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Evaluation of the 1943-to-1946 Liquid Effluent Discharge From the Linde Air Products Company Ceramics Plant

December 1981

Prepared for

Office of Operational Safety
Assistant Secretary for Environmental Protection,
Safety, and Emergency Preparedness
U.S. DEPARTMENT OF ENERGY

Prepared by

Environment and Conservation Directorate
Eastern Technical Division
THE AEROSPACE CORPORATION
Germantown, Maryland

Contract No. DE-AC01-81EV10532

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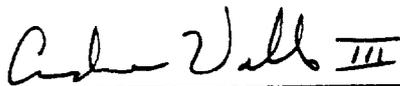
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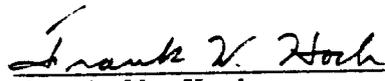
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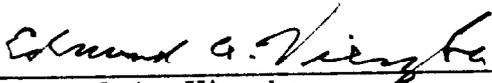
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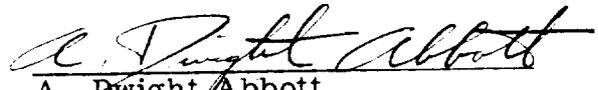
Andrew Wallo III
Project Engineer
Control Technology Section



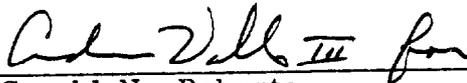
Frank W. Hoch
Systems Director
Environmental Controls
and Analysis Office



Edmund A. Vierzba
Control Technology Section



A. Dwight Abbott
Principal Director
Environment and Conservation
Directorate



Gerald N. Roberts
Technology Assessments Section

FOREWORD

This report presents an evaluation of the consequences of using onsite wells and sewers for disposing of liquid wastes from a uranium extraction process operated from 1943 to 1946 by the Linde Air Products Company, Tonawanda, New York. The process, known as Step I, was designed to extract uranium oxide (U_3O_8) from uranium ores supplied by the Manhattan Engineer District. The primary emphasis of the report is on the liquid effluents discharged from the process. In addition, the chemical process, its history, and operations are discussed; uranium and radium mass flow balances are calculated; and uranium and radium concentrations in residues are estimated. A brief description of the Linde Step I, II, and III processes (three independent production steps for converting uranium ore to uranium tetrafluoride) are also presented.

This report was prepared under contract to the Department of Energy (Office of Operational Safety) as part of its program to reassess the radiological condition of sites formerly used by the Manhattan Engineer District and Atomic Energy Commission in the development of nuclear energy. It is purely technical in nature and makes no attempt to detail or judge the policy decisions that resulted in the use of the wells. The estimates of effluent volumes and constituents made in this report are based on an analysis of historical data assembled by The Aerospace Corporation with assistance from Oak Ridge National Laboratory and the Oak Ridge Operations Office of the Department of Energy. The radiological data discussed in the report are from analyses completed by Oak Ridge Associated Universities, Argonne National Laboratory, and Oak Ridge National Laboratory. The Department of Energy's Office of Operational Safety, Environmental and Safety Engineering Division, provided support and guidance during preparation of the report and coordinated the activities of all contractors throughout the program.

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EVALUATION OF THE 1943-TO-1946 LIQUID EFFLUENT DISCHARGE FROM THE LINDE AIR PRODUCTS COMPANY CERAMICS PLANT

EXECUTIVE SUMMARY

BACKGROUND

In the early years of the Nation's nuclear energy program, the Linde Air Products Company (now Linde Air Products Division of Union Carbide Corporation) constructed and operated a uranium extraction facility for the Manhattan Engineer District. The plant received various uranium ores, concentrates, and residues and produced various uranium products, including uranium oxide (U_3O_8), uranium dioxide (UO_2), and uranium tetrafluoride (UF_4). There were two primary types of wastes that resulted from these operations: a solid, sludge-like material and a liquid effluent. Both were generated during the production of the U_3O_8 concentrate (Figure ES-1), which continued from mid-1943 to mid-1946.

The solid wastes resulted from the ore digestion and purification portions of the processes. These were, for the most part, the residue that remained after the uranium had been dissolved and removed from the ore. The amount of waste produced approximated the amount of the ore processed. The residues from domestic ores and concentrates were moved to the former Haist property, Tonawanda, New York, and residues from the foreign ore were stored at the former Lake Ontario Ordnance Works site. The history and radiological condition of these disposal and storage sites have been documented in a number of other reports.¹ The liquid waste is the primary subject of this study.

The effluent was the liquid that remained after the uranium initially dissolved from the ore was precipitated and filtered from the solution. It is estimated that over 500×10^6 l (about 130×10^6 gal) of the effluent were discharged over the course of the operation of the

¹ U.S. Department of Energy, "Radiological Survey of the Ashland Oil Company (Former Haist Property), Tonawanda, New York," DOE/EV-0005/4, Washington, D.C., May 1978, and "A Background Report for the Formerly Utilized Manhattan Engineer District/Atomic Energy Commission Sites Program," DOE/EV-0097A, Washington, D.C., September 1980. The Aerospace Corporation, "Background and Resurvey Recommendations for the Atomic Energy Commission Portion of the Lake Ontario Ordnance Works," ATR-82(7963-04)-1, Germantown, Maryland, to be published.

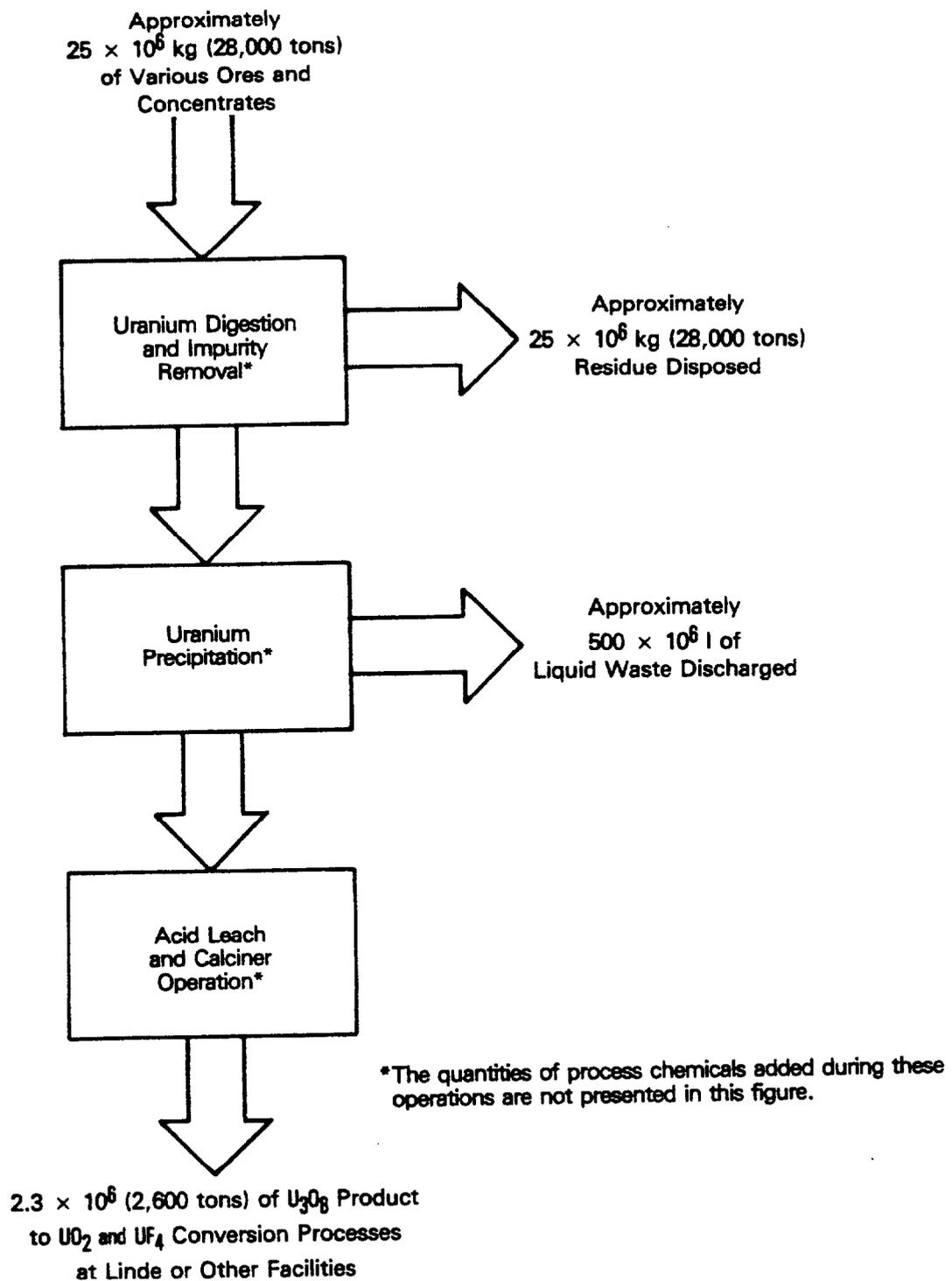


Figure ES-1. Simplified Schematic of the Linde Uranium Extraction Process and Approximate Amounts of Material

uranium refinery. Initially, the effluent was released into the sanitary sewer system. However, process changes completed early in 1944 that caused the pH of the effluent to increase made it unacceptable for the sanitary sewer system. As of April 1944, approximately 100×10^6 l (about 30×10^6 gal) of effluent were disposed of through the sanitary sewer system.

In April 1944, the company, with approval from the Manhattan Engineer District, began disposing of the wastes in onsite wells. The water from these wells was previously determined to be unacceptable for Linde's use due to its poor chemical quality. These wells would periodically plug and have to be cleaned or replaced. During these periods, the effluent was released into a storm sewer that flowed into Two Mile Creek, which in turn flowed to the Niagara River (Figure ES-2). New York State authorities were aware of these disposal practices and, due to the stream's already polluted condition, had no objection. It is estimated that slightly over 200×10^6 l (about 50×10^6 gal) were discharged into a total of seven 40m-deep wells, with an equal amount going into Two Mile Creek.

Uranium concentrations in the effluents discharged to the sanitary sewer averaged less than 8×10^{-5} $\mu\text{Ci/ml}$, which is below the current Nuclear Regulatory Commission (NRC) limit for natural uranium in water discharged to a sewer (1×10^{-3} $\mu\text{Ci/ml}$).² The concentration of uranium in wastes discharged to the wells and creek (about 1.5×10^{-5} $\mu\text{Ci/ml}$) was below the current Nuclear Regulatory Commission limit for natural uranium concentrations in waters discharged to unrestricted areas (3×10^{-5} $\mu\text{Ci/ml}$). Radium concentration in the effluent was maintained below 2.6×10^{-5} $\mu\text{Ci/ml}$ and was believed to average around 2.5×10^{-6} $\mu\text{Ci/ml}$. This level, giving credit for dilution, was below the current Nuclear Regulatory Commission limit for concentrations of radium in water released to a sewer; however, if a significant fraction of the radium was soluble,³ a dilution factor of 100 to 1000 would have

2 A microcurie (μCi) is one-millionth of a curie. A curie of any given radionuclide per unit volume of material is directly proportional to the amount of the radionuclide present in the material. For instance, 1 Ci of natural uranium is about 1.5 metric tons (about 1.6 tons) of natural uranium, and 1 Ci of radium-226 is about 1g or 0.035 oz of radium.

3 Radiation standards for allowable concentrations of radionuclides are given for soluble and insoluble fractions separately. Soluble radionuclides are those dissolved in the wastes; insolubles are those that are not dissolved but may be in the wastes as suspended solids. Standards for soluble fractions are lower or more restrictive because soluble elements are more easily taken up by the body. It is believed that the radium concentration presented here as average includes both soluble and insoluble fractions.

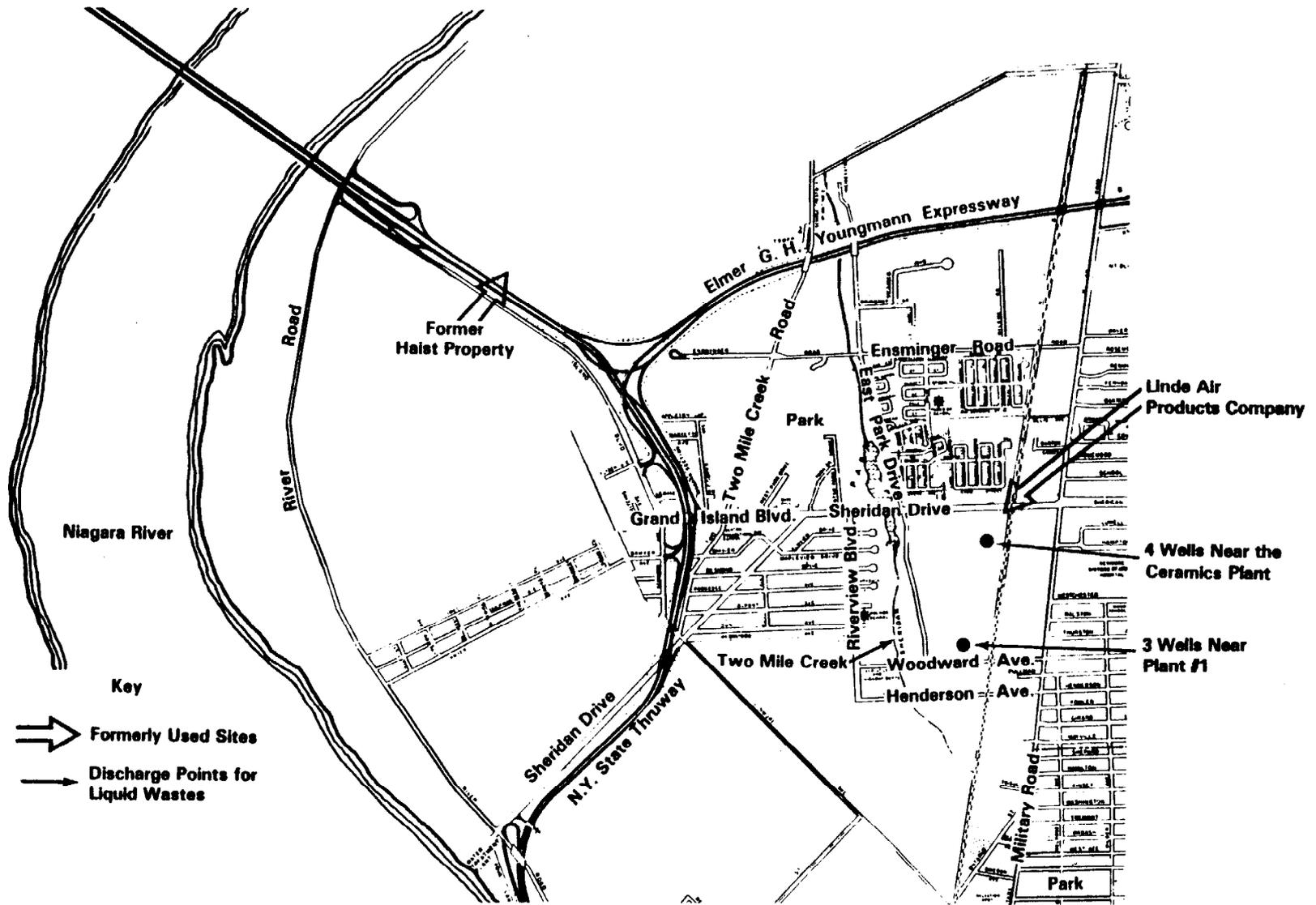


Figure ES-2. Location of the Linde Site, Disposal Wells, and Two Mile Creek

been necessary for the wastes discharged into the stream or wells to meet present-day limits.⁴

It is possible that these dilution factors were met, but it is also possible that present-day limits for radium were exceeded at least periodically. It should be noted that, except for the Linde action point (2.6×10^{-5} $\mu\text{Ci/ml}$), the radium concentration at which the company would not discharge the effluent, no criterion existed for radium in water during the time the refinery was operating. Furthermore, the chemical quality of both the creek and well water at the time of discharge precluded its use for many industrial and residential purposes such as drinking water. As a result, it is very unlikely that these discharge operations could have posed a radiological hazard to members of the general public. Furthermore, the low concentrations of radionuclides in the creek and wells during operation and the known use of the waters from termination of operations to the present time suggest that any residue remaining after operations ceased would have posed virtually no radiological hazard.

RADIOLOGICAL SURVEY AND PRESENT CONDITION

To verify that no hazard exists today and to determine if any remedial actions are warranted, the Department of Energy had numerous samples of soil, water, and sediment collected and analyzed. Oak Ridge Associated Universities, Argonne National Laboratory, and Oak Ridge National Laboratory performed these analyses on samples taken from areas used during the disposal operation, such as Two Mile Creek and the disposal wells. For comparison, samples were also collected in areas not involved with the disposal of the effluent. Samples were analyzed for various radioactive isotopes of radium, uranium, thorium, cesium, and lead. The data for these radionuclides are discussed in more detail in the body of this report, and the measured concentrations for uranium-238 and radium-226 at various locations are summarized in Table ES-1. Concentrations of all radionuclides in the water from Two Mile Creek at and downstream of the waste discharge point averaged from about 100 to 10,000 times lower than the appropriate

⁴ Code of Federal Regulations, Title 10, Part 20, indicates that discharge into a sewer is acceptable if insoluble and soluble radium concentrations in the sewer are less than 9×10^{-4} and 7×10^{-7} $\mu\text{Ci/ml}$, respectively, and discharge to an unrestricted area is acceptable if insoluble and soluble radium concentrations are 3×10^{-5} and 3×10^{-8} $\mu\text{Ci/ml}$, respectively. Credit can be given for dilution.

Nuclear Regulatory Commission limits⁵ for release of waters from a licensed facility to an unrestricted area. Sediment and soil samples collected in Two Mile Creek contained concentrations of radionuclides comparable to concentrations found in nearby creeks and in Two Mile Creek upstream of the Linde facility.

Table ES-1. Existing Concentrations of Radium and Uranium in Two Mile and Nearby Creeks

Sampling Location	Range and Average of Concentrations Measured	
	Radium-226	Uranium-238
Water Samples From Two Mile Creek at and Downstream of Linde	<0.01-0.3x10 ⁻⁸ μCi/ml	<150x10 ⁻⁸ μCi/ml
Water Samples Upstream of Linde and From Nearby Creeks in Tonawanda	0.01-0.04x10 ⁻⁸ μCi/ml	<210x10 ⁻⁸ μCi/ml
NRC Limits for Unrestricted Use*	3x10 ⁻⁸ μCi/ml	4000x10 ⁻⁸ μCi/ml
Soil Samples From the Banks of Two Mile Creek at and Downstream of Linde	0.2-0.9 pCi/g Average 0.6 pCi/g	<0.4-5.4 pCi/g Average 1.9 pCi/g
Soil Samples Upstream of Linde and in Other Creeks in Tonawanda	0.4-0.6 pCi/g Average 0.5 pCi/g	0.9-2.8 pCi/g Average 1.6 pCi/g
Sediment Samples From Two Mile Creek at and Downstream of Linde	0.4-0.8 pCi/g Average 0.6 pCi/g	<0.6-3.6 pCi/g Average ≤1.3 pCi/g
Sediment Samples Upstream of Linde and in Other Creeks in Tonawanda	0.6-0.7 pCi/g Average 0.7	0.8-4.3 pCi/g Average 2.0 pCi/g

* Code of Federal Regulations, Title 10, Part 20, Appendix B, Table II, Column 2.

An analysis of groundwater samples indicated that groundwater beneath the Linde facility contains slightly above-background concentrations of some of the radionuclides; however, concentrations in the

⁵ Code of Federal Regulations, Title 10, Part 20, Appendix B, Table II, Column 2.

groundwater were below the Nuclear Regulatory Commission limits (Table ES-2). Onsite soil and sediment samples and samples of material removed from test wells drilled near the disposal wells also contained uranium, radium, and thorium in excess of offsite baseline samples.

Table ES-2. Existing Radium-226 and Uranium-238 Concentrations in Groundwater

Sampling Location	Concentration x 10 ⁻⁸ μ Ci/ml of water			
	Radium-226		Uranium-238	
	Soluble	Insoluble	Soluble	Insoluble
Two Test Wells Drilled Near Disposal Wells	<0.1*	2.7	0.2**	320**
Old Gas Well on Linde Property	1.7	4.0	3.8**	76.3**
Industrial Well 2 km West of Linde Property	<0.004	0.2	<0.02**	<0.002**
Residential Well 2 km North of Linde Property	0.09		<53***	
NRC Limits for Unrestricted Use****	3	3000	4000	4000

* The value was less than (<) the detectable limit (in this case, 0.1) of the analytical procedure used. The actual value would range anywhere from 0 to the detectable limit.

** Argonne National Laboratory measured total uranium. The fraction of uranium-238 was calculated assuming the uranium isotopes are in equilibrium.

*** Analyses of the residential well waters were done by Oak Ridge Associated Universities; all other data presented were based on analyses by Argonne National Laboratory.

**** Code of Federal Regulations, Title 10, Part 20, Appendix B, Table II, Column 2.

No standards for radionuclides in soil applicable to this specific situation currently exist. The concentrations of the radionuclides in the soil and sediment are low and, under the conditions that exist at this site, any radiation exposures to workers or members of the public would be far below Nuclear Regulatory Commission radiation dose guidelines.

The survey data clearly demonstrated that the concentrations of radionuclides in groundwater were well below the levels established by the Nuclear Regulatory Commission as acceptable for release to an unrestricted area. Due to the very high natural mineral content of the groundwater, it is not acceptable as drinking water or for many other industrial or residential uses; hence, no significant pathway exists for exposure of the general public.

COMPARISON OF DOSE TO BACKGROUND RADIATION AND GUIDELINES

Radionuclides present at the Linde site are also naturally present in varying but small concentrations throughout our environment. Everyone is exposed to radiation from these naturally occurring radionuclides; such exposure is termed background exposure. Background exposure also includes exposure to cosmic radiation, to which individuals are continuously subjected. Background exposure is not dependent on man and is largely uncontrollable. An individual can reduce the level of background radiation he is receiving only by moving to a location with a lower level of background radiation. The typical person in the United States receives a radiation dose of about 100 mrem/yr from exposure to background radiation.

Scientifically based guidelines have been established for the protection of radiation workers and the general public from unacceptable increases in radiation dose received through the use of radioactive materials for scientific, industrial, or medical purposes. The limit established for the general public (500 mrem/yr for an individual) is much lower than the limit for radiation workers and is used internationally as well as by the U.S. Nuclear Regulatory Commission, Environmental Protection Agency, and Department of Energy. Because of the low levels of radioactive materials found and the limited use of the water, there is no significant pathway for exposure of the general public from contamination on the Linde site. However, to establish a maximum potential dose for the purpose of comparison, a worst-case exposure was calculated.

The case considered was that of an individual who obtains 100 percent of his vegetables from soil irrigated by waters containing concentrations of radionuclides equivalent to those measured in the most highly contaminated well. The increase in radiation dose to this individual would be less than 3 mrem/yr whole body dose equivalent. This increased dose would be less than 3 percent of the average background dose in the United States and less than 0.6 percent of the guideline established for the general public (Figure ES-3). This scenario is not very likely to occur. Furthermore, if it did occur, the concentration of radionuclides resulting from the effluent discharge in any well off the Linde site would be lower than the highest levels measured during these surveys. Therefore, 3 mrem/yr can be considered the worst possible exposure case and can be used to place an upper limit on any possible risk.

PRELIMINARY ESTIMATE OF RADIATION RISK

Exposure to radiation does not result in inevitable induction of cancer; rather, the result of exposure is an increased probability that an individual may contract cancer. For the purposes of radiation pro-

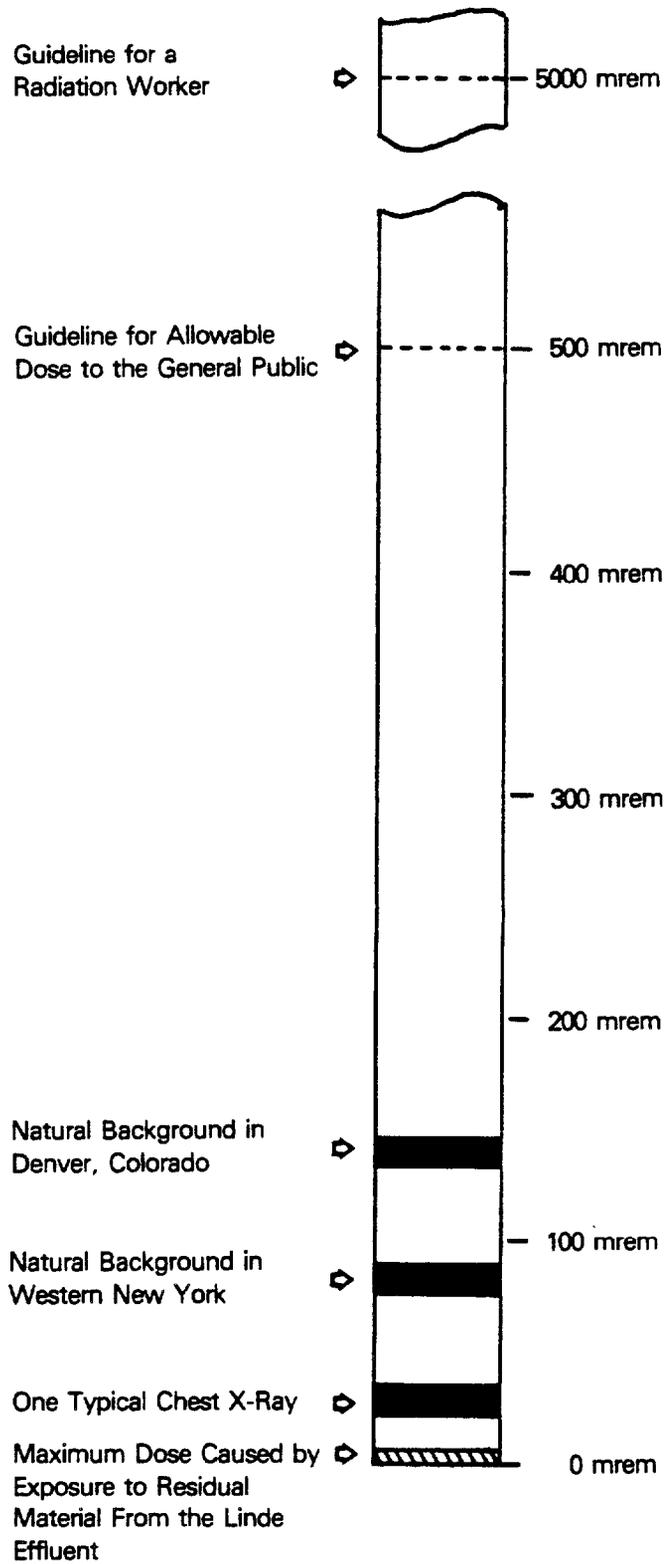


Figure ES-3. Comparison of Annual Dose From Linde Effluent Residue to Guideline and Background Doses

tection, exposure to even low levels of radiation is assumed to increase an individual's risk of contracting cancer. This risk can be affected by many variables, including age at the onset of exposure; variability in latency period (time between exposure and disease); genetic traits, personal habits, and general state of health; and previous or concurrent exposure to other cancer-causing agents. As a result of these many variables, no precise numerical value for a given individual's risk can be assigned. It is only with very large population groups that such a risk can be estimated with any certainty. However, to compare the effect of increased exposure to other risks, risk-estimating factors developed from data on large population groups are used to estimate the magnitude of the increased risk of radiation-induced cancer to an exposed individual.

The increased risk of contracting a fatal cancer for an individual who received 3 mrem/yr over his entire lifetime (70 yr) is estimated to be 0.002 percent (2 chances per 100,000 of eventually dying of cancer). For comparison, in 1977, 21.6 percent of all deaths (21,600 per 100,000) in New York State and Erie County, New York, were attributed to cancer. Hence, the risk of cancer associated with exposure to residual radiation caused by the discharge of the process effluent would be insignificant in comparison to normal risk.

SUMMARY

The uranium processing facility operated for the Manhattan Engineer District by the Linde Air Products Company discharged large volumes of effluent containing above-background concentrations of radionuclides. There is no indication that the discharge resulted in any radiation-associated hazards during or following the operation of the facility. Radiological surveys of the disposal pathways at the facility and vicinity properties did identify some above-background concentrations of uranium, radium, and thorium and their decay products. However, the concentrations are so low that any incremental radiation exposure to workers or the general public, as well as any added health risk, is negligible.

HISTORY OF LINDE OPERATIONS

The Linde Air Products Company operated, for the Manhattan Engineer District, a facility known as the Ceramics Plant. The plant performed three processes: in Step I, ores and occasionally residues¹ were processed to produce uranium oxide; in Step II, uranium oxide was converted to uranium dioxide; and in Step III, uranium dioxide was converted into uranium tetrafluoride. A brief description of the three processes is presented in Appendix A. This report is concerned solely with the Step I process because residues from Steps II and III were recycled, whereas Step I discharged large amounts of liquid and solid residue. Figure 1 is a conceptual flow diagram of the Step I process for African ore; the process varied slightly depending on the ore processed. Figure 2 is a similar diagram for a domestic ore.

Step I began shakedown operations in June/July 1943 and continued operations until mid-July 1946. Table 1 gives approximate production rates on a yearly basis over the life of the process. The data in the table were developed from estimates calculated on a monthly basis (Appendix B) using information obtained from various sources, including weekly progress reports. The estimates of effluents discharged and associated radioactive material losses were based on the same sources and monthly process estimates. A comparison of the uranium oxide production estimated from these sources with the reported Linde production from the Manhattan Engineer District files shows an agreement within 5 percent. As a result, the estimates of effluent discharges made in this report should be a fair representation of the actual Step I process discharges. Appendix B presents uranium and radium mass balance flow estimates of the amount of radioactive material lost in the effluent and residues for each 110 metric tons (100 tons) of uranium produced from African and domestic ores (Figures B-1, B-2, B-3).

ORES AND PRODUCTS FROM STEP I

The Step I process produced uranium oxide concentrates from seven different types of ore: four African ores (three low-grade pitchblendes and a torbernite) and three domestic ores. The African ores were unprocessed ores containing between 3 and 15 percent uranium oxide. These ores contained radium in secular equilibrium with uranium. The domestic ores, however, were generally tailings from vanadium

¹ Residues processed were from the Step II operation and other Manhattan Engineer District processes.

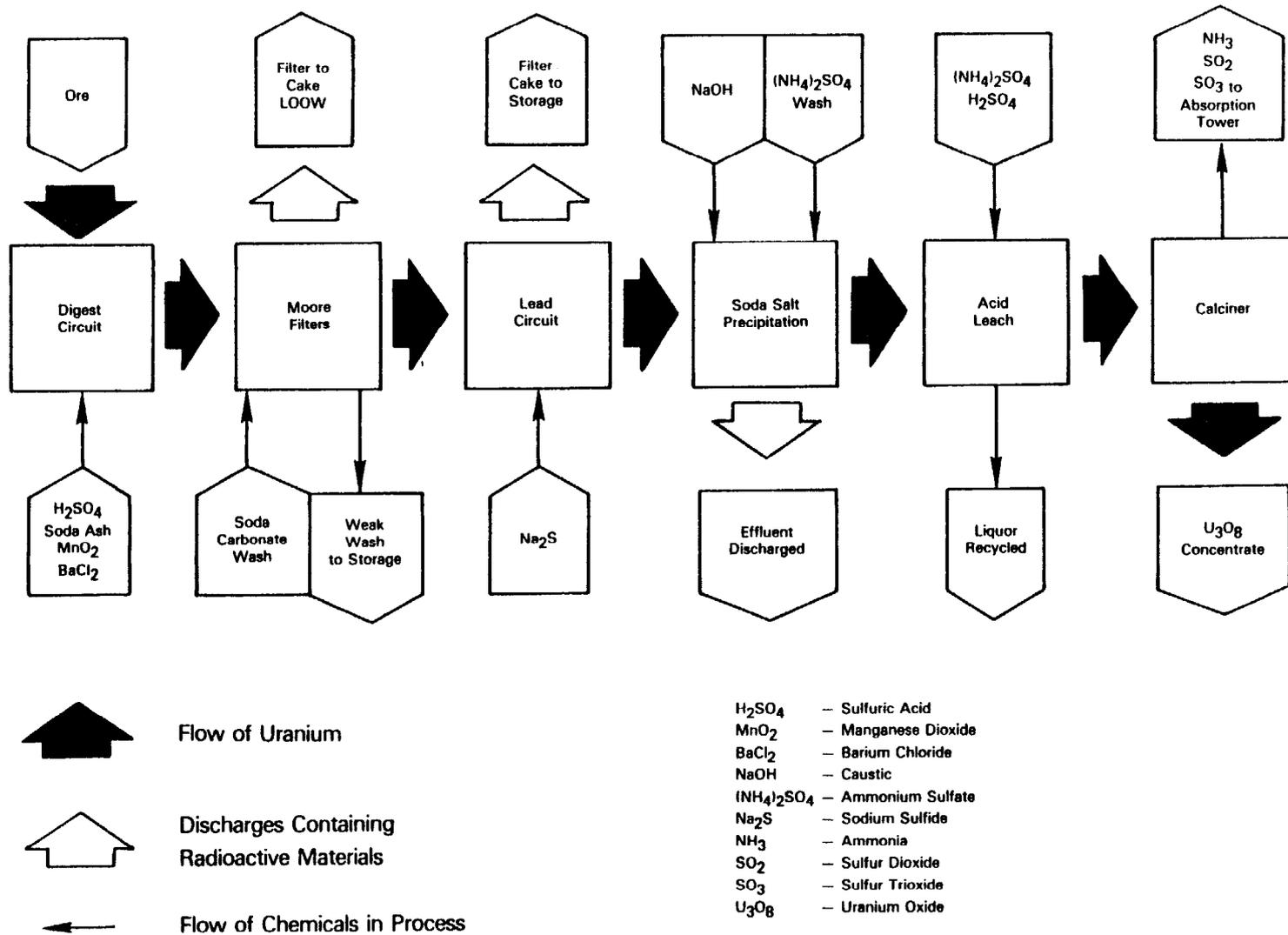
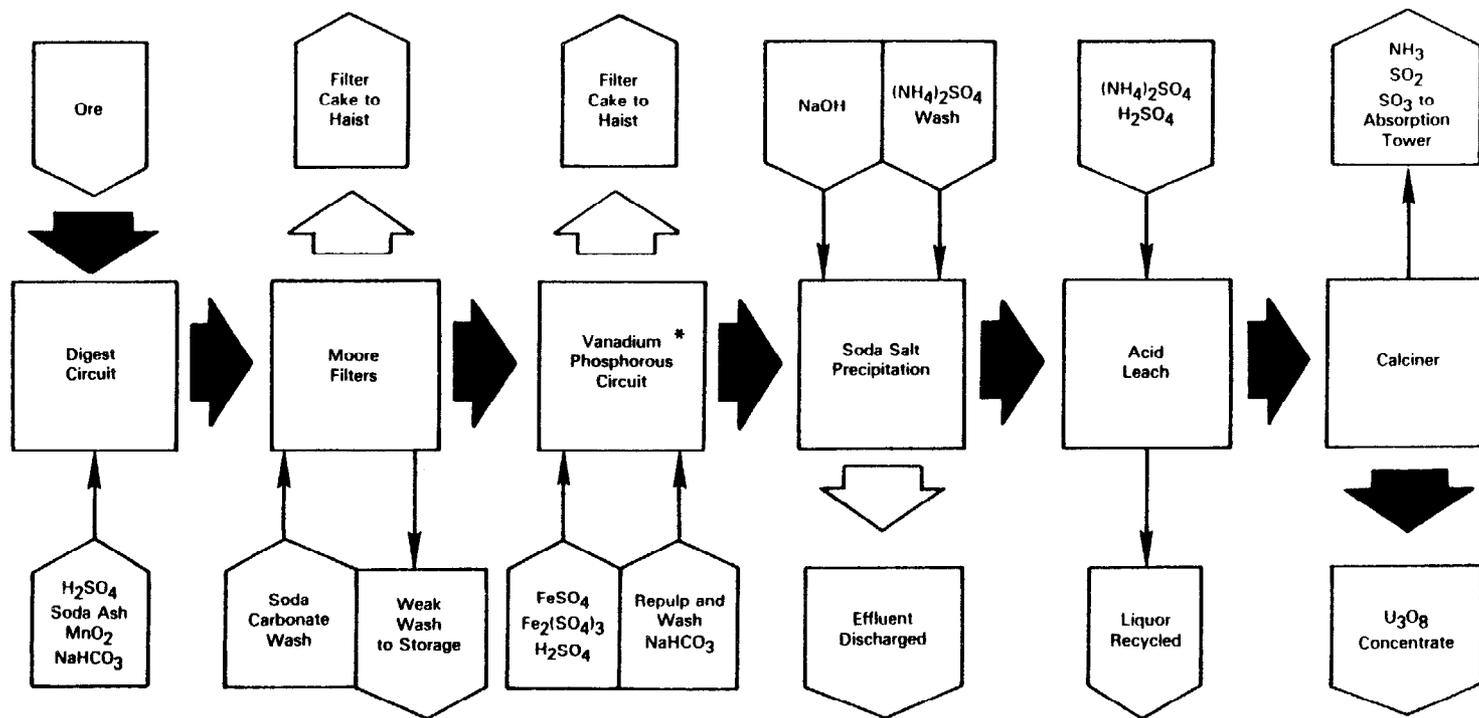
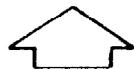


Figure 1. Sample Processing Flow Sheet (African Ore)



Flow of Uranium



Discharges Containing Radioactive Materials



Flow of Chemicals in Process

- | | | | |
|---|----------------------|---|-------------------------------|
| H ₂ SO ₄ | – Sulfuric Acid | Na ₂ CO ₃ | – Sodium Carbonate (soda ash) |
| MnO ₂ | – Manganese Dioxide | NH ₃ | – Ammonia |
| BaCl ₂ | – Barium Chloride | SO ₂ | – Sulfur Dioxide |
| NaOH | – Caustic | SO ₃ | – Sulfur Trioxide |
| (NH ₄) ₂ SO ₄ | – Ammonium Sulfate | U ₃ O ₈ | – Uranium Oxide |
| Na ₂ S | – Sodium Sulfide | FeSO ₄ | – Ferrous Sulfate |
| NaHCO ₃ | – Sodium Bicarbonate | Fe ₂ (SO ₄) ₃ | – Ferric Sulfate |

* Initially, vanadium was removed by adding lead sulfate, and the soda salt precipitation was an acid-caustic method that involved the addition of H₂SO₄ prior to the caustic; the revised procedure used the direct caustic method.

Figure 2. Sample Processing Flow Sheet (Domestic Ore Revised)*

Table 1. Uranium Oxide Production at Linde*

Year	Type of Ore	Ore Used in Metric Tons** (Tons)		Product in Metric Tons (Tons)		Waste Discharge	
						Residue	Effluent
1943	American Ore ($\approx 15\%$ U_3O_8) and Residue	1,000	(1,103)	171	(189)	to Haist Property	to sanitary sewer
	African Ore ($\approx 10\%$ U_3O_8)	450	(496)	42	(46)	to LOOW***	to sanitary sewer
	Subtotal	1,450	(1,599)	213	(235)	990-1,220 metric tons (1,091-1,345 tons)	$36.6-55.2 \times 10^6$ l ($10-15 \times 10^6$ gal)
1944	African Ore ($\approx 10\%$ U_3O_8)	9,550	(10,529)	944	(1,041)	to LOOW	to sanitary sewer until April and afterward to Linde wells or storm sewer
	American Ore ($\approx 14\%$ U_3O_8)	93	(103)	9	(10)	to Haist Property	
	Subtotal	9,643	(10,632)	953	(1,051)	9,184 metric tons (10,125 tons)	164.1×10^6 l (43×10^6 gal)
1945	American Ore ($\approx 14\%$ U_3O_8)	5,419	(5,974)	737	(813)	to Haist Property	to Linde wells or storm sewer
	Subtotal					5,219 metric tons (5,754 tons)	192.5×10^6 l (51×10^6 gal)
1946	American Ore ($\approx 14\%$ U_3O_8)	590	(650)	78	(86)	to Haist Property	to Linde wells or storm sewer
	African Ore ($\approx 4\%$ U_3O_8)	8,574	(9,453)	299	(330)	to LOOW	
	Subtotal	9,164	(10,103)	377	(416)	7,878-9,788 metric tons (8,685-10,791 tons)	107.6×10^6 l (28×10^8 gal)
	Total	25,676	(28,308)	2,280	(2,515)	23,271-25,411 metric tons (25,655-28,015 tons)	$500.8-519.4 \times 10^6$ l ($132-137 \times 10^6$ gal)

* Estimates of product and ore processed were made from data extracted from various sources and were calculated from estimates made for monthly production rates (given in Table B-1 in Appendix B.). Manhattan Engineer District history indicates that Linde processed a total of 2203 metric tons (2428 tons) of U_3O_8 , which is within 5 percent agreement with the value of calculated from the monthly data.

** One metric ton = 1000 kg or 2205 lb. The metric ton is about 1.1 short tons.

*** Lake Ontario Ordnance Works.

processing and were all preprocessed in the western states in order to concentrate the uranium prior to shipment to Linde. The major portion of the radium in these ores remained in the tailings left in the western states. These domestic ores, after initial processing, contained less than 20 percent uranium oxide. Linde also processed small amounts of sludges (containing up to 70 percent U_3O_8) resulting from the conversion of uranium oxide to uranium dioxide in the Step II process and from other Manhattan Engineer District operations. This material contained very little, if any, radium.

The product from Step I contained more than 97 percent uranium oxide and less than 3 percent insoluble acids and salts. Appendix C contains some estimates of the concentrations of the major constituents of the ores and product.

RESIDUES AND EFFLUENTS FROM STEP I

The principal residue from the Step I process was the Moore filter cake (Figures 1 and 2), a gelatinous cake remaining from the filtration of the uranium carbonate solutions. The Moore filter cake from the processed domestic ore was stored at the former Haist Property in Tonawanda, New York, and that from the foreign ore was shipped to the former Lake Ontario Ordnance Works (LOOW) in Lewiston, New York.

Because of the addition of sulfuric acid to dissolve the uranium and subsequent sodium carbonate neutralization, the cake consisted of insoluble oxides, sulfates, carbonates, and hydroxides, with hydrating and absorbed water that contained some soluble materials from the dissolving and sodium carbonate wash (5 percent Na_2CO_3 filter wash). Cake solids expected in large concentrations included silicon dioxide (some may have been added as filter aid); iron hydroxide; calcium hydroxide; calcium carbonate; aluminum hydroxide; lead sulfate (and perhaps carbonate), particularly from the African ores; barium sulfate and carbonate, where barium chloride was added to the African material to aid radium recovery; and magnesium hydroxide and carbonate (some $MgCO_3$ was used to enhance the filtration). In addition, the presence of some vanadium from the American concentrates and a variety of the minor metals in the African ores, such as nickel, cobalt, molybdenum, copper, and manganese (added later as an oxidant to increase dissolution), would be expected to some extent in the cake as hydroxides and carbonates. Complicated chemical reactions (difficult to predict) would have occurred because of hydration and multiple salt formation coupled with complexing. Some variation in chemistry would also have resulted from the differing feeds.

Other residue (of smaller volume) included the lead, phosphate, and vanadium cakes. The lead cake was a precipitate (from African ores, Figure 1) that resulted from the addition of sodium sulfide to the uranium carbonate solution to force precipitation of lead as lead sulfide. Other metals in the solution, such as molybdenum, iron,

cobalt, nickel, and copper, would also have been present in the cake, along with some small amounts of radium. Because it contained some radium and contracts for this ore required that the radium be accounted for, this material was generally barrelled.

The phosphate cake was a similar cake that resulted from the precipitation of phosphorous and lead (during the processing of 3-percent pitchblende ores) by the addition of sodium sulfide and ferric sulfate. Cobalt, nickel, and molybdenum compounds and small amounts of radium were present in the cake in addition to the phosphate.

The vanadium cake (domestic ore processing, Figure 2) was produced from the addition of lead sulfate to precipitate the vanadium as lead vanadate. Liquids (containing the uranium) from the precipitation went to the lead removal tanks, and the slurry was transferred to the lead recovery tanks before disposal. The process was revised in 1945, when ferrous and ferric sulfate were added to the domestic ore solutions to remove the vanadium and phosphorous. These wastes were stored at the Haist property.

The principal liquid effluent was from the filtration of the sodium diuranate cake (Figures 1 and 2), which followed the addition of caustic (sodium hydroxide) to the uranium carbonate solution. The resulting effluent had a high pH and was discharged to the sanitary sewer, storm sewer, and onsite wells over various periods during the plant's operation.

Some additional data regarding the ore, solid waste, and product from the Linde refinery are presented in Appendix C. The remainder of this report will deal primarily with the liquid effluents and the radioactive content of this filtrate.

LIQUID EFFLUENTS FROM THE STEP I PROCESS

During initial operations, uranium was precipitated from solution using a procedure that involved adding sulfuric acid to the uranium tricarbonate-rich solution and heating it to drive off carbon dioxide; this was followed by adding relatively small amounts of caustic to cause the precipitation. The effluent from this procedure had a pH that allowed its disposal into the sanitary sewer. This method of precipitation was abandoned at the end of 1943, however, because it was relatively slow and allowed more molybdenum and other impurities to contaminate the product than the direct caustic method of precipitation (Anderson, 1944). Linde developed the direct caustic method, which resulted in a better product in much less time. The method was essentially the brute-force removal of the uranium through the direct addition of caustic to the pregnant solution, driving the pH to levels as high as 11.5. This forced the uranium to precipitate as a diuranate, despite the presence of the carbonate.

One drawback to this method was that the effluent had a high pH and was no longer acceptable for direct disposal into the sanitary sewer system (Holmes, 1944a). As an alternative, two options considered were the use of disposal wells or discharge into Two Mile Creek. Although the discharge into the creek was approved by the State of New York, a decision was made to use disposal wells wherever possible and to rely on the Two Mile Creek option as a last resort. The effluent disposal wells (Figure 3) were approximately 40m (150 ft) deep and passed through a clay formation, into a gravel and sand layer and a variegated carbonate formation, possibly a mixture of magnesite and dolomite or limestone. Well logs for three of the disposal wells are presented in Appendix D. The groundwater in a section of the carbonate formation was identified as saltwater (Linde, 1949), and the water from the particular aquifer involved was found unacceptable for Linde Air Product Company's use. It was believed by the company to have been contaminated prior to 1944 and before the discharge of any Step I effluent into it (Holmes, 1944b). However, groundwaters from these formations are generally very high in dissolved minerals and, as a result, are unacceptable for many industrial uses. It may have been this natural mineral content that made these waters appear contaminated.

Two Mile Creek flows through the Linde facility and a park, where it is dammed to create a pond, and then into the Niagara River. The storm sewer discharged into the creek via a storm drainage ditch that entered the creek downstream of the dam (Figure 3). One memorandum (Ferry, 1944) suggests that the creek may have diluted the effluent 10

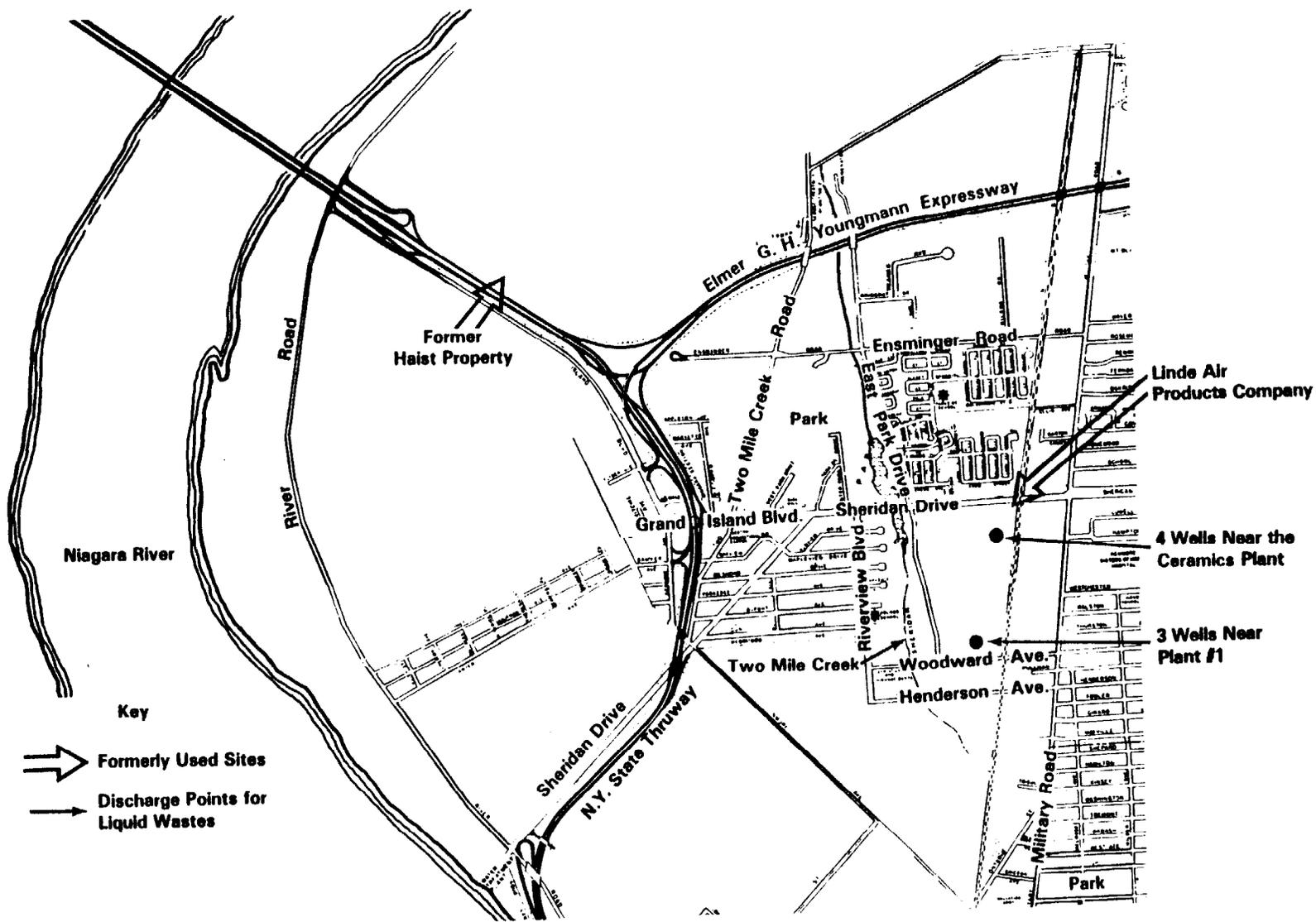


Figure 3. Linde Processing Facility and Discharge Points for Effluents

to 1; however, analyses of pH data² from other memoranda suggest that the creek may have had a flow rate up to 100 times that of the effluent discharge rate. Recent estimates of creek flow rates in the summer (see Present Condition and Survey Results) suggest that, at a minimum, creek flow rates would have been 15 to 40 times the average effluent discharge rates; the flow rates in the creek were much greater in the 1940s because industrial operations discharged plant water into the creek.

BASIC CHARACTERISTICS OF THE FILTRATE

The filtrate discharged to the sewers or wells was a high-pH solution (above pH 10)³ consisting mainly of ions from excess sodium sulfate, sodium carbonate, and sodium hydroxide. In addition, some chloride ions from the barium chlorides added to enhance radium recovery would also have been present, along with a small amount of a variety of complex anions of the many minor elements such as vanadium, nickel, and cobalt. (Appendix C lists the constituents of the ores.) Ammonium sulfate from the wash of the uranium precipitate would be expected to react rapidly with the caustic and release some ammonia. This was probably the cause of incidents in which the pump house operators were bothered by ammonia emissions from the wells located in the pump house (Cullen, 1945).

This complex solution would also contain small quantities of uranium and radium. At the low concentration found in these complex effluents, it is difficult to project which uranium and radium compounds would be favored and what their solubility would be. The uranium and radium would be present in solution as well as in colloidal form, and the relative amount of each is difficult to assess. The impact of this is not significant for uranium because standards for insoluble and soluble uranium are the same. However, standards for soluble and insoluble radium differ by a factor of 1000. It is believed that the analytical techniques used at that time would not have differentiated between the soluble and insoluble fractions; hence, the concentrations of uranium and radium in the effluents (based on the techniques used) would be total uranium and radium. An analysis of the solubility of various radium compounds suggests that a significant portion of the radium and probably the uranium in the effluent would be soluble.

² The average pH of the creek, measured over an 8-day period in March 1946, was about 8.3 upstream from the storm sewer discharge and 10.3 downstream from the discharge (Rehm, 1946).

³ Filtrate discharged between June 1943 and December 1943 was probably much closer to pH 7 (neutral).

VOLUME OF EFFLUENTS

As indicated previously, the liquid waste from the Step I process, the filtrate from the precipitation of the sodium diuranate which followed the addition of caustic soda, sodium hydroxide (Figure 1), was initially discharged into the sanitary sewer system. It appears that Linde began disposing of the effluents in onsite wells during or after April 1944 (Holmes, 1944) and that, from 1944 to 1946, three wells located in the area of Plant No. 1 and four wells located near the Ceramics Plant were used during various periods for this purpose. From time to time, the wells would plug, overflow, and have to be cleaned. During these periods, the effluents would be diverted to a storm sewer that connected with the Niagara River through Two Mile Creek. Based on information in progress reports and various operating memoranda, it is estimated that liquid waste volumes generated by the process during the period the wells were in use were as follows:

April to December 1944	121 x 10 ⁶ l (32 x 10 ⁶ gal)
January to December 1945	193 x 10 ⁶ l (51 x 10 ⁶ gal)
January to July 1946	<u>108 x 10⁶ l (28 x 10⁶ gal)</u>
Total	422 x 10 ⁶ l (111 x 10 ⁶ gal)

A 1948 Linde memorandum indicates that the volume of material pumped down the wells was about 140 x 10⁶ l (37 x 10⁶ gal) (Kent, 1948) and that it was dumped over the period from late February 1945 to July 1946. If the dates in the memorandum are correct, the volume given does not include the amount of material discharged into the wells in 1944 and early 1945. Based on the estimates of liquid effluent from ore processing from 1945 and 1946, it appears that about 50 percent of the effluent was dumped into the wells and the remainder into the storm sewer. Assuming that a similar dumping ratio existed in 1944 and early 1945, it appears that an additional 70 x 10⁶ l (18 x 10⁶ gal) may have been disposed of in the wells. It was therefore assumed that, during the period from April 1944 to July 1946, about 210 x 10⁶ l (55 x 10⁶ gal) of waste were disposed of in the wells and the remainder in the storm sewer to Two Mile Creek. All effluents prior to April 1944 (80 to 100 x 10⁶ l or 20 to 30 x 10⁶ gal) are assumed to have been discharged to the sanitary sewer.

URANIUM CONCENTRATION IN THE EFFLUENTS

Uranium losses in the effluents were monitored by Linde. From mid-1944 until 1946, the losses were reported on a regular basis in the weekly progress reports and, prior to that period, in memoranda or occasional progress reports. Reporting methods differed and, depending on the author, were reported as percent of uranium lost, pounds of uranium lost per batch or day, percent of uranium in effluent, and/or grams of uranium per 100 ml of effluent. The

concentration of uranium in the effluent or the percent of uranium lost varied depending on extraction efficiency; production rate (wash rates, filtering rates); and, to some extent, the type of ore processed.

During 1943 and the first two months of 1944, uranium extraction efficiencies generally ranged around 93 to 94 percent. Through the remainder of 1944, efficiencies generally exceeded a 96-percent uranium recovery rate and occasionally were as high as 98 percent. Extraction efficiencies over 1945 averaged about 98 percent and were somewhat lower in 1946, probably due to the lower grade ores being processed.

Uranium losses in the effluents in 1943 (during the lower extraction efficiency period) appear to be on the order of 2 to 3 percent of the uranium in the ore. This material was lost to the sewer system. In 1944, however, the data indicate that losses were generally below 1 percent of the total uranium, and the available progress reports indicate that later losses were maintained below 0.5 percent of the uranium in the ore.

The weekly averages of uranium oxide concentrations in the effluents analyzed from April 1944 to July 1946 (from progress reports) ranged between 0.011 and 0.064 gram of uranium oxide per liter of effluent, with the average being about 0.026 gram per liter (g/l). This would imply that the process lost an average of about 26 kg of uranium oxide per million liters or 220 lb of uranium oxide per million gallons of effluent during the period when the wells and storm sewer were being used.⁴ Concentrations of uranium oxide in the effluent during the period when the sanitary sewer was used for disposal of the effluent were somewhat higher. It is estimated that the concentrations averaged 0.15 g/l in 1943 and 0.03 g/l during the first 3 months of 1944, or about 1200 and 250 lb of uranium oxide per million gallons, respectively.

Assuming these loss rates and from 210×10^6 l (55×10^6 gal) of effluent disposed of in the wells, about 5.4×10^3 kg (12,000 lb) of uranium oxide (about 3 Ci of natural uranium⁵) were discharged to the wells. The remainder of the process effluents discharged to the storm sewer during this period, about 212×10^6 l (56×10^6 gal), would have contained about 5.6×10^3 kg (12,000 lb) of uranium oxide (about 3 Ci of natural uranium). Therefore, based on the available data, the total uranium oxide contained in the effluent released from April 1944 to July 1946 was about 11×10^3 kg (24,000 lb), or about 6 Ci of natural uranium.

⁴ Average uranium concentration (g/l) x 3.785 l/gal x 2.2×10^{-3} lb/g x 10^6 gal/ 10^6 gal.

⁵ Based on 6.77×10^{-7} Ci/g of natural uranium.

For comparison, the amount of uranium oxide discharged can also be estimated by assuming that Linde maintained uranium losses in the effluents to about 0.5 percent of the total uranium present and that 1812 metric tons (1990 tons) of uranium oxide (at about 97-percent extraction efficiency) were produced from April 1944 to July 1946. These data would also imply that about 9.3 metric tons (about 10 tons) of uranium oxide, or about 5 Ci of natural uranium, were lost in the effluents, which is in excellent agreement with the values estimated by using the effluent concentration data.

The available records contain little information on the uranium concentration in the effluents discharged to the sewer system during the period prior to the use of the waste wells. Anderson (1944) suggests that the effluents prior to 1944 contained 0.0001 to 0.032 percent uranium oxide by weight and averaged 0.015 percent (equivalent to 0.15 g/l⁶). Using the data in the progress reports, it is estimated that uranium oxide concentrations in the effluent during the first 3 months of 1944 averaged 0.03 g/l. Using these data and assuming that the discharge from July 1943 to April 1944 was between 80 and 100 x 10⁶ l (20 to 30 x 10⁶ gal)⁷, the uranium discharged to the sewers over that period ranged from 6.8 to 9.6 metric tons (7.5 to 10.5 tons) of uranium oxide, or from 4 to 5 Ci of natural uranium.

For comparison, some references (Van Horn, 1944; Thomas, 1944) indicated that as much as 3 percent⁸ and as little as 0.05 percent of the uranium oxide could have been lost in the effluent during some of the initial operations. This suggests that a maximum of 15 metric tons (about 17 tons) of uranium oxide, or less than 9 Ci of natural uranium, could have been discharged through the sanitary sewers during this period. The range estimated in the previous paragraph is in agreement with this maximum value.

The Code of Federal Regulations, Title 10, Part 20 (current Nuclear Regulatory Commission (NRC) regulations), states that the release of an effluent to a sewer is acceptable if the quantity, when diluted by the average daily quantity of sewage released into the sewer

6 This level is about six times higher than the average (0.026g U₃O₈/liter effluent) shown in the 1944 to 1946 progress reports.

7 These estimates are consistent with two 1944 memoranda (Anderson, 1944; Holmes, 1945) that indicated discharge rates of from 100,000 to 150,00 gal/day.

8 Losses were generally below this level (Anderson, 1944). They probably averaged below 2 percent of the total uranium oxide processed.

by the licensee, will result in an average concentration of less than 1×10^{-3} μCi of uranium per milliliter of solution (approximately 1.5 g/l). The regulations further state that a licensee shall not possess, use, or transfer licensed material so as to release to an unrestricted area radioactive material in concentrations that exceed 3×10^{-5} μCi of uranium per milliliter of solution (approximately 4.5×10^{-2} g/l). The regulations indicate that the release of higher concentrations could be approved if the licensee can demonstrate that the effluent will be diluted sufficiently after release to ensure that no individual will be exposed to concentrations above the given limit.

The average uranium concentrations in effluents from the Linde plant from April 1944 to July 1946 (those disposed of in the wells and storm sewer) were below both of the above limits. Even the maximum concentrations, based on weekly averages from the Linde progress reports, would have met these limits if credit was given for some dilution.⁹ The uranium concentrations for effluents released to the sanitary sewer during initial operations (June 1943 to April 1944) may have at times been in excess of the limits for release to an unrestricted area; they were, however, within the limits allowed for uranium concentrations in effluents discharged to a sanitary sewer (Table 2).

Table 2. Uranium Concentrations in Effluents at Time of Release

Description of Release	Concentrations in Step I Effluent		NRC* Limit ($\mu\text{Ci}/\text{ml}$)
	Range ($\mu\text{Ci}/\text{ml}$)	Average ($\mu\text{Ci}/\text{ml}$)	
Effluent Discharged to the Sanitary Sewer	$6 \times 10^{-7} - 1.84 \times 10^{-4}$	$\leq 8.1 \times 10^{-5}$	1×10^{-3}
Effluent Discharged to the Wells or Storm Sewer	$6.3 \times 10^{-6} - 3.7 \times 10^{-5}$	1.5×10^{-5}	3×10^{-5}

* Code of Federal Regulations, Title 10, Part 20, Appendix B, Table II, Column 1 indicates that the effluent must be less than or diluted to less than these values.

⁹ The maximum concentration found need only have been diluted by less than one-third to meet the current limits. It is highly unlikely that any of the effluents would have gotten into an accessible unrestricted area without being diluted by many times this amount.

RADIUM CONCENTRATION IN THE EFFLUENTS

Only limited data are available regarding the radium concentration in the effluents. Records indicate that consideration was given at one time to the use of a continuous monitoring method¹⁰ (Thomas, 1945) for monitoring the uranium lost in the effluent; it was felt, however, that the amount of radium present in the effluent from one type of ore (presumably African L-30 ore) would interfere with the method, although a second type of ore (American) would not. The American ores were generally concentrated in the western states prior to their shipment to Linde and, as a result, had a low radium-to-uranium ratio (Appendix B). The African ores (L-30, L-50, and R-10), however, generally had the radium and uranium in equilibrium. It is therefore assumed in this evaluation that the radium concentration in the effluent from the African ore processing would represent the worst of the two cases.

Some estimates of the maximum amount of radium discharged during the processing of the L-30 and L-50 ores can be made, based on the fact that contracts with African Metals called for the return of at least 95 percent of the radium in the processed ore. Actual processing operations supposedly held the losses to less than 3 percent (97 percent of the radium remained in the residues). Assuming a total of 986 metric tons or about 1080 tons of U_3O_8 produced from the L-30 and L-50 ores and a uranium extraction efficiency of about 97 percent, there were 862 metric tons or about 950 tons of uranium, or less than 595 Ci of natural¹¹ uranium (about 290 Ci of uranium-238), in the ore. This would imply about 290 Ci of radium-226 (in equilibrium with uranium-238¹²) and maximum effluent losses amounting to 8.5 Ci of radium-226. A similar analysis for the R-10 ore, but assuming a 95-percent extraction efficiency, would suggest that a maximum of 2.7 Ci of radium was lost during the processing of this ore.

Using effluent volumes calculated from data in the progress reports, it is estimated that from 169 to 173 x 10⁶ l (about 45 x 10⁶ gal) of effluent were released during L-30 and L-50 processing and about 77 x 10⁶ l (about 20 x 10⁶ gal) during R-10 processing. This would imply that the maximum concentration of radium in the effluent is about 4.6 x

10 This particular method involved the use of a Geiger tube and was not employed due to the obvious lack of sufficient sensitivity and background interference.

11 Assumes 6.77×10^{-7} Ci/g of natural uranium.

12 Assumes that 99.27 percent of the natural uranium is uranium-238 and that uranium-238 is 3.33×10^{-7} Ci/g.

10^{-5} $\mu\text{Ci/ml}$. This value represents a maximum radium content in the uranium carbonate solutions, based on the amount of radium that was not precipitated on the Moore filters following the digest circuit. Of the radium not precipitated at this stage, a portion probably precipitated during the liquor purification (lead, vanadium, and/or phosphorus removal step); however, the largest fraction of the remaining radium would be expected to have precipitated with the uranium after the addition of the caustic during the soda salt precipitation step.¹³ The remainder would be discharged with the effluents.

A letter from the University of Rochester (Bale, 1945) to the Manhattan Engineer District suggested that the radium content of the filtrates from the processing of R-10 ore was "on the order of 0.25×10^{-8} grams/liter" (2.5×10^{-6} $\mu\text{Ci/ml}$).¹⁴ This value is less than 1/18 times the maximum value estimated in the analysis above, suggesting, as expected, that much of the radium escaping the Moore filters may have been precipitated during the liquor purification or the uranium precipitation and not discharged to the wells or storm sewer. The implications are that the radium released in the effluents from the African ore may be on the order of 0.6 Ci rather than 11.2 Ci.

Some Linde progress reports indicated that an "action point" of 2.6×10^{-8} g/l or 2.6×10^{-5} $\mu\text{Ci/ml}$ for radium concentrations in the effluent was used to determine if the liquid waste could be discharged. This figure is in order with the maximum value calculated above and, as would be expected, is greater than the University of Rochester's estimated concentration.

The Code of Federal Regulations, Title 10, Part 20 (current Nuclear Regulatory Commission regulations), states that the release of an effluent to a sewer is acceptable if the quantity, when diluted by the average daily quantity of sewage released to the sewer by the licensee, will result in an average concentration of less than 7×10^{-7} μCi of soluble radium-226 per milliliter of solution and 9×10^{-4} μCi of insoluble radium-226 per milliliter of solution. It is further stated that a licensee shall not possess, use, or transfer licensed material so as to release to an unrestricted area radioactive material in concentrations that exceed 3×10^{-8} μCi of soluble radium-226 per milliliter of solution or 3×10^{-5} μCi of insoluble radium-226 per milliliter. The regulations also indicate that the release of higher

¹³ Merrit (1971) indicated that in an alkaline circuit, nearly all the radium dissolved in the pregnant solution is precipitated with the product.

¹⁴ This assumes that 1 gram of radium-226 equals 1 curie. The actual specific activity for radium-226 is about 0.98 Ci/g.

concentrations could be approved if the licensee can demonstrate that the effluent will be diluted sufficiently after release to ensure that no individual will be exposed to concentrations above the given limits.

Both the University of Rochester's order-of-magnitude estimate and the maximum concentration levels calculated from available data suggest the effluents would meet the standard for insoluble radium if some credit is given for dilution in the creek or aquifer. However, if a significant portion of the radium was soluble, a dilution factor on the order of 1000 for maximum possible radium levels or 100 for the assumed average radium levels in the effluent would be required to ensure that the current standards were met at the time of release.

PRESENT CONDITION AND SURVEY RESULTS

This section reviews the survey and analyses of the effluent disposal pathways completed by Department of Energy survey contractors and presents an evaluation of the results in terms of potential for exposure of the general public. The survey results described in this section pertain to the liquid effluent disposal site pathways. The radiological conditions of the sites used for storing the solid residue and the Linde facility (grounds and buildings) are discussed in other reports (ORNL, 1978; Aerospace, 1981; DOE, 1978).

TWO MILE CREEK

Two Mile Creek flows north along the west side of East Park Drive. It flows into a small pond located on both sides of Sheridan Drive and then on to the Niagara River. On June 24, 1981, measurements of flow rate downstream of the pond ranged from 70 to 250 l/s. The flow is probably less now than it was years ago because the creek was previously used for disposal of industrial (cooling and such) waters from facilities upstream of the Linde facility. The radiological condition of the creek was evaluated in two separate surveys, one conducted in 1976 by Oak Ridge National Laboratory (ORNL, 1978), and one conducted in 1981 by Oak Ridge Associated Universities (ORAU, 1981). Concentrations of radionuclides measured in the creek were all below Nuclear Regulatory Commission limits¹⁵ for discharge to an unrestricted area. The initial water samples taken in 1976 suggested that water downstream from the Linde outfall may have had slightly elevated uranium and radium concentrations; however, the 1981 survey did not identify any significant differences in either uranium or radium concentrations upstream or downstream of the Linde discharge point. Furthermore, all the uranium and radium concentrations measured in Two Mile Creek were comparable to those measured in samples from nearby streams in the Tonawanda area. Thorium-230 and -232 concentrations in the water samples of the creek were found well below applicable limits¹⁵ in both surveys. Concentrations in only two of the samples exceeded the range of concentrations found in background samples taken in the area, and the highest concentration was less than 1.8 percent of the Nuclear Regulatory Commission limit. Table 3 lists the water analyses results of the 1976 and 1981 surveys.

¹⁵ Code of Federal Regulations, Title 10, Part 20.

Table 3. Concentrations of Radionuclides in Stream Waters

Results From 1976 Survey (ORNL, 1978)							
Sampling Location	Number*	Concentration in $\mu\text{Ci}/\text{ml} \times 10^{-8}$					
		Radium-226	Uranium-234	Uranium-238	Thorium-230		
<u>Two Mile Creek</u>							
Southern End of the Pond	2	<0.018	0.054	0.048	<0.0005		
Northern End of Pond	4	0.333	0.157	0.148	<0.013		
300m Downstream of the Pond	6	0.027	0.117	0.123	<0.009		
Results From 1981 Survey (ORAU, 1981)							
Sampling Location	Number*	Concentration in $\mu\text{Ci}/\text{ml} \times 10^{-8}$					
		Radium-226	Uranium-234	Uranium-238	Thorium-230	Uranium-235	Thorium-232
<u>Two Mile Creek</u>							
1000m Upstream of the Linde Outfall	1	0.037		<59	<0.07	<0.5	0.01
Southern End of the Pond	2	0.031		<51	3.56	<0.4	0.09
In Pond South of Sheridan Drive	3	0.033		86	<0.05	<0.4	0.01
Northern End of the Pond	4	0.056		<150	0.01	2	0.01
In Creek Just Downstream of the Pond	5	<0.007		<51	0.5	<0.4	0.01
500m Downstream of the Pond	7	0.017		<53	0.03	<0.4	0.01
1500m Downstream of the Pond	8	0.083		<55	0.11	1	0.04
Other Creeks in the Area		0.008-0.034		<58-<214	<0.06-0.07	<0.4-2	<0.01-<0.03
MRC Limit for Unrestricted Use**		3	3000	4000	200	3000	200

* Sampling location number indicates the location on Figure 4.

** Code of Federal Regulations, Title 10, Part 20, Appendix B, Table II, Column 2.

Soil samples were also collected from the banks of Two Mile Creek. Table 4 lists the results of these analyses along with the range of values measured in samples from nearby creeks. In general, the soil of Two Mile Creek contains radionuclides in concentrations similar to the concentrations found in the baseline samples from other creeks and in Two Mile Creek upstream of the Linde facility. Sediment samples from Two Mile Creek (Table 5) also contain concentrations of radionuclides similar to the ranges found in the baseline samples.

LINDE WELLS AND GROUNDWATER

The seven Linde disposal wells were located in two general areas: Three were in the area of Plant No. 1 near the present Building 8, and the remaining wells were in the area of the former Ceramics Plant near the present Buildings 30 and 38 (Figure 5). To characterize the condition of groundwater in the area of the disposal wells, a number of samples were collected by Linde and analyzed by Argonne National Laboratory in April 1981. Samples were taken from two test wells -- one drilled near Building 8 and the other near Building 38 -- and from an old gas well near Building 77 (Figure 5). Samples were also taken from an industrial well located approximately 2 km (1.25 mi) west of the Linde facility.

Table 6 summarizes the results obtained from the Argonne analysis. The complete results are presented in Appendix E. The average concentrations measured in each well were below limits set by the Nuclear Regulatory Commission for the release of water to an unrestricted area.¹⁶ Concentrations of all radionuclides dissolved in the water were lowest in the well 2 km west of the site. Dissolved uranium and radium concentrations were highest in the well near the southeast corner of the site (near Building 77).

Concentrations of radionuclides in suspended solids were, with the exception of radium, also the lowest in waters from the well located to the west of the Linde site. Uranium concentrations were highest in samples taken from the test well near Building 8, at the location of the disposal wells near Plant No. 1. The test well in the area of the other four disposal wells at the former Ceramics Plant contained concentrations greater than the offsite well, but did not have nearly as high a uranium or thorium concentration as the test well near Plant No. 1 (Building 8). However, the disposal wells near the Ceramics Plant were further apart, and the test well samples may not be completely representative of groundwater in the disposal wells (Appendix D).

¹⁶ Code of Federal Regulations, Title 10, Part 20.

Table 4. Radionuclide Concentrations* in Soil on Creek Banks (ORAU, 1981)

Sampling Location	Concentration in pCi/g				
	Radium-226	Uranium-238	Uranium-235	Thorium-232	Thorium-230
Six Sampling Locations in Two Mile Creek at and Downstream of Linde (Numbers 3 through 8, Figure 4)					
- Range	0.20-0.85	<0.36-5.42	0.01-0.08	0.03-0.88	0.03-2.51
- Average	0.55	1.86	0.04	0.47	0.87
Two Mile Creek Upstream of Linde	0.50	0.85	0.03	0.59	1.72
Two Nearby Creeks	0.42-0.62	1.12-2.83	0.04-0.05	0.51-0.56	0.59-0.73

* The values given have 2-sigma (about 95%) confidence levels based only on counting statistic errors that can range from $\pm 3\%$ to $\pm 140\%$ of the value. In general, the lower concentrations have larger error ranges.

Table 5. Radionuclide Concentrations* in Sediment From Two Mile and Other Nearby Creeks (ORAU, 1981)

Sampling Location	Concentration in pCi/g				
	Radium-226	Uranium-238	Uranium-235	Thorium-232	Thorium-230
Six Sampling Locations in Two Mile Creek at and Downstream of Linde (Numbers 3 through 8, Figure 4)					
- Range	0.41-0.81	<0.59-3.60	0.03-0.09	0.2-0.56	0.02-1.56
- Average	0.58	1.34	0.05	0.41	0.81
Two Mile Creek Upstream of Linde Facility (One Sample)	0.69	4.30	0.10	0.01	0.92
Two Creeks in Tonawanda					
- Range	0.55-0.70	0.82-0.95	0.05	0.70-0.80	0.60-0.70
- Average	0.63	0.89	0.05	0.75	0.65

* The values presented have 2-sigma (about 95%) confidence intervals that range from $\pm 2\%$ to $\pm 50\%$ and are based only on counting statistic errors. In general, lower concentrations have larger error ranges.

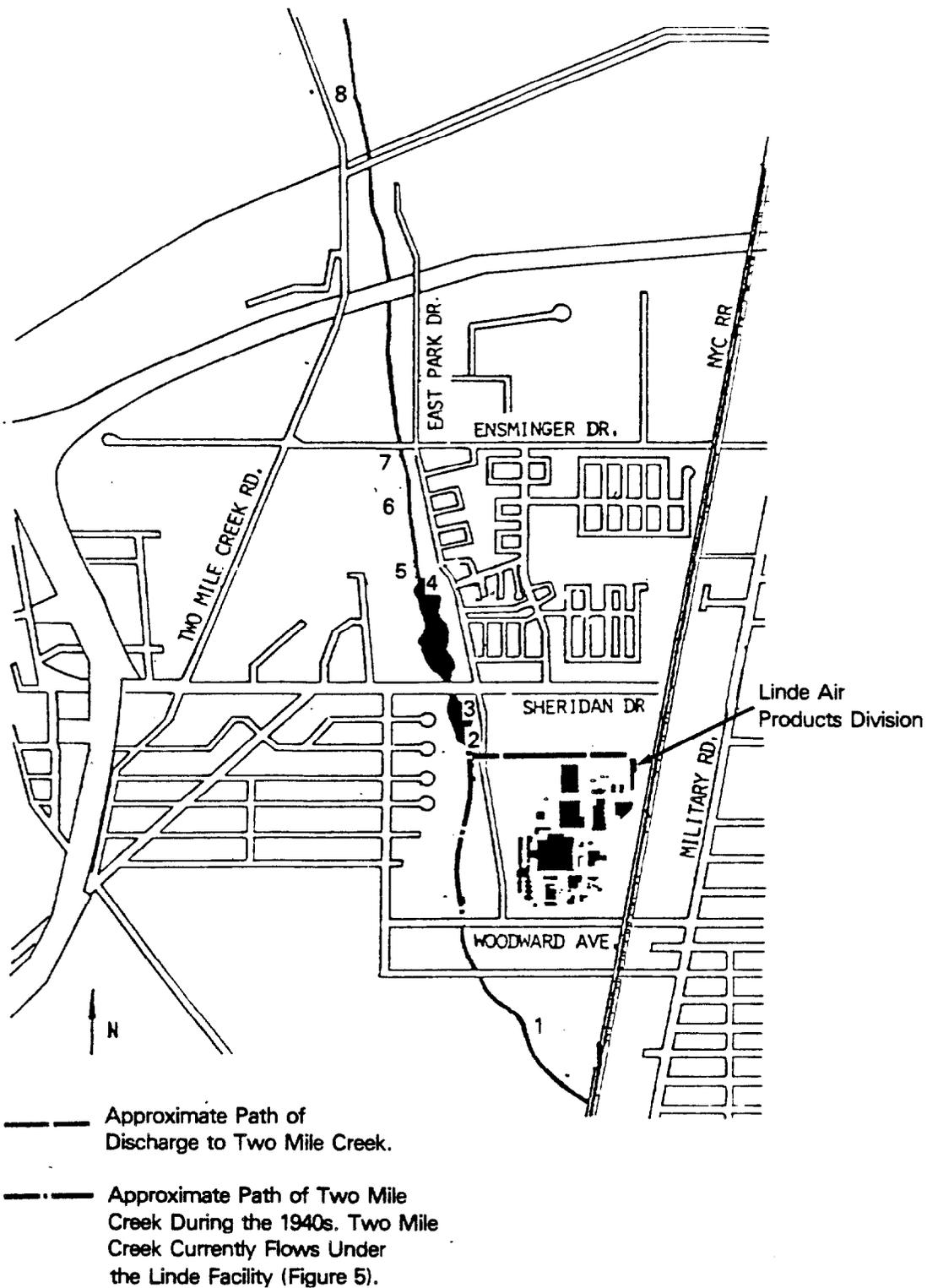
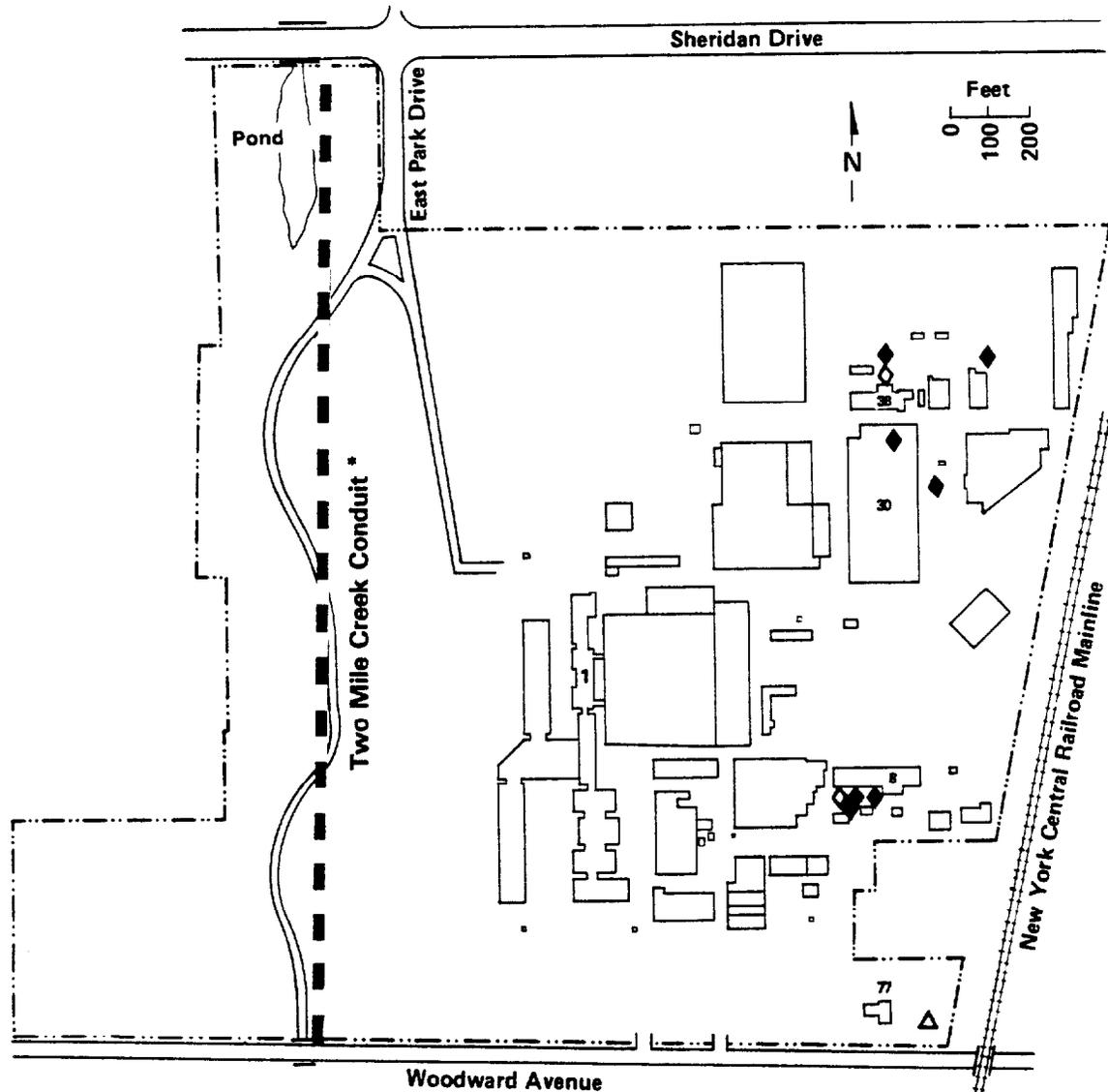


Figure 4. Samping Locations Along Two Mile Creek (ORAU, 1981)



- Legend:
- 38 — Building Number
 - ◆ — Location of Disposal Wells
 - ◇ — Location of Test Wells
 - △ — Location of Gas Well Tested

*Two Mile Creek currently runs under the Linde property from Woodward Avenue north to the dam.

Figure 5. Plan View of Linde Air Products Division Showing Original Disposal Wells, Test Wells, and Groundwater Sample Collection Sites

Table 6. Concentrations of Selected Radionuclides in Groundwater Samples Collected by Linde Division and Processed by Argonne National Laboratory*

Sampling Location	Average Concentrations of Radionuclides Dissolved in Solution x 10 ⁻⁸ μCi/ml						
	Radium-226	Uranium-238**	Uranium-235**	Uranium-234**	Thorium-232	Thorium-230	Thorium-228
Test Well Near Building 38 (Former Ceramics Plant)	0.026	0.23	0.01	0.23	0.074	0.017	0.049
Test Well Near Building 8 (Plant No. 1)	0.125	0.078	0.004	0.078	0.003	0.205	0.004
Gas Well Near Building 77	1.68	3.83	0.17	3.83	0.070	0.319	0.07
Well 2 km West of Linde Property	0.004	Total Uranium <0.03			0.002	0.008	0.002
NRC Limit for Unrestricted Use***	3	4000	3000	3000	200	200	700

Sampling Location	Average Concentrations of Radionuclides Suspended in Solution x 10 ⁻⁸ μCi/ml of Groundwater (pCi/g of Suspended Solid)						
	Radium-226	Uranium-238**	Uranium-235**	Uranium-234**	Thorium-232	Thorium-230	Thorium-228
Test Well Near Building 38 (Former Ceramics Plant)	1.40 (0.54)	5.09 (3.25)	0.23 (0.15)	5.09 (3.25)	0.94 (0.50)	1.04 (0.60)	1.20 (0.59)
Test Well Near Building 8 (Plant No. 1)	4.03 (3.08)	625.9 (629.8)	28.16 (28.33)	625.4 (629.8)	0.92 (0.58)	55.78 (52.47)	0.89 (0.55)
Gas Well Near Building 77	1.36 (0.94)	76.28 (52.65)	3.43 (2.37)	76.28 (52.65)	0.07 (0.05)	0.32 (0.22)	0.4 (0.3)
Well 2 km West of Linde Property	0.04 (15)	Total Uranium <0.003 (<1.4)			0.00007 (0.03)	0.0002 (0.08)	0.0005 (0.2)
NRC Limit for Unrestricted Use***	3000	4000	3000	3000	4000	3000	1000

* A complete listing of Argonne's results is presented in Appendix E.

** The analysis conducted by Argonne was for total uranium. Concentrations of the uranium isotopes were calculated assuming 1 Ci of natural uranium contains 0.489 Ci of uranium-238, 0.489 Ci of uranium-234, and 0.022 Ci of uranium-235.

*** Code of Federal Regulations, Title 10, Part 20, Appendix B, Table 11, Column 2.

The old gas well, near Building 77, had a greater-than-expected concentration of uranium in the suspended solids of nearly 110 pCi/g. The concentration of uranium in carbonate rocks is relatively constant and averages about 1.5 pCi/g, with a maximum of about 12 pCi/g (Rogers, 1969). This particular well is deep and extends through a number of formations; however, it is also unlikely that these shales or sandstones would approach any concentration near 100 pCi/g. It is possible that the high concentrations of uranium in the suspended solids could be the result of selective dissolution of the major constituents in the rocks. The dissolved uranium concentration measured in this well (about 8×10^{-8} $\mu\text{Ci/ml}$), as noted earlier, is higher than levels in the other wells and higher than levels measured in other carbonate aquifers elsewhere in the country (0.3 to 0.006×10^{-8} $\mu\text{Ci/ml}$) (Cowart, 1978). It is, however, within concentrations measured in mineralized aquifers (0.1 to 31×10^{-8} $\mu\text{Ci/ml}$) (Rogers, 1969).

Additional measurements of groundwater were made by ORAU in June 1981. Samples were collected and analyzed from the two test wells and a residential well located 2 km north of the Linde facility. In general, the analysis confirmed the previous results and indicated that concentrations of radionuclides are within Nuclear Regulatory Commission limits¹⁷ (Table 7).

OTHER ONSITE MEASUREMENTS

ORAU also collected samples of water and silt sediment from the storm sewer and sanitary sewer (Tables 8 and 9). Silt and sediment samples did have elevated concentrations of uranium, radium, and thorium. One water sample taken from the storm sewer had a slightly higher uranium concentration than other surface water samples; however, the concentration was not significantly higher than concentrations found in other creeks in the area and was below Nuclear Regulatory Commission limits for release to an unrestricted area.¹⁷

Soil samples were also collected on the Linde site near the former disposal wells and from spoil remaining from the drilling of the two test wells. The ranges are also shown in Table 9. The concentrations in soil were above those of offsite locations, and concentrations in the spoil from the subsurface samples contained uranium well above levels expected in carbonate rocks such as magnesite and limestone, indicating, as expected, that the subsurface in the areas of the wells is contaminated.

A complete characterization of the Linde facility, excluding the liquid effluent pathways, is presented in DOE/EV-0005/5 (ORNL, 1978).

¹⁷ Code of Federal Regulations, Title 10, Part 20.

Table 7. Radionuclide Concentrations in Groundwater Samples Collected and Analyzed by Oak Ridge Associated Universities

Sampling Location	Concentration of Radionuclides x 10 ⁻⁸ μCi/ml				
	Radium-226	Uranium-238	Uranium-235	Thorium-232	Thorium-230
Linde Test Well Near Building 38 (near the former Ceramics Plant)	0.031	<59	1	<0.01	0.06
Linde Test Well Near Building 8 (near the site of Plant No. 1)	0.016	<64	<0.5	<0.01	0.02
Residential Well 2 km North of Linde	0.091	<49	<0.4	<0.01	
NRC Limit for Unrestricted Use*	3	4000	3000	200	200

* Code of Federal Regulations, Title 10, Part 20, Appendix B, Table II, Column 2.

Table 8. Radionuclide Concentrations in Water Samples Collected Onsite

Sampling Location	Concentration of Radionuclides x 10 ⁻⁸ μCi/ml				
	Radium-226	Uranium-238	Uranium-235	Thorium-232	Thorium-230
<u>Onsite</u>					
Sanitary Sewer	<0.007-0.016	<65	<0.4-1	≤0.02	0.06-0.20
Storm Sewer	0.32	<160	3	0.03	0.01
<u>Background*</u>					
City Water	0.008	<57	1	<0.03	<0.06

* For surface water background levels, see Table 3, "Other Creeks in Area" column.

Table 9. Concentrations of Radionuclides in Onsite Silt, Sediment, and Soil Samples (ORAU, 1981)

Sample Description	Concentration in pCi/g				
	Radium-226	Uranium-238	Uranium-235	Thorium-232	Thorium-230
Sediment Samples From Linde Storm Sewer	0.64-6.93	4.5-116	0.17-4.57	0.34-0.62	0.2-17.7
Sediment Samples From Sanitary Sewer	0.38-1.94	<0.51-362	0.05-12.93	0.11-0.21	0.34-1.33
Samples of Surface Soil Near Disposal Wells	0.93-2.74	11.40-15.80	0.35-0.57	0.66-0.72	1.72-3.55
Samples of Subsurface Soil Near Disposal Wells	0.94-5.53	11.20-24.05	0.36-0.84	0.78-0.92	3.30-5.90
Samples of Soil From Test Wells Near Disposal Wells	0.82-1.93	10.96-26.40	0.33-1.09	0.51-0.74	3.53-8.79

Note: For typical background concentrations, see Table 4, Radionuclide Concentrations in Soil on Creek Banks.

EXPOSURE AND HEALTH EFFECTS EVALUATION

The naturally occurring radionuclides present on the Linde property and in Two Mile Creek are also present in minute quantities throughout our environment. Concentrations of these radionuclides in normal soils, air, water, food, etc., are referred to as background concentrations. Radiation exposures resulting from this environmental radioactivity are referred to as background exposures. These background exposures are not caused by any human activity and, to a large extent, can be controlled only through man's moving to areas with lower background exposures. Each and every human receives some background exposure daily.

Workers and, to a lesser extent, members of the general public can also be exposed to radiation levels above normal background levels from the use of radioactive materials for scientific, industrial, and medical purposes. Scientifically based guidelines have been established as upper limits on these additional exposures. The generally accepted guideline for exposure received by an individual member of the general public is 500 mrem/yr whole body dose.

There are three primary pathways of human exposure to radioactivity: direct exposure to gamma radiation, inhalation of radionuclides in the air, and ingestion of food or water contaminated with radionuclides. The discharge of liquid effluents did not result in any residual contamination that would produce significant exposures through the first two pathways. It is only the third pathway that could produce any measurable exposure.

Concentrations of radionuclides in water from Two Mile Creek were not significantly greater than levels found at other nearby creeks; thus, any use of the creek water would not cause exposures measurably different from background exposures. The analysis of water from the Linde test wells did indicate above-background (but below-guideline) concentrations of radionuclides. Under present use conditions, no one is receiving exposures in excess of background exposure; however, the concentration of the radionuclides in the groundwater could possibly result in a slight increase if conditions were to change. To determine an upper limit for potential increases in exposure to persons using the water, a worst-case scenario was evaluated.

As discussed earlier and in Appendix D, the very high mineral content of this groundwater makes the water unacceptable for drinking or for most industrial purposes. Therefore, such uses are not considered as possible pathways of exposure. Groundwater in the area is applied to some industrial purposes, such as cooling water, and for residential uses such as washing automobiles and watering lawns. It is

therefore conceivable, though not probable, that the water could also be used for crop irrigation, possibly in large gardens. This exposure pathway would represent a worst-case scenario. To determine the potential dose from this scenario, the following were assumed:

- 100 percent of the vegetables consumed were grown on soil irrigated with contaminated water.
- Radionuclide concentrations in the water were equivalent to the highest concentrations measured during the Department of Energy surveys.
- Approximately 0.25 kg of vegetables was consumed by the individual daily.

The annual organ doses were then estimated using procedures presented in ORNL/OEPA-7 (Hill, 1979) and were converted to the annual equivalent whole body dose by applying the weighting factors recommended in the International Commission on Radiation Protection Publication 26 (ICRP, 1977).

The annual incremental dose received under this scenario would be less than 3 mrem/yr whole body dose equivalent. Such a dose would result in a lifetime incremental increase of 210 mrem, assuming a 70-yr lifetime. In the United States, a person receives an average of about 100 mrem/yr, or 7000 mrem per lifetime from natural background radiation. The worst-case exposure resulting from residues in the groundwater would therefore result in less than a 3-percent increase in background radiation. This level would be less than 0.6 percent of the internationally accepted guideline of 500 mrem/yr for exposure to the general public.

Although the estimated exposure is very low, for the purposes of radiation protection, all radiation exposures are assumed capable of increasing the risk of contracting cancer. Exposure to radiation does not result in inevitable induction of cancer; rather, the result of exposure is an increased probability that an individual may contract cancer. Precise numerical values for an individual's increased risk cannot be determined, however, with any certainty. Such factors as the individual's personal habits, state of health, previous or concurrent exposure to cancer-causing agents other than radiation, genetic history, age at onset of exposure, and variability in latency period (time between radiation exposure and physical evidence of disease) can cause wide variations in the risk of cancer. There are thus large uncertainties in any estimates of the expected number of increased cancers in a small population group.

With these uncertainties in mind, an estimate of the risk of contracting cancer can be made using the estimated 3 mrem/yr and 210 mrem/70-yr accumulated whole body dose equivalent, and assuming 100

cancer deaths per million persons exposed per rem of radiation exposure (BEIR, 1980; ICRP, 1977). The total increased risk due to radiation-induced cancer from the consumption of crops grown on soil irrigated by the contaminated groundwater is 0.002 percent (2 deaths per 100,000). This increase is insignificant when compared to the normal risk of dying from cancer. In 1977 for instance, 21.6 percent of all deaths (21,600 per 100,000) in New York State and in Erie County, New York, were attributed to cancer (DHHS, 1980).

CONCLUSIONS

The concentrations of radionuclides and radiation levels in Two Mile Creek at and downstream of the Linde facility are comparable to those measured upstream of the Linde facility and in other nearby streams. The radionuclide concentrations in soil and water around the disposal wells are above background levels, but the concentrations in the groundwater are well below the applicable Nuclear Regulatory Commission limits for water released to an unrestricted area. An evaluation of the worst-case potential exposure determined that doses would be very low, about 0.6 percent of the guideline values, and that the associated radiation-induced risk of cancer would be insignificant when compared to the normal risk of contracting cancer.

A review and evaluation of process operating data indicated that the uranium concentrations in the effluents met even present-day Nuclear Regulatory Commission limits. The radium concentration, depending on dilution rates and solubility, may have exceeded present-day limits, but was maintained below an "action point" established for the operation. The evaluation showed that no hazard due to the radioactive materials in the effluent existed at the time of the uranium facility's operation or from the termination of operations to the present time. Neither the groundwater nor the creek water were used for drinking, and the concentration of radionuclides in the effluent was sufficiently low that radiation exposures from other uses of the water would be negligible.

Based on the information in this report and the data collected through Department of Energy radiological surveys, there appears to be no need for additional radiological or hydrological studies of this site and the associated effluent disposal pathways.

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APPENDIX A. DESCRIPTION OF LINDE CERAMICS PLANT OPERATIONS

The ceramics plant operations included three separate processes: Step I processed domestic and African ore and produced uranium oxide (U_3O_8), Step II produced uranium dioxide (UO_2) from the uranium oxide, and Step III converted the uranium dioxide into uranium tetrafluoride (UF_4).

STEP I, DOMESTIC ORE TREATMENT

Sulfuric acid was added to the ore slurry until a pH of 0.7 to 0.8 was reached. Pyrolusite or magnasite (MnO_2) was added to oxidize any reduced uranium. The mixture was digested at $90^{\circ}C$ for 3 hours and then cooled with weak wash solution at $60^{\circ}C$. The uranium was in solution as uranyl sulfate, and many of the impurities (iron, silica, phosphorus, vanadium, alumina) were also partially in solution.

Soda ash was added until the pH reached about 9.2. Some sodium bicarbonate was also added, which precipitated most of the impurities and left the uranium in solution as sodium uranyl tricarbonate. The slurry was filtered in the Moore filters, and the cake hauled to a tailings pile.

The liquors contained vanadium and phosphorus as objectionable impurities. These were removed by the addition of ferrous and ferric sulfates, respectively. The resultant iron cake was filtered off in plate and frame presses and hauled to the tailings pile. The liquors were treated with caustic soda and precipitated the uranium as sodium diuranate.¹ The filtrate from this step was discharged as waste effluent.

The sodium diuranate cake was treated with sulfuric acid and ammonium sulfate and was converted to an ammonium uranyl sulfate complex. This was removed in a filter press. The cake (acid leach cake) was fed to a calciner to drive off the ammonia, sulfur dioxide and trioxide, and water, leaving the black oxide of uranium.

¹ Initial domestic ore processing used a precipitation involving the addition of sulfuric acid followed by caustic, but the revised process used the direct caustic method.

STEP I, AFRICAN ORE TREATMENT

The treatment of African ore was very similar to that of domestic ore. The digestion step required more pyrolusite because more of the uranium was in a reduced state. Also, barium chloride had to be added to act as a "gatherer" for the radium. The African ore contained little vanadium or phosphorus, so the iron sulfate step was omitted. Instead, sodium sulfide was added to remove the lead. The remainder of the process was the same. The molybdenum stayed in solution when the uranium was precipitated.

STEP II, OPERATIONS

The black oxide from Step I was dissolved in nitric acid. Certain insoluble impurities were filtered off, and the solution was evaporated to "molten uranyl nitrate hexahydrate."² This was added to cold ether. The impurities remained in solution in the water, and the uranium went into the ether layer. The layers were separated, and the uranium was extracted from the ether with water.³ The solution was concentrated and decomposed with heat to uranium trioxide. The trioxide was treated with hydrogen in a rotary furnace and reduced to a very pure uranium dioxide. The residues from this process were recycled to the Step I process.

STEP III, OPERATIONS

The uranium dioxide from Step II was loaded on magnesium trays and treated at 1000°F with anhydrous hydrofluoric acid. This converted the uranium dioxide to uranium tetrafluoride.

² Linde Corporation, Construction, Process, and Operation Reports, Contract W-7401-Eng-14, Step I, Step II, and Step III, October 1, 1946.

³ Metallic salts are more soluble in water than ether, and the uranyl nitrate is equally soluble in both. The solubility of the uranyl nitrate was shifted by changes in the ether/water ratios.

APPENDIX B. PROCESSING RECORDS AND RADIOACTIVE MATERIAL FLOW ANALYSES

Table B-1 lists the monthly estimates of production from the Linde Step I operations. These estimates were based on Ceramics Plant progress reports that were identified during records searches of the Oak Ridge Operations Office archives. The searches were conducted by personnel from Oak Ridge National Laboratory and The Aerospace Corporation.

Figures B-1, B-2, and B-3 present the radioactive material balance in the Step I process for the various ores processed. These figures are also based on available data and reflect the best estimates to date. The balance is based on an assumed 110 metric tons or 100 tons of uranium in a final product.

In Table B-1, it was assumed, for domestic ore processed in 1943, that extraction efficiencies were greater than or equal to 93 percent. Extraction efficiencies for domestic ore after 1945 and for the African ores were generally greater and are stated in the table. There were sufficient data to make some reasonable estimates regarding radium in the African ores; the major uncertainty in this area is that there is no information regarding radium in the product or in the liquor purification cakes. The radium balance from the domestic ore processing has many more uncertainties; the only available information is that the radium in the domestic ore was significantly less than that in the African ores.

Much of the domestic ore received by Linde was from the Manhattan Engineer District plant at Grand Junction, Colorado, which received "green sludge" from the United States Vanadium Corporation plants at Uravan and Durango, Colorado. United States Vanadium Corporation extracted the uranium by acid percolation leaching and then precipitated the vanadium from the tailings by adding iron to the solution. This was followed by neutralization. The precipitated green sludge was then roasted with soda ash at Grand Junction. Water leaching of the calcines removed the vanadium, and the uranium was recovered in the residue as sodium diuranate. This material (containing an equivalent of 10 to 15 percent U_3O_8) was processed at Linde.

The major portion of the radium would be expected to remain with the residues left from the acid percolation leaching.¹ In modern acid leaching operations, the amount of radium dissolved in solution can vary. Estimates of from 0.25 to 5 percent of the radium in the ore

¹ R.C. Merrit, "The Extractive Metallurgy of Uranium," Colorado School of Mines, 1971.

Table B-1. Linde Step I Production and Discharge Estimates by Month

Year/ Month	Ore		Extraction Efficiency (%)	Product		Liquid Effluent		Residue		
	Type	Approximate Amount (metric tons)		U ₃ O ₈ (metric tons)	Amount (10 ⁶ liters)	U ₃ O ₈ Content (x100 kilograms)	Location	Amount (metric tons)	U ₃ O ₈ Content ^a	Location
1943										
June	American ^b			-	-		Sewer			Haist
July	(10-20% U ₃ O ₈)			10.83	1.9- 2.9 ^d		↕			↕
August	↕			35.39	6.1- 9.2		↕			↕
September				47.26	8.1-12.1		↕			↕
October				34.92	6.0- 9.0		↕			↕
November	American			42.41	7.3-11.0		↕			Haist
December	African L-30	450 ^c		41.94	7.2-11.0		Sewer			LOOW
Subtotal		≈1450	≈93	212.75	36.6-55.2	54.9-82.8		990-1220	10.09-10.31	
	American	≈590		Amer. 170.81	Amer. 29.4-44.2	(avg. 0.15g/l)		Amer. 540-820	8.84	
	Scrap	≈410		Afric. 41.94	Afric. 7.2-11.0			Afric. 400 ^e	1.25- 1.47	
	African	≈450								
1944										
January	African L-30	549	93	55.07	9.47	2.844	Sewer			LOOW
February	(8-12% U ₃ O ₈)	821	93	86.14	14.19	4.259	Sewer	0.87%		↕
March	assumed 10.8%	1098	97	115.04	19.79	5.937	Sewer	0.44%		↕
April	↕	962	97	100.81	15.61	3.741	Wells or	0.41%		↕
May		942	97	98.69	17.36	3.279	Storm Sewer	0.37%		↕
June		962	97	100.81	16.17	4.381	↕	0.35%		↕
July		671	98	71.03	11.29	3.193	↕	0.25%		↕
August		863	98	91.42	17.65	5.410	↕	0.26%		↕
September		795	98	85.85	15.12	5.941	↕	0.21%		↕
October	African L-30 & L-50 (6.7% U ₃ O ₈)	680	97	60.17	11.98	5.773	↕	0.25%		↕
November	African L-50	1207	98	79.24	13.18	3.782	Wells or	0.20%		LOOW
December	American L-19	93	96	8.89	2.31	0.499	Storm Sewer	0.52%		Haist
Subtotal		9643		953.16	164.12	49.039		9184	27.90-31.59	
	African	9550		944.27	161.81	48.540		9120 ^b	27.57-31.26	
	L-30	8064		846.89				7532	24.70-27.88	
	L-50	1486		97.38				1588	2.87- 3.38	
	American L-19	93		8.89	2.31	0.499		64	≈0.33	
						(avg. 0.030g/l)				

B-2

Table B-1. Linde Step I Production and Discharge Estimates by Month
(Continued)

Year/ Month	Ore		Extraction Efficiency (%)	Product		Liquid Effluent		Residue	
	Type	Approximate Amount (metric tons)		U ₃ O ₈ (metric tons)	Amount (10 ⁶ liters)	U ₃ O ₈ Content (x100 kilograms)	Location	Amount (metric tons)	U ₃ O ₈ Content ^a
1945									
January	American	249	0.98	34.19	5.17	1.855	Wells or Storm Sewer ↑ ↓ Wells or Storm Sewer	0.50%	Haist ↑ ↓ Haist
February	L-19 (mostly 10- 16.5% U ₃ O ₈ assumed 14% avg.)	361	0.97	49.03	8.17	1.723		0.60%	
March		523	0.97	70.97	12.64	2.499		0.53%	
April		392	0.97	53.22	9.12	2.009		0.55%	
May		583	0.98	79.99	20.45	3.324		0.38%	
June		495	0.97	67.31	20.63	5.887		0.43%	
July		436	0.97	59.21	20.22	4.771		0.45%	
August		559	0.97	75.80	20.97	3.565		0.38%	
September		499	0.97	67.77	16.87	2.943		0.50%	
October		461	0.97	62.56	20.21	5.093		0.57%	
November		484	0.98	66.41	22.49	4.553		0.47%	
December	American	377	0.96	50.77	15.54	3.388		0.66%	
Subtotal		5419		737.22	192.48	41.610 (avg. 0.022g/l)	5219	27.14	
1946									
January	American	443	0.95	58.89	28.14	7.057	Wells or Storm Sewer ↑ ↓ Wells or Storm Sewer	0.87%	Haist Haist LOOW LOOW LOOW & Mdx Haist LOOW Middlesex
February	Amer. & Afric.	687	0.94	37.11 ^f	8.87	2.721		0.24%	
March	African R-10	1935	0.95	64.32	14.88	5.909		0.18%	
April	(≈3.5% U ₃ O ₈)	2048	0.94	67.39	16.24	3.356		0.21%	
May	R-10	2446	.95	95.62	18.87	4.045		0.18%	
June	R-10&Q-20 (17.7%)	1319	.95	49.34 ^g	14.67	2.825		0.49%	
July	R-10&Q-20&Ash	286	≈.95	14.90 ^g	5.91	2.322		≈0.8%	
Subtotal		9164		377.57	107.58	28.236			
	L-19	590		78.25	31.04	7.946	548	2.85	Haist
	R-10	8492		285.72	76.54	20.290	7250-9100 ^h	16.41-19.31	LOOW
	Q-20	82		≈13.60			80-140 ⁱ	0.6-1.1	Middlesex
						(avg. 0.026g/l)			

Notes:

- Annual amounts (subtotals) given in metric tons; monthly amounts given in percent.
- Also processed scrap.
- Assumed 5% of the L-30 was processed in 1943.
- Assumed 78 to 118 liters of effluent generated per pound of product.
- The Aerospace Corporation, "Background and Resurvey Recommendations for the Atomic Energy Commission, "Background and Resurvey Recommendations for the Lake Ontario Ordnance Works," ATR-82(7963-04)-1, Germantown, Maryland, July 1981.

- 52.2% of product from domestic ore.
- ≈6.8 metric tons of product from Q-20.
- Includes R-10 iron cake (Aerospace, 1981).
- Based on 220 metric tons of sludge (wet weight).

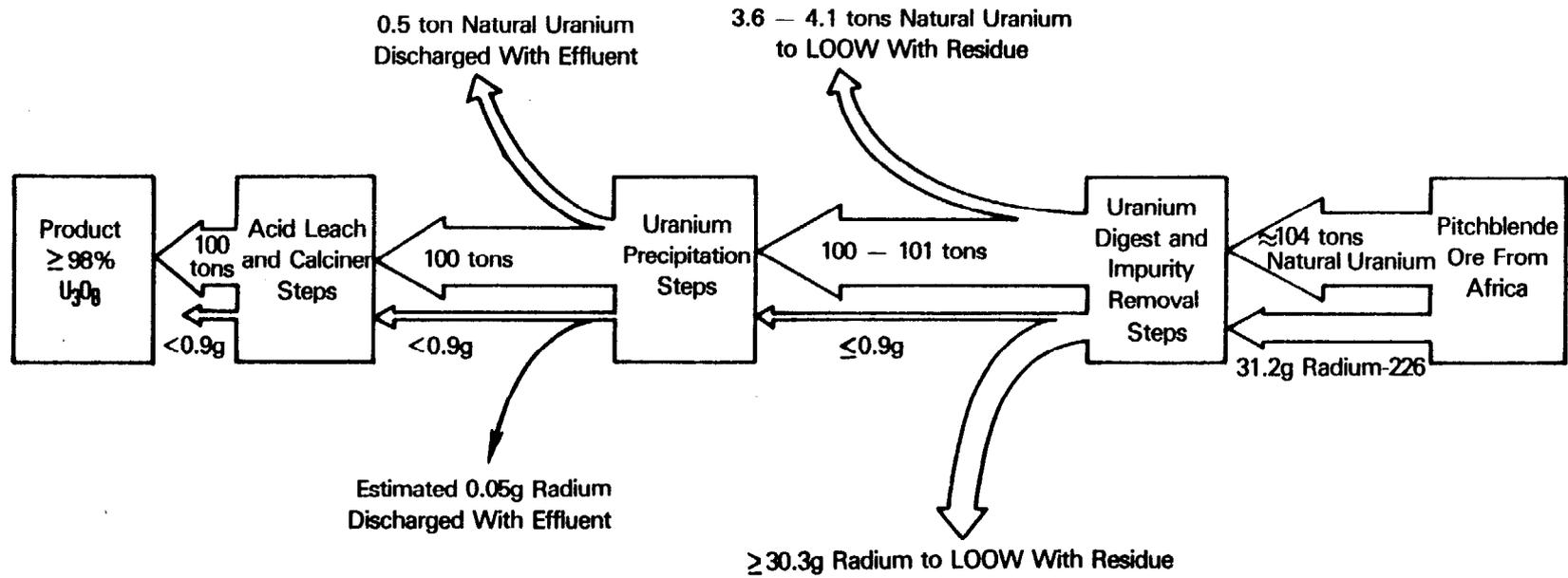
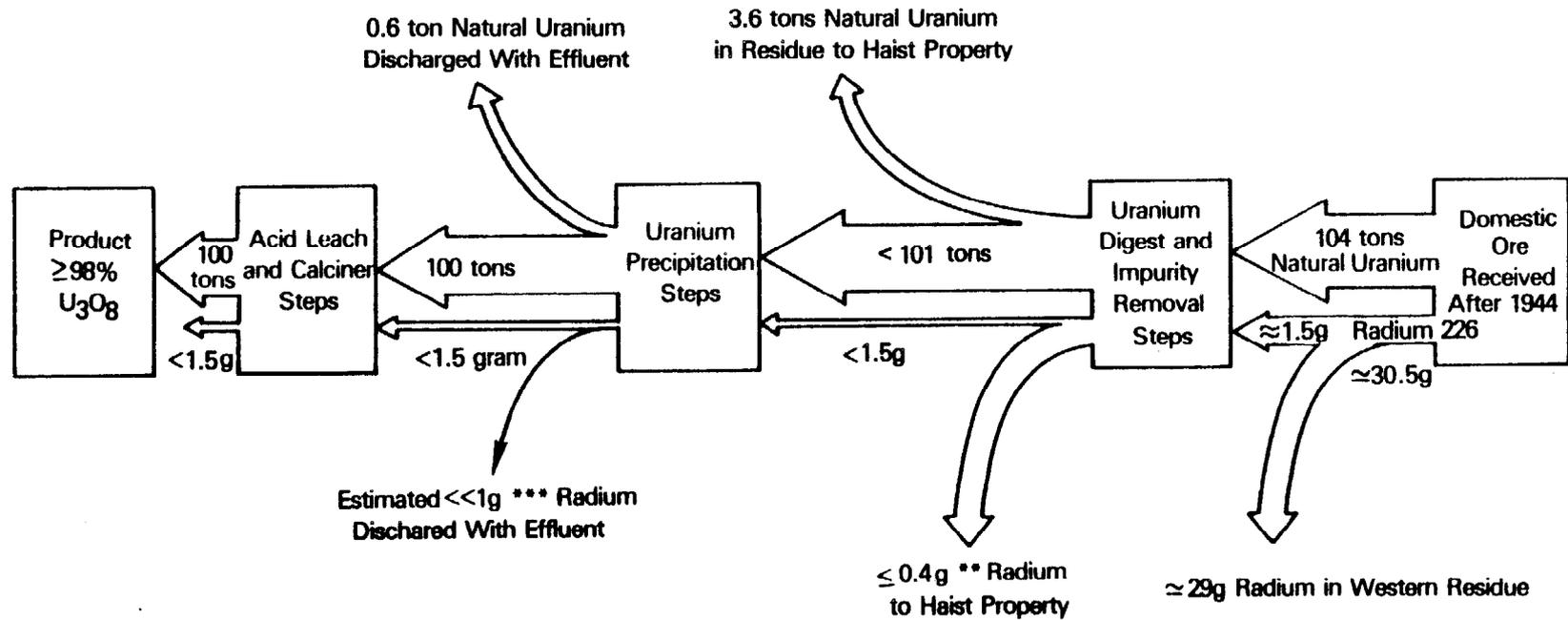


Figure B-1. Uranium and Radium Mass Balance for Processing Belgium Congo Pitchblende Ores at the Linde Step I Facility

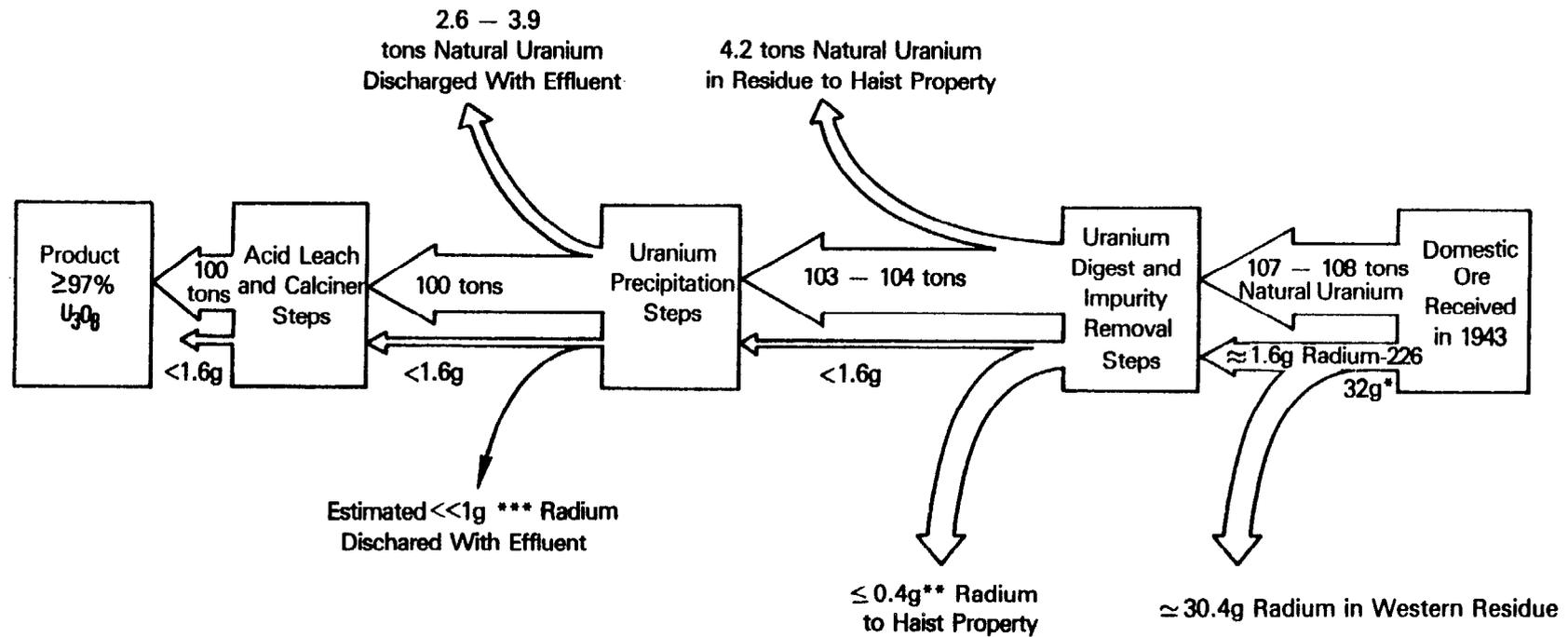


*The mass balance for radium in these ores is very uncertain.

**Based on radium in residue samples from the former Haist property; however, this maximum could be higher if there was any dilution with soil or significant dissolution by rainwater of the radium in material stored at Haist.

***This was probably much less than 0.5g, which is the maximum level allowable by a Linde action point for radium. W. Thomas (Corps of Engineers, 1945) suggested that radium in the American ore processing effluent was much less than the concentration in the African ore processing effluent.

Figure B-2. Uranium Mass Balance for Domestic Ore Processed at the Linde Step I Facility After November 1944



*The mass balance for radium in these ores is very uncertain.

**Based on radium in residue samples from the former Haist property; however, this maximum could be higher if there was any dilution with soil or significant dissolution by rainwater of the radium in material stored at Haist.

***This was probably much less than 0.5g , which is the maximum level allowable by a Linde action point for radium. W. Thomas (Corps of Engineers, 1945) suggested that radium in the American ore processing effluent was much less than the concentration in the African ore processing effluent.

Figure B-3. Uranium Mass Balance for Domestic Ore Processed at the Linde Step I Facility in 1943

dissolved in the leaching have been made by various sources. A major portion of this dissolved fraction of the radium would be expected to have stayed with the uranium² and been present in the concentrate (10 to 15 percent U₃O₈ equivalent) that was shipped to Linde from Grand Junction.

Therefore, if all the radium in the domestic ore shipped to Linde were in solution, the maximum radium-226 concentration in solution would have been between 0.4 and 7.5×10^{-8} g/l.³ However, a very significant portion of the radium would be expected to remain in the residue after the digestion of the uranium. Further, much of the radium dissolved in the uranium-rich solution after the digest would be expected to precipitate with the uranium in the final product. Therefore, the radium-226 in the effluent from the domestic ore processing would be expected to have been much less than the calculated maximum.

Estimates of the radium in the pitchblende ore were based on the assumption that the uranium-238 and radium-226 in the ore were in secular equilibrium when received. Available data suggested that Linde precipitated 97 percent of the radium in the filter cakes sent to the Lake Ontario Ordnance Works.⁴ If all the radium-226 remained in the solution (approximately 3 percent of the total radium or 0.009 Ci of radium-226 per ton of uranium produced) after this step, the radium concentration in the process solution would have been from 2.8 to 4.3×10^{-8} g/l.⁵ However, as with the domestic ore, some of the radium remained with the purification cake, and the significant portion would have remained in the product. Only a fraction of the radium was discharged. One reference⁶ suggests that the actual concentration of radium in the effluent was on the order of 2.5×10^{-9} g/l.

2 This is estimated to be from 0.003 to 0.05 Ci of radium-226 per Ci of uranium-238, or less than 0.015 gram per ton of natural uranium. This is an approximation based on an initial concentration in the material of 0.25 percent uranium in equilibrium with radium.

3 Assumes about 200,000 liters of effluent per ton of uranium produced.

4 Linde Corporation, Construction, Process, and Operation Reports, Contract W-7401-Eng-14, Step I, Step II, and Step III, October 1, 1946.

5 Assumes approximately 200,000 liters of effluent are produced per ton of uranium produced.

6 Letter from W. Bale to Major VanHorn, "Disposal of Tailings for 3.2% Ore," October 18, 1945.

APPENDIX C. CHEMICAL CONSTITUENTS IN ORES, RESIDUES, EFFLUENTS, AND PRODUCTS

Tables C-1 through C-3 present the results of the assay of typical ores and products from the Linde plant as well as the results of selected analyses of residues. These values are from historical records¹ and are all pre-1955. The analysis of solids from the liquid effluent gave the following values (based on one set of samples):

- Sodium, 43.64 percent
- Sulfates, 37.21 percent
- Calcium, 1.05 percent
- Carbon dioxide, 6.74 percent
- Iron, 0.67 percent
- Water, 9.04 percent

Data regarding the current condition of the residue from the processing of Africian ores can be obtained from the Battelle report, "A Comprehensive Characterization and Hazard Assessment of the DOE-Niagara Falls Storage Site," June 1981.

¹ Linde Corporation, Construction, Process, and Operation Reports, Contract W-7401-Eng-14, Step I, Step II, and Step III, October 1, 1946. Memorandum from S.H. Brown to R.L. Kirk, "Summation of Residue Sampling Program at Tonawanda and St. Louis," Atomic Energy Commission, May 28, 1953. Memorandum from R.L. Kirk to M. Eisenbud, "Waste Residues Handling," Atomic Energy Commission, September 22, 1953.

Table C-1. Typical* Analyses of Selected Ores Processed by Linde

Percent of Compound	Domestic Ores		Foreign Ores Pitchblende			Torbernite Q-20
	L-19	GUI	L-30	L-50	R-10	
U ₃ O ₈	15.8	12.5-2.0	10.54	6.7	3.53	17.72
Y ₂ O ₅	2.5	2.35	0.2	2.2	0.26	0.40
MoO ₃	0.02		0.35	0.3	0.3	0.31
PbO	0.01		0.9			
P ₂ O ₅	2.5	2.65	0.2	0.14	0.1	4.62
SiO ₂	13.0	28.6	50.0	51.4	55.8	51.14
CaO	17.0		1.0	1.0	.52	
NiO						
MgO	0.3		13.0	13.53	11.41	5.16
CoO				0.56	0.2	0.23
Fe ₂ O ₃	12.0	20.2	2.2	1.97	1.74	1.92
Al ₂ O ₃	5.0		9.0	9.42	13.61	6.45
CuO						2.78
CO ₂				2.88	2.29	
Na ₂ O				Ni1	0.25	
Ra			(23.7 mg/ton)			

* These values are typical assays and do not necessarily indicate an average. The L-30 and L-50 are very similar ores and as such were not separated in the tables in Appendix B. Similarly, L-19 and GUI are not separated.

Table C-2. Typical Analyses of Product From the Linde Step I Operation

Percent of Compound	Product From Processing		
	L-19 (1943)*	L-19 (1944)**	L-30**
U ₃ O ₈	97.0 (min)	98.2	97.7
Acid Solubles	0.5 (max)		
SiO ₂	0.05 (max)	0.52	
Acid Sulfide Metals	0.6 (max)		0.058
(NH ₄) ₂ CO ₃ Insoluble	0.5 (max)		.027
HNO ₃ Insoluble			0.42
Al ₂ O ₃	0.3 (max)		} 0.1
Fe ₂ O ₃	0.2 (max)	0.31	
P ₂ O ₅	0.3 (max)	0.3	0.63
Na ₂ O		0.08	0.24
V ₂ O ₅	0.05 (max)	0.11	0.054
SO ₄	0.05 (max)	0.036	0.29 (SO ₃)
Ag	0.0010 (max)		
B	0.0002 (max)		
Cd	0.0005 (max)		
Cl	0.05 (max)		
Mn	0.005 (max)		
Rare Earths	0.0015 (max)		

* The values of L-19 (1943) are specifications for the product while the other two are "typical" values that may not represent an average.

** L-19 was not differentiated from GUI, and L-30 was typical of L-50.

Table C-3. Typical Analyses of Residues at Lake Ontario Ordnance Works and Haist Property*

Ore Residues	Concentrations in g/g of Residues of					
	Uranium Oxide	Cobalt	Nickel	Copper	Radium	Vanadium Oxide
L-19 (Haist)						
Actual	1.1x10 ⁻³ (2.8x10 ⁻³)**			1.3x10 ⁻³		
Dry	2.5x10 ⁻³ (6.3x10 ⁻³)	-	-	2.8x10 ⁻³		
L-30						
Actual	1.4x10 ⁻³ (2.2x10 ⁻³)	3.4x10 ⁻³	1.1x10 ⁻²	1.1x10 ⁻³	1.6x10 ⁻⁵	
Dry	2.5x10 ⁻³ (4x10 ⁻³)	6.2x10 ⁻³	2x10 ⁻²	2x10 ⁻³	3x10 ⁻⁵	
L-50						
Actual	0.7x10 ⁻³ (1.3x10 ⁻³)	3.7x10 ⁻³	1.2x10 ⁻²	1.5x10 ⁻³	2.1x10 ⁻⁵	
Dry	1.1x10 ⁻³ (2x10 ⁻³)	5.9x10 ⁻³	1.9x10 ⁻²	2.4x10 ⁻³	1.9x10 ⁻⁵	
R-10						
Actual	0.9x10 ⁻³ (1.8x10 ⁻³)	3.9x10 ⁻³	1.0x10 ⁻²	2.7x10 ⁻³	7.0x10 ⁻⁶	
Dry	1.2x10 ⁻³ (2.3x10 ⁻³)	5.3x10 ⁻³	1.3x10 ⁻²	3.7x10 ⁻³	9.6x10 ⁻⁶	
R-10 Iron Cake (Phosphate Cake)						
Actual	1.6x10 ⁻³ (4.3x10 ⁻³)	1.9x10 ⁻³	3.1x10 ⁻²	-		
Dry	3.4x10 ⁻³ (9.1x10 ⁻³)	4x10 ⁻³	6.5x10 ⁻²	-		
P-78 (Haist) (Phosphate & Vanadium Cake)						
Actual	1.5x10 ⁻³ (3x10 ⁻³)			1.2x10 ⁻³		
Dry	3.4x10 ⁻³ (6.7x10 ⁻³)	-	-	2.5x10 ⁻³		(4-7x10 ⁻²)

* Values in this table are based on 1953 data collected to evaluate reprocessing feasibility and operating data. Data from recent surveys have not been presented.

** The first value is an estimate made for the reprocessing study, the second is based on initial analyses from the operating records.

APPENDIX D. GENERAL GEOLOGY OF THE TONAWANDA AREA

This appendix contains a brief overview of the geology in the Tonawanda region and a discussion of water quality. Some data regarding uranium concentrations in similar formations and groundwater are also presented.

Tonawanda, New York, is located on the Huron plain, which is bounded on the north by the Niagara Escarpment and on the south by the Onondaga Escarpment (Figure D-1¹). It is approximately 170m (570 ft) above sea level. The convoluted rock beds in this area have a slight southerly slope, about 5.3m/km (28 ft/mi). Kindle and Taylor¹

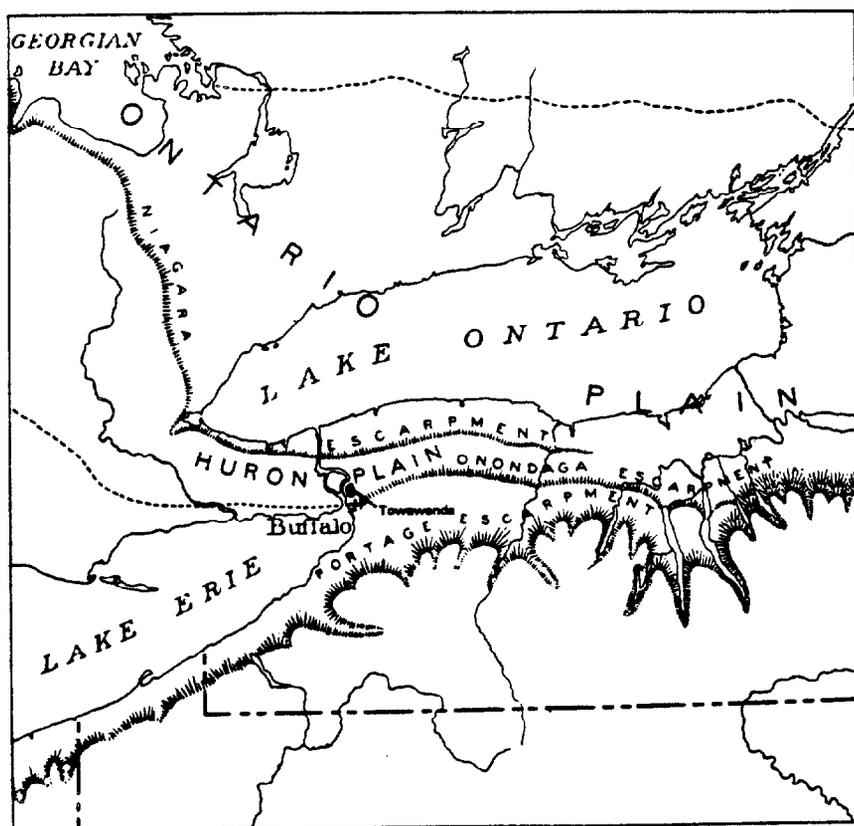


Figure D-1. Sketch Map of the Vicinity of Tonawanda Showing Physiographic Divisions¹

¹ E.M. Kindle and F.B. Taylor, Geologic Atlas of the United States, Niagara Falls, New York, U.S. Geological Survey, Washington, D.C., 1973.

indicate that the uppermost formations in the area consist of magnesium limestone and shale with layers of dolomite and gypsum. The bedrock in the area of the Linde plant is covered by about 25 to 30m (80 to 100 ft) of red clay. The direction and the flow rate of water in these formations are difficult to predict. In carbonate rock, the solution openings (caused by water dissolving the carbonate rock) are irregularly distributed both horizontally and vertically, and wells that are drilled near one another to similar depths can produce significantly different yields.²

Groundwater in the Buffalo-Niagara Falls area was investigated by Reck and Simmons.³ In general, they found the groundwaters to have a very high mineral content (dissolved solids) averaging about 800 ppm over the entire region. Water from formations located in the Grand Island and Tonawanda areas averaged over 4000 ppm. The high mineral content of these waters requires extensive treatment to make the water suitable for most industrial purposes. As a result, the waters are used only for cooling water and purposes for which the chemical quality is not important. Similar problems are encountered in residential use of the water. For instance, the only residential well identified in the Department of Energy evaluations was a general-purpose well. The owner indicated that it was not acceptable for drinking; it could be used only for washing equipment or watering the lawn.

It was because of the poor water quality that the Linde wells were suggested and used for disposal of the uranium process effluent. Linde had found the water in the wells unacceptable for its use and believed the wells to have been contaminated by other industrial operations. It may very well have been the naturally high mineral content of the water that caused the unacceptable condition; however, it was not uncommon in western New York for industries to use wells for disposing of their wastes.³ The disposal wells usually became clogged, losing their ability to absorb wastes, as did the Linde wells. Figures D-2, D-3, and D-4 are drawings of three of the seven Linde disposal wells, and Figure D-5 is the well driller's drawing of one of the Linde gas wells.⁴ The gas well was not used for waste disposal; the drawing is presented to detail the local strata and because it indicates that the most probable aquifer (at about 40m or 150 ft) used for disposal was described as being

² W.C. Waller, Ground Water Resource Evaluation, McGraw-Hill, New York, New York, 1970.

³ C.W. Reck and E.T. Simmons, "Water Resources of the Buffalo-Niagara Falls Region," Geological Survey Circular 173, U.S. Geological Survey, Washington, D.C., 1952.

⁴ Figures D-3, D-4, and D-5 were redrawn from 1943 drawings and well logs obtained from the Linde Air Products Division files.

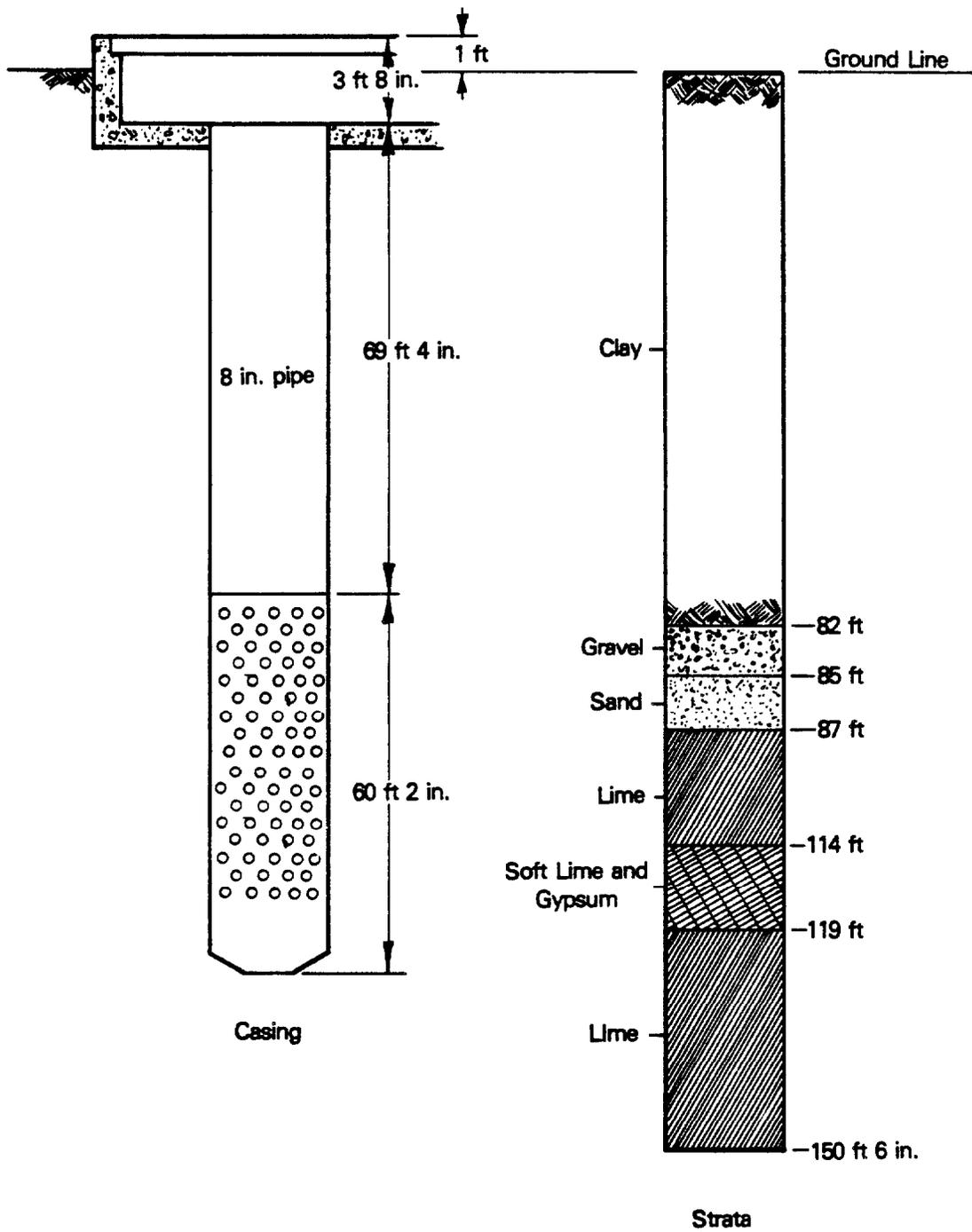


Figure D-2. Drawing of No. 2 Well Used for Disposal (at Plant No. 1)

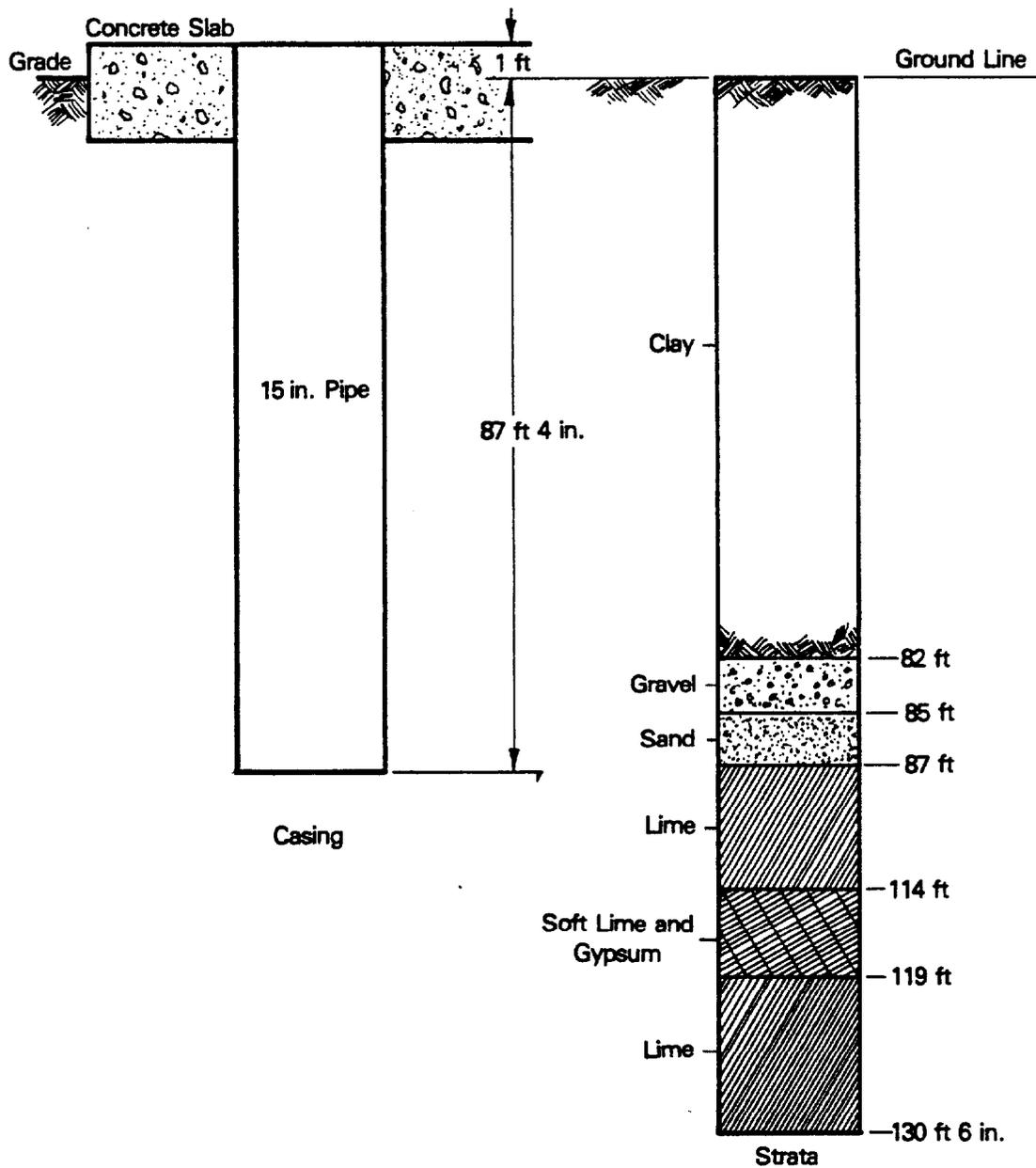


Figure D-3. Drawing of No. 3 Well Used for Disposal (at Plant No. 1)

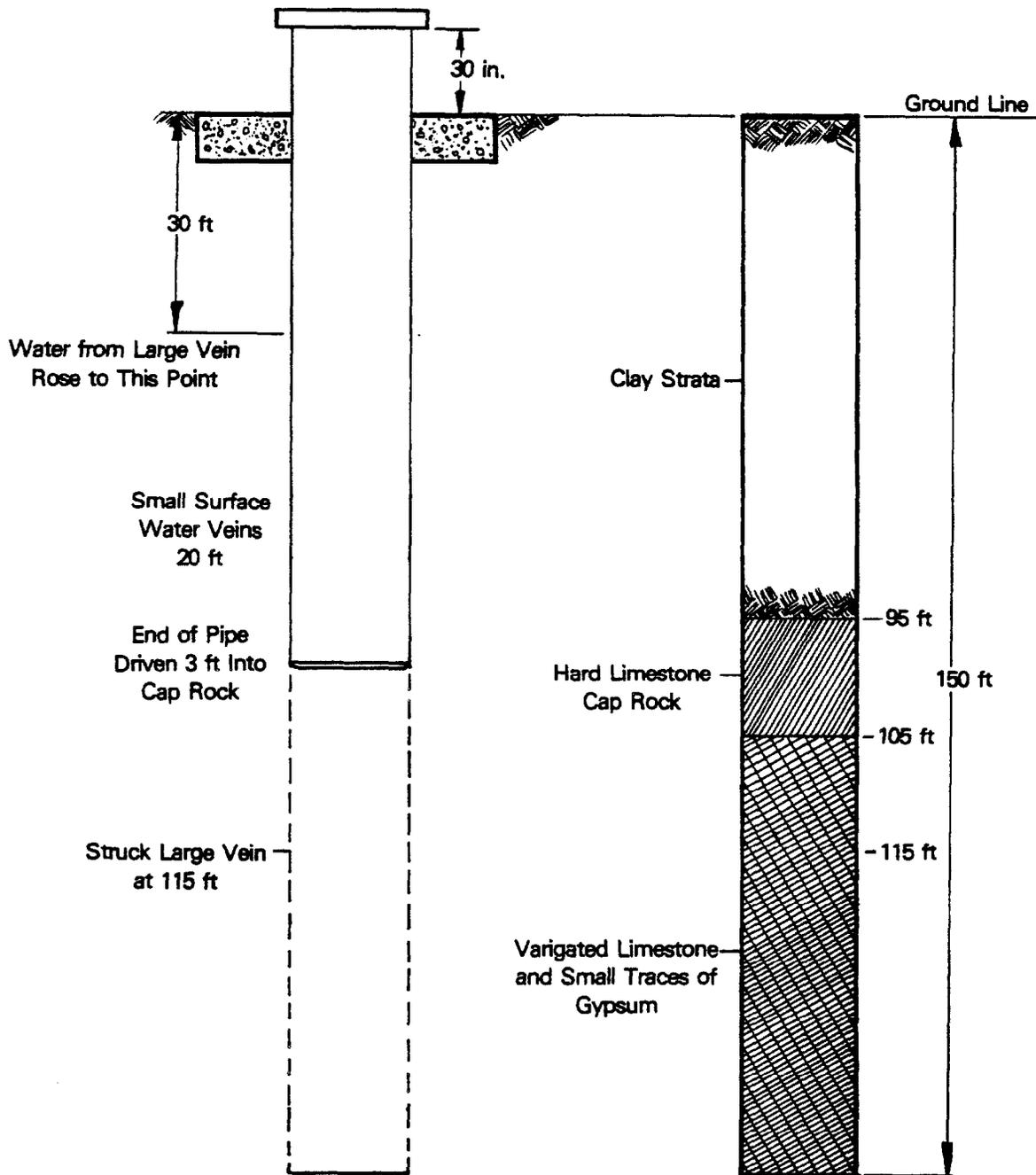


Figure D-4. Drawing of Disposal Well Drilled at Ceramics Plant

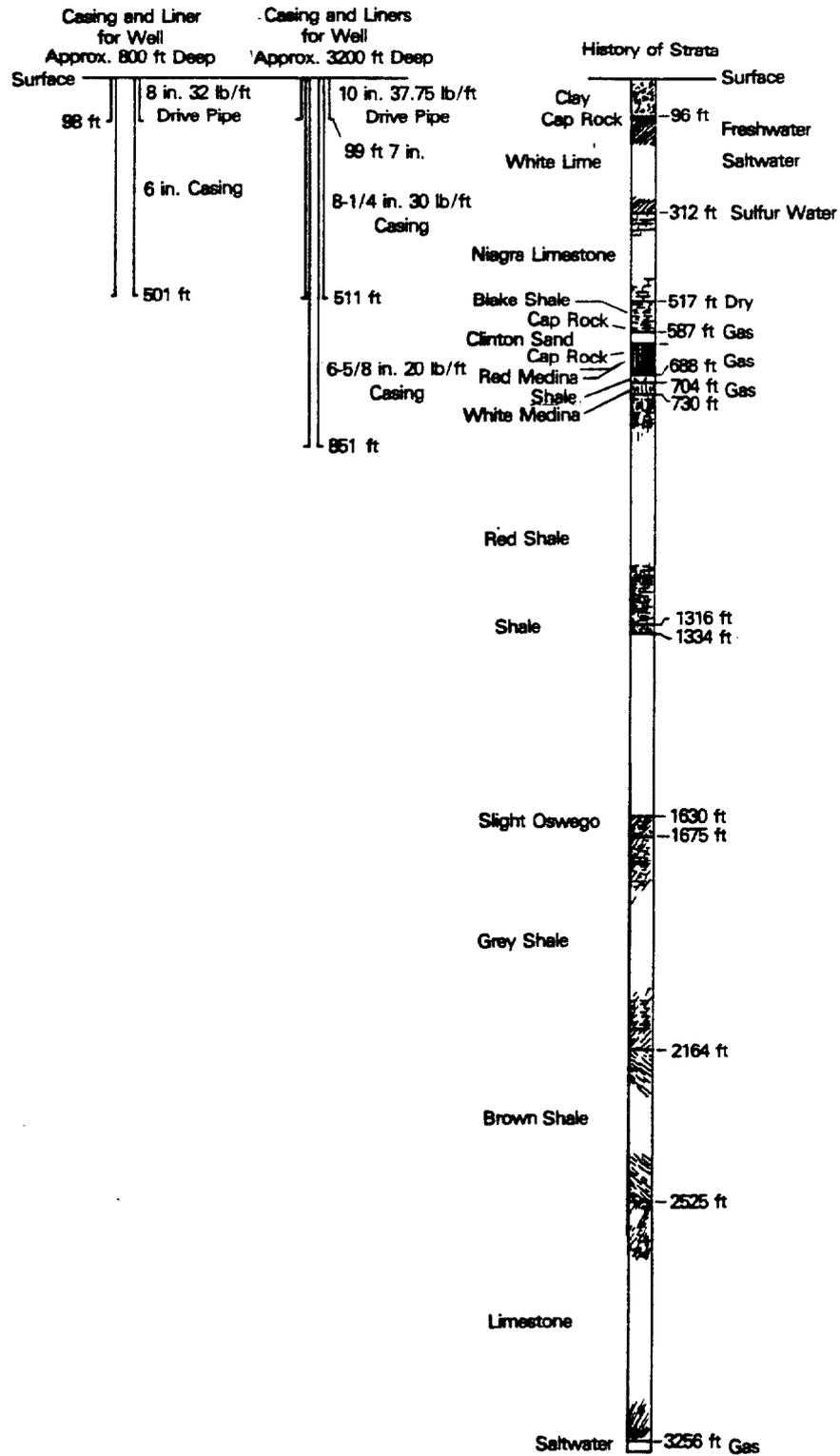


Figure D-5. Gas Well Drawing Showing Substrata

saltwater. It is also possible that the upper aquifer noted as freshwater could have been affected by the process effluents.

The practice of using wells for disposal of industrial wastes has been discouraged by local health officials since the early 1950s. Some studies suggest that this region is well suited for deep well disposal,⁵ but these wells would probably be deeper than the 150-ft Linde wells.

No information regarding uranium concentrations in groundwater or rocks in the Tonawanda area was identified. However, Rogers and Adams⁶ indicated that uranium concentrations in carbonate rocks were fairly consistent and averaged about 2.2 ppm (1.5 pCi/g). The highest concentration reported was 18 ppm, but almost all the samples had a concentration below 4 ppm. Uranium concentrations in water from carbonate aquifers were reported by Cowart.⁷ Values range from 0.1 to 4.8 $\mu\text{g}/\text{l}$ (0.07 to 3.2 pCi/l). Concentrations in groundwater samples measured off the Linde site were below or within this range; some onsite samples exceeded this range. Rogers and Adams reported that uranium concentrations in waters from mineralized aquifers were measured and ranged from 1 to 310 pCi/l. Concentrations of dissolved uranium in all groundwater samples taken during the Linde surveys were within this range.

⁵ J.J. Geraghty et al., Water Atlas of the United States, Water Information Center, 1973.

⁶ J.J.W. Rogers and J.A.S. Adams, "Uranium," Handbook of Geochemistry, Vol. II, Springer Verlaney, Inc., New York, New York, 1969.

⁷ J.B. Cowart, "Variation of Uranium Isotopes in Some Carbonate Aquifers," National Radiation Environment III, Department of Energy, Washington, D.C., 1978.

APPENDIX E. GROUNDWATER ANALYSIS BY ARGONNE NATIONAL LABORATORY

This appendix contains the results of an analysis of Linde well samples obtained by the Argonne National Laboratory Radiological Survey Group. The tables were prepared by Argonne in September 1981.

Table E-1 presents a description of the samples and total solids and measured radionuclides in the samples. The values in Table E-2 are the results presented in Table E-1 converted to microcuries per milliliter of water. Argonne included the Department of Energy guidelines from DOE 5480.1, change 2, attachment XI-1, "Concentrations in Air and Water Above Natural Background," at the end of Table E-2. The samples coded with an A are the suspended solid samples filtered from the water, and the B samples are dissolved solids (soluble material).

The ^{137}Cs , $^{137\text{m}}\text{Ba}$, Th series, and ^{226}Ra series results were obtained by gamma spectral analysis. Fluorometric analysis was performed for the uranium. The results were reported in micrograms of uranium per gram of material and converted to picocuries per gram assuming 0.699 pCi per gram of uranium.

The thorium isotopes were determined by thorium separation chemistry followed by alpha spectrometry. The analysis of ^{210}Pb involved lead separation chemistry, followed by a 3- to 4-month buildup period for ^{210}Pb , and then additional ^{210}Pb separation and alpha spectral analysis.

Table E-1. Total Solids and Measured Radionuclides in Samples

Sample No. and Description	Total Solids -(gms)	Total H ₂ O (gals.)	Total H ₂ O (liters)	Results pCi/gram			Uranium Fluorometric		Thorium Alpha Spec pCi/gram			²¹⁰ Pb pCi/gram
				¹³⁷ Cs- ^{137m} Ba	²³² Th Series (²²⁸ Ac)	²²⁶ Ra Series (²¹⁴ Bi)	ug/gram	pCi/gram	²³² Th	²³⁰ Th	²³² Th	
1A - Total Suspended Solids Linde Gas Well - Bldg. 77 14,786-85	362	6.6	24.98	.03 ± .02	< .06	.94 ± .07	154	107.66	.05 ± .02	.3 ± .1	.22 ± .065	6.8 ± 0.68
1B - Total Dissolved Solids Linde Gas Well - Bldg. 77, 14,786-85	1750	6.6	24.98	.03 ± .02	.18 ± .05	.24 ± .04	1.6	1.12	.01 ± .005	.01 ± .005	.04 ± .01	0.0055 ± .0011
2A - Total Suspended Solids Dunlop Well 14,786-85	0.628	7.0	26.5	< .03	< .06	15 ± 8	< 2.0	< 1.4	.03 ± .02	.2 ± .1	.08 ± .03	0.10 ± 0.01
2B - Total Dissolved Solids Dunlop Well 14,786-85	35.8	4.6	17.4	< .03	< .06	< .02	< 0.2	< 1.4	.01 ± .005	.01 ± .005	.04 ± .01	0.0061 ± .0016
3A - Total Suspended Solids Linde Well "C" 84.5' 14,786-84 #10	252	1.5	5.68	< .03	.62 ± .06	.8 ± .07	8.5	5.94	.5 ± .1	.5 ± .1	.5 ± .1	0.51 ± 0.05
3B - Total Dissolved Solids Linde Well "C" 84.5' 14,786-84 #10	24	1.5	5.68	.32 ± .06	< .06	< .02	2.1	1.47	.12 ± .06	.31 ± .05	.01 ± .005	0.0051 ± .0010
4A - Total Suspended Solids Linde Well "C" 89' 14,786-84 #12	378	3.0	11.35	< .03	.36 ± .07	.43 ± .04	3.3	2.31	.2 ± .1	.5 ± .1	.3 ± .1	0.24 ± 0.02
4B - Total Dissolved Solids Linde Well "C" 89' 14,786-84 #12	102	3.0	11.35	.10 ± .03	< .06	< .02	0.74	.52	< .01 ± .005	< .01 ± .005	< .01 ± .005	0.0072 ± .0015
5A - Total Suspended Solids Linde Well "C" 114' 14,786-84 #16	319	8.0	30.28	< .03	.61 ± .06	.58 ± .06	8.7	6.1	.77 ± .04	.62 ± .05	.85 ± .04	0.46 ± 0.05
5B - Total Dissolved Solids Linde Well "C" 114' 14,786-84 #16	389	8.0	30.28	< .03	.09 ± .04	.04 ± .02	0.49	.34	.007 ± .001	.018 ± .002	.014 ± .002	0.0080 ± .0016

Table E-1. Total Solids and Measured Radionuclides in Samples (Continued)

Sample No. and Description	Total Solids (gms)	Total H ₂ O (gals)	Total H ₂ O (liters)	Results pCi/gram			Uranium Fluorometric		Thorium Alpha Spec pCi/gram			²¹⁰ Pb pCi/gram
				¹³⁷ Cs- ^{137m} Ba	²³² Th Series (²²⁸ Ac)	²³⁶ Ra Series (²¹⁴ Bi)	µg/gram	pCi/gram	²³² Th	²³⁰ Th	²³⁴ Th	
6A - Total Suspended Solids Linde Well "C" 150' 14,786-84 #20	28	8.0	30.28	< .03	< .06	.34 ± .06	17.5	12.23	.51 ± .03	.55 ± .03	.75 ± .03	0.41 ± 0.04
6B - Total Dissolved Solids Linde Well "C" 150' 14,786-84 #20	316	8.0	30.28	< .03	< .06	< .02	0.41	.28	.018 ± .002	.027 ± .002	.023 ± .002	0.013 ± .0013
7A - Total Suspended Solids Well Near Wells #1, 2 and 3 87' 14,786-93	422	8.0	30.28	< .03	.84 ± .06	1.17 ± .08	12.5	8.74	.71 ± .06	.69 ± .06	.87 ± .08	0.52 ± 0.05
7B - Total Dissolved Solids Well Near Wells #1, 2 and 3 87' 14,786-93	44	8.0	30.28	< .03	< .06	< .06	3.1	2.17	.012 ± .003	.020 ± .005	.03 ± .01	0.014 ± .0017
8A - Total Suspended Solids Well Near Wells #1, 2 and 3 94' 14,786-93	207	3.0	11.35	< .03	1.03 ± .07	5.0 ± .35	2780.0	1943.5	.52 ± .08	.43 ± .1	82 ± 4	11.6 ± 1.2
8B - Total Dissolved Solids Well Near Wells #1, 2 and 3 94' 14,786-93	23	3.0	11.35	< .03	< .06	1.2 ± .3	136.0	95.1	.013 ± .004	.013 ± .004	1 ± .1	0.36 ± .036
9A - Total Suspended Solids Well Near Wells #1, 2 and 3 115' 14,786-93	588	5.0	18.92	< .03	.28 ± .08	1.32 ± .09	407.0	284.53	.52 ± .06	.54 ± .06	18 ± 2	1.8 ± 1.8
9B - Total Dissolved Solids Well Near Wells #1, 2 and 3 115' 14,786-13	43	5.0	18.92	.2 ± .1	.6 ± .2	.5 ± .2	87.0	60.82	.022 ± .003	.026 ± .003	1 ± .1	0.16 ± .016
10A - Total Suspended Solids Well Near Wells #1, 2 and 3 150' 14,786-93	79	8.0	30.28	< .03	.09 ± .05	4.8 ± .34	3740.0	2614.6	.55 ± .09	.55 ± .09	109 ± 5	16 ± 1.6
10B - Total Dissolved Solids Well Near Wells #1, 2 and 3 150' 14,786-93	39	8.0	30.28	< .03	< .06	1.07 ± .1	339.0	237.0	.023 ± .003	.035 ± .003	3 ± .1	0.26 ± .026

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1
CS

Table E-2. Analysis Results in Microcurie Per Milliliter of Water

Sample No.	Constant	¹³⁷ Cs- ^{137m} Ba	²³² Th Series (²²⁸ Ac)	²²⁶ Ra Series (²¹⁴ Pb)	U Flour.	²³² Th	²³⁰ Th	²³⁰ Th	²¹⁰ Pb
1A	.0144915933	4 x 10 ⁻¹⁰	8.69 x 10 ⁻¹⁰	1.36 x 10 ⁻⁹	1.56 x 10 ⁻⁶	7.24 x 10 ⁻¹⁰	4.35 x 10 ⁻⁹	3.19 x 10 ⁻⁹	9.85 x 10 ⁻⁸
1B	.0700560448	2.1 x 10 ⁻⁹	1.26 x 10 ⁻⁹	1.68 x 10 ⁻⁹	7.84 x 10 ⁻⁹	7.00 x 10 ⁻¹⁰	≤ 7.00 x 10 ⁻¹⁰	3.19 x 10 ⁻⁹	3.85 x 10 ⁻¹⁰
2A	.0009236981	< 7.11 x 10 ⁻¹³	< 1.42 x 10 ⁻¹²	3.55 x 10 ⁻¹⁰	3.31 x 10 ⁻¹¹	7.11 x 10 ⁻¹³	4.74 x 10 ⁻¹²	1.89 x 10 ⁻¹²	2.37 x 10 ⁻¹²
2B	.0020574713	< 6.17 x 10 ⁻¹¹	< 1.23 x 10 ⁻¹⁰	< 4.11 x 10 ⁻¹¹	< 2.87 x 10 ⁻¹⁰	2.05 x 10 ⁻¹¹	≤ 2.05 x 10 ⁻¹¹	8.23 x 10 ⁻¹¹	1.25 x 10 ⁻¹¹
3A	.0443661972	< 1.33 x 10 ⁻⁹	2.75 x 10 ⁻⁹	3.54 x 10 ⁻⁹	2.64 x 10 ⁻⁷	2.22 x 10 ⁻⁹	2.22 x 10 ⁻⁹	2.22 x 10 ⁻⁹	2.26 x 10 ⁻⁹
3B	.0042253521	1.35 x 10 ⁻⁹	< 2.54 x 10 ⁻¹⁰	< 8.45 x 10 ⁻¹¹	6.20 x 10 ⁻⁹	5.07 x 10 ⁻¹⁰	1.31 x 10 ⁻⁹	4.23 x 10 ⁻¹¹	2.15 x 10 ⁻¹¹
4A	.0333039648	< 9.99 x 10 ⁻¹⁰	1.20 x 10 ⁻⁹	1.43 x 10 ⁻⁹	7.68 x 10 ⁻⁹	6.66 x 10 ⁻⁹	1.66 x 10 ⁻⁹	9.9 x 10 ⁻⁹	7.99 x 10 ⁻⁹
4B	.0089867841	8.98 x 10 ⁻¹⁰	< 5.39 x 10 ⁻¹⁰	1.79 x 10 ⁻¹⁰	4.65 x 10 ⁻⁹	< 8.99 x 10 ⁻¹¹	< 8.99 x 10 ⁻¹¹	< 8.99 x 10 ⁻¹¹	6.47 x 10 ⁻¹¹
5A	.0105350066	< 3.16 x 10 ⁻¹⁰	6.43 x 10 ⁻⁹	6.11 x 10 ⁻⁹	6.41 x 10 ⁻⁹	8.11 x 10 ⁻⁹	8.64 x 10 ⁻⁹	8.95 x 10 ⁻⁹	4.85 x 10 ⁻⁹
5B	.0128467635	< 3.85 x 10 ⁻¹⁰	1.16 x 10 ⁻⁹	5.14 x 10 ⁻¹⁰	4.40 x 10 ⁻⁹	8.99 x 10 ⁻¹¹	2.31 x 10 ⁻¹⁰	1.80 x 10 ⁻¹⁰	1.03 x 10 ⁻¹⁰
6A	.0009247028	< 2.77 x 10 ⁻¹¹	< 5.55 x 10 ⁻¹¹	3.14 x 10 ⁻¹⁰	1.13 x 10 ⁻⁹	4.72 x 10 ⁻¹⁰	5.09 x 10 ⁻¹⁰	6.94 x 10 ⁻¹⁰	3.79 x 10 ⁻¹⁰
6B	.0127476882	< 3.82 x 10 ⁻¹⁰	< 7.65 x 10 ⁻¹⁰	< 2.55 x 10 ⁻¹⁰	3.65 x 10 ⁻⁹	2.29 x 10 ⁻⁹	3.44 x 10 ⁻¹⁰	2.93 x 10 ⁻¹⁰	3.57 x 10 ⁻¹⁰
7A	.0139366918	< 4.18 x 10 ⁻¹⁰	1.17 x 10 ⁻⁹	1.63 x 10 ⁻⁹	1.22 x 10 ⁻⁷	9.90 x 10 ⁻⁹	9.61 x 10 ⁻⁹	1.21 x 10 ⁻⁹	7.25 x 10 ⁻⁹
7B	.0014531044	< 4.36 x 10 ⁻¹¹	< 8.72 x 10 ⁻¹¹	< 2.90 x 10 ⁻¹¹	3.15 x 10 ⁻⁹	1.74 x 10 ⁻¹¹	2.91 x 10 ⁻¹¹	4.36 x 10 ⁻¹¹	2.03 x 10 ⁻¹¹
8A	.0182378855	< 5.47 x 10 ⁻¹⁰	1.88 x 10 ⁻⁹	9.12 x 10 ⁻⁹	3.54 x 10 ⁻⁵	9.48 x 10 ⁻⁹	7.84 x 10 ⁻⁹	1.50 x 10 ⁻⁶	2.12 x 10 ⁻⁷
8B	.0020264317	< 6.08 x 10 ⁻¹¹	< 1.22 x 10 ⁻¹⁰	2.43 x 10 ⁻⁹	1.93 x 10 ⁻⁷	2.63 x 10 ⁻¹¹	2.63 x 10 ⁻¹¹	2.03 x 10 ⁻⁹	7.30 x 10 ⁻¹⁰
9A	.0310782241	< 9.32 x 10 ⁻¹⁰	8.70 x 10 ⁻⁹	4.10 x 10 ⁻⁹	8.84 x 10 ⁻⁶	1.62 x 10 ⁻⁹	1.68 x 10 ⁻⁹	5.59 x 10 ⁻⁷	5.72 x 10 ⁻⁸
9B	.0022727273	4.55 x 10 ⁻¹⁰	1.36 x 10 ⁻⁹	1.14 x 10 ⁻⁹	1.38 x 10 ⁻⁷	5.00 x 10 ⁻¹¹	5.91 x 10 ⁻¹¹	2.27 x 10 ⁻⁹	3.64 x 10 ⁻¹⁰
10A	.0026009828	< 7.83 x 10 ⁻¹¹	2.35 x 10 ⁻¹⁰	1.25 x 10 ⁻⁹	6.82 x 10 ⁻⁶	1.43 x 10 ⁻⁹	1.43 x 10 ⁻⁹	2.8 x 10 ⁻⁷	4.17 x 10 ⁻⁸
10B	.0012879789	< 3.86 x 10 ⁻¹¹	< 7.73 x 10 ⁻¹¹	1.38 x 10 ⁻⁹	3.05 x 10 ⁻⁷	2.96 x 10 ⁻¹¹	4.51 x 10 ⁻¹¹	3.86 x 10 ⁻⁹	3.35 x 10 ⁻¹⁰
		¹³⁷ Cs 5-2 x 10 ⁻⁵ 1-4 x 10 ⁻⁵	^{Thnat.} 5-1 x 10 ⁻⁶ 1-1 x 10 ⁻⁵	²²⁶ Ra 5-3 x 10 ⁻⁶ 1-3 x 10 ⁻⁵	^{U_{nat.}**} 5-6 x 10 ⁻⁷ 1-2 x 10 ⁻⁵	²³² Th 5-2 x 10 ⁻⁶ 1-4 x 10 ⁻⁵	²³⁰ Th 5-7 x 10 ⁻⁶ 1-1 x 10 ⁻⁵	²³⁰ Th 5-2 x 10 ⁻⁶ 1-3 x 10 ⁻⁵	²¹⁰ Pb 5-1 x 10 ⁻⁷ 1-2 x 10 ⁻⁶

*Natural Thorium Curie (4.44 x 10¹² dpm/Curie)

**Natural Uranium Curie (4.45 x 10¹² dpm/Curie)