be elevated. The ranges in these concentrations were 11.2 to 757 pCi/g and 0.8 to 77 pCi/g, respectively. Holes dug by insectivores* and rabbits were found in five locations in the fenced area. Elevated radioactivity was found at one of these locations. The concentrations of ^{226}Ra, ^{238}U and ^{227}Ac were 1260, 1076 and 1084 pCi/g, respectively. Measurements in the vicinity of Coldwater Creek revealed near normal background conditions except near the two outfalls (north and south) from the storage site. An analysis of sediments from the creek indicated

*Moles, shrews, hedgehogs, etc.

S 165

^{238}U concentrations of 26.2 pCi/g at the south outfall and 12.3 pCi/g at the north outfall. The concentration of ^{226}Ra at the south outfall was 11.5 pCi/g; and at this same point, the gamma-ray exposure rate was 18 μR/hr.

In the drainage ditches on either side of Brown Road which bounds the storage site on the north side, external gamma-ray exposure rates were found to range from 3 to 89 μR/hr.

Water samples were collected in Coldwater Creek and in six bore holes. Only ^{238}U and ^{226}Ra were sought in the samples from Coldwater Creek. The concentrations of these nuclides were 1.04×10^{-3} pCi/ml and 1.69×10^{-4} pCi/ml. The most restrictive nuclide concentration is for ^{226}Ra in the soluble form and is 30 pCi/liter.* It is seen that the concentration of ^{226}Ra in Coldwater Creek is more than two orders of magnitude lower than this permissible concentration. An analysis of groundwater reveals measurable quantities of several nuclides to depths of 35 ft below the surface. Although many of the concentrations exceed normal background levels (as seen in Table 14), only ^{210}Pb is seen to exceed the RCG (100 pCi/liter) for continuous exposure.

One of the most significant potential sources of radiation exposure is the contaminated soil in the west section of the site. At the present time, most of the contamination is covered with earth in varying thicknesses. In some places, this earth cover has eroded up to three feet in the short span of six years after it was placed over some barium sulfate cake residuals left on the storage site when stored material was sold and removed. Holes which were drilled through the area of known

* 10 CFR 20

S 160

contaminated deposits revealed ^{226}Ra concentrations to 1438 pCi/g and corresponding ^{238}U concentrations to 1.9% by weight. The contaminated layers ranged in thickness from four to seven feet with the maximum contamination occurring from 2.5 to 5.5 ft below the surface. This corresponds to an elevation (msl) of the contaminated layer ranging from 521 to 524 ft.

A complete topographical survey was made of the site. A comparison was made between the site elevations measured in 1971 and those measured by the same company in 1977. It was seen that there is a slight decrease in elevation due to natural settling and erosion. In some cases the elevation change averaged six inches per year.