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RADIOLOGICAL SURVEY OF THE LIQUID EFFLUENT DISPOSAL PATHWAYS FORMERLY USED BY LINDE AIR PRODUCTS DIVISION (Union Carbide Corporation) Tonawanda, New York

P.W. Frame, J.D. Berger, C.F. Riemke, L.A. Young W.O. Helton, R.D. Condra, C.F. Weaver

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INTRODUCTION

From mid-1943 through mid-1946 Linde Air Products Corporation, Tonawanda, New York, under contract with the Manhattan Engineer District, separated uranium from ores to produce uranium oxide $(U_{3}O_{8})$. The $U_{3}O_{8}$ was then converted to uranium dioxide and uranium tetrafluoride, but it was only the first step, the production of $U_{3}O_{8}$, that resulted in any significant quantities of radioactive waste. This waste consisted primarily of a gelatinous residue from the filtering of uranium carbonate, and a liquid effluent from the filtration of sodium diuranate.

The solid residue was disposed of according to the source of the ore being processed. Domestic ores handled by Linde had been pre-processed so that their Ra-226 concentrations were relatively low. Residues generated by the processing of these domestic ores were stored at the Haist Property, Tonawanda. African ores handled by Linde had not undergone pre-processing. As a result, these ores and their waste residues possessed comparatively high radium concentrations. The waste residues from African ores were stored at the Lake Ontario Ordnance Works in Lewiston, New York.

The liquid effluents were caustic solutions (pH approximately 10) primarily due to high concentrations of dissolved sodium carbonate, sodium hydroxide, and sodium sulphate. Initial discharges of this liquid waste (an estimated 80 million to 100 million liters)¹ were directed into the sanitary sewer system. Due to problems created at the Tonawanda sewage treatment facility by the caustic solutions, this disposal procedure was halted in April 1944. Subsequent wastes were discharged into several on-site wells (an estimated 207 million liters).¹ Seven different wells were used for this purpose between April 1944 and July 1946. These wells would frequently become plugged or overflow, in which case, the effluents (an estimated 214 million liters)¹ were diverted to a storm drainage ditch, no longer existent, which flowed into Two Mile Creek.

Current estimates indicate that the concentrations of uranium in effluents released into the sanitary sewer system were between 6×10^{-7} and $1.8 \times 10^{-4} \ \mu$ Ci/ml (average: $8.6 \times 10^{-5} \ \mu$ Ci/ml).¹ These estimated values are within the present NRC limit of $1 \times 10^{-3} \ \mu$ Ci/ml for disposal into a sanitary sewer.² Effluents released into the disposal wells and storm sewers are estimated to have had uranium concentrations ranging between 6×10^{-6} and $4 \times 10^{-5} \ \mu$ Ci/ml (average: $1.6 \times 10^{-5} \ \mu$ Ci/ml).¹ The maximum estimated concentration is above the NRC guideline of $3 \times 10^{-5} \ \mu$ Ci/ml for release to unrestricted areas, however, the average concentration is within this limit.

The concentrations and chemical form of the radium in the liquid effluents are not as well known as in the case of uranium. Estimates of the total radium concentrations in the wastes generated from the processing of African ores are between 2.5 x 10^{-6} and 4.3 x 10^{-5} µCi/ml. Concentrations in effluents from the processing of domestic ores would probably have been lower.

Current NRC limits for release into a sanitary sewer are 7 x 10^{-7} µCi/ml for soluble Ra-226 and 9 x 10^{-4} µCi/ml for insoluble Ra-226.² For release into an unrestricted area, these limits are 3 x 10^{-8} µCi/ml and 3 x 10^{-5} µCi/ml for soluble and insoluble Ra-226

respectively. The solubility of radium in these effluents is unknown and the present concentration guidelines may have been exceeded if the radium was in a soluble form.

Recent sampling of several test wells and a portion of Two Mile Creek on the Linde property has indicated water and soil radionuclide concentrations within the NRC guidelines for unrestricted use.^{2,3,5} To further confirm these findings, a survey of all pathways utilized by Linde for disposal of the liquid effluent from the uranium process was performed on June 24-25, 1981 by the Radiological Site Assessment Program of Oak Ridge Associated Universities, Oak Ridge, Tennessee.

SURVEY OBJECTIVES

This survey was performed to 1) obtain further information concerning radionuclide concentrations in existing portions of the aforementioned liquid waste pathways, 2) characterize the composition of current residues in the disposal pathways, and 3) determine the baseline levels of these radionuclides in the Tonawanda area for comparison.

SITE DESCRIPTION

Linde Air Products Corporation is located in the town of Tonawanda, New York. The plant occupies a portion of the property bounded by East Park Drive on the west, Sheridan Drive on the north, New York Central Railroad on the east, and Woodward Avenue on the south (Figure 1). Two Mile Creek runs in a northerly direction essentially parallel to and on the west side of East Park Drive. As seen in Figure 1, this creek expands into a small pond on either side of Sheridan Drive. Between 1944 and 1946, liquid waste carried by the storm sewer system discharged via a storm drainage ditch into Two Mile Creek at a point near the southern tip of this pond.

Many sections of the original pathways followed by the liquid wastes no longer exist. The on-site wells employed for waste disposal have been sealed and extensive portions of the original sanitary and storm sewer systems, including the ditch leading to Two Mile Creek, have been abandoned, removed, or filled in. Furthermore, the sewer systems have undergone periodic cleanings since 1946 when the wells were last used for disposal.

The nearest residential area is on the western side of Two Mile Creek. Mr. P. Sarno who lives approximately two kilometers north of the Linde site at 538 Two Mile Creek Road, is the only local resident utilizing well water. He has been obtaining general purpose water from the well (depth approximately 30 m) for the past twenty years, however high mineral concentrations prohibit its use for drinking water.

Due to the lack of wells in the Tonawanda area, there is not adequate data to establish the direction of ground water movement. Conflicting opinions of local engineers suggest the direction to be either westerly or northerly. It has been estimated that it would cost approximately \$100,000 to resolve this question, and this expense is felt to be beyond the scope of this survey.

SURVEY PROCEDURES

Samples collected during this survey included soil, sediment (from both the bed of Two Mile Creek and the Linde sewer system) and water.

Sampling Techniques

1. Soil and Sediment Sampling

Surface (to 5 cm) soil samples of approximately 1 kilogram each were collected by trowel after removing vegetation

cover. Subsurface soil samples were collected at a depth of between 10 cm and 15 cm. Sediment samples from the sewer systems were collected with the aid of a long handled scoop. Sediment samples from Two Mile Creek were collected from the upper six inches of sediment using a post-hole digger or trowel, depending on the depth of the water and accessibility to the creek bank.

2. Water Sampling

Water samples of approximately 3.5 liters were collected by dipper and stored in plastic collapsible containers. Caution was used to avoid disturbance and collection of sediments. Immediately after collection, the temperature and pH of the water were monitored. Where appropriate, flow rates were estimated.

Sample Locations

The following samples were collected from various effluent pathways using the methods described above:

Off-site Samples

- Surface soil, sediment, and water samples at one site upstream of the Linde discharge point, at the discharge point, and at five sites downstream from the discharge point (see Figure 1).
- Baseline surface soil, sediment, and water samples from Ellicott Creek and an unnamed creek (i) near the intersection of Niagara Falls Boulevard and Interstate 290 (see Figure 2).
- 3. Water from the Tonawanda city water supply, Tonawanda Creek at N. Ellicott Creek Road, a small unnamed creek (ii) on the

north side of North French Road, and a small unnamed creek (iii) north of the intersection of Millersport Highway and Miller Road (see Figure 2).

4. Water from the well of Mr. P. Sarno at 538 Two Mile Creek Road (see Figure 1).

On-site Samples

- Surface and subsurface soil samples near the two original disposal wells where sampling was possible (see Figure 3),
- Water samples from a depth of approximately 16 meters in both test wells (see Figure 3),
- 3. Surface soil samples from spoil discarded during drilling of the two test wells (see Figure 3),
- 4. Water samples from one storm sewer and three sanitary sewers that were part of the sewer system that originally existed in the vicinity of the disposal wells (see Figures 4, 5),
- 5. Sediment samples from five storm sewers and two sanitary sewers that were part of the sewer system that originally existed and in the vicinity of the disposal wells (see Figures 4, 5).

Sample Analysis and Interpretation of Data

Soil and sediment samples were analyzed for U-238, U-235, Ra-226, K-40 and Cs-137 by gamma spectrometry. Alpha spectrometry was employed for the evaluation of Th-230 and Th-232 concentrations. Water samples initially were analyzed for U-238, U-235, K-40, Cs-137 and gross alpha concentrations. However, large quantities of suspended solids created high MDA's* for the gross alpha analysis.

*Minimum Detectable Activities

It was therefore decided to perform radon emanation analyses for Ra-226 and alpha spectrometry for Th-230 and Th-232 on all water samples. The five soil and sediment samples with the highest U-238 concentrations were also analyzed for Ac-227 and Pa-231 by gamma spectrometry. Analytical procedures are described in further detail in Appendix A.

RESULTS

Baseline Samples

Baseline soil samples collected from the banks of Ellicott Creek (site 23) and the first unnamed creek (site 24) contained radionuclide concentrations in the following ranges; 0.04 to 0.05 pCi/g for U-235, 0.42 to 0.64 pCi/g for Ra-226, 0.15 to 0.27 pCi/g for Cs-137, 0.51 to 0.56 pCi/g for Th-232, 1.12 to 2.83 pCi/g for U-238, 5.75 to 12.71 pCi/g for K-40, and 0.59 to 0.73 pCi/g for Th-230 (Table 1). Radionuclide concentrations in the sediment samples from these sites contained 0.05 pCi/g for U-235, 0.55 to 0.70 pCi/g for Ra-226, 0.08 to 0.13 pCi/g for Cs-137, 0.70 to 0.80 pCi/g for Th-232, 0.82 to 0.95 pCi/g for U-238, 12.51 to 19.73 pCi/g for K-40, and 0.60 to 0.70 Ci/g for Th-230 (Table 2).

Baseline water samples were collected from the two sites mentioned previously, Ellicott Creek (site 23) and the unnamed creek (site 24), plus, Tonawanda Creek (site 26) and two other unnamed creeks (sites 27, 28). Concentrations of radionuclides in these samples ranged from <0.4 x 10^{-8} to 2.0 x 10^{-8} µCi/ml for U-235, 0.008 x 10^{-8} to 0.034 x 10^{-8} µCi/ml for Ra-226, <0.7 x 10^{-8} to 1 x 10^{-8} µCi/ml for Cs-137, <0.01 x 10^{-8} to <0.03 x 10^{-8} µCi/ml for Th-232, <214 x 10^{-8} µCi/ml for U-238 and 0.03 x 10^{-8} to 0.07 x 10^{-8} µCi/ml for Th-230,(Table 3).*

^{*&}lt; symbol denotes that the concentration determined was less than the minimum detectable.

Off-Site Samples

Two Mile Creek

Soil from the banks of Two Mile Creek generally contained radionuclide concentrations within the range found in the baseline samples. The maximum U-235, Ra-226, and Th-232 concentrations (0.08, 0.85 and 0.88 pCi/g respectively) are from the bank near the outfall of the former storm drainage ditch. The other maximum concentrations, 5.42 pCi/g for U-238 and 0.33 pCi/g for Cs-137, and 2.51 pCi/g for Th-230, were found near Ensminger Road (approximately 1.0 km downstream from the outfall).

Sediment obtained from the various locations along Two Mile Creek generally contained radionuclide levels comparable to sediments found in Ellicott Creek and the first unnamed creek. Highest values for U-235, Cs-137, and U-238 (0.10, 0.47, and 4.30 pCi/g respectively) were found in the sample collected upstream from the outfall.

Water samples contained U-235, Ra-226, Cs-137 and U-238 concentrations similar to the five baseline water samples from Ellicott Creek, Tonawanda Creek and the three small unnamed creeks. Three of the locations sampled along Two Mile Creek (site 3, site 6, and site 7) correspond to the three locations sampled in the Oak Ridge National Laboratory (ORNL) survey.⁴ Ra-226 concentrations at site 3 $(0.031 \times 10^{-8} \mu \text{Ci/ml})$ and site 7(0.017 x $10^{-8} \mu \text{Ci/ml})$ are comparable to the values reported for these sites by ORNL (<0.018 x 10^{-8} µCi/ml and 0.027 x 10^{-8} µCi/ml respectively). ORNL's analysis of 0.33 x 10^{-8} μ Ci/ml for Ra-226 at site 6 is higher than our determination of $<0.007 \times 10^{-8}$ uCi/ml, however, ORNL did not report a range of error for their results. Furthermore, radionuclide concentrations in Two Mile Creek will be dependent on a number of variables including water level, flow rate, and temperature in the creek. U-238 concentrations at these sites have been determined to be below the MDA's of the present survey. Since the ORNL values were also below our MDAs, a comparison of U-238 concentrations between the two studies is not possible.

Private Well and City Water Samples

U-235, Cs-137 and U-238 concentrations in the city water sample and water from the private well of Mr. P. Sarno showed little difference from those characterizing the baseline samples discussed in the previous section. The Ra-226 concentration in the well water $(0.091 \times 10^{-8} \mu \text{Ci/ml})$ was approximately three times the highest level in the baseline samples but still well within the NRC guideline² of $3\times10^{-8} \mu \text{Ci/ml}$. The Ra-226, Th-232, and Th-230 concentrations in city water were comparable to that of the baseline samples.

On-Site Samples

Disposal and Test Wells

Surface and subsurface soil samples collected close to the disposal wells contained radionuclide concentrations above concentrations noted at off-site locations. The ranges of levels measured near these wells were U-235, 0.36 to 0.84 pCi/g; Ra-226, 0.93 to 5.53 pCi/g; Cs-137, 0.19 to 0.47 pCi/g; Th-232, 0.66 to 0.92 pCi/g; U-238, 11.20 to 24.05 pCi/g; and Th-230, 1.72 to 5.90 pCi/g. The maximum concentrations of all of these radionulides except Cs-137 were found in the subsurface soil near the test well north of Building 38. During the ORNL survey of Linde Air Products, the closest sample collected to that same location (site 11) was reported to have comparable surface soil concentrations, 34.7 pCi/g for U-238 and 3.3 pCi/g for Ra-226.⁴

Small quantities of spoil were left behind from the drilling of the two test wells. Samples of this spoil had U-238 concentrations of 10.96 pCi/g and 26.4 pCi/g for the wells north of Building 38 (site 12) and south of Building 8 (site 13) respectively. These same locations also had concentrations of all other radionuclides of concern in excess of the concentrations in the baseline samples. The

maximum Th-230 concentration in soils (8.79 pCi/gm) was noted in the debris from the test well at site 12.

Radionuclide concentrations in water from the two test wells were $\langle 0.5 \times 10^{-8}$ to 1 x 10^{-8} µCi/ml for U-235, 0.016 x 10^{-8} to 0.031 x 10^{-8} µCi/ml for Ra-226, 1 x 10^{-8} µCi/ml for Cs-137, $\langle 0.1 \times 10^{-8}$ µCi/ml for Th-232, $\langle 59 \times 10^{-8}$ to $\langle 64 \times 10^{-8}$ µCi/ml for U-238 and 0.2 x 10^{-8} to 0.6 x 10^{-8} µCi/ml for Th-230. These values are in agreement with the previous analyses of the well performed by Argonne National Laboratory.³

Sanitary and Storm Sewers

The highest Ra-226 concentration encountered in the survey, 6.9 pCi/g, was in the sediment from a storm sewer near the northeast corner of Building 30 at collection site 19 (U-238, 99.2 pCi/g). This sediment also had the highest Th-230 concentrations encountered, 17.7 pCi/g. Silt from a sanitary sewer at collection site 15, immediately south of Building 19, recorded the highest U-238 concentration 362 pCi/g (Ra-226, 1.94 pCi/g). Only twenty-five meters separates these two sites. It was also in this same general area that the ORNL study⁴ noted the highest levels of Ra-226 in soil. Sediment samples from another storm sewer, just south of Building 8 at collection site 21, also contained elevated levels of U-238 (116 pCi/g). Water samples from the latter collection site had the highest U-235, Cs-137 levels (3 x 10^{-8} µCi/ml and 2 x 10^{-8} µCi/ml respectively). Th-232 concentrations from sewer samples were not significantly different from the baseline concentrations; however, the Th-230 concentrations in the storm sewer sediments from locations 19 and 21 were the highest noted during this survey, i.e. 17.7 and 9.89 pCi/g respectively.

The five soil and sediment samples with the highest U-238 levels had the following Ac-227 and Pa-231 concentrations (Table 5): 14.25 pCi/g Ac-227 and 0.73 pCi/g Pa-231 for subsurface soil near the disposal well at site 11 (0.84 pCi/g U-235), 2.10 pCi/g Ac-227 and

<0.29 pCi/g Pa-231 for soil from the test well spoil at site 13 (1.09 pCi/g U-235), 5.54 pCi/g Ac-227 and 0.95 pCi/g Pa-231 for sediment from the sanitary sewer at site 15 (12.93 pCi/g U-235), 14.29 pCi/g Ac-227 and 1.14 pCi/g Pa-231 for sediment from the storm sewer at site 19 (4.57 pCi/g U-235), and 2.07 pCi/g Ac-227 and <0.39 pCi/g Pa-231 for sediment from the storm sewer at site 21 (4.10 pCi/g U-235). In all cases the Ac-227 concentration was greater than the Pa-231 concentration. This increase in the Ac-227 to Pa-231 ratio has been noted in residues from other Uranium processing operations and is thought to be due to selective leaching during the chemical separations procedures.

COMPARISON OF SURVEY RESULTS WITH GUIDELINES

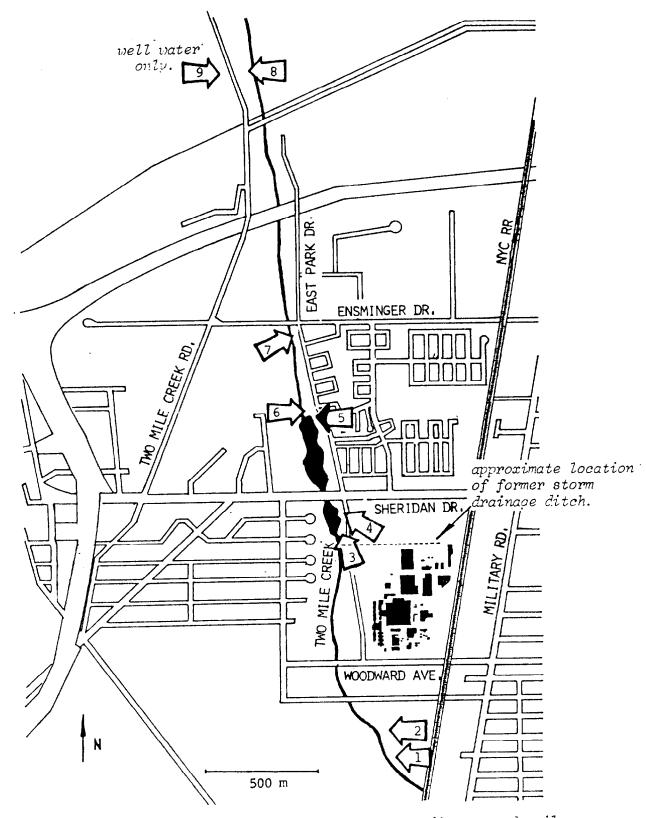
Guidelines for permissible concentration of radionuclides in water released into unrestricted areas are given in 10 CFR 20, Appendix B, Table II, Column 2. The applicable concentrations for soluble forms are 3 x 10^{-5} µCi/ml for U-235, 3 x 10^{-8} µCi/ml for Ra-226, 2 x 10^{-5} µCi/ml for Cs-137, 2 x 10^{-6} µCi/ml for Th-232, 4 x 10^{-5} µCi/ml for U-238, and 2 x 10^{-6} µCi/ml for Th-230. The measured concentrations in all water samples analyzed in this survey were below these levels.

The Environmental Protection Agency has proposed an interim limit of $\langle 5 \text{ pCi/g}$ for Ra-226 in surface soil.⁵ Guidelines for permissible soil concentrations of the rest of these radionuclides have not yet been adopted. Levels which have been applied by the Nuclear Regulatory Commission for decommissioning specific sites are 32.5 pCi/g for U-235, 31.0 pCi/g for Th-230, 35.0 pCi/g for U-238 and 23.1 pCi/g for Th-232.⁶ Surface soil concentrations from all the off-site samples are below these levels. While these proposed soil guidelines are not intended to apply to restricted areas, it should be noted that only one soil sample (5.53 pCi/g of Ra-226) exceeded these levels.

Guidelines for radionuclide concentrations in sediment have not been proposed. It was noted, however, that none of the levels for radionuclide concentrations in soil were exceeded by the off-site sediment samples. Three on-site sediment samples (sites 15, 19, and 21) exceeded these guidelines for U-238, and one of these (site 19) exceeded the Ra-226 concentration guideline.

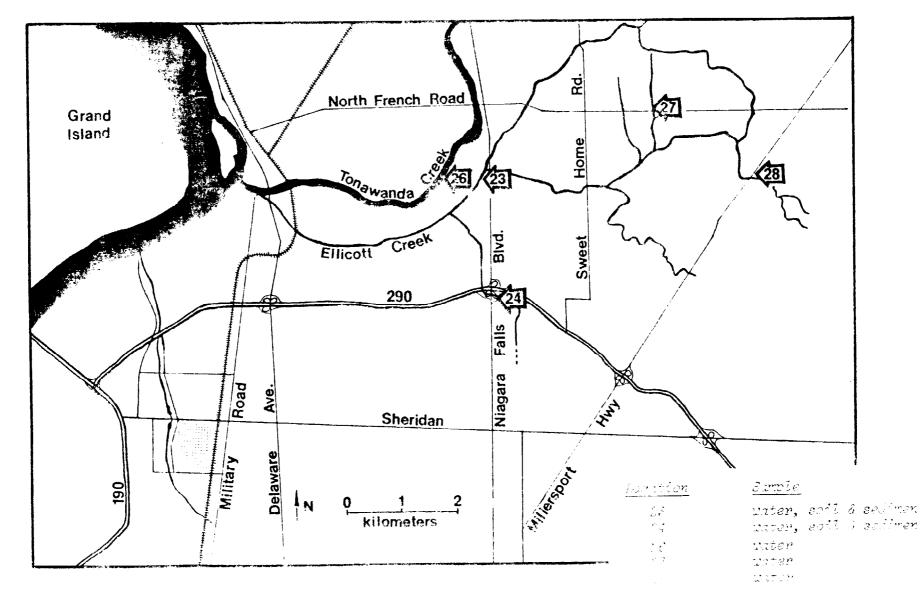
SUMMARY

Concentrations of radionuclides, associated with uranium processing operations at the Linde Air Products facility in Tonawanda, New York, during the mid 1940's were measured in various liquid effluent pathways utilized by that organization. Several soil and sediment samples, collected from isolated locations on the Linde facility property, contain uranium processing residues which exceed the guidelines for unrestricted areas. However, these do not present a radiological hazard to either the on-site employees or the general public. The concentrations of radionuclides in the water, soil, and sediment of off-site pathways are in the range of the normal environmental levels in the Tonawanda area. Therefore radiation exposure to the public resulting from the previous operations at the Linde facility are negligible.



💭 indicates sampling sites for water, sediment, and soil.

FIGURE 1. View of Linde Air Products Division and Vicinity Indicating Sample Collection Sites Along Two Mile Creek.



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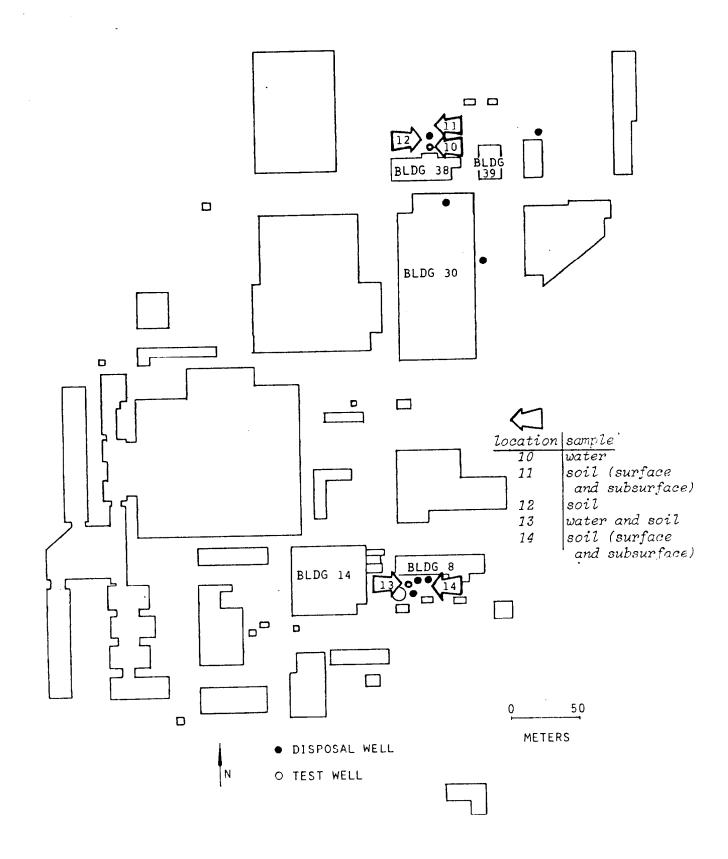


FIGURE 3. Plan View of Linde Air Products Division Indicating Original Disposal Wells, Test Wells, and Sample Collection Sites.

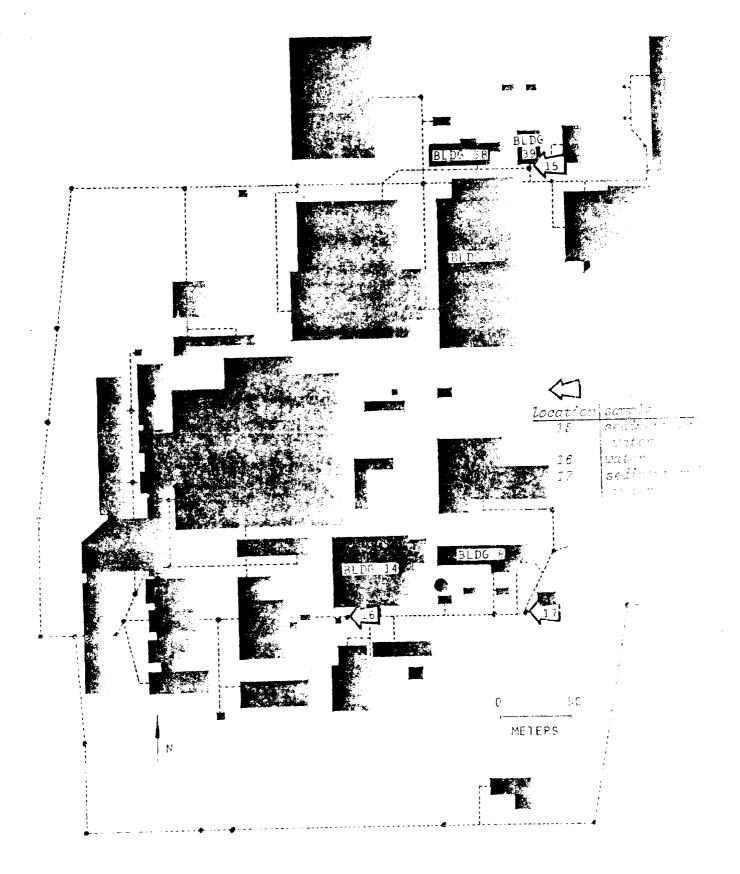


FIGURE 4. Plan View of Linde Air Products Division Indicating Present Sanitary Sewer System and Sample Collection Sites.

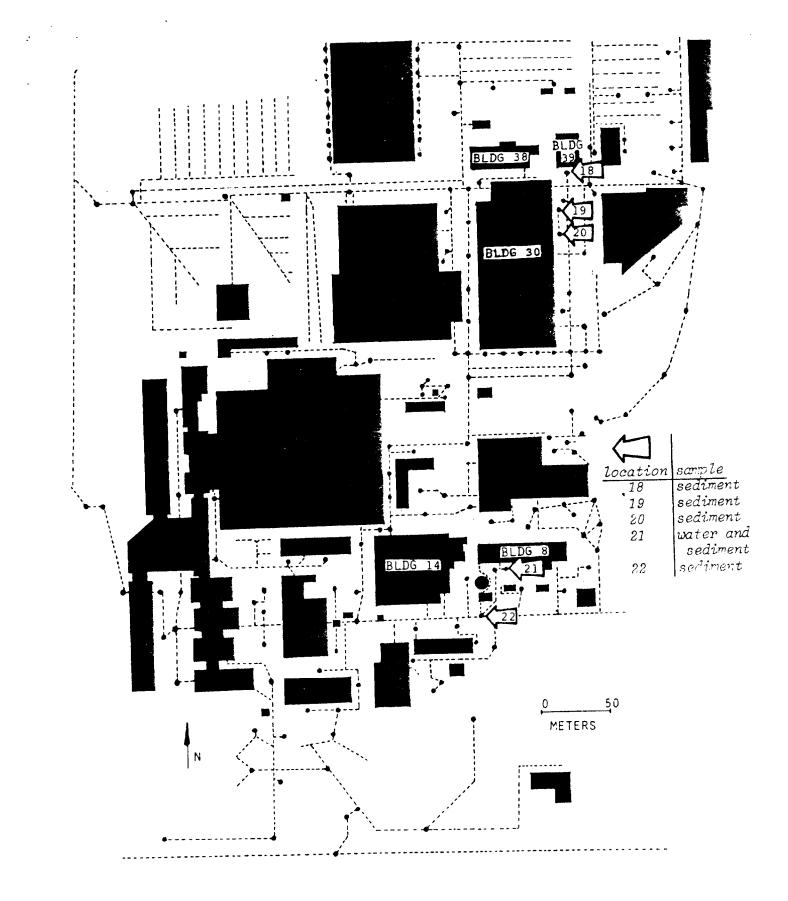


FIGURE 5. Plan View of Linde Air Products Division Indicating Present Storm Sewer System and Sample Collection Sites.

Collection Site	Sample Description	U-235 (pCi/g)	Ra-226 (pCi/g)	Cs-137 (pCi/g)	Th-232 (pCi/g)	Ŭ-238 (pCi/g)	K-40 (pCi/g)	Th-230 (pCi/g)
2	Two Mile Creek - Upstream	0.03±0.01 ^b	0.50±0.02	0.03±0.01	0.59±0.09	0.85±0.10	11.72±0.33	1.72±0.1
3	Two Mile Creek	0.08±0.02	0.85±0.03	0.08±0.01	0.03±0.01	1.33±1.28	15.65±0.41	0.03±0.0
4	Two Mile Creek	0.01±0.01	0.20±0.01	0.20±0.01	0.07±0.08	<0.36	2.24±0.16	0.10±0.
5	Two Mile Creek	0.04±0.01	0.53±0.02	0.08±0.01	0.88±0.15	0.78±0.97	13.51±0.32	0.76±0.
6	Two Mile Creek	0.05±0.01	0.54±0.02	0.19±0.01	0.65±0.12	2.10±0.98	9.77±0.30	1.10±0.
7	Two Mile Creek	0.01±0.01	0.69±0.03	0.33±0.02	0.60±0.12	5.42±1.72	14.9 ±0.039	2.51±0.
8	Two Mile Creek	0.07±0.01	0.48±0.03	0.31±0.02	0.57±0.17	1.15±1.00	10.56±0.31	0.74±0.
11	Near Disposal Well - Surface	0.57±0.06	2.74±0.13	0.47±0.05	0.72±0.12	15.80±4.20	6.07±0.65	3.55±0.
11	Near Disposal Well-Subsurface	0.84±0.09	5.53±0.19	0.24±0.05	0.92±0.19	24.05±6.14	3.48±0.69	5.90±0
12	Test Well Debris	0.33±0.06	1.93±0.11	0.13±0.04	0.74±0.15	10.96±4.07	13.55±0.96	8.79±0.
13	Test Well Debris	1.09±0.06	0.82±0.08	1.00±0.04	0.51±0.10	26.40±4.80	12.33±0.91	3•53±0.
14	Near Disposal Well-Surface	0.35±0.04	0.93±0.08	0.20±0.03	0.66±0.10	11.40±3.30	4.09±0.51	1.72±0
14	Near Disposal Well-Subsurface	0.36±0.05	0.94±0.08	0.19±0.03	0.78±0.06	11.20±3.60	5.99±0.60	3.30±0
23	Ellicott Creek	0.05±0.01	0.64±0.03	0.15±0.01	0.56±0.07	2.83±1.36	12.71±0.36	0.59±0
24	Unnamed Creek (1)	0.04±0.01	0.42±0.02	0.27±0.01	0.51±0.13	1.12±0.95	5.75±0.24	0.73±0

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TABLE 1 RADTONUCLIDE CONCENTRATIONS IN SOIL (pCi/g)

a Refer to Figures 1, 2, 4 & 5 for location of samples. b Errors given are 2 σ due to counting statistics only.

Collection Site	Samile Sactivian	0-225 () ()	Pa 226 (1012/g)	02-137 (p01/g)	Th-232 (p ^r i/g)	U-238 (pC1/z)	K-40 (ptt/g)	ಡಿ∽ನ3೦ (p೧೮/g)
1	Two Mile Creek-Upstream	0.1010.020	0.6910.05	47+0.03	0.01±0.04	4.30±2.10	11.2110.51	0.9250.24
3	Two Mile Creek	0.0610.01	0.52±0.02	0.02±0.01	0.02±0.01	0 .71 ±0 . 95	14.23±0.33	0.0250.01
4	Two Mile Creek	0.04±0.01	0.62:0.02	0.06±0.01	0.55±0.09	0.59	10.05+0.29	0.5710.11
5	Two Mile Creek	0.04±0.01	0.5450.02	0.0450.01	0.40-0.05	1.53+0.90	12.85±0.33	1.5510.11
6	Two Mile Creek	0.09±0.03	0.81+0.07	0.13±0.03	0.46:0.16	3.50:2.80	4.21±0 .51	1.11:0.27
7	Two Mile Creek	0.03±0.01	0.58+0.03	0.18±0.01	0.40:0.06	0.98±0.99	6.38:0.25	0.64±0.09
8	Two Mile Creek	0.04:0.01	0.41+0.02	0.2010.01	0.56±0.19	<0.65	8,91±0,31	0.93±0.28
15	Sanitary Sewer	12.93±0.25	1.94:0.18	0.43±0.10	0.11±0.06	362±20	4.83±0.88	1.33±0.20
17	Sanitary Sewer	0.05:0.01	0.38:0.02	0.07±0.01	0.21±0.05	<0.51	2.72±0.17	0.34+0.0
18	Storm Sewer	0.1910.01	1.35:0.04	0.32:0.01	0.62:0.10	6.4711.38	4.73-0.22	1.41±0.1
19	Storm Sewer	4.57±0.17	6.93:0.26	0.4510.06	0.51±0.09	99.20±11.27	5.05±0.79	17.7 +0.6
20	Storm Sewer	0.52±0.05	1.5910.10	0.2920.04	0.65±0.13	12.80±3.70	2.16:0.41	2.0350.2
21	Stona Sever	4,10-0,12	0.89±0.11	0.1010.05	0.34±0.04	116± 13	13.80±1.20	3.89+0.2
22	Storm Sewer	0.17**.03	0.5910.07	0,12±0.03	0.39±0.09	4,50±2.76	6-0510-63	0.2010.0
23	Ellicott Creek	0.05:0.01	0.35+0.02	0.13±0.01	0.70+0.02	0.82±1.09	12-5110-33	0.60±0.1
24	Unnamed Cceck (i)	0.0310.01	0.70±0.03	0.08±0.01	0.8010.18	0.95±1.13	19.73+0.41	0.70+0.1

TABLE : ADMONIONIDE COMMENTER DAS IN CEDIMENT (DUL/g)

a Refer to Figures 1, 2, 1 & 5 for locations of complex.
 b Errors gives see 2. Sub to counting a sticker body.

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.(. 0)	(.5 80)	(14-5) 1.34 ·	(8.)
105 de	55 - 17 10	191-11, 182 43	
6.1	$\int_{\mathcal{O}} z ^{-1} = - z $	1,5° 4/ 2,623	(747)

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Collection Site	Sample Description	U-235	Ra-226	Cs-137	Th-232	V-238	Th-230
Site a 1 3 4 5 6 7 8 9 10 13 12 15 16 17 21 23 24 25 26 27 28	Description Two Mile Creek-Upstream Two Mile Creek Two Mile Creek Unde Test Well Sanitary Sewer Sanitary Sewer Sanitary Sewer Storm Sewer Ellicott Creek Unnamed Creek (1) City Water Tonawanda Creek (11) Unnamed Creek (11)	$\begin{array}{c} 0-235 \\ <0.5 \\ <0.4 \\ <0.4 \\ 2 \pm 2 \\ <0.4 \\ <0.4 \\ 1 \pm 1 \\ <0.5 \\ 1 \pm 1 \\ <0.5 \\ 1 \pm 1 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <1 \pm 2 \\ <0.4 \\ <0.4 \\ <0.4 \\ <1 \pm 2 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ <0.4 \\ $	$\begin{array}{c} \text{Ra-226} \\ \hline 0.037 \pm 1.020 \\ \hline 0.031 \pm 0.024 \\ 0.033 \pm 0.020 \\ 0.056 \pm 0.025 \\ \hline 0.007 \\ 0.017 \pm 0.019 \\ 0.083 \pm 0.025 \\ \hline 0.091 \pm 0.029 \\ 0.031 \pm 0.023 \\ 0.016 \pm 0.022 \\ \hline 0.032 \pm 0.04 \\ 0.034 \pm 0.022 \\ 0.026 \pm 0.024 \\ 0.034 \pm 0.022 \\ 0.030 \pm 0.020 \\ 0.010 \pm 0.017 \\ 0.008 \pm 0.017 \\ \hline 0.008 \pm 0.018 \\ \hline 0.018 $	$\begin{array}{c} 1 \pm 1 \\ < 0.3 \\ < 0.3 \\ < 1 \\ < 0.4 \\ < 0.4 \\ < 0.4 \\ < 0.4 \\ 1 \pm 1 \\ < 0.7 \\ < 0.4 \\ < 1 \\ < 1 \\ < 1 \end{array}$	$Th-232$ 0.01 ± 0.02 0.09 ± 0.03 0.01 ± 0.02 0.01 ± 0.02 0.01 ± 0.02 0.01 ± 0.02 0.04 ± 0.03 <1.6 <0.01 <0.01 <0.02 0.02 ± 0.02 0.02 ± 0.02 0.03 ± 0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03 <0.03	<pre></pre>	<0.07 3.56 ± 0.20 ~
	Limit for Release of Soluble Radionuclide Into Unrestricted Areas	3000	3	2000	200	4000	200

TABLE 3 RADIONUCLIDE CONCENTRATIONS IN WATER (μ Ci/ml x 10⁻⁸)

a Refer to Figures 1-5 for locations of samples.
 b Errors given are 2σ due to counting statistics only.
 c Insufficient sample for alpha spectrometry analysis; gamma spectrometry performed for Th-232.

2.4 - 2 2.57 - .59 0.02.8 .7.9 .059 - 1037 ... (...24) 101 - .09 251.92 .71 2.06 - .07 12.05

TABLE 4

APPROXIMATE VOLUME FLOW RATES OF TWO MILE CREEK^a

Collection Site	Volume Flow Rate (1/sec)
6	70
7	160
8	250

^a As estimated at the time of sampling, June 24, 1981, based on cross sectional area and rate of surface movement.

TABLE 5

Ac-227 AND Pa-231 CONCENTRATIONS IN SELECTED SOIL AND SEDIMENT SAMPLES (pCi/g)

Collection Site ^a	Sample Description	Ac-227 (pCi/g)	Pa-231 (pCi/g)
11	Disposal Well - Subsurface Soil	14.25 ± 0.51 ^b	0.73 ± 0.57
13	Test Well Debris - Soil	2.10 ± 0.24	<0.29
15	Sanitary Sewer - Sediment	5.54 ± 0.64	0.95 ± 1.02
19	Storm Sewer - Sediment	14.29 ± 0.55	1.14 ± 0.88
21	Storm Sewer - Sediment	3.07 ± 0.37	<0.39

a Refer to Figures 3-5 for location of samples.

b Errors given are 2σ due to counting statistics only.

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- 2. Title 10, Code of Federal Regulations, U.S. Government Printing Office (1981), <u>Standards for Protection Against Radiation.</u>
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- 4. R.W. Legget, W.D. Cottrell, J. Burden, M.T. Ryan, <u>Radiological</u> <u>Survey of the Former Linde Uranium Refinery. Tonawanda. NY</u>, Oak Ridge National Laboratory, Oak Ridge, TN, DOE/EV-0005/5 (1978).
- 5. Title 40, Code of Federal Regulations, Part 192, <u>Proposed Cleanup</u> <u>Standards for Inactive Uranium Processing Sites. Invitation for</u> <u>Comment.</u> Federal Register (April 22, 1980).
- 6. E.Y. Shum, Nuclear Regulatory Commission, Division of Fuel Cycle and Material Safety, to Nuclear Fuel Services, Inc. <u>Evaluation</u> of NFS Proposed Remedial Actions and Other Alternatives of Soil <u>Contamination</u>, Private Communication (June, 1980).

APPENDIX A

INSTRUMENTATION AND ANALYTICAL PROCEDURES

Soil amd Sediment Samples

Soil and sediment samples were dried at 120° C, finely ground, mixed, and a portion placed in a one-liter Marinelli beaker. The quantity placed in each beaker was chosen to reproduce the calibrated counting geometry and ranged from 400 to 600 grams of soil. The beakers were capped but not sealed. Net soil weights were determined and the samples counted using a 23% Ge(Li) detector (Princeton Gamma Tech) coupled to a pulse height analyzer (Nuclear Data, model ND66).

The following energy peaks were used for determination of the radionuclides of concern:

Ra-226	0.609 MeV from Bi-214
U-235	0.185 MeV
U-23 8	1.001 MeV from Pa-234m (secular equilibrium assumed)
K-40	1.460 MeV
Cs-137	0.662 MeV
Pa-231	0.300 MeV
Ac-227	0.351 MeV from Bi-211 (secular equilibrium assumed)

The background plus Compton continuum was "stripped" by hand calculations from each of the photopeaks of interest prior to applying appropriate calibration and correction factors.

To evaluate the effect of possible radon losses on the equilibrium of Bi-214 with Ra-226, two soil samples were sealed in counting beakers. The relative photopeak intensities of various Ra-226 decay products were noted and compared to the relative intensities of capped, but unsealed, samples over a time period necessary for the Bi-214 peak intensity to stabilize. From this

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comparison it was determined that radon losses resulted in a 20% decrease in the Bi-214 concentration and that this condition reached an equilibrium state in the unsealed sample within approximately three days after sample preparation (drying, grinding, and placing into the beakers). Sufficient time to reach this equilibrium state was therefore allowed between sample preparation and analysis and, a correction factor was applied to all Ra-226 calculations (based on the Bi-214 photopeak intensity) to allow for the 20% decrease due to radon loss.

For U-235 analysis, contributions in the 0.185 MeV photopeak area from the 0.186 MeV Ra-226 gamma ray were subtracted. The ratio of the 0.186 MeV to 0.609 MeV peak intensities in a soil sample containing Ra-226, but no U-235, was determined and this ratio was multiplied by the intensity of the 0.609 MeV photopeak in each of the samples to determine the magnitude of this contribution.

Alpha spectrometry was performed for Th-230 and Th-232 by an outside analytical laboratory.

<u>Water Samples</u>

All water samples were rough filtered through Whatman No. 2 filter paper. Remaining suspended solids were removed by a subsequent filtration through 0.45 μ millipore filters. The filters, together with attached solids, were discarded; the filtrate was acidified by the addition of 20 ml of concentrated nitric acid.

Gamma spectrometry was performed on 500 ml samples in Marinelli beakers in the same manner as was the case for the soil.

The EPA Radon Emanation technique (EPA - 600/4 - 75 - 008, revised) was employed on all water samples for the evaluation of Ra-226 concentrations.

Alpha spectrometry analysis for Th-230 and Th-232 was performed by an outside analytical laboratory,

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APPENDIX B

EVALUATION OF RADIATION EXPOSURES ASSOCIATED WITH FORMER LIQUID EFFLUENT PATHWAYS AT LINDE AIR PRODUCTS, TONAWANDA, NEW YORK

Low concentrations of U-235, Ra-226, Th-232, U-238 and Th-230 are present in water, soil, and sediment samples collected in the vicinity of the Linde Air Products facility. These particular radionuclides were present in the uranium ores used at the Linde facility during the mid-1940s, however these same radionuclides are naturally occuring and present in water and soil throughout the world. The fission product Cs-137 noted on these samples is also present in the environment as a result of fallout from nuclear weapons tests.

Concentrations of these radionuclides in water and soil collected in the Linde facility property are also within the limits for off-site, general public use. Several sediment samples from sewers which existed during the uranium processing operations contained radionuclide concentrations above the limits for unrestricted areas. However, these sediments are present in limited quantities and in physically isolated locations. Furthermore, the lack of significant concentrations in water samples from these same locations indicates that the radionuclides are in an insoluble form.

Concentrations of the above radionuclides in the off-site effluent pathways formerly used by Linde Air Products are in the range of the normal environmental (baseline) levels in soil, sediment and water from the Tonawanda, New York, area. These concentrations are also well within the scientifically - based guidelines for unrestricted areas. It can therefore be concluded that no significant residues from the previous uranium processing operations at Linde Air Products are present in these effluent pathways and that any radiological exposure to the public resulting from these operations is negligible.

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