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ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY

ANNUAL REPORT FOR 1983

by

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TABLE OF CONTENTS

TABLE OF CONTENTS

LIST OF TABLES

LIST OF TABLES

LIST OF FIGURES

ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY
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ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1983 are presented and discussed. To evaluate the effect of Argonne operations on the environment, measurements were made for a variety of radionuclides in air, surface water, soil, grass, bottom sediment, and milk; for a variety of chemical constituents in air, surface water, ground water, and Argonne effluent water; and of the environmental penetrating radiation dose. Sample collections and measurements were made at the site boundary and off the Argonne site for comparison purposes. Some on-site measurements were made to aid in the interpretation of the boundary and off-site data. The potential radiation dose to off-site population groups is also estimated. The results of the program are interpreted in terms of the sources and origin of the radioactive and chemical substances (natural, fallout, Argonne, and other) and are compared with applicable environmental quality standards.

I. INTRODUCTION

A. General

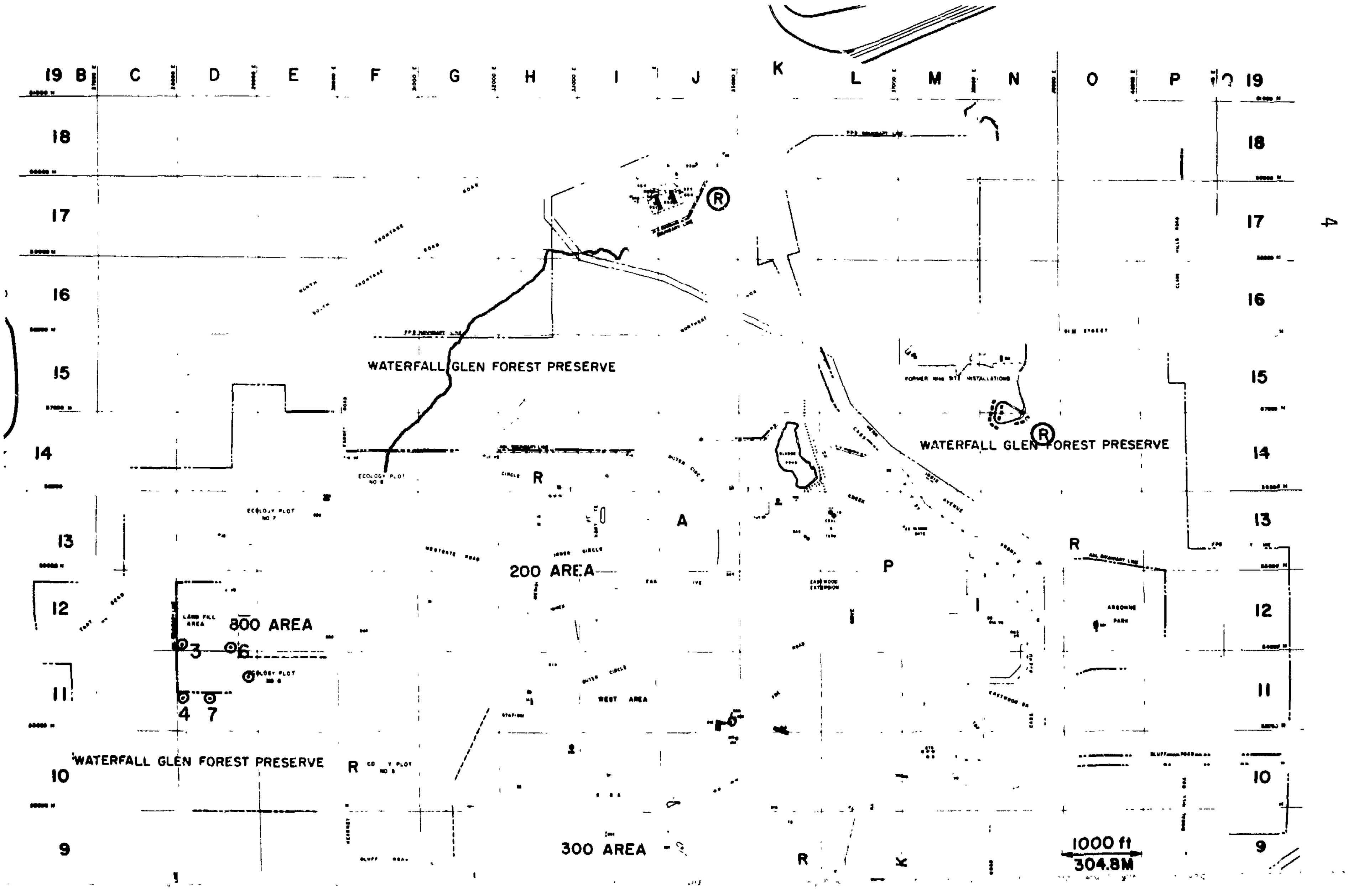
This report is prepared to provide the U. S. Department of Energy (DOE) and the public with information on the levels of radioactive, chemical, and biological pollutants in the environment of Argonne National Laboratory (ANL) and on the amounts, if any, added to the environment as a result of Argonne operations. The report follows the guidelines given in DOE Order 5484.1, Chapter III.[1] The Laboratory conducts a continuous environmental monitoring program on and near the Argonne site whose primary purpose is to determine the magnitude, origin, and identity of radioactive or toxic chemical substances in the environment. Of special interest is the detection of any such materials released to the environment by Argonne. One important function of the program is to verify the adequacy of Argonne's pollution controls.

Argonne is a DOE energy research and development laboratory with several principal objectives. It conducts a broad program of research in the basic energy and related sciences (physical, chemical, material, nuclear, biomedical, and environmental), and serves as an important engineering center for the study of nuclear and non-nuclear energy sources. Some of the energy-related research projects conducted during 1983 were safety studies for light water and breeder reactors, development of components and materials for fission and fusion reactors, improvements in the utilization of coal for power production (particularly high sulfur coal), electrochemical energy sources including fuel cells and batteries for vehicles, solar energy utilization, ocean thermal energy conversion (OTEC) using ammonia and freon as the working fluids, evaluation of heat exchangers, and operation of the National Battery Test Laboratory. Other areas of research are the use of superconducting magnets for improved nuclear particle accelerators and magnetohydrodynamics coal technology, magnetic confinement fusion, the immobilization of radioactive waste products for safe disposal, medical radioisotope technology, carcinogenesis, and the biological effects of small amounts of radiation. Environmental research studies include biological activity of energy-related mutagens and carcinogens, chemistry of actinides in natural waters, the effect of sulfur dioxide on crop growth, and land reclamation. A significant portion of these laboratory studies requires the use of radioactive and chemically-toxic substances.

The principal nuclear facilities at the Laboratory are a 200 kW light-water cooled and moderated biological research reactor (Janus) fueled with enriched uranium; the Argonne Thermal Source Reactor (ATSR), a 10 kW *research reactor fueled with enriched uranium; a superconducting heavy ion linear accelerator (Argonne Tandem Linac Accelerating System, ATLAS); a 60-inch cyclotron; several other charged particle accelerators (principally of the Van de Graaff and Dynamitron type); a large fast neutron source (Intense Pulsed Neutron Source, IPNS) in which high energy protons strike a heavy metal target to produce the neutrons; cobalt-60 irradiation sources; chemical and metallurgical plutonium laboratories; and several hot cells and laboratories designed for work with multicurie quantities of the actinide elements and with irradiated reactor fuel elements. Two major facilities, a 12.5 GeV proton accelerator (Zero Gradient Synchrotron, ZGS) and a 5 MW heavy-water enriched-uranium reactor (CP-5) were not in operation during 1983. The CP-5 facility is awaiting decontamination and decommissioning, while this work has been completed at the ZGS. Also located on the ANL site is the DOE New Brunswick Laboratory, a safeguards plutonium and uranium measurements and analytical chemistry laboratory. The principal non-nuclear activities at Argonne that may produce a measurable impact on the environment are a coal-fired boiler (No. 5) and the OTEC studies. The boiler is designed to burn high sulfur (3.5%) Illinois coal to produce export steam for Laboratory use. It is equipped with a slaked lime spray scrubber and bag collector to reduce sulfur dioxide and particulate emissions. The OTEC system has the potential for water pollution since it uses large amounts of ammonia. Coal conversion and carcinogen studies are a secondary potential pollution source because the quantities involved are small.*

B. Description of Site

Argonne National Laboratory (Illinois site) occupies the central 688 hectares (1,700 acres) of a 1514-hectare (3,740-acre) tract in DuPage County, 43 km (27 mi) southwest of downtown Chicago, and 39 km (24 mi) due west of Lake Michigan. It lies in the Des Plaines River Valley, south of Interstate Highway 55 and west of Illinois Highway 83. Figures 1 and 2 are maps of the site and of the surrounding area. The 826-hectare (2,040-acre) area surrounding the site (Waterfall Glen Forest Preserve) was formerly Argonne property, but was deeded to the DuPage County Forest Preserve



19 B C D E F G H I J K L M N O P Q 19
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WATERFALL GLEN FOREST PRESERVE
WATERFALL GLEN FOREST PRESERVE
WATERFALL GLEN FOREST PRESERVE
ECOLOGY PLOT NO 8
ECOLOGY PLOT NO 7
ECOLOGY PLOT NO 6
R A P R K R
200 AREA
800 AREA
300 AREA
LAND FILL AREA
WEST AREA
FORMER NIKE SITE INSTALLATIONS
ARGONNE PARK
ELM STREET
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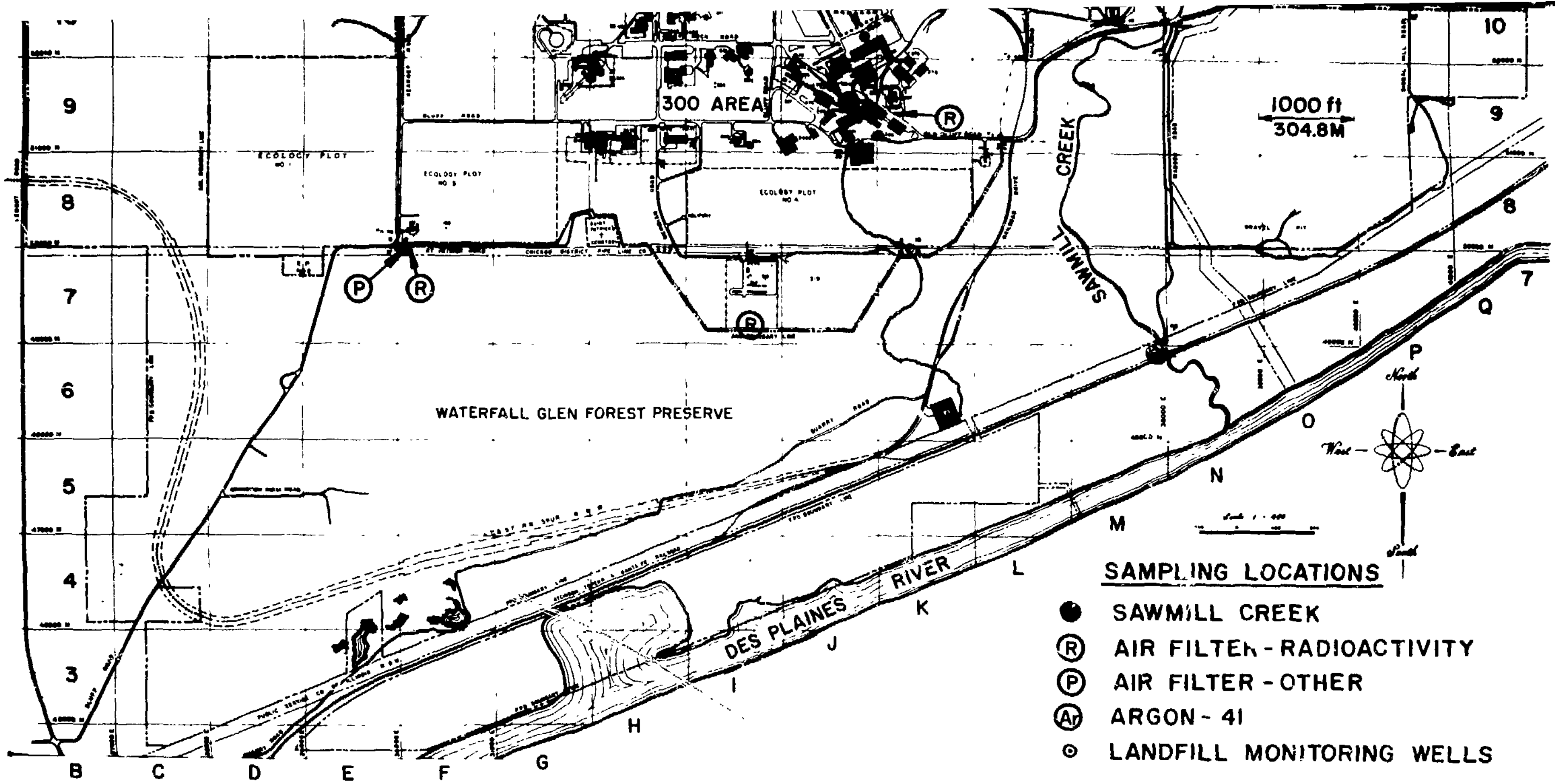

Fig. 1. Sampling Locations at Argonne National Laboratory

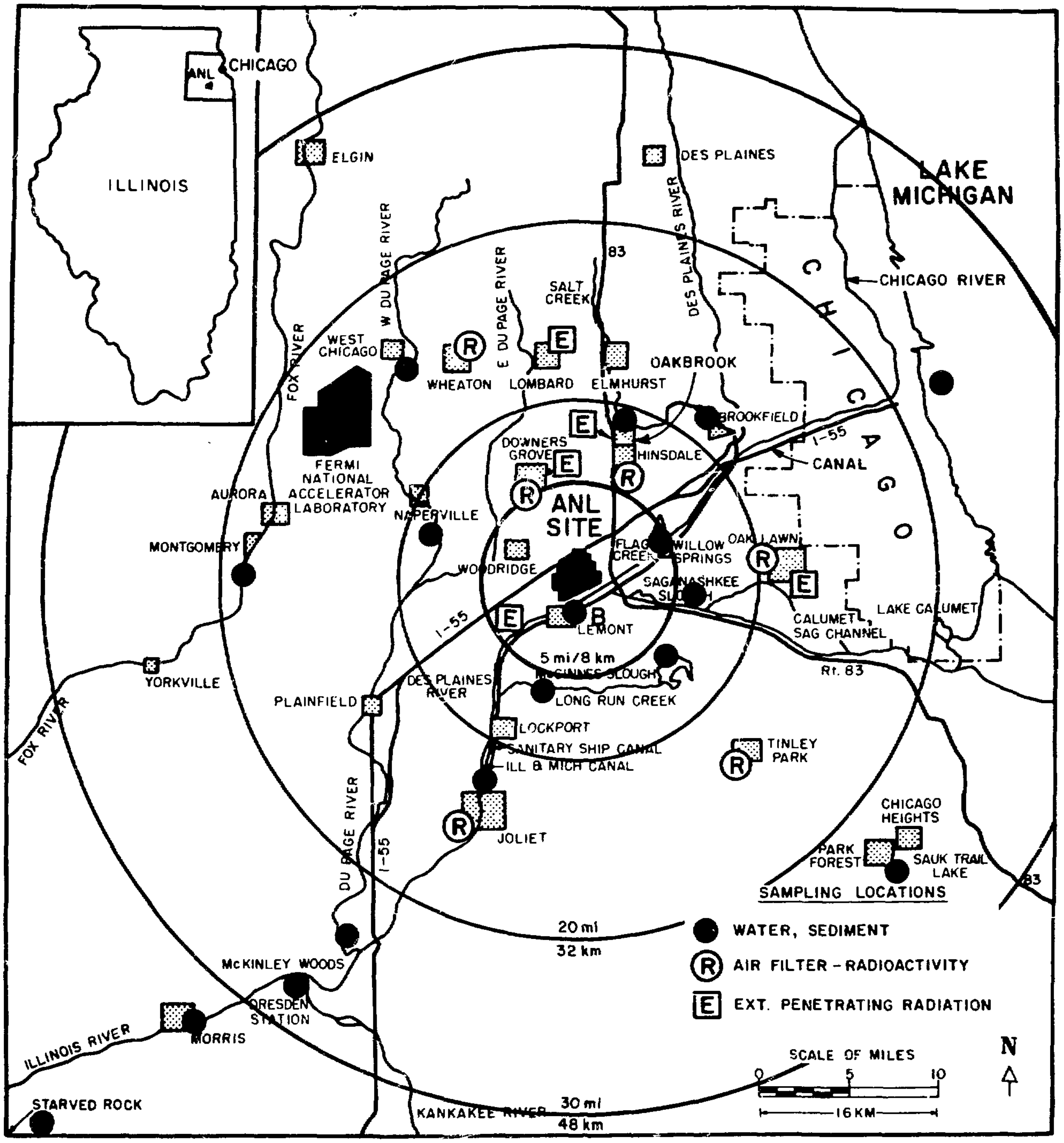

Fig. 2. Sampling Locations Near Argonne National Laboratory

District in 1973 for their use as a public recreational area, nature preserve, and demonstration forest.

The terrain is gently rolling, partially-wooded, former prairie and farmland. The grounds contain a number of small ponds and streams, the principal one being Sawmill Creek, which runs through the site in a southerly direction and enters the Des Plaines River about 2.1 km (1.3 mi) southeast of the center of the site. The land is drained primarily by Sawmill Creek, although the extreme southern portion drains directly into the Des Plaines River, which flows along the southern boundary of the Forest Preserve. This river flows southwest until it joins the Kankakee River about 48 km (30 mi) southwest of the Laboratory to form the Illinois River.

The largest topographical feature is the Des Plaines River channel, about 1.6 km (1 mi) wide. This channel contains the River, the Chicago Sanitary and Ship Canal, and the Illinois and Michigan Canal. Their presence extends the uninhabited area about 1.6 km (1 mi) south of the site. The elevation of the channel surface is 180 m (590 ft) above sea level. Bluffs, which comprise the south border of the site, rise from the channel at varying slope angles of 15° to 60°, reaching an average elevation of 200 m (650 ft) above sea level at the top. The land then slopes gradually upward reaching the average site elevation of 220 m (725 ft) above sea level at 940 m (3,000 ft) from the bluffs. Several large ravines oriented in a north-south direction are located in the southern portion of the site. The bluffs and ravines generally are forested with mature deciduous trees. The remaining portion of the site changes in elevation by no more then 7.6 m (25 ft) in a distance of 150 horizontal m (500 ft). In the southern portion of the Forest Preserve, the Chicago District Pipe Line Co. and the Atchison, Topeka, and Santa Fe Railroad have rights-of-way. Additional information about the site is given in the Argonne Environmental Assessment.[2]

C. Population

The area around Argonne has exhibited a large population growth in the past 20 years. Large areas of farmland have been converted into housing. A directional and annular 80-km (50-mi) population distribution for the area, which is used for the population dose calculations later in this report, is shown in Table 1. The population distribution was obtained by modifying a distribution for 1981 prepared by Urban Decision Systems, Inc. and which was based on the 1980 census. The values for distances within 8 km (5 mi) of the site were modified by using quarter-section population data supplied by the Northeastern Illinois Planning Commission, as adjusted on the basis of local observations.

D. Meteorology

The climate of the area is that of the upper Mississippi Valley, as moderated by Lake Michigan. A summary of the meteorological data collected on the site from 1950 to 1964 is available[3] and provides an adequate sample of the climate.

The most important meteorological parameters for the purposes of this report are wind direction, wind speed, temperature, and precipitation. The wind data are used to calculate radiation doses from air emissions and in selecting air sampling locations and distances. Temperature and precipitation data are useful in interpreting some of the environmental results. Since on-site meteorological data are no longer available in suitable form, measurements taken in 1983 at the nearest U. S. Weather Bureau station, O'Hare Airport, 31 km (18 mi) north of Argonne, were used. The 1983 average monthly and annual wind roses are shown in Figure 3. The wind roses are polar coordinate plots in which the lengths of the radii represent the percentage frequency of wind speeds in classes of 2.01-6 m/s (4.5-13.4 mph), 6.01-10 m/s (13.4-22.4 mph), and greater than 10.01 m/s (22.4 mph). The number in the center represents the percent of observations of wind speed less than 2 m/s (4.5 mph) in all directions. The direction of the radii represents the direction from which the wind blows. Sixteen radii are shown on each plot at 22.5° intervals; each

TABLE 1

Incremental Population Data in the Vicinity of ANL, 1981

						In Thousands				
Distance, miles Distance, km	0 - 1 0-1.6	1 - 2 1.6-3.2	2 - 3 3.2-4.8	3 - 4 4.8-6.4	4 - 5 6.4-8.0	5-10 8-16	10-20 16-32	20-30 32-48	30-40 48-64	40-50 64-80
<u>Direction</u>										
N	0	344	1504	863	4115	37.2	179.2	312.1	133.3	202.1
NNE	0	188	2086	14685	5882	38.8	290.7	493.4	95.9	0
NE	0	528	6544	1450	1219	44.0	710.1	940.7	0	0
ENE	0	2630	3640	1854	985	35.6	630.5	240.8	0	0
E	0	14	212	20	15	34.4	514.9	249.4	10.7	25.2
ESE	0	0	85	275	120	11.3	206.2	291.9	271.0	69.0
SE	0	5	155	225	68	29.0	69.5	119.2	24.4	13.3
SSE	0	44	2299	1422	120	1.9	21.7	9.3	9.2	20.0
S	0	100	574	2114	725	5.5	18.5	1.8	33.0	39.5
SSW	0	60	4407	1928	705	19.1	100.9	9.4	17.7	7.5
SW	0	620	1304	50	915	13.1	31.5	6.5	15.0	7.8
WSW	0	492	50	409	12261	3.3	7.1	2.1	6.3	9.4
W	0	2853	905	14000	16464	4.1	58.7	19.6	15.0	6.6
WNW	0	1007	140	5100	5960	39.8	85.5	8.7	7.7	50.3
NW	0	215	2032	3367	7741	28.5	65.2	87.2	10.5	16.6
NNW	0	323	987	2156	7710	41.1	151.2	167.1	107.7	79.5
Total	0	9423	26924	49918	65005	386.7	3141.4	2959.2	757.4	546.8
Cumulative Total	0	9423	36347	86265	151270	538.0	3679.4	6638.6	7396.0	7942.8

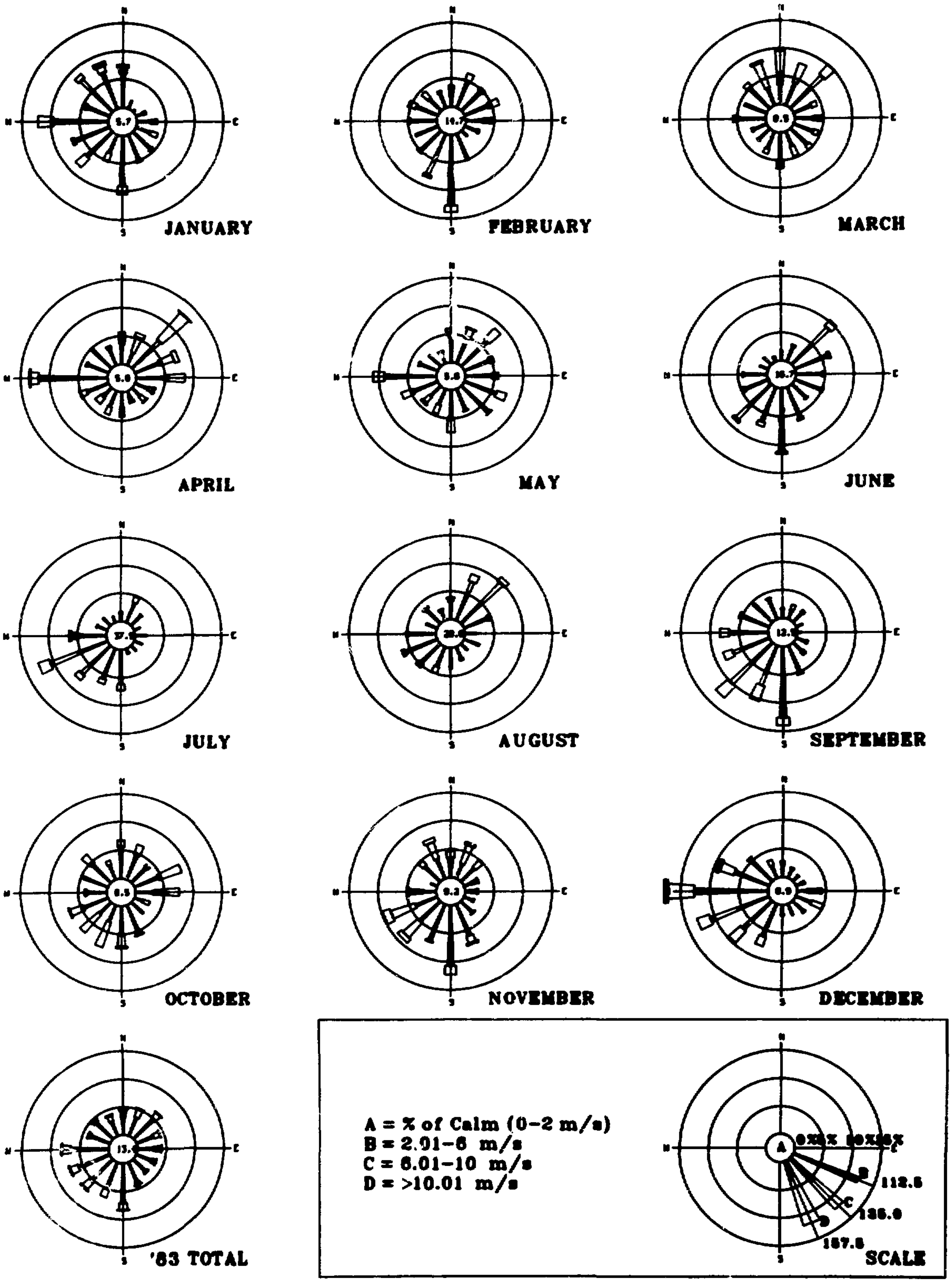

Fig. 3. Monthly and Annual Wind Roses Near Argonne National Laboratory, 1983

radius represents the average wind speed for the direction covering 11.25° on either side of the radius.

A comparison of the monthly wind roses indicates that the winds are sufficiently variable so that monitoring for airborne releases must be carried out in all directions from the site. For example, the dominant wind direction in July is west-southwest while in August it is northeast. The annual average wind rose for 1983 is consistent with the long term average wind direction, which is usually from the west to south.

The precipitation and temperature data for 1983 are shown in Table 2. Temperatures were normal in 1983 compared to the long term average. Significantly above normal amounts of precipitation in April, May, September, October, and November resulted in a total precipitation in 1983 of 122.5 cm (48.2 in), about 43% higher than the long term annual average.

E. Geohydrology

The geology of the Laboratory area consists of about 30 m (100 ft) of glacial till overlying dolomite bedrock. The bedrock is the Niagaran and Alexandrian dolomite of Silurian age. These formations are underlain by Maquoketa shale of Ordovician age, and older dolomites and sandstones of Ordovician and Cambrian age. The beds are nearly horizontal.

Two principal aquifers are used as water supplies in the vicinity of the Laboratory area. The upper aquifer is the Niagaran-Alexandrian dolomite, which is about 60 m (200 ft) thick in the Laboratory area, and has a piezometric surface between 15 and 30 m (50 and 100 ft) below the ground surface over much of the site. The lower aquifer is the Galesville sandstone, which lies between 150 and 450 m (490 and 1,500 ft) below the surface. The Maquoketa shale separates the upper dolomite aquifer from the underlying sandstone aquifer. This shale retards hydraulic connection between the upper and lower aquifers.

The four domestic water wells now in use are about 90 m (300 ft) deep in the Niagaran dolomite, about 300 feet from the ground surface. One

TABLE 2

ANL Weather Summary, 1983

	Precipitation (cm)			Temperature (°C)		
Month	Amount	Average		Monthly Average	Average	
		(1)	(2)		(3)	(4)
January	1.68	4.88	4.32	-3.2	-4.2	-6.6
February	5.23	4.57	3.20	-0.8	-2.7	-3.9
March	9.04	6.93	6.76	3.0	2.5	2.3
April	19.53	7.98	9.73	6.3	8.8	9.2
May	15.90	8.81	8.15	11.8	14.6	15.1
June	10.44	9.47	10.36	20.9	20.2	20.1
July	10.80	8.66	9.40	24.8	23.2	22.6
August	5.28	8.10	9.25	25.2	22.5	22.1
September	13.74	8.08	9.53	18.1	18.7	18.0
October	11.20	6.55	5.21	11.6	12.5	11.7
November	12.60	5.88	5.87	5.6	4.7	4.4
December	7.03	5.30	6.10	-9.8	-1.5	-2.6
Total	122.5	85.20	87.88			

U. S. Weather Bureau Averages
(1) 1873-1977 (2) 1957-1982
(3) 1871-1977 (4) 1957-1982

well, in the Galesville sandstone 490 m (1,600 ft) deep, is not used because the water table has dropped below the pumping level. The water level in the Niagaran dolomite has remained reasonably stable under Laboratory pumping, dropping about 3.7 m (12 ft) between 1960 and 1980. The aquifer appears to be adequate for future Laboratory use, but this ground water source is used throughout the area. There are also several monitoring wells and small capacity water wells used for laboratory experiments, fire protection, and sanitary facilities.

F. Water and Land Use

The principal stream that drains the site is Sawmill Creek. It carries effluent water continuously from a sewage treatment plant (Marion Brook Treatment Plant) located a few kilometers north of the site. The plant has an operating capacity of about 11.4 megaliters (3 million gallons) per day. In addition, the residential development in the area has resulted in the collection and channeling of additional runoff water into the Creek. Treated sanitary and laboratory waste water from Argonne are combined and discharged into Sawmill Creek at location 7M in Figure 1. This effluent averaged 3.1 megaliters (0.82 million gallons) per day, while the water flow in the Creek upstream of the waste-water outfall averaged about 38 megaliters (10 million gallons) per day during 1983. The combined Argonne effluent consisted of 70% laboratory waste water and 30% sanitary waste water.

Sawmill Creek and the Des Plaines River above Joliet, about 21 km (13 mi) southwest of Argonne, receive very little recreational or industrial use. A few people fish in these waters downstream from Argonne and some duck hunting takes place on the Des Plaines River. Water from the Chicago Sanitary and Ship Canal is used by Argonne for cooling towers and by others for industrial purposes, such as hydroelectric generators and condensers, and for irrigation at the State prison near Joliet. The Argonne usage is about 0.4 megaliter (100,000 gallons) per day. The Canal, which receives Chicago Metropolitan Sanitary District effluent water, is used for industrial transportation and some recreational boating. Near Joliet, the River and Canal are combined into one waterway, which continues until *it joins the Kankakee River to form the Illinois River about 48 km (30 mi) southwest of Argonne. The Dresden Nuclear Power Station complex is located at the confluence of the Kankakee, Des Plaines, and Illinois Rivers. This Station uses water from the Kankakee River for cooling, and discharges the water into the Illinois River. The first use of water for drinking is an indirect one at Alton, on the Mississippi River about 710 km (370 mi) downstream from Argonne, where water is used to replenish ground water supplies by infiltration. In the vicinity of the Laboratory, only subsurface water (from both shallow and deep aquifers) and Lake Michigan water are used for drinking purposes.*

The principal recreational area near Argonne is Waterfall Glen Forest Preserve, which surrounds the site as described in Section I.B. and is shown in Figure 1. The area is available for hiking, skiing, and equestrian sports. Sawmill Creek flows through the south portion of the Preserve on its way to the Des Plaines River. Several large forest preserves of the Cook County Forest Preserve District are located east and southeast of Argonne and the Des Plaines River. The preserves include the two sloughs shown in Figure 2, McGinnis and Saganashkee, as well as other smaller lakes. These areas are used for picnicking, boating, fishing, and hiking. A small park located in the eastern portion of the Argonne site (12-0 in Figure 1) is for the use of Argonne and Department of Energy employees only.

The most recently available information on milk and principal agricultural production in a 10,600 sq km (4,100 sq mi) area around Argonne is shown by County in Table 3. These estimates were obtained from the Illinois Crop Reporting Service of the State Department of Agriculture. For oats, the data are for 1982; for milk, corn, soybeans, and wheat, the data are for 1983.

TABLE 3

Agricultural Production Near ANL, 1982, 1983

	Milk		Corn	Soybeans	Wheat	Oats
County	No. of Cows	Million Pounds	Million Bushels			
DuPage	200	2.3	1.16	0.65	0.062	0.24
Cook	200	2.3	0.90	0.75	0.056	0.045
Will	3,100	35.8	11.66	5.01	0.34	0.24
Kane	5,600	64.6	8.94	2.63	0.17	0.21
Kendall	600	6.9	6.30	2.54	0.088	0.079
Grundy	700	8.1	5.33	3.17	0.50	0.053
Lake	1,300	15.0	1.41	0.93	0.30	0.13

Note: To convert pounds into kilograms, multiply by 0.45; to convert bushels into cubic meters, multiply by 0.035.

II. SUMMARY

This is one in a series of annual reports prepared to provide the U. S. Department of Energy (DOE) and the public with information on the level of radioactive and chemical pollutants in the environment and on the amounts of such substances, if any, added to the environment as a result of Argonne operations. The previous report in this series is ANL-83-26.[4] Included in this report are the results of measurements obtained in 1983 for a number of radionuclides in air, surface water, ground water, soil, grass, bottom sediment, and milk; for a variety of chemical constituents in air and water, and for the external penetrating radiation dose.

Total alpha and beta activities, fission and activation products, plutonium, thorium, and uranium were measured in air-filter samples collected continuously at the site perimeter and off the site. All of these samples contained only radionuclides from natural sources and nuclear test detonations. Almost all of the radioactive materials injected into the air by previous atmospheric nuclear tests have either decayed or have been deposited on the ground. No activity attributable to Argonne operations could be detected.

The major airborne radionuclides released from the Laboratory were carbon-11, argon-41, and krypton-85. The maximum dose from these three nuclides at the site boundary was 0.04 mrem/yr and in the north direction, as calculated from an atmospheric dispersion model. The calculated dose to the closest full-time resident, who is located about 0.5 km (0.3 mi) north of the site boundary, was 0.02 mrem/yr. These releases constitute an insignificant addition to the dose received from the natural background radiation, which is about 95 mrem/yr. The total 80-km population dose from these radionuclides was less than 0.3 man-rem for 1983.

Argonne waste water is discharged into Sawmill Creek, and this stream was sampled above and below the site to evaluate the effect of Argonne operations on its radioactive content. The nuclides (for which analyses were made) added to the Creek in the waste water, and the percent of the

Concentration Guide (CG)* of their net average Creek concentrations, were hydrogen-3, 0.003%; strontium-90, < 0.02%; neptunium-237, 0.00011%; plutonium-239,240, 0.00017%; americium-241, 0.00017%; and curium-244 and/or californium-249, 0.00003%. Although Sawmill Creek is not a source of potable water, the dose to an individual using water at these concentrations as his sole source of drinking water would be less than 0.6 mrem/yr.

Sawmill Creek flows into the Des Plaines River, which in turn flows into the Illinois River. The radioactivity levels in the latter two streams were similar to those in other streams in the area, and the activity added to the Creek by Argonne waste water had no measurable effect on the radioactive content of either the Des Plaines or Illinois Rivers.

Plutonium concentrations in soil showed the same general range and average at the site perimeter and off the site as in the past years. The average plutonium-239,240 content of the top 5 cm (2 in) of soil was 1.0×10^{-3} µCi/m^2 at the site perimeter and 0.8×10^{-3} µCi/m^2 off the site. The corresponding plutonium-238 averages were 0.04×10^{-3} µCi/m^2 and 0.04×10^{-3} µCi/m^2, respectively. The plutonium content of grass was similar to that found in previous years and was about a factor of 10^4 less than soil from the same locations. The results were within the range reported by other laboratories for fallout from test detonations, and the plutonium found in soil and grass is attributed to this source. The plutonium content of samples from beds of off-site streams and ponds ranged from 1×10^{-9} µCi/g to 12×10^{-9} µCi/g of plutonium-239,240, a range found in previous years to be normal for fallout plutonium in such materials. Higher concentrations of some gamma-ray emitters and transuranic nuclides were found in the sediment just below the

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*Unless otherwise indicated, the hazard due to a given concentration of a radioactive nuclide is assessed in this report by comparison with the Concentration Guides (CG) and annual dose limits, or Radiation Protection Standards, for uncontrolled areas, specified by the U. S. Department of Energy Order 5480.1, Chapter XI.[5] The pertinent CGs are listed in the Appendix, Section V.B. Comparison with other standards is made where appropriate. The modified dose assessment methods introduced recently by the International Commission on Radiological Protection have not been used in this report for the reasons given in Section III.A.

Laboratory waste water outfall as a result of their presence in Argonne waste water.

Milk from a dairy farm near the Laboratory was analyzed for several fission products, including hydrogen-3. Hydrogen-3 concentrations averaged 182 x 10^{-9} µCi/mL. The strontium-90 concentration (3.8 x 10^{-9} µCi/mL), and the cesium-137 concentration (0.9 x 10^{-9} µCi/mL) were similar to 1982 results. These radionuclides resulted from nuclear test detonations.

Measurements of penetrating radiation were made at several locations at the site boundary and off the site. The off-site results averaged 95 mrem/yr with a standard deviation of 9 mrem/yr, which is in the normal range for the area. At two site boundary locations, above-normal readings were recorded that were attributable to Argonne operations. At the south fence (grid 7I in Figure 1), the dose rate averaged about 200 mrem/yr above normal as a result of radiation from an on-site temporary storage facility for radioactive waste. About 300 m (0.2 mi) south of the fence, the measured dose rate dropped to 105 ± 19 mrem/yr, which is slightly above the normal range. Along the north side of the site, the dose at the fence at location 14I was about 21 mrem/yr above normal due to radiation from cobalt-60 sources in Building 202. Since all of these locations are unoccupied, there are no individuals receiving these measured doses. The calculated outdoor dose rate from these sources to the residents closest to the south boundary, about 1.6 km (1 mi) from the fence line, was less than 0.03 mrem/yr. Similarly, the dose rate to the residents closest to the north boundary, about 0.75 km (0.5 mi) from the fence, was about 0.04 mrem/yr. Thus, doses to individuals near the site from these sources will not exceed 0.01% of the 500 mrem/yr limit.

Concentrations of chemical constituents and other water quality parameters were measured in Argonne waste and effluent water and in Sawmill Creek. The results were compared to the standards adopted by the State of Illinois as well as National Pollution Discharge Elimination System (NPDES) permit limits. Concentrations of mercury, hexavalent chromium, iron, and zinc in the Des Plaines River were measured to determine whether any contribution from Argonne waste water could be seen.

Results from the nine NPDES permit locations indicated general compliance, with the exception of mercury at location 001 (7M), pH at locations 006 (8J) and 009 (14K), and suspended solids at 004 (14J), 005 (14F), 006 (8J), and 009 (14K). The average mercury level of 1 μg/L (twice the permit level) resulted from releases in Argonne waste water which occurred even though reasonable care was taken to prevent them. The pH values that exceeded the limit (pH > 9.0) resulted from the alkaline water conditioning process used for Argonne domestic water and/or extremely low water levels and stagnant conditions. The high pH water has no impact on the receiving stream since the relative volume and buffering capacity of the effluent are low.

The average values in Sawmill Creek for dissolved oxygen and chemical constituents were within the State of Illinois Water Quality Standards. The average levels of copper and iron were 131 and 117% of the State standards, and individual values exceeded the standards 53 and 37% of the time. Mercury concentrations exceeded the State standard 2% of the time and averaged 24% of the standard. The mercury was released in Argonne water; the iron and copper were not related to Argonne operations. Concentrations of mercury and iron in the Des Plaines River were not affected by the amounts in Argonne effluent water.

Monitoring wells in the ANL sanitary landfill area were examined for selected inorganic and organic constituents. Levels of manganese in two of the wells were elevated as was the pH in one and arsenic in another. Organic compounds (benzene, toluene, and xylene) were not detected.

An unusual event took place during 1982 when about 7.6 megaliters (2 million gallons) of water containing about 26 mCi of tritium drained from a large excavation by a subsurface route over a period of a few days. It is believed that the water loss occurred by hydrologic piping of the till into a solutionally-enlarged opening in the underlying Niagara dolomite.

Tritium was detected in a monitoring well and a seep southwest of the excavation after the water loss. The concentrations increased, reaching a maximum about 10 days after the drainage, providing evidence on the presence and direction of movement of the water in the underlying dolomite aquifer. The maximum concentration in the well was 1.7 nCi/L, about 8% of the EPA standard for drinking water. Continued monitoring of the affected wells was conducted during 1983. Tritiated water concentrations continued to decrease and fell below the detection limit by summer. Periodic monitoring will be maintained to determine if the tritium appears at some other subsurface location.

III. MONITORING RESULTS

A. Radiological

The radioactivity of the environment was determined by measuring the concentrations of radioactive nuclides in naturally-occurring materials and by measuring the external penetrating radiation dose. Sample collections and measurements were made at the site perimeter and off the site principally for comparison purposes. Some on-site results are also reported when they are useful in interpreting perimeter and off-site results. Since radioactivity is usually spread by air and water, the sample collection program has concentrated on these media. In addition, soil, plants, foodstuffs, precipitation, and materials from the beds of lakes and streams were also collected and analyzed.

The results of radioactivity measurements are expressed in this report in terms of microcuries per milliliter (µCi/mL) for water, air, and milk and microcuries per gram (g) and/or square meter (m^2) for soil, bottom sediment, and vegetation. When a nuclide was not detected, the result is given as less than (<) the minimum amount detectable (detection limit) by the analytical method used. Averages, including individual results that were less than the detection limit, were calculated by one of the following two methods. If a large fraction (usually 50% or more) of the individual results was less than the detection limit, the average was calculated with the assumption that such results were equal to the detection limit, and the resulting average value is expressed as less than (<) the computed average. If only a small fraction of the individual results was less than the detection limit, the average was calculated with the assumption that such results were actually one-half of the detection limit, and the average is given as a definite value. The former technique probably overestimates the average concentration in those samples below the detection limit and gives an upper limit for the average of all the samples in the group, since it is unlikely that all concentrations not detectable are at the detection limit. The latter method is based on the assumption that the values below the detection limit are distributed between zero and the detection limit with a frequency such that the average value is one-half of the detection limit. The averages that are obtained by using these two methods under the conditions indicated are believed to give an adequate picture of the average concentration at locations where the concentrations not only varied greatly, but were at times not detectable. Penetrating radiation measurements are reported in units of millirem (mrem) per year and population dose in man-rem.

Average values are usually accompanied by a plus-or-minus (±) limit value. Unless otherwise stated, this value is the standard error at the 95% confidence level calculated from the standard deviation of the average, and is a measure of the range in the concentrations encountered at that location. It does not represent the conventional uncertainty in the average of repeated measurements on the same or identical samples. Since many of the variations observed in environmental radioactivity are not random but occur for specific reasons (e.g., nuclear testing), samples collected from the same location at different times are not replicates. The more random the variation in activity at a particular location, the closer the confidence limits will represent the actual distribution of values at that location. The averages and confidence limits should be interpreted with this in mind. When a plus-or-minus figure accompanies an individual result in this report, it represents the statistical counting error at the 95% confidence level.

The measured concentration or radiation dose is compared with appropriate standards as a means of assessing the hazard. The standards used in this report are usually the Concentration Guides (CGs) and annual dose limits (Radiation Protection Standards) for uncontrolled areas given in DOE Order 5480.1, Chapter XI.[5] The pertinent CGs as well as the detection limits are given in the Appendix, Section V.B. Although the CGs apply to concentrations above natural levels, the percent of CG is sometimes given in this report for activities that are primarily of natural origin for comparison purposes. Such values are enclosed in parentheses to indicate this. Where other standards are used, their source is identified in the text.

The new dose assessment and evaluation procedures and systems recommended by the International Commission on Radiological Protection (ICRP) in their reports ICRP-26,[6] et seq, are not used in this report for the following reasons. These recommendations have not yet been officially adopted by DOE, although it is anticipated that they will be used in some form by DOE and all regulatory bodies in the future. The annual whole body dose limit recommended by ICRP-26 for both occupational and public exposures has not changed from their earlier recommendations, and these are already in effect as DOE standards; and the dose to off-site individuals and the population from Argonne operations, as will be shown, is very small and entirely whole body. In addition, whole body equivalent doses calculated from the annual limit of intake and the corresponding derived air and water concentrations are not different in any significant way from the organ doses calculated from the Concentration Guides and dose conversion factors used in this report.

1. Air

The radioactive content of particulate matter was determined by collecting and analyzing air-filter samples. The sampling locations are shown in Figures 1 and 2. Separate collections were made for specific radiochemical analyses and for alpha, beta, and gamma counting. The latter measurements were made on samples collected continuously on glass fiber filters changed weekly at eight locations at the Argonne site perimeter and at five locations off the site. The site perimeter samplers are placed at the nearest location to the site boundary fence that provides electrical power and shelter. Measurements were made at the perimeter because comparison between perimeter and off-site concentrations is necessary in evaluating and establishing the normal environmental concentration. If only off-site radioactivity were reported, their normality or origin could not be evaluated. Higher activities at the site perimeter may indicate radioactivity released by Argonne if the differences are greater than the error in sampling and measurement. Such results require investigation to determine the cause of the difference. The relative error is between 5 and 20% for most results, but approaches 100% at the detection limit.

The total alpha and beta activities in the individual weekly samples are summarized in Table 4. These measurements were made in low-background gas flow proportional counters, and the counting efficiencies used to convert counting rates to disintegration rates were those measured for radon decay products on filter paper. The average concentrations of a number of gamma-ray emitters, as determined by gamma-ray spectrometry performed on composite weekly samples are given in Table 5. The gamma-ray detector is a shielded germanium diode calibrated for each gamma-ray emitting nuclide measured.

The alpha activities, principally due to naturally-occurring nuclides, averaged the same as the past several years and were in their normal range. The average beta activity for the year, 2.7×10^{-14} *µCi/mL, was identical to 1982 average. Except for cesium-137, the fission products from previous nuclear tests found and reported in the past were not detected in 1983 due to decay and/or deposition. The most recent atmospheric nuclear test took place on October 16, 1980, by the People's Republic of China. The stratospherically produced beryllium-7 and the cesium-137 exhibit the same spring increase in concentration, indicating their stratospheric origin. The lead-210 in air is due to the radioactive decay of the naturally-occurring uranium decay series, principally from the decay of gaseous radon-222 in air.*

Samples for radiochemical analyses were collected at perimeter locations 12N and 7I (Figure 1) and off the site in Downers Grove (Figure 2). Collections were made on polystyrene filters. The total air volume filtered for the monthly samples was about 25,000 m^3*. Samples were ignited at 600°C to remove organic matter and prepared for analysis by vigorous treatment with hot hydrochloric, hydrofluoric, and nitric acids.*

Plutonium and thorium were separated on an anion exchange column and the uranium was extracted from the column effluent. Following the extraction, the aqueous phase was analyzed for radiostrontium by a standard radiochemical procedure. The separated plutonium, thorium, and uranium fractions were electrodeposited and measured by alpha spectrometry. The chemical recoveries were monitored by adding known amounts of plutonium-242,

TABLE 4

TOTAL ALPHA AND BETA ACTIVITIES IN AIR-FILTER SAMPLES, 1983*
(CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

		NO. OF	ALPHA ACTIVITY			BETA ACTIVITY		
MONTH	LOCATION	SAMPLES	AV.	MIN.	MAX.	AV.	MIN.	MAX.
JANUARY	PERIMETER	29	2.2	1.2	3.3	31	18	43
	OFF-SITE	15	2.0	0.5	8.5	22	11	36
FEBRUARY	PERIMETER	32	2.6	1.6	4.2	29	22	39
	OFF-SITE	17	1.6	0.9	2.7	23	14	33
MARCH	PERIMETER	29	2.6	1.5	4.2	26	17	37
	OFF-SITE	23	1.8	0.6	3.4	22	12	33
APRIL	PERIMETER	34	2.4	0.8	5.1	23	8	39
	OFF-SITE	22	1.8	0.6	3.4	19	7	30
MAY	PERIMETER	34	2.3	1.6	4.5	21	11	31
	OFF-SITE	21	1.9	0.7	4.2	20	12	33
JUNE	PERIMETER	32	2.8	1.1	4.1	26	13	42
	OFF-SITE	20	2.1	0.5	3.9	25	13	35
JULY	PERIMETER	36	3.0	1.2	5.2	28	14	64
	OFF-SITE	21	2.4	1.1	3.9	27	13	42
AUGUST	PERIMETER	34	2.6	1.2	4.5	27	18	34
	OFF-SITE	21	2.0	0.5	4.0	27	17	59
SEPTEMBER	PERIMETER	34	3.2	1.4	5.9	28	16	61
	OFF-SITE	21	2.5	1.4	5.0	30	17	64
OCTOBER	PERIMETER	33	2.3	1.2	5.7	26	14	56
	OFF-SITE	21	1.9	0.6	4.8	26	13	53
NOVEMBER	PERIMETER	35	2.0	0.8	4.8	26	14	37
	OFF-SITE	21	2.1	0.8	4.6	25	19	34
DECEMBER	PERIMETER	24	2.6	1.3	4.0	38	24	54
	OFF-SITE	18	2.5	0.7	5.5	37	26	52
ANNUAL SUMMARY	PERIMETER	386	2.5 ± 0.1	0.8	5.9	27 ± 1	8	64
	OFF-SITE	241	2.1 ± 0.1	0.5	8.5	25 ± 1	7	64
% CG	PERIMETER	-	(0.003)	(0.0008)	(0.006)	0.027	0.008	0.064
% CG	OFF-SITE	-	(0.002)	(0.0005)	(0.008)	0.025	0.007	0.064

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* THESE RESULTS WERE OBTAINED BY MEASURING THE SAMPLES FOUR DAYS AFTER THEY WERE COLLECTED TO AVOID COUNTING THE NATURAL ACTIVITY DUE TO SHORT-LIVED RADON AND THORON DECAY PRODUCTS. THIS ACTIVITY IS NORMALLY PRESENT IN THE AIR AND DISAPPEARS WITHIN FOUR DAYS BY RADIOACTIVE DECAY.

NOTE: (1E-15 = 10^{-15})

TABLE 5

GAMMA-RAY ACTIVITY IN AIR-FILTER SAMPLES, 1983
(CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

MONTH	LOCATION	BE7	CS137	PB210
JANUARY	PERIMETER	48	0.1	45
	OFF-SITE	49	0.1	51
FEBRUARY	PERIMETER	65	0.1	36
	OFF-SITE	61	0.1	44
MARCH	PERIMETER	84	0.2	27
	OFF-SITE	66	0.2	28
APRIL	PERIMETER	84	0.1	21
	OFF-SITE	81	0.2	23
MAY	PERIMETER	88	0.2	19
	OFF-SITE	87	0.3	23
JUNE	PERIMETER	90	0.2	23
	OFF-SITE	104	0.3	31
JULY	PERIMETER	91	0.2	29
	OFF-SITE	120	0.3	42
AUGUST	PERIMETER	89	0.2	37
	OFF-SITE	110	0.3	47
SEPTEMBER	PERIMETER	75	< 0.1	31
	OFF-SITE	94	0.1	49
OCTOBER	PERIMETER	68	< 0.1	35
	OFF-SITE	77	0.1	47
NOVEMBER	PERIMETER	73	< 0.1	34
	OFF-SITE	74	< 0.1	40
DECEMBER	PERIMETER	59	0.1	63
	OFF-SITE	60	0.1	68
ANNUAL SUMMARY	PERIMETER	76 ± [illegible]	0.1 ± 0.1	33 ± 7
	OFF-SITE	82 ± [illegible]	0.2 ± 0.1	41 ± 8
% CG (X 1E-3)	PERIMETER	(.19)	0.03	(417)
	OFF-SITE	(.20)	0.04	(513)

thorium-234, and uranium-232 tracers prior to ignition. Since alpha spectrometry cannot distinguish between plutonium-239 and plutonium-240, it should be understood that when plutonium-239 is mentioned in this report, the alpha activity due to the plutonium-240 isotope is also included. The results are given in Table 6.

The average strontium-90 and plutonium-239 concentrations decreased by a factor of about two compared to 1982 and are consistent with the decrease observed in the annual average of the long-lived nuclide cesium-137 in Table 5. Strontium-89 was not observed above the detection limit of 1 x 10^{-16} µCi/mL. Figure 4 shows the monthly plutonium-239 air concentrations for the past eleven years. The arrows indicate the approximate dates of atmospheric nuclear tests. An examination of Figure 4 indicates that plutonium in air exhibits significant increases in concentration in the spring following an atmospheric nuclear test. The magnitude of the increase is related to the size of the test.

The similarity of the concentrations and monthly variation of airborne alpha, beta, gamma, plutonium, and strontium activities at the site perimeter and off the site indicates that these activities originated in a widespread source - fallout from nuclear test detonations and naturally-occurring materials - and not a localized source such as Argonne.

The thorium and uranium concentrations are in the same range found during the past several years and are considered to be of natural origin. The amounts of thorium and uranium in a sample were proportional to the mass of inorganic material collected on the filter paper, and the bulk of these elements in the air was due to resuspension of soil. In contrast, the amount of plutonium in the air samples contributed by soil, if the resuspended soil has the same plutonium concentration as the first centimeter on the ground, ranged from about 3% in November to 40% in May of the total plutonium in the samples.

With the termination of the CP-5 reactor operation (Building 330, location 9H) on September 28, 1979, the principal source of tritiated water vapor and argon-41 in air was eliminated. The only measurable

TABLE 6

STRONTIUM, THORIUM, URANIUM AND PLUTONIUM CONCENTRATIONS IN AIRFILTER SAMPLES, 1983
(CONCENTRATIONS IN 1E-18 MICROCURIES/ML)

MONTH	LOCATION[1]	SR-90	TH-228	TH-230	TH-232[2]	U-234	U-235	U-238[2]	PU-238	PU-239[3]
JANUARY	7I	30 ± 10	11 ± 1	10 ± 1	5 ± 1	16 ± 2	1.4 ± 0.9	15 ± 2	0.4 ± 0.3	0.8 ± 0.3
	12N	20 ± 10	12 ± 1	21 ± 2	10 ± 1	32 ± 2	2.9 ± 1.0	34 ± 2	< 0.3	1.0 ± 0.4
	OFF-SITE	< 10	6 ± 2	1 ± 1	1 ± 1	2 ± 1	0.4 ± 0.9	2 ± 1	0.3 ± 0.4	0.5 ± 0.4
FEBRUARY	7I	40 ± 10	9 ± 1	10 ± 1	4 ± 1	13 ± 1	1.3 ± 0.8	14 ± 1	< 0.3	1.2 ± 0.4
	12N	40 ± 10	16 ± 2	21 ± 2	9 ± 1	26 ± 2	1.3 ± 0.8	26 ± 2	< 0.3	1.7 ± 0.5
	OFF-SITE	20 ± 10	6 ± 1	3 ± 1	2 ± 1	4 ± 1	< 0.3	4 ± 1	0.3 ± 0.2	0.4 ± 0.3
MARCH	7I	40 ± 20	13 ± 1	13 ± 1	6 ± 1	13 ± 1	0.8 ± 0.8	12 ± 1	0.3 ± 0.3	2.0 ± 0.5
	12N	60 ± 40	16 ± 1	17 ± 1	10 ± 1	18 ± 2	1.4 ± 0.9	16 ± 2	0.5 ± 0.4	1.8 ± 0.5
	OFF-SITE	110 ± 30	23 ± 3	19 ± 2	10 ± 2	16 ± 3	< 0.3	15 ± 3	0.3 ± 0.6	2.6 ± 0.8
APRIL	7I	70 ± 70	14 ± 1	11 ± 1	6 ± 1	10 ± 1	< 0.3	11 ± 1	0.4 ± 0.3	1.8 ± 0.5
	12N	50 ± 10	12 ± 1	10 ± 1	5 ± 1	11 ± 1	0.5 ± 0.8	11 ± 2	0.4 ± 0.4	2.9 ± 0.6
	OFF-SITE	40 ± 10	14 ± 2	6 ± 1	5 ± 1	6 ± 1	1.0 ± 0.9	6 ± 1	< 0.3	1.3 ± 0.5
MAY	7I	50 ± 30	39 ± 2	36 ± 2	28 ± 2	28 ± 2	0.4 ± 1.1	29 ± 2	0.3 ± 0.3	2.2 ± 0.5
	12N	60 ± 10	34 ± 2	30 ± 2	27 ± 2	24 ± 2	1.4 ± 0.8	27 ± 2	< 0.3	1.7 ± 0.6
	OFF-SITE	60 ± 10	26 ± 2	16 ± 1	13 ± 1	17 ± 1	1.3 ± 0.7	14 ± 1	< 0.3	2.3 ± 0.6
JUNE	7I	90 ± 40	19 ± 2	17 ± 1	25 ± 2	14 ± 2	0.8 ± 1.1	14 ± 2	< 0.3	2.2 ± 0.4
	12N	70 ± 50	8 ± 1	14 ± 2	8 ± 1	18 ± 2	0.9 ± 1.1	18 ± 2	< 0.3	2.0 ± 0.6
	OFF-SITE	80 ± 20	4 ± 1	28 ± 2	9 ± 1	12 ± 1	1.0 ± 0.8	12 ± 1	< 0.3	1.6 ± 0.7
JULY	7I	50 ± 20	12 ± 1	18 ± 1	13 ± 1	17 ± 1	1.3 ± 0.8	17 ± 1	< 0.3	2.2 ± 0.5
	12N	60 ± 40	12 ± 2	15 ± 2	10 ± 1	13 ± 2	0.7 ± 0.9	13 ± 2	< 0.3	2.7 ± 0.7
	OFF-SITE	70 ± 30	11 ± 2	18 ± 2	10 ± 2	20 ± 2	0.4 ± 1.3	15 ± 2	< 0.3	2.4 ± 0.7
AUGUST	7I	30 ± 10	16 ± 2	13 ± 1	7 ± 1	9 ± 1	< 0.3	10 ± 1	< 0.3	1.4 ± 0.4
	12N	30 ± 20	11 ± 1	8 ± 1	3 ± 1	13 ± 2	0.6 ± 1.0	11 ± 2	< 0.3	1.4 ± 0.6
	OFF-SITE	< 10	11 ± 2	15 ± 2	3 ± 1	9 ± 1	0.5 ± 0.8	7 ± 1	< 0.3	1.2 ± 0.5
SEPTEMBER	7I	30 ± 10	6 ± 2	9 ± 1	5 ± 1	10 ± 1	0.5 ± 0.5	9 ± 1	< 0.3	1.0 ± 0.3
	12N	20 ± 40	6 ± 1	12 ± 1	5 ± 1	15 ± 1	0.6 ± 0.7	14 ± 1	< 0.3	0.4 ± 0.4
OCTOBER	7I	< 10	1 ± 1	2 ± 1	1 ± 1	11 ± 1	0.6 ± 0.6	10 ± 1	< 0.3	0.9 ± 0.4
	12N	< 10	4 ± 2	8 ± 2	4 ± 1	18 ± 2	1.6 ± 0.8	10 ± 1	< 0.3	1.3 ± 0.5
	OFF-SITE	< 10	3 ± 1	4 ± 1	2 ± 1	4 ± 2	< 0.3	3 ± 2	< 0.3	1.0 ± 0.6
NOVEMBER	7I	< 10	6 ± 3	11 ± 1	5 ± 1	12 ± 1	< 0.3	13 ± 1	< 0.3	1.6 ± 0.5
	12N	< 10	5 ± 3	9 ± 1	4 ± 1	10 ± 1	< 0.3	10 ± 1	< 0.3	2.5 ± 0.6
	OFF-SITE	< 10	3 ± 3	5 ± 1	3 ± 1	6 ± 1	< 0.3	6 ± 1	< 0.3	2.3 ± 0.9
DECEMBER	7I	20 ± 10	9 ± 2	15 ± 1	8 ± 1	13 ± 1	0.4 ± 0.2	12 ± 1	< 0.3	0.8 ± 0.4
	12N	< 10	10 ± 3	14 ± 2	7 ± 1	16 ± 2	0.9 ± 0.4	17 ± 2	< 0.3	0.9 ± 0.5
	OFF-SITE	< 10	5 ± 2	9 ± 1	5 ± 1	7 ± 1	< 0.3	7 ± 1	< 0.3	0.8 ± 0.7
ANNUAL	7I	40 ± 10	13 ± 6	14 ± 5	9 ± 5	14 ± 3	0.7 ± 0.3	14 ± 3	< 0.3	1.5 ± 0.3
	12N	40 ± 10	12 ± 5	15 ± 4	9 ± 4	18 ± 4	1.1 ± 0.4	17 ± 5	< 0.3	1.7 ± 0.5
SUMMARY	OFF-SITE	40 ± 20	10 ± 5	11 ± 5	6 ± 2	10 ± 4	0.5 ± 0.3	8 ± 3	< 0.3	1.5 ± 0.5
%CG	7I	0.02	(6.5)	(4.6)	(0.9)	(0.35)	(0.02)	(0.27)	< 0.03	0.1
	12N	0.02	(6.0)	(4.9)	(0.9)	(0.45)	(0.03)	(0.34)	< 0.03	0.2
(X 1E-3)	OFF-SITE	0.02	(5.1)	(3.8)	(0.6)	(0.24)	(0.01)	(0.16)	< 0.03	0.1

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[1] PERIMETER LOCATIONS ARE GIVEN IN TERMS OF THE GRID COORDINATES IN FIGURE 1.

[2] THE CONCENTRATIONS IN UNITS OF MICROGRAMS/CUBIC METER CAN BE OBTAINED BY MULTIPLYING THE VALUE IN MICROCURIES/ML BY 2.96 X 10^{12} FOR URANIUM-238 AND BY 9 X 10^{12} FOR THORIUM-232. THE MASS OF THE OTHER THORIUM ISOTOPES IN COMPARISON TO THORIUM-232 AND THE OTHER URANIUM ISOTOPES IN COMPARISON TO URANIUM-238 IS NEGLIGIBLE.

[3] PLUTONIUM-240 IS INCLUDED (SEE TEXT).

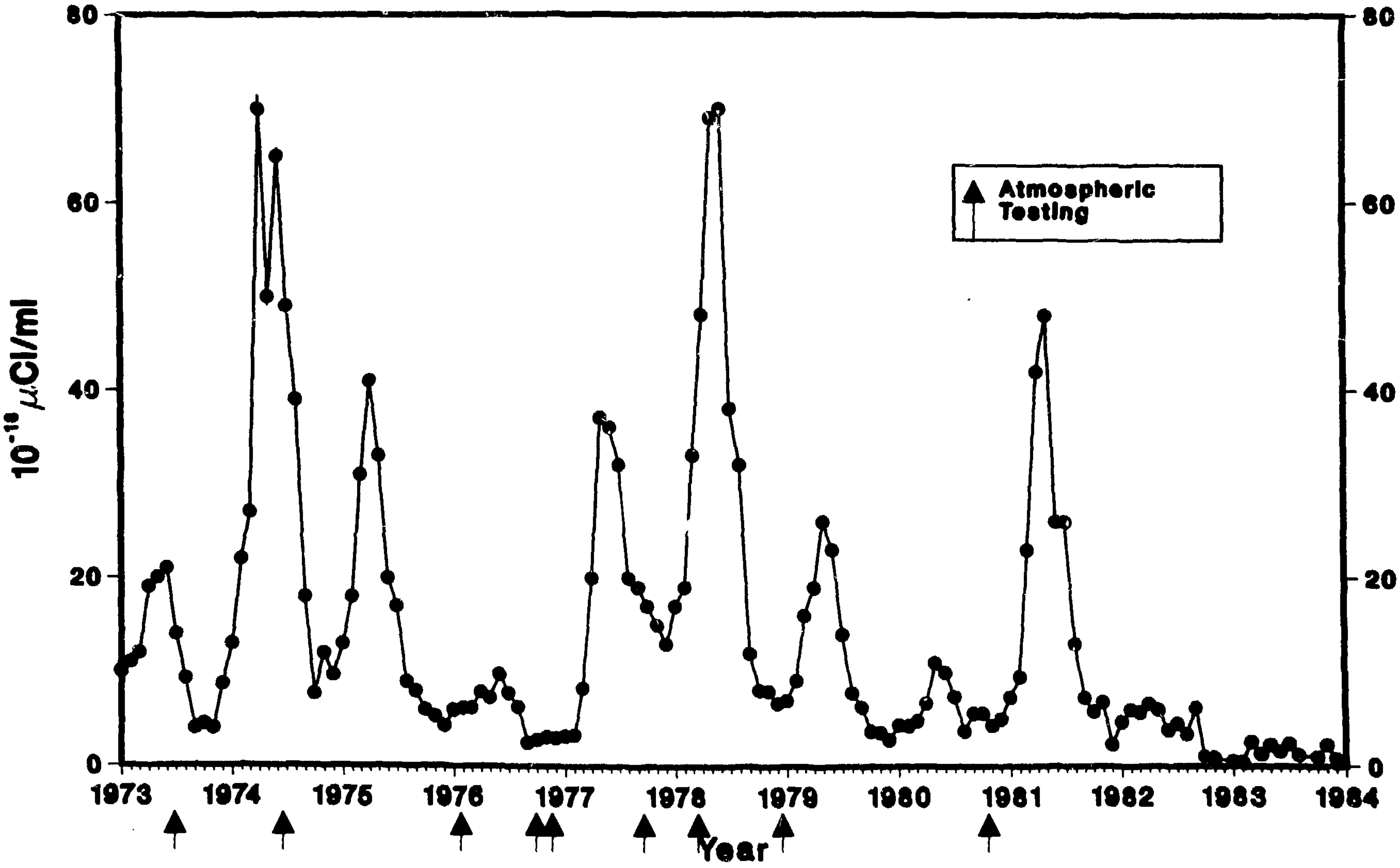

Fig. 4. Plutonium-239,240 Air Concentrations, 1973-1983

source of argon-41 was from the Janus reactor (Building 202, location 13I). Argon-41 was collected in the exhaust stack of this reactor by filling an evacuated "Marinelli-type" container with air and measuring the argon-41 concentration by gamma-ray spectrometry. The total amount released during 1983 was about 0.75 Ci. The major airborne effluents released were 119 Ci of carbon-11 from the IPNS facility (location 9J) and 2.2 Ci of krypton-85 from Building 212 (location 12I). Several other fission products were also released in millicurie or smaller amounts.

2. Surface Water

Total (nonvolatile) alpha and beta activities were determined by counting the residue remaining after evaporation of the water, and applying counting efficiency corrections determined for uranium-233 (for alpha activity) and thallium-204 (for beta activity) to obtain disintegration rates. Hydrogen-3 was determined on a separate sample, and this activity does not appear in the total nonvolatile beta activity. Uranium was determined fluorophotometrically, and the results calculated in terms of activity with the assumption that the isotopic composition was that of natural uranium. Analyses for other radionuclides were performed by specific radiochemical separations followed by appropriate counting. One liter aliquots were used for all analyses except hydrogen-3 and the transuranium nuclides. Hydrogen-3 analyses were performed by liquid scintillation counting of 10 mL in a gel system. Analyses for transuranium nuclides were performed on 10- or 50-liter samples by chemical separation methods followed by alpha spectrometry.[7,8] Plutonium-236 was used to determine the yields of plutonium and neptunium, which were separated together. A group separation of a fraction containing the transplutonium elements was monitored for recovery with americium-243 tracer.

Argonne waste water is discharged into Sawmill Creek, a small stream that runs through the Laboratory grounds, drains surface water from much of the site, and flows into the Des Plaines River about 500 m (0.3 mi) downstream from the waste-water outfall. Sawmill Creek was sampled upstream from the Argonne site and downstream from the waste-water outfall to determine if radioactivity was added to the stream by Argonne waste water or surface drainage. The sampling locations are shown in Figure 1. Below the waste-water outfall, daily samples were collected by a continuous sampler, which operated about 95% of the year. When the continuous sampling device was not functioning, a grab sample was collected each working day. Equal portions of the daily samples collected each week were combined and analyzed to obtain an average weekly concentration. Above the site, samples were collected once a month and were analyzed for the same radionuclides as the below-outfall samples.

Annual summaries of the results obtained for Sawmill Creek are given in Table 7. Comparison of the results, and 95% confidence levels of the averages, for the two sampling locations shows that the nuclides whose presence in the Creek water can be attributed to Argonne operations were hydrogen-3, neptunium-237, plutonium-239, americium-241, and occasionally strontium-90, plutonium-238, curium-242 and/or californium-252, and curium-244 and/or californium-249. The percentage of individual samples containing activity attributable to Argonne was 65% for hydrogen-3, 83% for neptunium-237, 95% for plutonium-239, and 96% for americium-241. The concentrations of all these nuclides were low compared to the CGs. The principal radionuclide added to the Creek by Argonne waste water, in terms of concentration, was hydrogen-3. Its average net concentration was equivalent to 0.003% of the CG. The decrease in the hydrogen-3 over the past few years is due to the cessation of the operation of the CP-5 reactor. The total concentration, regardless of source, must be used in assessing the hazard of a radionuclide not naturally present, so the percent CG in the table was calculated on this basis.

The hydrogen-3 in the Creek above the site was similar in concentration to levels found away from the Laboratory site and is characteristic of the current ambient level in surface water. During 1983, the hydrogen-3 content of 22 other lakes and streams ranged from $< 100 \times 10^{-9}$ µCi/mL to 244×10^{-9} µCi/mL and averaged 144×10^{-9} µCi/mL.

The total radioactive effluent discharged to the Creek in Argonne waste water can be estimated from the average net concentrations and the volume of water carried by the Creek. These totals are 1.4 Ci of

TABLE 7

RADIONUCLIDES IN SAWMILL CREEK WATER, 1983

TYPE OF ACTIVITY	LOCATION *	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/ML) AVG.	MIN.	MAX.	PERCENT CG AVG.	MIN.	MAX.
ALPHA (NONVOLATILE)	16K	12	2.0 ± 0.1	1.0	3.2	(0.067)	(0.035)	(0.107)
	7M	248	2.1 ± 0.2	0.9	5.5	(0.071)	(0.031)	(0.184)
BETA (NONVOLATILE)	16K	12	16 ± 1	8	31	(0.53)	(0.26)	(1.04)
	7M	248	17 ± 2	7	30	(0.57)	(0.24)	(0.98)
HYDROGEN-3	16K	12	123 ± 4	< 100	251	0.0041	< 0.003	0.0084
	7M	248	225 ± 60	< 100	1332	0.0075	< 0.003	0.0444
STRONTIUM-90	16K	12	< 0.26	< 0.25	0.35	< 0.083	< 0.08	0.12
	7M	248	0.26 ± 0.03	< 0.25	0.55	0.087	< 0.08	0.18
RADIUM-226	16K	12	0.75 ± 0.02	0.46	1.59	(2.49)	(1.53)	(5.29)
	7M	248	0.56 ± 0.08	0.22	1.52	(1.87)	(0.73)	(5.06)
URANIUM** (NATURAL)	16K	12	1.7 ± 0.1	0.4	2.5	(0.0042)	(0.0010)	(0.0063)
	7M	248	1.8 ± 0.2	0.7	3.4	(0.0045)	(0.0016)	(0.0084)
NEPTUNIUM-237	16K	12	-	-	< 0.001	-	-	< 0.000033
	7M	248	0.0042 ± 0.0015	< 0.001	0.037	0.00014	< 0.00003	0.0012
PLUTONIUM-238	16K	12	-	-	< 0.001	-	-	< 0.00002
	7M	248	< 0.0032	< 0.001	0.063	0.00006	< 0.00002	0.00125
PLUTONIUM-239	16K	12	< 0.00054	< 0.0005	0.0007	< 0.00001	< 0.00001	0.00001
	7M	248	0.0089 ± 0.0024	< 0.0005	0.04650	0.00018	< 0.000010	0.0009
AMERICIUM-241	16K	12	< 0.001	< 0.001	0.001	< 0.000025	< 0.000025	0.00003
	7M	248	0.0075 ± 0.0019	< 0.001	0.038	0.00019	< 0.000025	0.0010
CURIUM-242 AND/OR CALIFORNIUM-252	16K	12	-	-	< 0.001	-	-	< 0.000005
	7M	248	< 0.0012	< 0.001	0.0030	< 0.000006	< 0.000005	0.000015
CURIUM-244 AND/OR CALIFORNIUM-249	16K	12	-	-	< 0.001	-	-	< 0.000014
	7M	248	0.0026 ± 0.0007	< 0.001	0.0132	0.000038	< 0.000014	0.00019

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* LOCATION 16K IS UPSTREAM FROM THE ARGONNE SITE AND LOCATION 7M IS DOWNSTREAM FROM THE ARGONNE WASTE-WATER OUTFALL.

** URANIUM CONCENTRATIONS IN UNITS OF MICROGRAMS/L CAN BE OBTAINED BY MULTIPLYING THE CONCENTRATION GIVEN BY 1.48 X 10^9.

hydrogen-3, < 1.0 mCi of strontium-90, 0.05 mCi of neptunium-237, 0.05 mCi of plutonium-239, 0.09 mCi of americium-241, and < 0.05 mCi of curium and californium nuclides.

Since Sawmill Creek empties into the Des Plaines River, which in turn flows into the Illinois River, the radioactivity in the latter two streams is important in assessing the contribution of Argonne waste water to the environmental radioactivity. The Des Plaines River was sampled twice a month below, and monthly above, the mouth of Sawmill Creek to determine if the radioactivity in the Creek had any effect on the activity in the River.

Annual summaries of the results obtained for these two locations are given in Table 8. The average nonvolatile alpha, beta, and uranium concentrations in the River were very similar to past averages and remained in the normal range. Results were quite similar above and below the Creek for all radionuclides since the activity in Sawmill Creek was reduced by dilution so that it was not detectable as such in the Des Plaines River. The average nonvolatile alpha and beta activities, 1.6×10^{-9} µCi/mL and 8×10^{-9} µCi/mL, respectively, of 22 off-site surface water samples collected this year (excluding the Des Plaines River) were similar to the levels found in previous years.

The radioactivity in samples of Illinois River water, shown in Table 9, was similar to those found previously at these same locations. No radioactivity originating at Argonne could be detected in the Des Plaines or Illinois Rivers.

3. Soil, Grass, and Bottom Sediment

The radioactive content of soil, grass, and bottom sediment was measured at the site perimeter and off the site. The purpose of the off-site sampling was to measure deposition for comparison with perimeter samples, and with results obtained by other organizations for samples collected at large distances from nuclear installations. Such comparisons are useful in determining if the soil activity near Argonne is normal. For this

TABLE 8

RADIONUCLIDES IN DESPLAINES RIVER WATER, 1983

TYPE OF ACTIVITY	LOCATION *	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/ML) AVG.	MIN.	MAX.	PERCENT CG AVG.	MIN.	MAX.
ALPHA	A	12	1.8 ± 0.1	1.0	2.3	(0.059)	(0.034)	(0.076)
(NONVOLATILE)	B	24	1.7 ± 0.2	0.6	2.8	(0.055)	(0.019)	(0.092)
BETA	A	12	12 ± 1	6	26	(0.40)	(0.20)	(0.85)
(NONVOLATILE)	B	24	12 ± 2	5	23	(0.40)	(0.18)	(0.77)
HYDROGEN-3	A	12	115 ± 3	< 100	228	0.0038	< 0.003	0.0076
	B	24	< 114	< 100	186	< 0.0038	< 0.003	0.006
STRONTIUM-90	A	12	0.26 ± 0.01	< 0.25	0.38	0.09	< 0.08	0.13
	B	24	0.26 ± 0.05	< 0.25	0.60	0.09	< 0.08	0.20
RADIUM-226	A	12	0.41 ± 0.01	0.22	1.14	(1.38)	(0.73)	(3.)
	B	12	0.64 ± 0.06	0.18	3.80	(2.13)	(0.60)	(12.7)
URANIUM **	A	12	1.8 ± 0.1	0.5	4.0	(0.0045)	(0.0011)	(0.0099)
(NATURAL)	B	23	1.7 ± 0.3	0.3	2.7	(0.0042)	(0.0008)	(0.0067)
NEPTUNIUM-237	A	12	-	-	< 0.001	-	-	< 0.00003
	B	12	-	-	< 0.001	-	-	< 0.00003
PLUTONIUM-238	A	12	-	-	< 0.001	-	-	< 0.00002
	B	12	-	-	< 0.001	-	-	< 0.00002
PLUTONIUM-239	A	12	< 0.00060	< 0.0005	0.0010	< 0.000012	< 0.00001	0.000020
	B	12	< 0.00059	< 0.0005	0.0009	< 0.000012	< 0.00001	0.000019
AMERICIUM-241	A	12	-	-	< 0.001	-	-	< 0.000025
	B	12	< 0.0010	< 0.001	0.001	< 0.000026	< 0.000025	0.000036
CURIUM-242 AND/OR	A	12	-	-	< 0.001	-	-	< 0.000005
CALIFORNIUM-252	B	12	-	-	< 0.001	-	-	< 0.000005
CURIUM-244 AND/OR	A	12	< 0.0010	< 0.001	0.0011	< 0.000014	< 0.000014	0.000016
CALIFORNIUM-249	B	12	< 0.0010	< 0.001	0.0010	< 0.000014	< 0.000014	0.000015

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* LOCATION A, NEAR WILLOW SPRINGS, IS UPSTREAM AND LOCATION B, NEAR LEMONT, IS DOWNSTREAM FROM THE MOUTH OF SAWMILL CREEK. SEE FIGURE 2.

** URANIUM CONCENTRATIONS IN UNITS OF MICROGRAMS/L CAN BE OBTAINED BY MULTIPLYING THE CONCENTRATION GIVEN BY 1.48×10^{9}.

TABLE 9

Radionuclides in Illinois River Water, 1983
(Concentrations in 10^{-9} μCi/mL)

Date Collected	Location	Alpha*	Beta*	Hydrogen-3	Uranium** (natural)	Plutonium-239
June 16	McKinley Woods State Park	1.9 ± 0.3	12.1 ± 0.3	220 ± 110	0.9 ± 0.1	0.00049 ± 0.00020
June 16	Below Dresden Power Station	1.8 ± 0.3	8.3 ± 0.3	140 ± 100	1.2 ± 0.1	0.00012 ± 0.00010
June 16	Morris	0.9 ± 0.2	7.8 ± 0.3	< 100	1.3 ± 0.2	-
June 16	Starved Rock State Park	1.5 ± 0.2	6.7 ± 0.3	< 100	1.3 ± 0.1	-
October 26	McKinley Woods State Park	0.5 ± 0.1	5.7 ± 0.3	150 ± 90	0.4 ± 0.1	0.00032 ± 0.00011
October 26	Below Dresden Power Station	0.6 ± 0.1	7.2 ± 0.3	190 ± 90	0.5 ± 0.1	0.00032 ± 0.00010
October 26	Morris	0.6 ± 0.2	5.5 ± 0.2	150 ± 90	0.8 ± 0.2	-
October 26	Starved Rock State Park	1.4 ± 0.2	7.8 ± 0.4	160 ± 90	0.9 ± 0.1	-

*Nonvolatile activity.

**Uranium concentrations in units of μg/L can be obtained by multiplying the concentration by 1.48 x 10^9.

purpose, the ASTM site selection criteria, sample collection, and sample preparation techniques were used.[9] *Sites were selected in several directions and at various distances from the Laboratory. Each site was selected on the basis that the soil appeared, or was known to have been, undisturbed for a number of years. Attempts were made to select open, level, grassy areas that were mowed at reasonable intervals. Public parks were selected when available.*

Each soil sample consisted of ten cores totaling 864 cm^2 *in area by 5 cm deep. Through 1976, samples had been collected down to 30 cm to measure total deposition and as a result of five years of sample collection at this depth, the total deposition in the Argonne environment has been established. By reducing the sampling depth to 5 cm, the analysis will be more sensitive to changes in current deposition. The grass samples were obtained by collecting the grass from a 1* m^2 *area in the immediate vicinity of a soil sample. A grab sample technique was used to obtain bottom sediment. After drying, grinding, and mixing, 100 g portions of soil, bottom sediment, and grass were analyzed by the same methods described in Section III.A for air-filter residues. The plutonium and americium were separated from the same 100 gram aliquot of soil. Results are given in terms of the oven-dried (110°C) weight.*

The results for the gamma-ray emitting nuclides in soil are presented in Table 10. The antimony-125 is a residual from past atmospheric nuclear tests by the People's Republic of China. Other intermediate half-life fission products reported previously have decayed to below their detection limits. The cesium-137 levels are similar to those found over the past several years, and represent an accumulation from nuclear tests over a period of many years. The annual average concentrations for the perimeter and off-site samples are similar. The plutonium and americium concentrations are given in Table 11. The ranges and concentrations of plutonium and americium in soil are similar at both perimeter and off-site locations. The small differences between the perimeter and off-site americium-241 concentrations require further investigation to decide if they are within the range expected from fallout. For fallout americium-241 in soil, about 10% is due to direct deposition, while about 90% is from the

TABLE 10

Gamma-Ray Emitting Radionuclides in Soil, 1983
(Concentrations in 10^{-6} μCi/g)

Date Collected	Location	Antimony-125	Cesium-137	Radium-226 (Bismuth-214)	Thorium-228 (Lead-212)	Thorium-232 (Actinium-228)
	<u>Perimeter</u>*					
June 21	7E/F	0.41 ± 0.23	1.04 ± 0.07	1.61 ± 0.11	1.23 ± 0.08	1.19 ± 0.17
June 21	6J	0.55 ± 0.21	0.88 ± 0.07	1.49 ± 0.11	1.02 ± 0.07	1.14 ± 0.17
June 21	10P	0.57 ± 0.22	1.03 ± 0.07	3.87 ± 0.17	1.40 ± 0.08	1.00 ± 0.15
June 21	14N	0.48 ± 0.21	1.17 ± 0.07	1.27 ± 0.11	1.25 ± 0.07	0.99 ± 0.16
June 21	14F	0.32 ± 0.24	1.27 ± 0.07	2.48 ± 0.14	1.18 ± 0.07	0.89 ± 0.14
October 28	4E/F	0.30 ± 0.24	1.57 ± 0.09	1.37 ± 0.13	0.74 ± 0.07	0.68 ± 0.17
October 28	7E/F	0.31 ± 0.17	0.88 ± 0.06	0.80 ± 0.10	0.96 ± 0.07	0.92 ± 0.16
October 28	10P	0.36 ± 0.13	0.96 ± 0.04	1.36 ± 0.08	1.07 ± 0.05	0.83 ± 0.12
October 28	14N	0.46 ± 0.13	0.96 ± 0.04	0.94 ± 0.07	1.10 ± 0.05	0.92 ± 0.11
October 28	14E	0.42 ± 0.13	0.65 ± 0.04	1.31 ± 0.09	0.93 ± 0.05	0.81 ± 0.12
	Average	0.42 ± 0.06	1.03 ± 0.16	1.65 ± 0.57	1.09 ± 0.12	0.94 ± 0.10
	<u>Off-Site</u>					
June 15	Woodridge, IL	0.68 ± 0.28	1.04 ± 0.07	2.21 ± 0.14	1.29 ± 0.08	1.16 ± 0.17
June 15	Naperville, IL	0.51 ± 0.22	1.18 ± 0.07	2.79 ± 0.15	1.63 ± 0.08	1.74 ± 0.23
June 15	Romeoville, IL	0.49 ± 0.21	0.58 ± 0.05	1.92 ± 0.14	1.26 ± 0.08	1.26 ± 0.18
June 16	McKinley Woods, IL	0.34 ± 0.18	0.80 ± 0.06	1.82 ± 0.12	1.30 ± 0.07	1.00 ± 0.15
June 16	Dresden Lock & Dam, IL	0.63 ± 0.21	1.31 ± 0.07	2.17 ± 0.13	1.18 ± 0.07	0.95 ± 0.14
October 26	Channahon, IL	0.50 ± 0.22	1.05 ± 0.07	1.21 ± 0.11	0.99 ± 0.07	0.86 ± 0.16
October 26	Starved Rock State Park, IL	0.23 ± 0.23	0.76 ± 0.08	1.45 ± 0.12	0.88 ± 0.08	0.65 ± 0.14
October 26	Morris, IL	0.41 ± 0.20	0.26 ± 0.04	1.32 ± 0.11	0.91 ± 0.07	0.79 ± 0.15
October 27	Western Springs, IL	0.34 ± 0.26	1.92 ± 0.09	1.76 ± 0.13	0.91 ± 0.07	0.83 ± 0.15
October 27	Brookfield, IL	0.46 ± 0.20	0.71 ± 0.06	1.16 ± 0.11	0.96 ± 0.07	0.90 ± 0.16
	Average	0.46 ± 0.09	0.96 ± 0.29	1.78 ± 0.33	1.13 ± 0.15	1.01 ± 0.20

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*The perimeter locations are given in terms of the grid coordinates in Figure 1. activity.

TABLE 11

Transuranics in Soil, 1983

Date Collected	Location	Plutonium-238		Plutonium-239		$^{238}Pu/^{239}Pu$	Americium-241		$^{241}Am/^{239}Pu$
		10^{-9} µCi/g	10^{-3} µCi/m^2	10^{-9} µCi/g	10^{-3} µCi/m^2		10^{-9} µCi/g	10^{-3} µCi/m^2	
	<u>Perimeter</u>*								
June 21	7E/F	0.9 ± 0.2	0.043 ± 0.009	22.8 ± 1.2	1.04 ± 0.05	0.041	7.5 ± 0.4	0.343 ± 0.017	0.33
June 21	6J	0.8 ± 0.2	0.040 ± 0.011	16.8 ± 1.0	0.90 ± 0.05	0.045	3.5 ± 0.4	0.190 ± 0.022	0.21
June 21	10P	0.3 ± 0.1	0.016 ± 0.005	9.8 ± 0.5	0.51 ± 0.03	0.031	3.6 ± 0.3	0.186 ± 0.014	0.36
June 21	14N	0.8 ± 0.2	0.047 ± 0.013	19.7 ± 1.2	1.10 ± 0.06	0.042	5.4 ± 0.4	0.304 ± 0.022	0.28
June 21	14F	0.9 ± 0.2	0.039 ± 0.009	22.8 ± 1.2	0.95 ± 0.05	0.041	4.4 ± 0.3	0.184 ± 0.013	0.19
October 28	4E/F	1.0 ± 0.2	0.062 ± 0.012	26.6 ± 1.0	1.57 ± 0.06	0.039	-	-	-
October 28	7E/F	0.9 ± 0.2	0.054 ± 0.011	18.8 ± 0.8	1.12 ± 0.05	0.048	-	-	-
October 28	10P	0.6 ± 0.1	0.031 ± 0.007	14.9 ± 0.6	0.84 ± 0.04	0.037	-	-	-
October 28	14N	0.8 ± 0.2	0.034 ± 0.007	19.7 ± 0.8	0.84 ± 0.03	0.041	-	-	-
October 28	14E	0.4 ± 0.1	0.024 ± 0.007	11.4 ± 0.6	0.62 ± 0.03	0.039	-	-	-
	Average	0.7 ± 0.1	0.039 ± 0.009	18.3 ± 3.3	0.95 ± 0.19	0.040	4.9 ± 1.5	0.241 ± 0.068	0.27
	<u>Off-Site</u>								
June 15	Woodridge, IL	1.0 ± 0.2	0.044 ± 0.009	19.6 ± 1.0	0.83 ± 0.04	0.053	1.7 ± 0.5	0.147 ± 0.046	0.18
June 15	Naperville, IL	0.7 ± 0.2	0.026 ± 0.007	17.8 ± 1.0	0.70 ± 0.04	0.038	2.7 ± 0.9	0.213 ± 0.073	0.15
June 15	Romeoville, IL	0.5 ± 0.2	0.024 ± 0.008	13.2 ± 0.8	0.67 ± 0.04	0.036	1.6 ± 0.3	0.084 ± 0.015	0.12
June 16	McKinley Woods State Park, IL	0.8 ± 0.2	0.033 ± 0.008	15.7 ± 0.9	0.70 ± 0.04	0.048	3.2 ± 0.4	0.143 ± 0.018	0.20
June 16	Dresden Lock and Dam, IL	0.9 ± 0.2	0.038 ± 0.009	24.0 ± 1.2	1.03 ± 0.05	0.037	5.1 ± 1.6	0.218 ± 0.068	0.21
October 26	Channahon, IL	0.7 ± 0.2	0.040 ± 0.010	21.6 ± 0.9	1.21 ± 0.05	0.033	-	-	-
October 26	Starved Rock State Park, IL	0.5 ± 0.1	0.032 ± 0.009	9.4 ± 0.5	0.58 ± 0.03	0.055	-	-	-
October 26	Morris, IL	0.2 ± 0.1	0.008 ± 0.005	2.7 ± 0.3	0.14 ± 0.02	0.058	-	-	-
October 27	Western Springs, IL	1.7 ± 0.2	0.088 ± 0.013	33.3 ± 1.1	1.73 ± 0.06	0.051	-	-	-
October 27	Brookfield, IL	0.8 ± 0.2	0.043 ± 0.011	15.2 ± 0.8	0.84 ± 0.04	0.051	-	-	-
	Average	0.8 ± 0.3	0.038 ± 0.013	17.3 ± 5.3	0.84 ± 0.27	0.046	2.9 ± 1.3	0.161 ± 0.050	0.17

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*The perimeter locations are given in terms of the grid coordinates in Figure 1.

decay of the previously deposited plutonium-241. The measured deposition of americium-241 and the americium-241/plutonium-239 ratio is consistent with reported values.[10]

Composite monthly precipitation samples were analyzed for plutonium-239. The results are given in Table 12, along with results since 1973 for comparison. The 1983 deposition by precipitation was equivalent to 0.1% of the total plutonium deposited through 1976, which is 2.2×10^{-3} *µCi/*m^2*.*[11] *Deposition since 1976 has not changed this value significantly. The data in Table 12 is illustrated in Figure 5. The arrows at the bottom of the figure indicate the approximate dates of the Chinese atmospheric nuclear tests. Comparison of the results in Figure 5 with the plutonium air concentrations for the past eleven years in Figure 4 shows excellent correlation.*

The results of radionuclide concentrations measured in grass are given in Table 13. The annual averages and concentration ranges were similar at the perimeter and off-site locations as well as similar to previous years, indicating no contribution from ANL operations. In terms of deposition, the plutonium-239 concentration was a factor of about 10^4 *less in the grass than in the soil from the same location.*

Results of analysis of bottom sediment samples for gamma-ray emitters and transuranics are given in Table 14. The annual off-site averages are in the same range found in off-site samples collected in previous years. Plutonium results vary widely between locations and are strongly dependent on the retentiveness of the bottom material.

A set of samples was collected on August 11, 1983, from the Sawmill Creek bed, above, at, and at several locations below the point at which the Laboratory discharges its treated waste water (location 7M in Figure 1). The results are listed in Table 14 and indicate that the sample above the 7M outfall is similar to the off-site samples. The plutonium, americium, and cesium concentrations are the highest at the outfall and decrease rapidly with distance, indicating their origin in Argonne waste water. Similar sets of samples were collected on September 15, 1982, and

TABLE 12

Ground Deposition of Plutonium-239,240

(Units of 10^{-6} $\mu Ci/m^2$)

Month	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983
January	-	0.60	1.1	0.25	0.17	0.37	0.57	0.15	0.06	0.23	0.53
February	0.14	1.1	1.7	0.38	0.15	0.12	0.43	0.21	0.43	< 0.1	0.23
March	0.27	2.0	1.5	0.58	0.74	4.7	-	0.50	0.48	< 0.1	0.27
April	0.40	5.2	4.0	0.21	1.0	3.3	1.78	0.56	3.70	< 0.1	0.10
May	0.50	4.6	1.8	0.58	1.8	4.7	0.58	0.81	1.43	1.10	0.26
June	0.16	3.5	1.9	0.65	1.2	3.0	0.73	0.54	1.11	0.69	0.11
July	0.32	5.6	0.48	0.26	0.71	1.7	0.45	0.67	0.51	-	0.27
August	0.34	1.0	0.38	0.15	1.6	0.54	0.54	0.62	0.40	0.55	0.11
September	0.27	0.25	0.10	0.06	1.5	0.69	0.21	0.65	0.29	0.37	0.13
October	0.30	0.45	0.12	0.30	0.85	0.34	0.53	0.53	0.36	0.16	0.18
November	0.22	0.73	0.08	0.05	0.72	0.74	0.09	0.13	0.24	0.13	0.33
December	0.46	0.71	0.21	0.12	0.43	0.40	0.13	0.12	0.07	0.13	0.19
Average Monthly Deposition	0.31 ± 0.07	2.1 ± 1.2	1.1 ± 0.7	0.30 ± 0.13	0.91 ± 0.31	1.7 ± 1.0	0.55 ± 0.27	0.46 ± 0.14	0.76 ± 0.26	0.33 ± 0.13	0.23 ± 0.12
Annual Deposition	3.38	25.74	13.37	3.59	10.87	20.60	6.04	5.49	9.08	3.66	2.71
Percent Added to Existing	0.2	1.2	0.6	0.2	0.5	0.9	0.3	0.3	0.4	0.2	0.1

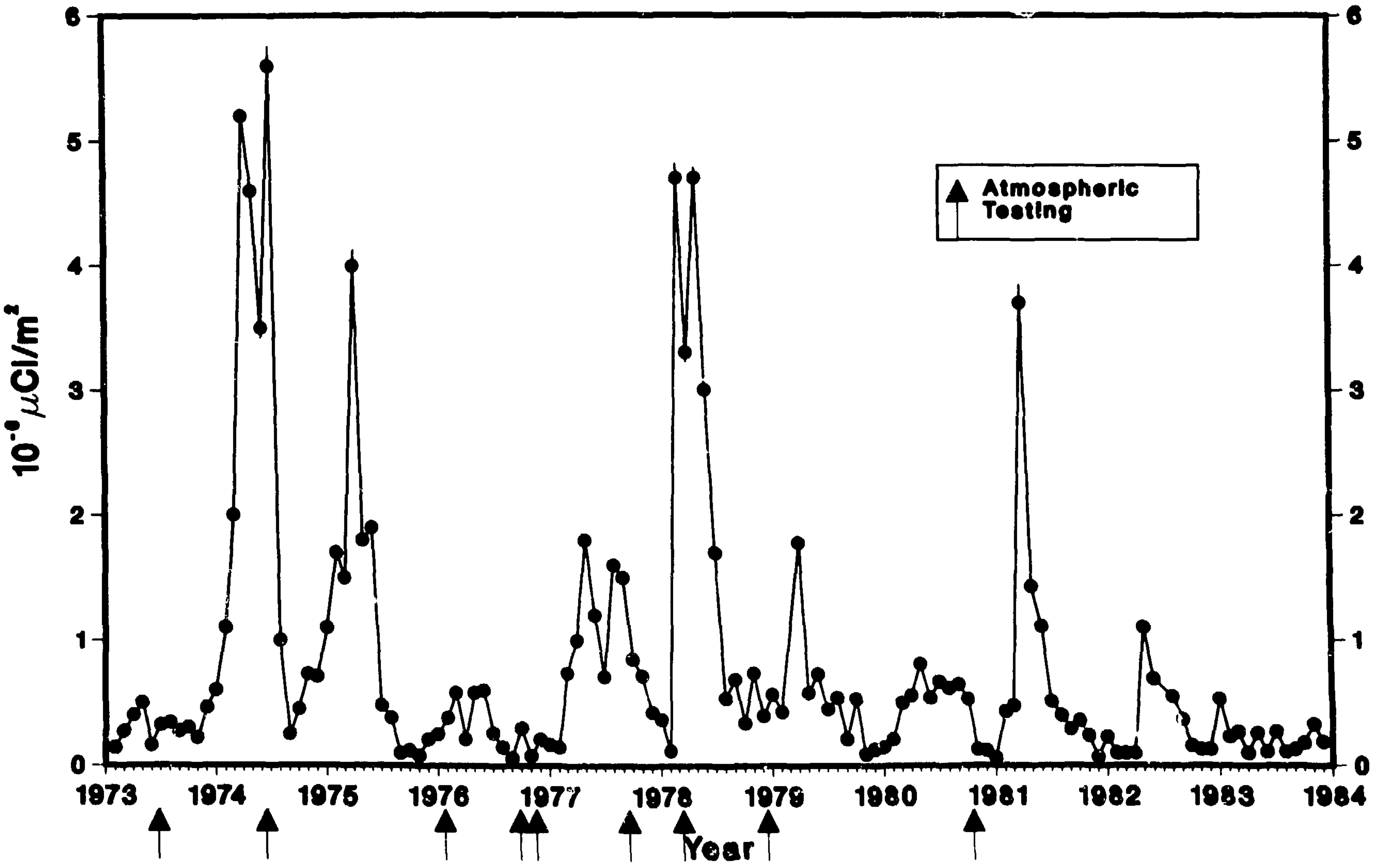

Fig. 5. Plutonium-239,240 Surface Deposition, 1973-1983

TABLE 13

Radionuclides in Grass, 1983

Date Collected	Location	Concentrations in 10^{-9} µCi/g					10^{-6} µCi/m^2
		Cesium-137	Thorium-228	Thorium-230	Thorium-232	Plutonium-239	Deposition of Plutonium-239
	Perimeter[*]						
June 21	7E/F	50 ± 20	0.6 ± 0.3	0.9 ± 0.2	0.5 ± 0.1	0.50 ± 0.11	0.097 ± 0.022
June 21	6J	50 ± 20	0.8 ± 0.1	1.5 ± 0.1	0.9 ± 0.6	0.65 ± 0.12	0.094 ± 0.017
June 21	10P	40 ± 20	0.8 ± 0.2	1.3 ± 0.3	0.9 ± 0.2	0.08 ± 0.06	0.009 ± 0.007
June 21	14N	30 ± 20	1.0 ± 0.5	1.7 ± 0.2	1.1 ± 0.2	0.08 ± 0.06	0.012 ± 0.009
June 21	14F	60 ± 20	1.9 ± 1.0	2.1 ± 0.4	1.8 ± 0.4	0.35 ± 0.07	0.097 ± 0.021
October 28	4E/F	20 ± 20	-	-	-	-	-
October 28	7E/F	< 10	-	-	-	-	-
October 28	10P	< 10	-	-	-	-	-
October 28	14N	< 10	-	-	-	-	-
October 28	14E	10 ± 20	-	-	-	-	-
	Average	29 ± 12	1.0 ± 0.5	1.5 ± 0.4	1.0 ± 0.4	0.33 ± 0.23	0.062 ± 0.042
	Off-Site						
June 15	Woodridge, IL	70 ± 30	1.9 ± 0.4	3.7 ± 0.3	1.6 ± 0.2	0.09 ± 0.05	0.017 ± 0.010
June 15	Naperville, IL	60 ± 30	2.1 ± 0.3	7.9 ± 0.7	2.0 ± 0.2	0.06 ± 0.06	0.007 ± 0.009
June 15	Romeoville, IL	80 ± 30	2.6 ± 0.7	5.0 ± 0.7	2.7 ± 0.5	0.12 ± 0.05	0.017 ± 0.008
June 16	McKinley Woods State Park, IL	40 ± 40	6.0 ± 1.2	14.1 ± 1.0	6.4 ± 0.6	0.19 ± 0.11	0.009 ± 0.005
June 16	Dresden Lock and Dam, IL	50 ± 20	2.9 ± 0.4	4.0 ± 0.3	2.7 ± 0.3	-	-
October 26	Channahon, IL	80 ± 50	-	-	-	0.28 ± 0.08	0.045 ± 0.013
October 26	Starved Rock State Park, IL	20 ± 20	-	-	-	0.10 ± 0.06	0.036 ± 0.020
October 26	Morris, IL	< 10	-	-	-	0.12 ± 0.06	0.013 ± 0.006
October 27	Western Springs, Il	30 ± 10	-	-	-	0.08 ± 0.06	0.009 ± 0.007
October 27	Brookfield, IL	80 ± 20	-	-	-	-	-
	Average	52 ± 16	3.1 ± 1.5	6.9 ± 3.9	3.1 ± 1.7	0.13 ± 0.05	0.019 ± 0.010

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[*] The perimeter locations are given in terms of the grid coordinates in Figure 1.

TABLE 14

Radionuclides in Bottom Sediment, 1983

Date Collected	Location	Concentrations in 10^{-6} µCi/g		Concentrations in 10^{-9} µCi/g		
		Antimony-125	Cesium-137	Plutonium-238	Plutonium-239	Americium-241
	Perimeter[*]					
August 11	50 m Above 7M Outfall	0.21 ± 0.09	0.06 ± 0.02	0.1 ± 0.1	1.6 ± 0.3	0.4 ± 0.1
August 11	7M Outfall	0.16 ± 0.09	0.70 ± 0.04	5.9 ± 0.4	62.2 ± 2.2	3.9 ± 0.8
August 11	50 m Below 7M Outfall	< 0.1	0.22 ± 0.04	0.7 ± 0.2	8.8 ± 0.7	1.0 ± 0.1
August 11	100 m Below 7M Outfall	0.34 ± 0.11	0.23 ± 0.02	0.8 ± 0.2	12.4 ± 0.8	0.1 ± 0.1
August 11	500 m Below 7M Outfall	0.16 ± 0.11	0.32 ± 0.03	2.3 ± 0.3	26.2 ± 1.3	0.4 ± 0.1
	Off-Site					
June 15	DuPage River, Naperville, IL	0.54 ± 0.13	0.15 ± 0.02	0.2 ± 0.1	2.6 ± 0.3	0.1 ± 0.1
June 15	Des Plaines River, Romeoville, IL	0.49 ± 0.12	0.37 ± 0.03	0.7 ± 0.2	12.5 ± 0.8	0.2 ± 0.2
June 16	DuPage River, Channahon, IL	0.41 ± 0.13	0.31 ± 0.03	0.5 ± 0.2	5.6 ± 0.5	0.1 ± 0.1
June 16	Illinois River, McKinley Woods State Park, IL	0.62 ± 0.15	0.19 ± 0.02	0.4 ± 0.2	5.8 ± 0.7	-
June 16	Illinois River, Dresden Lock & Dam, IL	0.16 ± 0.04	0.12 ± 0.01	0.1 ± 0.1	1.6 ± 0.2	< 0.1
October 26	Illinois River, Starved Rock State Park, IL	0.63 ± 0.26	0.04 ± 0.03	< 0.1	0.9 ± 0.2	0.6 ± 0.2
October 26	Illinois River, Morris, IL	0.38 ± 0.32	0.36 ± 0.06	0.3 ± 0.1	11.7 ± 0.6	-
October 27	Long Run Creek, Lemont, IL	0.40 ± 0.25	0.14 ± 0.04	0.1 ± 0.1	2.6 ± 0.3	-
October 27	Salt Creek, Western Springs, IL	0.19 ± 0.17	0.02 ± 0.02	0.1 ± 0.1	1.8 ± 0.2	-
October 27	Des Plaines River, Brookfield, IL	0.51 ± 0.20	0.35 ± 0.06	0.1 ± 0.1	2.0 ± 0.3	-
	Average	0.43 ± 0.10	0.21 ± 0.08	0.3 ± 0.1	4.7 ± 2.7	0.2 ± 0.2

[*] The perimeter locations are given in terms of the grid coordinates in Figure 1.

September 24, 1980. Comparison of plutonium concentrations indicates that the 1982 results were an order of magnitude higher at the outfall, but similar farther downstream, while the 1980 results were more like the 1983 set. The changes in concentrations of the various nuclides with time indicates the dynamic nature of the sediment material in this area.

4. Milk

Raw milk was collected monthly from a local dairy farm south of Lemont and analyzed for several radioactive nuclides. Iodine-131 was analyzed with a detection limit of 1×10^{-10} µCi/mL by an ion-exchange separation followed by beta counting. Cesium-137 was analyzed with a detection limit of 5×10^{-10} µCi/mL by an ion-exchange separation followed by gamma-ray spectrometry. The other nuclides were analyzed by the same methods used for water and with the same detection limits. The results are given in Table 15. The average strontium-90 and cesium-137 concentrations were similar to the 1982 results, the hydrogen-3 concentrations were somewhat higher. These nuclides are fission products from nuclear tests and their presence in milk is not related to Argonne operations. No short-lived fission products, i.e., strontium-89, iodine-131, or barium-140 were detected in milk.

The concentrations given in Table 15 may be compared to the CGs for drinking water given in the Appendix, Part B. The drinking water CGs are based on an intake of 2.2 liters per day. The consumption of one liter of milk per day would result in an average intake of 0.5% of the strontium-90 and 0.003% of the cesium-137 and hydrogen-3 Concentration Guides.

5. External Penetrating Radiation

Measurements were made with calcium fluoride and lithium fluoride thermoluminescent dosimeter (TLD) chips. Each measurement was the average of four chips exposed in the same packet. The response of the chips was determined with a National Bureau of Standards standard radium-226 source, and the results calculated in terms of air dose. Dosimeters were exposed at a number of locations at the site boundary to determine any dose that

TABLE 15

Radionuclides in Milk, 1983
(Concentrations in 10^{-9} µCi/mL)

Date Collected	Hydrogen-3	Strontium-90	Cesium-137
January 5	< 100	3.0 ± 0.5	0.4 ± 0.7
February 2	101 ± 94	3.2 ± 0.2	0.6 ± 0.2
March 2	118 ± 95	3.0 ± 0.5	0.6 ± 0.3
April 6	< 100	2.5 ± 0.4	0.6 ± 0.2
May 4	249 ± 94	2.8 ± 0.3	0.8 ± 0.3
June 1	228 ± 93	5.9 ± 0.6	1.0 ± 0.4
July 5	223 ± 93	6.1 ± 0.6	1.2 ± 0.5
August 3	266 ± 94	5.2 ± 0.9	1.1 ± 0.2
September 7	317 ± 95	4.6 ± 2.1	1.4 ± 0.5
October 12	176 ± 92	2.8 ± 0.6	0.8 ± 0.2
November 2	208 ± 93	3.9 ± 1.4	0.5 ± 0.3
December 7	< 100	2.1 ± 0.2	1.3 ± 0.3
Average	182 ± 44	3.8 ± 0.8	0.9 ± 0.2

might be due to Argonne operations at the closest uncontrolled approaches to the Laboratory, and at several locations on the site. The latter were chosen for two principal purposes: to determine where abnormal doses might be encountered, [illegible] where the results might be useful in determining the origin of any abnormal dose readings obtained at the boundary. Readings were also taken at five off-site locations for comparison purposes. These locations are shown in Figure 2.

The results are summarized in Tables 16 and 17, and the site boundary and on-site readings are also shown in Figure 6. Measurements were made for four successive exposure periods that varied in length from 85 to 104 days, and in total covered the period from January 17, 1983, to January 24, 1984. The results for each period were calculated in terms of annual dose for ease in comparing measurements made for different elapsed times, and were weighted according to their exposure times in calculating the annual average at each location. The uncertainty given in the tables for an average is the 95% confidence limit calculated from the standard deviation of the average.

The off-site results averaged 95 ± 11 mrem/yr. From 1973 to 1982, the corresponding values varied from 89 ± 7 mrem/yr (1979) to 100 ± 23 mrem/yr (1973). The ten-year average was 94 ± 3 mrem/yr. Thus, the background radiation has been quite consistent and the 1983 average is within the range found previously. The variations from year to year have also been consistent at each location. The annual averages at Downers Grove, Lemont, and Lombard have been within 5 mrem/yr of the average of all five off-site locations (80% have been within 3 mrem/yr). At Oak Lawn, the results have been from 6 to 13 mrem/yr (average, 10 mrem/yr) below average and at Oakbrook, 3 to 11 mrem/yr (average, 8 mrem/yr) above average.

If the off-site locations are an accurate sample of the radiation background in the area, then annual averages at the site boundary in the range of 95 ± 11 mrem/yr may be considered normal with a 95% probability, or 95% of the normal boundary results should have averages in this range. To compare boundary results for individual sampling periods, the standard deviation of the 19 individual off-site results is useful. This

TABLE 16

Environmental Penetrating Radiation at Off-Site Locations, 1983

	Dose Rate (mrem/year)				
	Period of Measurement				
Location	1/17-4/12	4/12-7/6	7/6-10/18	10/18-1/24	Average
Downers Grove	89	96	96	100	95 ± 7
Lemont	91	93	96	107	97 ± 13
Lombard	97	93	98	104	98 ± 8
Oakbrook	100	100	109	109	104 ± 10
Oak Lawn	83	80	80	90	83 ± 9
Average	92 ± 9	92 ± 10	96 ± 14	102 ± 10	95 ± 11

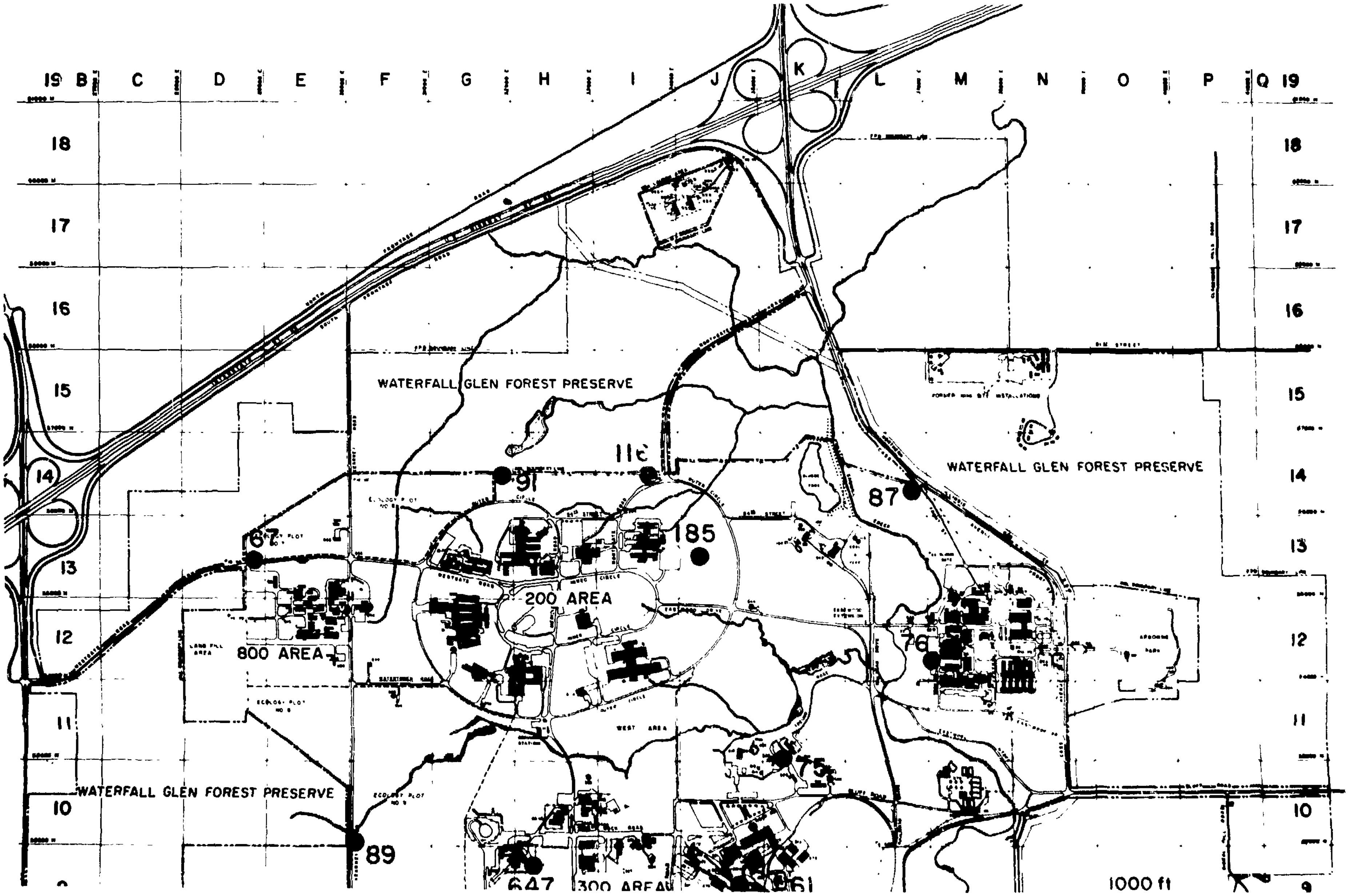
value is 8.5 mrem/yr, so that individual results in the range of 95 ± 17 mrem/yr may be considered normal with a 95% probability, unless there are known reasons to the contrary.

At two site boundary locations, 7I (south) and 14I (north), the dose rates were consistently above the normal range. At 7I this was due to radiation from a Radioactive Waste Storage Facility in the northern half of grid 7I. Waste is packaged and temporarily kept in this area prior to removal for permanent storage elsewhere. The net above-normal dose at this location was about 200 mrem/yr, half of the corresponding 1982 average. In previous years, this value has ranged from 560 mrem/yr in 1980 to 114 mrem/yr in 1977. About 300 m (0.2 mi) south of the fence in grid 6I, the measured dose dropped to 105 ± 19 mrem/yr, which is not statistically different from normal levels. The dose rates at these locations were relatively constant throughout the year, and similar to the last quarter of 1982. Earlier in 1982 these dose rates were three times greater, but decreased to current levels after shipment of a substantial portion of the stored waste to disposal sites. The above-normal dose at the 7I boundary was about 5% of the dose at the center of the area. The above-normal dose at the 8H/I location, 20 m NW of the Waste Storage Area

TABLE 17

Environmental Penetrating Radiation at ANL, 1983

	Dose Rate (mrem/year)				
	Period of Measurement				
Location	1/17-4/12	4/12-7/6	7/6-10/18	10/18-1/24	Average
14L - Boundary	88	83	89	87	87 ± 5
14I - Boundary	119	112	113	122	116 ± 9
14G - Boundary	89	85	95	93	91 ± 8
13D - Boundary	70	62	68	67	67 ± 6
9/10EF - Boundary	82	85	92	97	89 ± 12
8H - Boundary	101	95	109	117	105 ± 18
8H - Boundary, Center, St. Patrick's Cemetery	101	102	110	116	108 ± 13
7I - Boundary	310	197	273	379	292 ± 139
6I - 200 m N of Quarry Road	97	95	106	118	105 ± 19
9L - Boundary	-	72	69	75	72 ± 13
9H - 50 m SE of CP-5	652	598	600	735	647 ± 118
8H - 65 m S of Building 316	90	88	97	103	95 ± 13
8H/I - 200 m NW of Waste Storage Area	115	111	115	124	116 ± 10
7I - Center, Waste Storage Area	5,120	2,950	3,600	4,700	4,090 ± 1,830
10/11K - Lodging Facilities	72	70	78	80	75 ± 9
9J - Between ZGS Condenser and Building 370	58	57	60	67	61 ± 9
13J - 135 m E of Building 202	187	183	181	188	185 ± 6
12M - 30 m W of Building 55	70	75	73	87	76 ± 14
9I - 65 m NE of Building 350, 230 m NE of Building 316	88	77	88	89	86 ± 10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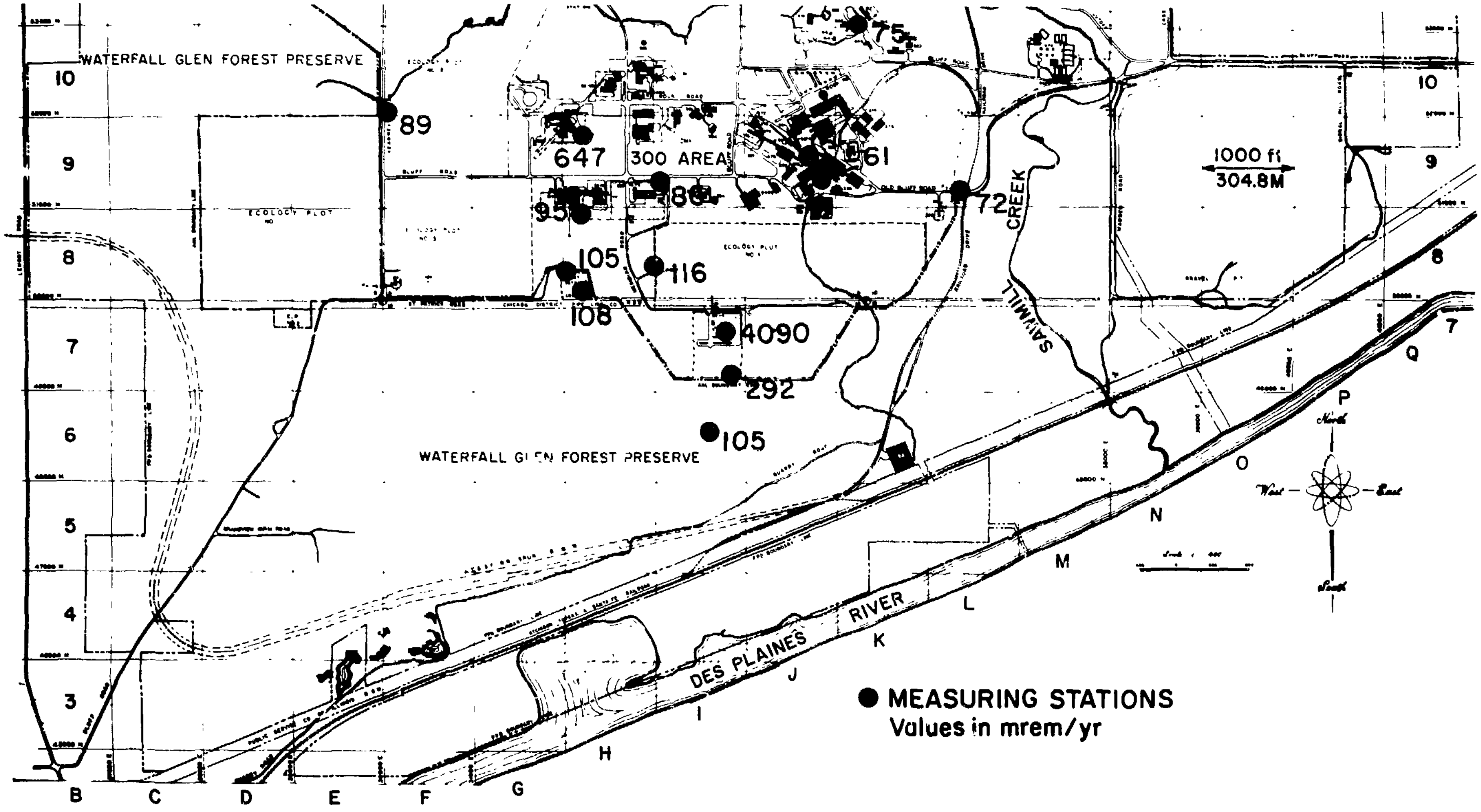


Fig. 6. Penetrating Radiation Measurements at the ANL Site, 1983

also responded to the removal of stored waste in the same manner as the 7I readings, indicating that the Storage Area was also responsible for the elevated dose rates at that location. At location 14I, the north boundary, the dose rate was 21 mrem/yr above normal, about the same as during 1982. This dose is due to the use of cobalt-60 irradiation sources in Building 202.

The dose in the south portion of grid 8H is of interest since this area includes St. Patrick's Cemetery, which was in use before Argonne was constructed and is open to visitors. In 1983, as in previous years, this dose is estimated to be 5 to 10 mrem/yr above normal, and was formerly attributed to radiation from the Storage Area at location 7I. This conclusion appeared reasonable, in view of the off-site averages, the topography of the area, and the lower dose rates north of the Cemetery and south of radiation sources in grid 9H. However, the dose at the Cemetery did not decrease as did the dose rates at other locations where above-normal results were attributed to the 7I Storage Area. The 8H averages were compared with the off-site average with the use of standard statistical tests, and differences were not significant at the 95% confidence level. In addition, the highest off-site location, Oakbrook, consistently exhibits dose rates very close to those at the Cemetery. Possible explanations are that the 8H dose rates are natural or that the granite stones in the Cemetery produce the elevated rates. This matter is under investigation.

6. Potential Radiation Dose Estimates

a. Radionuclides in Air and Water

The radiation doses at the site boundary and off the site that could have been received by the public from radioactive materials leaving the site were calculated by two methods. Where measured radionuclide concentrations in water were available, conversion of concentration to dose was based on the ratio of environmental concentrations to the Concentrations Guides given in Table 18. This table gives the annual radiation doses (after 70 years of intake for long-lived nuclides in bone) that would re-

TABLE 18

Concentration-to-Dose Conversion Factors

Nuclide	Medium	Concentration* (μCi/mL)	Dose* (rem)	Critical Organ
Americium-241	Water	4×10^{-6}	1.5	Kidney
	Water	5×10^{-6}	3	Bone
Argon-41	Air	4×10^{-8}	0.5	Whole Body
Californium-249	Water	4×10^{-6}	3	Bone
Californium-252	Water	7×10^{-6}	1.5	GI (LLI)
	Water	2×10^{-5}	3	Bone
Carbon-11	Air	6×10^{-8}	0.5	Whole Body
Curium-242	Water	2×10^{-5}	1.5	GI (LLI)
	Water	2×10^{-4}	3	Bone
Curium-244	Water	7×10^{-6}	3	Bone
Hydrogen-3 (H_2O)	Air	2×10^{-7}	0.5	Whole Body
	Water	3×10^{-3}	0.5	Whole Body
Krypton-85	Air	3×10^{-7}	0.5	Whole Body
Neptunium-237	Water	3×10^{-6}	3	Bone
Plutonium-238	Water	5×10^{-6}	3	Bone
Plutonium-239	Water	5×10^{-6}	3	Bone
Strontium-90	Water	3×10^{-7}	3	Bone

*The concentrations and doses, except for carbon-11, are the Concentration Guides and Radiation Protection Standards (RPS), respectively, specified in DOE Order 5480.1, Chapter XI for individuals in uncontrolled areas, except for americium-241 (bone), californium-252 (GI and bone), and curium-242 (bone). Since RPS values for these nuclide-organ combinations are not given in DOE Order 5480.1, Chapter IX, the concentrations used were one-tenth of the 168 hour occupational values specified by the ICRP.[12] For carbon-11, the values are taken from reference 13.

sult from continuous exposure at the specified concentrations for those nuclides whose presence in the environment are attributable to Argonne.

Carbon-11, argon-41, and krypton-85 are the major gaseous radioactive effluents released from the Laboratory. These nuclides also produce the principal exposure pathway, directly from air to man by submersion, for radioactive substances released from Argonne. The concentrations and dose rates were calculated for those three nuclides by a computer program based on an atmospheric dispersion model.[14,15] The following parameters were used in the calculations:

a) annual release rates: carbon-11, 119 Ci; argon-41, 0.75 Ci; and krypton-85, 2.2 Ci.
b) meteorological data: the wind speed and direction data shown in Figure 3.
c) the usual parameters for stack height, building, wake, plume, momentum, temperature, etc.

The calculations were carried out to 80 km (50 mi) using the population distribution of 16 segments and ten distance increments given in Table 2. The dose rate was calculated at the midpoint of each interval and integrated over the entire area to give the annual cumulative dose.

The highest perimeter dose rates are in the north to east sectors. The closest full-time resident, who would receive the largest dose, is located approximately 0.5 km (0.3 mi) north of the site boundary. The results are summarized in Table 19.

The population data in Table 2 was used to calculate the cumulative population dose from carbon-11, argon-41, and krypton-85. The results are given in Table 20, together with the natural external radiation dose. The natural radiation dose was that measured at the off-site TLD locations, and it is assumed that this dose is r resentative of the entire area within an 80 km (50 mi) radius.

TABLE 19

Dose From Airborne Emissions, 1983
(Millirems)

		Maximum Perimeter		Maximum Individual	
Nuclide	Pathway	Annual	50-Year	Annual	50-Year
Carbon-11	Submersion	0.04	0.04	0.02	0.02
Argon-41	Submersion	0.0005	0.0005	0.0002	0.0002
Krypton-85	Submersion	0.0002	0.0002	0.0001	0.0001

TABLE 20

80 km Population Dose, 1983

	Man-Rems	
Source	Annual	50-Year
Carbon-11	0.292	0.292
Argon-41	0.012	0.012
Krypton-85	0.011	0.011
Natural	7.65×10^5	-

The only location where radionuclides attributable to Argonne operations could be found in off-site water was Sawmill Creek below the wastewater outfall. Although this water is not used for drinking purposes, the dose that would be received by an individual ingesting water at the concentrations found in the Creek was calculated. A minor exposure route is from water to man by ingestion 710 km (370 mi) downstream from Argonne (Section I.E.), but the dilution of Sawmill Creek water at this point is so great that the dose calculation is meaningless. No other exposure pathways are significant. Those nuclides added to Sawmill Creek by Argonne waste water, their net concentrations in the Creek, and the corresponding dose rates, if water at these concentrations were used as the sole water supply by an individual, are given in Table 21. The dose rates were all well below the standards for individuals in uncontrolled areas. It should be emphasized that Sawmill Creek is not used for drinking, swimming, or boating. Inspection of the area shows there are few fish in the stream, and they do not constitute a significant source of food for any individual.

TABLE 21

Radionuclide Concentrations and Dose Estimates for Sawmill Creek Water, 1983

Nuclide	Conc. (net avg.) 10^{-9} µCi/mL	Dose mrem/year	Percent of Standard*
Hydrogen-3	101	0.017	0.0034
Strontium-90	< 0.05	< 0.5	< 0.017
Neptunium-237	0.0033	0.0033	0.00011
Plutonium-239	0.0085	0.0051	0.00017
Americium-241	0.0067	0.0025 (kidney)	0.000084
		0.0040 (bone)	0.00013
Curium-244	0.0019	0.0008 (bone)	0.00003
Californium-249	0.0019	0.0014	0.00005

*DOE Order 5480.1 Standard.[5]

As indicated in Table 7, occasional Sawmill Creek samples (less than 10) contained traces of plutonium-238 and curium-242 and/or californium-252 nuclides, but the averages were only slightly greater than the detection limit. The annual dose due to an individual consuming water at these concentrations can be calculated as was done for those nuclides more commonly found in Creek water, but the method of averaging probably overestimates the true concentration. These annual doses range from 1×10^{-3} to 2×10^{-5} mrem/yr for these nuclides.

The Environmental Protection Agency (EPA) has established drinking water standards based on a dose of 4 mrem/yr for man-made beta particle and photon emitting radionuclides.[16] For hydrogen-3, the EPA standard is 2×10^{-5} μCi/mL and for strontium-90, it is 8×10^{-9} μCi/mL. The concentrations in Table 22 correspond to 0.5% for hydrogen-3 and < 0.6% for strontium-90 of the EPA standards. No specific EPA standards exist for the transuranic nuclides.

b. External Penetrating Radiation Dose

Above normal fence-line doses attributable to Argonne operations were found at the south boundary near the Waste Storage Facility (location 7I) and at the north boundary near Building 202 (14I). The results are discussed in Section III.A.5.

At location 7I, the fence-line dose from Argonne was about 200 mrem/yr. Approximately 300 m (0.3 mi) south of the fence line (grid 6I), the measured dose averaged 105 ± 19 mrem/yr, approximately the upper end of the normal range, 106 mrem/yr. There are no individuals living in this area. The closest residents are about 1.6 km (1 mi) south of the fence line. At this distance, the calculated dose rate from the Waste Storage Area is 0.005 mrem/yr, if the energy of the radiation were 0.66 MeV, and 0.026 mrem/yr, if the energy were 1.3 MeV. The energy spectrum of the radiation varies with the composition of the stored waste and is not known, so it is necessary to assume an energy to make the calculations. Since cesium-137 and cobalt-60 are common radionuclides, the energies of the gamma-rays from these nuclides were used in the calculations. In the area north of the site, where the fence-line radiation dose from the cobalt-60 sources in Building 202 was measured at about 21 mrem/yr, the nearest residents are 750 m (0.47 mi) to the north-northwest. The calculated dose at that location was about 0.04 mrem/yr.

TABLE 22

Water Quality Standards and Detection Limits
(Concentrations in mg/L)

Constituent	State Standard Stream	State Standard Effluent	Detection Limit
Ammonia Nitrogen (as N)	1.5	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	0.1
Arsenic	1.0	0.25	0.005
Barium	5.0	2.0	0.005
Cadmium	0.05	0.15	0.0004
Chromium(VI)	0.05	0.3	0.003
Chromium(III)	1.0	1.0	0.003
Copper	0.02	1.0	0.0008
Cyanide	0.025	0.025	0.020
Fluoride	1.4	15	0.02
Iron	1.0	2.0	0.05
Lead	0.1	0.1	0.002
Manganese	1.0	1.0	0.001
Mercury	0.0005	0.0005	0.0001
Nickel	1.0	1.0	0.003
pH	6.5-9.0	5.0-10.0	-
Selenium	1.0	1.0	0.005
Silver	0.005	0.1	0.0002
Sulfate	500	-	1.0
Temperature	-	-	0.1°C
Total Dissolved Solids	1000	-	-
Zinc	1.0	1.0	0.01

The applicable Radiation Protection standards for whole body external radiation dose to the general population is a maximum of 500 mrem/yr to critical individuals, or if individual doses are not known, 170 mrem/yr to a suitable sample of the exposed population.[5] The latter criterion assumes that the maximum dose to individuals in the sample will not exceed the average by more than a factor of three. Thus, the external penetrating radiation dose to individuals living near the site will not exceed 0.01% of the 500 mrem/yr limit or 0.03% of the "suitable sample" limit. At the fence line, where higher doses were measured, the land is wooded and unoccupied.

All of these dose calculations are based on full-time, outdoor exposure. Actual exposures to individuals are substantially less since they are inside (which provides shielding) or away from their dwelling much of the time.

c. Summary

The total dose received by off-site residents was a combination of the separate pathways that contribute to this total: carbon-11, argon-41, and krypton-85 submersion dose and cobalt-60 external radiation dose. The highest dose was about 0.06 mrem/yr to individuals living north of the site if they were outdoors at that location during the entire year. The total annual population dose to the entire area within an 80 km (50 mi) radius is < 0.3 man-rem.

B. Chemical and Biological Pollutants

1. Air

The environmental air data reported for the past several years have been obtained to determine concentrations of total suspended particulates (TSP), trace metals, and polynuclear aromatic compounds prior to the burning of coal at Argonne as a replacement fuel for oil and gas. Coal use began on February 10, 1981, at Boiler No. 5, and monitoring was directed toward demonstrating compliance with the USEPA Order No. EPA-5-80-A.[17]

The Order provides that samples for TSP be collected for 24-hour periods at six-day intervals and that these results be reported monthly. For this purpose, samplers were maintained at location 8F, generally sampling air upwind of the boiler, and at location 12M, generally sampling air downwind of the boiler during the heating season. In 1982, the boiler became operative as designed and the Order was no longer in effect. In 1983, samples were obtained during the period January through August at which time the sampling was discontinued. The average geometric mean for the sample obtained at 12M was 55 $\mu g/m^3$ and the average geometric mean obtained for the sample at 8F was 50 $\mu g/m^3$. On May 6, 1983, there was a large scale dust storm in the midwest. Samples were collected on this day and results of 480 $\mu g/m^3$ and 560 $\mu g/m^3$ were obtained at 8F and 12M, respectively. If these two values are eliminated from the overall average, values of 45 and 52 $\mu g/m^3$ are obtained at the two locations.

Results obtained since the initiation of this program in 1977 have been consistent at all locations and no change in the total suspended particulates attributable to Argonne operations has been observed.

2. Water

The environmental water data contained in this section were collected to ascertain the effectiveness of Argonne's effluent pollution controls and determine compliance with state regulations and NPDES permits. The appropriate stream and effluent standards are listed in the ILLINOIS POLLUTION CONTROL BOARD RULES AND REGULATIONS. Chapter 3.[18] Stream Quality Standards appear in Part II and Effluent Quality Standards appear in Part IV of that document. All of the results are compared to the State standards, which are listed in Table 22. Minimum detectable amounts are included for comparison. The detection limits for atomic absorption methods represent twice the background variation, which is commonly used for this purpose. Detection limits for ion selective methods are those listed by the manufacturer. The lower limit of detection for x-ray fluorescence is based on the appropriate counting statistics.

The analytical methods have been described in previous reports in this series. The results of measurements for chemical constituents are expressed as milligrams (mg) or micrograms (μg) per liter (L). Averages were calculated as described in Section III.A. Yearly averages are reported with a (±) limit value. This value is the standard error at the 95% confidence limit and is calculated from the standard deviation of the yearly average. Only when sample concentrations are random does this value represent the actual distribution occurring at the sampling location.

As in the past, emphasis has been placed on Sawmill Creek, since this is the principal route for waste water leaving the Argonne site, and on the NPDES permit locations.

a. Waste Treatment Plant Effluent Water

The major discharge of waste water from Argonne operations is by way of the waste treatment plant. The water volume from this source was approximately 3.1 megaliters (0.82 million gallons) per day in 1983 and was comprised of 30% sanitary waste water and 70% water from laboratory operations. Laboratory waste is directed to a large holding pond with an overflow which controls the flow to about 1300 liters/minute (350 gallons/minute). The discharge occurs over a 24-hour period. Since the sanitary system is fairly constant, the overall output is constant.

Water from the waste treatment plant is collected on a continuous basis during the work week. A flow proportional 24-hour sample of the combined sanitary and laboratory effluent was obtained each day and was analyzed for constituents of interest.

The results obtained for chemical constituents in the effluent water from the treatment plant are shown in Table 23. All of the average concentrations were below the State standards, except for mercury. The average value for mercury was 116% of the State standard and 39% of the samples exceeded this value. The corresponding values for 1982 were 84% and 27%. The increase over previous years was due primarily to a sample obtained

TABLE 23

CHEMICAL CONSTITUENTS IN EFFLUENTS FROM ANL TREATMENT PLANT, 1983
(CONCENTRATIONS IN MICROGRAM/LITER)

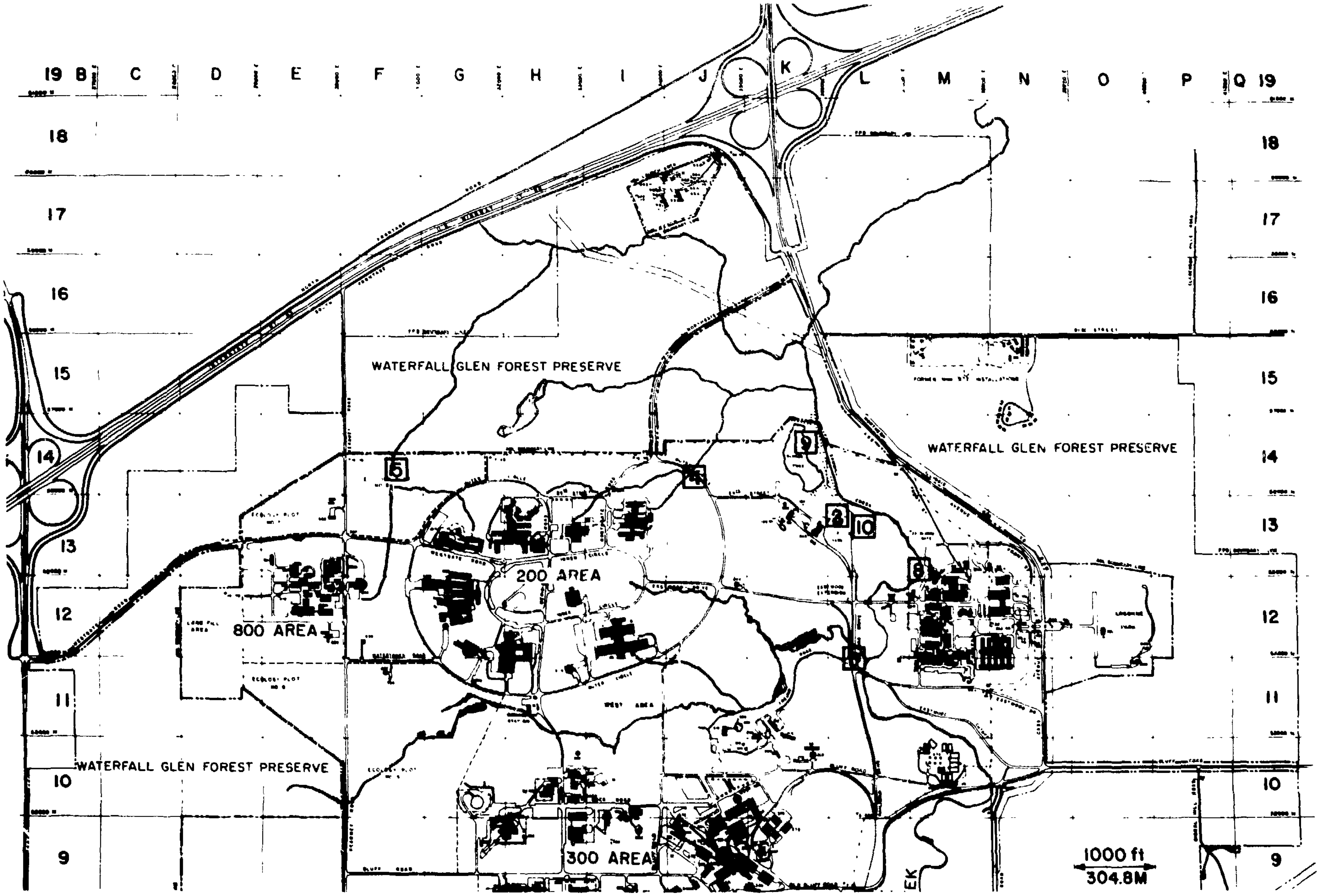
CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	MIN.	MAX.	PERCENT OF STANDARD (AVG.)	PERCENT EXCEEDING STATE STANDARD
ARSENIC	12	-	-	< 5	< 2.0	0
BARIUM	51	18 ± 2	6	35	0.9	0
BERYLLIUM	12	0.05 ± 0.02	0.01	0.09	-	-
CADMIUM	51	0.6 ± 0.1	< 0.2	2.1	0.4	0
COPPER	51	57 ± 25	11	484	5.7	0
FLUORIDE	51	420 ± 62	234	1150	2.8	0
IRON	51	226 ± 32	100	733	11	0
LEAD	51	2.5 ± 0.5	< 2.0	9.6	2.5	0
MANGANESE	51	22 ± 3	7	51	2.2	0
MERCURY	51	0.58 ± 0.17	0.11	3.44	116	39
NICKEL	51	9 ± 1	3	26	0.9	0
PH	241	-	6.4	7.8	-	0
SELENIUM	12	-	-	< 5	< 0.5	0
SILVER	51	2.2 ± 0.2	0.8	5.5	2.2	0
ZINC	51	94 ± 29	40	684	9.4	0

the first week of November which contained 3.4 μg/L. Efforts are continuing to lower these levels by monitoring the waste water retention tanks most likely to contain mercury, and treating those tanks containing elevated concentrations to remove most of the mercury. Concentration of all the other constituents are similar to last year's values, probably a measurement of ambient levels, and are not due to Argonne operations.

b. National Pollution Discharge Elimination System (NPDES)

The NPDES permit program provides for 10 sampling locations, which are shown in Figure 7. The permit is currently being considered for reissuance since the expiration date was October 1983. Discharge point 001 is located at the waste treatment plant and discharge point 002 is located at 13L and contains cooling water from the power plant. Other locations cited in previous reports as cooling tower effluents are related to current permit locations as follows. Permit location 003 monitors water from the combined 200 Area blowdown channels and is former location 12L. Location 004 (formerly 14J) monitors water from Building 202 and contains cooling water. Location 005 monitors water from former locations 12F and 14G and now includes water from the entire 800 Area. Location 006 monitors the canal water treatment plant outfall and is former location 8J. Locations 007, 008, and 009 are new sampling points and monitor water from ZGS cooling, East Area cooling, and the water plant and pond overflow, respectively. Permit location 010 is designed to monitor coal pile runoff, but contained no water in 1983. Monitoring at locations 002-009 was carried out on a once per month basis, and were grab samples. Sampling at location 001 was performed once per week.

The results obtained for the permit parameters for NPDES locations 001-009 are listed in Tables 24 to 32. At location 001, there was one coliform bacteria value that exceeded the limit and this was traced to low chlorine residual and was quickly alleviated. The samples for mercury analysis at 001 are obtained from the laboratory waste before it is combined with the sanitary waste water. The permit condition of 0.5 μg/L for mercury was exceeded 70% of the time by various amounts. In December, 1981, new effluent regulations were adopted by the Illinois Pollution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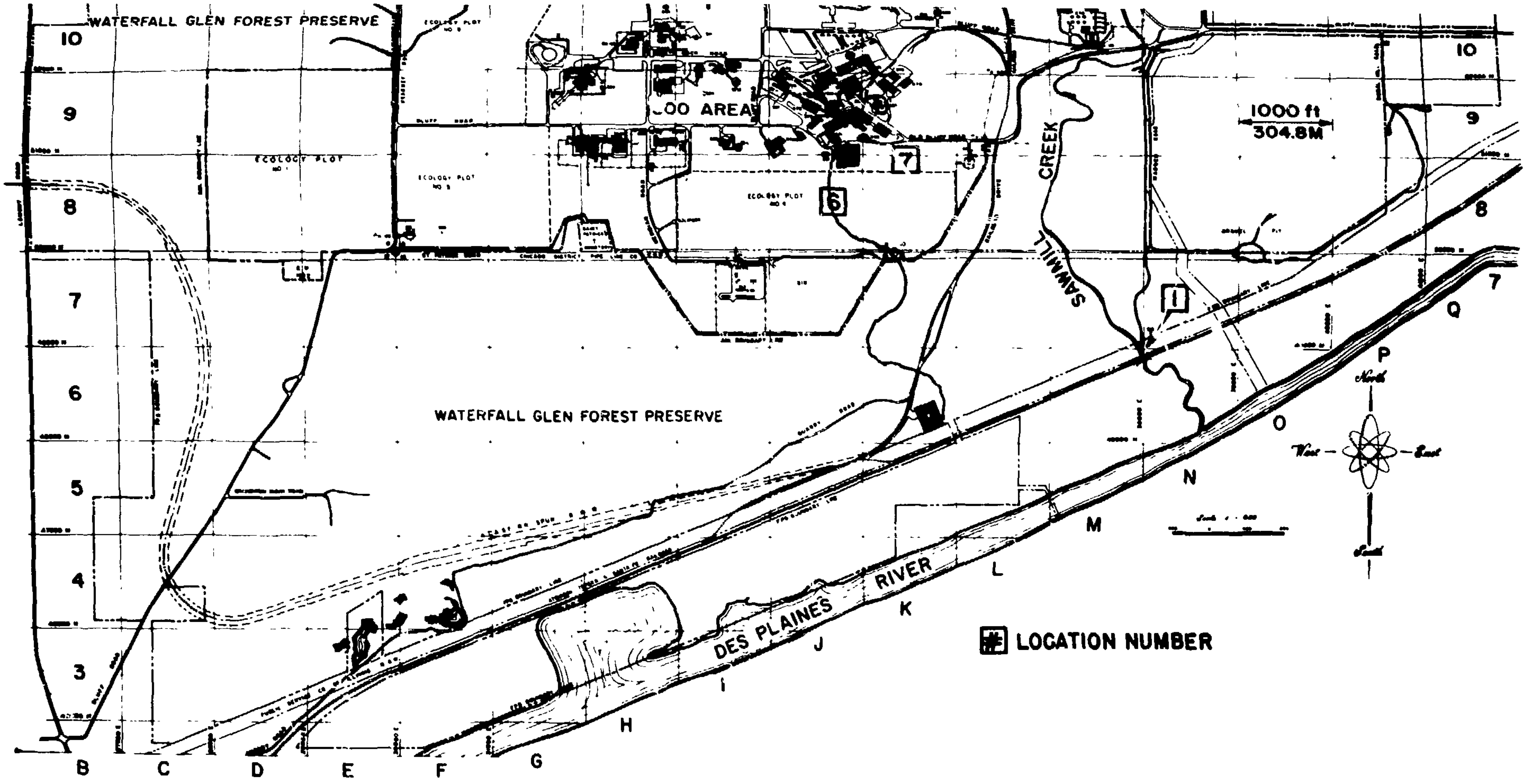

Fig. 7. NPDES Permit Locations

TABLE 24

ANL National Pollution Discharge Elimination System Data Summary, Location 001 (7M), 1983

Constituent	No. of Samples	Quantity or Conc. Avg.	Min.	Max.	Units	Permit Limit	Number Exceeding
B.O.D.	38	2.9 ± 0.4	1.6	7.6	mg/L	75	0
Flow	12	0.82 ± 0.06	0.70	1.00	MGD*	-	-
Mercury (total)	50	1.70 ± 1.21	0.20	30.20	μg/L	0.5	35
pH	37	-	6.1	7.7	Units	6-9	0
Total Suspended Solids	40	4.4 ± 1.2	1.0	19.0	mg/L	75	0
Gross Beta	52	17 ± 2	7	30	pCi/L	100	0
Strontium-90	52	0.26 ± 0.03	< 0.25	0.55	pCi/L	2	0
Radium-226	52	0.56 ± 0.08	0.22	1.52	pCi/L	1	6

*Million Gallons/Day.

TABLE 25

ANL National Pollution Discharge Elimination System Data Summary, Location 002 (13L), 1983

Constituent	No. of Samples	Quantity or Conc.			Units	Permit Limit	Number Exceeding
		Avg.	Min.	Max.			
pH	8	-	7.0	8.9	Units	6-9	0
Total Suspended Solids	7	40 ± 41	14	130	mg/L	15	3

TABLE 26

ANL National Pollution Discharge Elimination System Data Summary, Location 003 (12L), 1983

Constituent	No. of Samples	Quantity or Conc. Avg.	Min.	Max.	Units	Permit Limit	Number Exceeding
Total Chlorine Residual	12	-	-	< 0.1	mg/L	0.75	0
Total Dissolved Solids	12	545 ± 82	413	763	mg/L	3500	0
pH	12	-	7.9	8.7	Units	6.5-9.0	0
Total Suspended Solids	12	9.3 ± 2.1	3.9	17.1	mg/L	15	1

TABLE 27

ANL National Pollution Discharge Elimination System Data Summary, Location 004 (14J), 1983

Constituent	No. of Samples	Quantity or Conc. Avg.	Min.	Max.	Units	Permit Limit	Number Exceeding
Total Dissolved Solids	12	563 ± 62	438	802	mg/L	3500	0
Flow	12	0.06 ± 0.02	0.01	0.12	MGD*	-	-
pH	12	-	7.7	8.5	Units	6-9	0
Total Suspended Solids	12	7.8 ± 4.0	1.0	22.7	mg/L	15	1

*Million Gallons/Day.

TABLE 28

ANL National Pollution Discharge Elimination System Data Summary, Location 005 (14F), 1983

Constituent	No. of Samples	Quantity or Conc. Avg.	Min.	Max.	Units	Permit Limit	Number Exceeding
Flow	12	0.10 ± 0.04	0.01	0.20	MGD*	-	-
Oil	8	-	-	< 5.0	mg/L	30	0
pH	12	-	7.5	8.2	Units	6-9	0
Total Suspended Solids	12	45 ± 54	1.	312	mg/L	15	7

*Million Gallons/Day.

TABLE 29

ANL National Pollution Discharge Elimination System Data Summary, Location 006 (8J), 1983

Constituent	No. of Samples	Quantity or Conc. Avg.	Min.	Max.	Units	Permit Limit	Number Exceeding
Flow	12	0.10 ± 0.05	0.02	0.33	MGD*	-	-
pH	12	-	4.8	12.3	Units	6-9	2
Total Suspended Solids	12	26 ± 22	4	127	mg/L	15	5

*Million Gallons/Day.

TABLE 30

ANL National Pollution Discharge Elimination System Data Summary, Location 007 (9K), 1983

Constituent	No. of Samples	Quantity or Conc. Avg.	Min.	Max.	Units	Permit Limit	Number Exceeding
Flow	12	0.01 ± 0.01	0.01	0.02	MGD*	-	-
pH	12	-	7.9	11.4	Units	6-9	6

*Million Gallons/Day.

TABLE 31

ANL National Pollution Discharge Elimination System Data Summary, Location 008 (13M), 1983

Constituent	No. of Samples	Quantity or Conc. Avg.	Min.	Max.	Units	Permit Limit	Number Exceeding
pH	12	-	7.8	8.3	Units	6-9	0

TABLE 32

ANL National Pollution Discharge Elimination System Data Summary, Location 009 (14K), 1983

Constituent	No. of Samples	Quantity or Conc. Avg.	Min.	Max.	Units	Permit Limit	Number Exceeding
Flow	12	0.09 ± 0.02	0.01	0.16	MGD*	-	-
pH	12	-	7.8	10.8	Units	6-9	11
Total Suspended Solids	12	15 ± 18	1	106	mg/L	15	1

*Million Gallons/Day.

Control Board which provides a monthly limit, by exemption, of 3.0 μg/L for mercury discharges from research facilities. If this limit were in effect, all the results except one would be in compliance. Violations of a few parameters occurred at other locations due to a variety of temporary conditions, such as suspended solids due to runoff from heavy precipitation. At location 006, the suspended solids were frequently above the limit of 15 mg/L, probably as a result of Argonne operations. Attempts to resolve this problem are underway. At locations 007 and 009, the pH was usually above the upper limit of 9.0. This is probably the result of the use of alkaline water softening compounds at 009 and stagnant water conditions at 007. The high pH water has no impact on the receiving stream, Sawmill Creek, since the relative volume and buffering capacity of the effluent are low.

c. Sawmill Creek

Samples collected for evaluation of the effect of the sanitary waste on stream quality were obtained once per week using specially constructed sampling bottles. The sampling bottles were designed to provide temperature measurements as well as to minimize changes in oxygen content during collection. These samples were collected 15 m (50 ft) upstream of the Argonne wastewater outfall [7M (up)] and 60 m (200 ft) downstream of the outfall [7M (down)].

The data from these studies are in Table 33. Average levels for ammonia nitrogen were below the State limit at both locations for the first time since studies began. This is due, in part, to the mild weather in January and February, when the stream did not freeze as it usually does. The distribution of ammonia is highly temperature dependent and, when the surface is frozen, diffusion is prevented. Hence, normal loss of ammonia to air is prevented when the surface is covered with ice. The dissolved oxygen levels obtained during 1983 were all above the State minimum of 5 mg/L. The total dissolved solids above the outfall exceeded the State standard in about half of the samples, and increased the solids content of the downstream sample slightly. The dissolved solids averaged about 25%

TABLE 33

SAWMILL CREEK - EFFECT OF SANITARY WASTE, 1983

CONSTITUENT	LOCATION *	NO. OF SAMPLES	CONCENTRATION (MG/L) AVG.	MIN.	MAX.	AVG. PERCENT OF STANDARD	PERCENT EXCEEDING STATE STANDARD
AMMONIA NITROGEN	7M (UP)	37	0.6 + 0.3	0.1	4.3	40	5
	7M (DOWN)	37	0.6 ± 0.2	0.1	3.9	39	5
CHLORIDE	7M (UP)	10	352 + 106	157	600	-	-
	7M (DOWN)	10	324 ± 94	158	520	-	-
CYANIDE	7M (DOWN)	40	-	-	< 0.01	40	0
DISSOLVED OXYGEN	7M (UP)	41	11.2 + 0.4	7.8	14.9	-	-
	7M (DOWN)	41	11.1 ± 0.4	7.8	14.4	-	-
DISSOLVED SOLIDS	7M (UP)	41	1010 + 120	490	1810	101	49
	7M (DOWN)	41	972 ± 104	491	1650	97	46
PH **	7M (UP)	41	-	7.8	8.8	-	-
	7M (DOWN)	41	-	7.7	8.8	-	-
SULFATE	7M (UP)	10	150 + 25	81	212	30	0
	7M (DOWN)	10	149 ± 28	78	208	30	0
TEMPERATURE ***	7M (UP)	41	14.1 + 2.4	2.7	26.7	-	-
	7M (DOWN)	41	13.8 ± 2.3	3.0	26.3	-	-

---

* LOCATION 7M (UP) IS 15 M (50 FT) UPSTREAM FROM THE WASTE-WATER OUTFALL. ALL OTHER SAMPLES WERE COLLECTED 60 M (200 FT) DOWNSTREAM FROM THE OUTFALL.

** UNIT

*** DEGREES CENTIGRADE

lower in 1983 than in 1982, probably due to the larger amount of precipitation in 1983.

Samples to evaluate the effect of combined sanitary and laboratory waste on the concentrations of chemical constituents in Sawmill Creek were collected by a continuous sampler in the Creek five times per week. These were the same samples taken for radioactivity analyses. The results are summarized in Table 34.

None of the individual samples for arsenic, barium, cadmium, chromium, fluoride, lead, nickel, selenium, silver, and zinc exceeded the State standards. The levels of mercury averaged 24% of the State limit and exceeded this limit 2% of the time, a factor of two improvement over 1982. As in the past, high iron levels reflect high turbidity conditions and are not related to Argonne releases. The levels of copper exceeded State limits 53% of the time, but they are not due to ANL operations since the average stream and effluent copper levels differ by a factor of only two, but the effluent water is diluted by about a factor of 10 with stream water.

d. Des Plaines River

The effect of Sawmill Creek on the Des Plaines River was evaluated by collecting samples at Willow Springs (upstream of Argonne) and at Lemont (downstream of Argonne). These samples were analyzed for total mercury, hexavalent chromium, total iron, and total zinc. The results are in Table 35. In no case was there any indication that hexavalent chromium, mercury, or zinc levels were affected by the Argonne effluent.

TABLE 34

CHEMICAL CONSTITUENTS IN SAWMILL CREEK LOCATION 7M,* 1983
(CONCENTRATIONS IN MICROGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION			PERCENT OF STANDARD (AVG.)	PERCENT EXCEEDING STATE STANDARD
		AVG.	MIN.	MAX.		
ARSENIC	12	5 ± 1	< 5	10	0.5	0
BARIUM	51	54 ± 6	10	96	1.1	0
BERYLLIUM	12	0.07 ± 0.03	0.01	0.13	-	-
CADMIUM	51	1.0 ± 0.2	0.3	3.1	2.0	0
CHROMIUM(III)	51	12 ± 5	3	96	1.2	0
CHROMIUM(VI)	51	-	-	< 3	< 6.0	0
COPPER	51	26 ± 5	8	93	131	53
FLUORIDE	51	455 ± 40	240	712	32	0
IRON	51	1170 ± 360	34	6300	117	37
LEAD	51	6 ± 1	1	20	5.9	0
MANGANESE	51	161 ± 36	27	642	16	0
MERCURY	51	0.12 ± 0.03	< 0.05	0.60	24	2
NICKEL	51	10 ± 1	3	23	1.0	0
PH	247	-	7.0	8.3	-	0
SELENIUM	12	-	-	< 5	< 0.5	0
SILVER	51	1.2 ± 0.2	0.3	3.4	23	0
ZINC	51	50 ± 8	10	149	5.0	0

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* LOCATION 7M IS 60 M (200FT) DOWNSTREAM FROM THE WASTE-WATER OUTFALL.

TABLE 35

Chemical Constituents in the Des Plaines River, 1983

Constituent	Location*	No. of Samples	Concentration (mg/L) Avg.	Min.	Max.
Chromium(VI)	A	12	-	-	< 0.01
	B	24	-	-	< 0.01
Iron	A	12	1.6 ± 0.7	0.6	3.9
	B	24	1.0 ± 0.3	0.3	3.5
Mercury	A	12	-	-	< 0.0001
	B	24	-	-	< 0.0001
Zinc	A	12	0.06 ± 0.03	0.03	0.18
	B	24	0.04 ± 0.01	0.01	0.07

*Location A, near Willow Springs, is upstream and Location B, near Lemont, is downstream from the mouth of Sawmill Creek. See Figure 2.

3. Ground Water

A sanitary landfill for non-radioactive waste is located on the western edge of the Argonne site (Figures 1 and 8). This landfill operates under Illinois EPA permit No. 1981-29-OP and is further described in Reference 2. Operation of the ANL landfill began in July, 1966. Its use was restricted to the disposal of waste generated by normal Laboratory activities. Disposal of radioactive materials was prohibited and there is no evidence that any have been deposited at this site. However, during the period 1969-1978, substantial quantities of liquid organic waste were added to a "French drain" which was located in the northeast sector of the landfill.

In 1979, an investigation was begun to determine the subsoil characteristics of the site and to place ground-water monitoring wells at appropriate points in and around the landfill. Since the topography suggested a south-southwesterly water flow, Wells No. 1 and No. 5 were located outside the landfill and were meant to measure water entering and leaving the site. Wells No. 2, No. 3, and No. 4 were placed in the landfill itself.

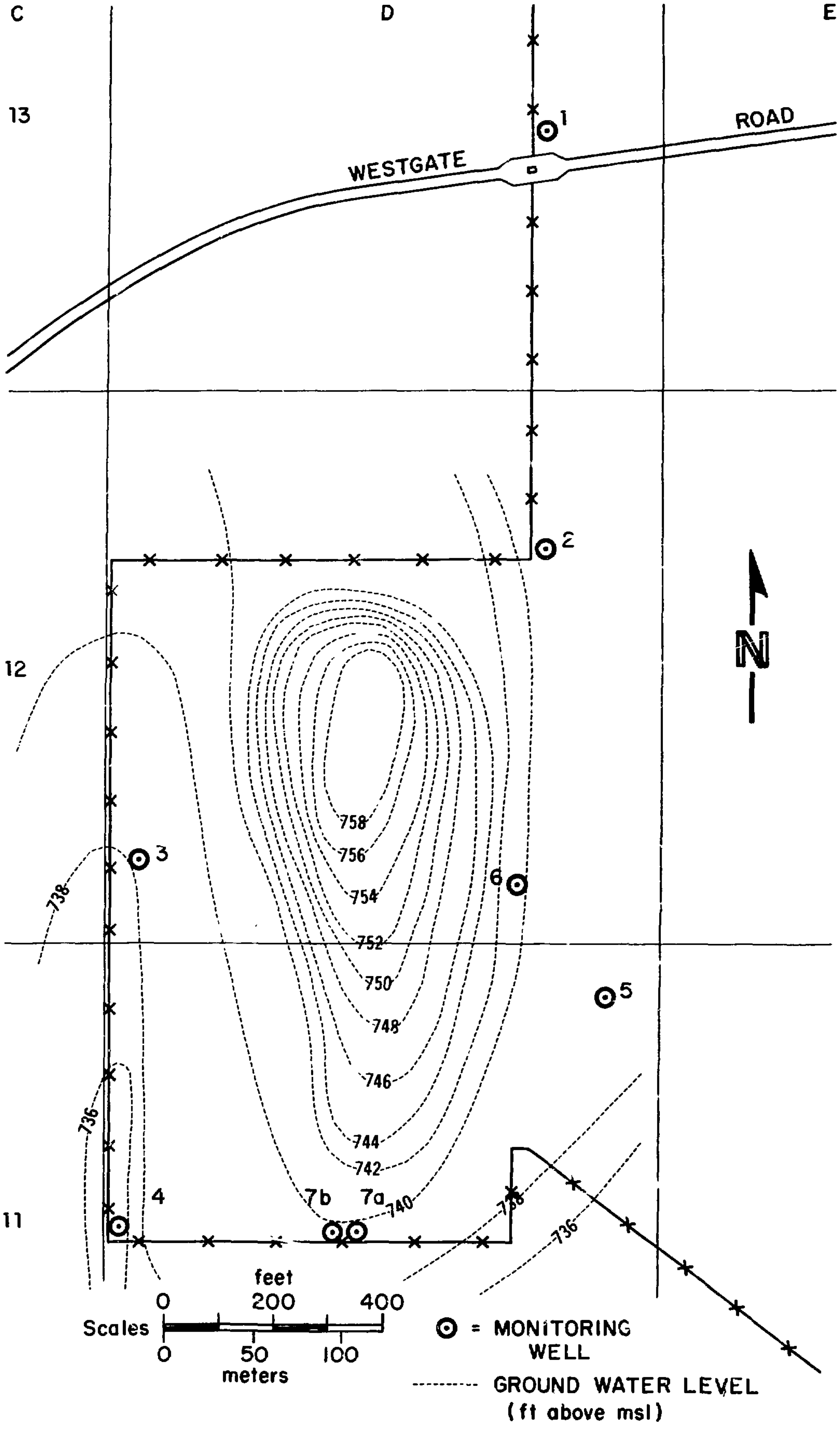

Fig. 8. Monitoring Well Locations for Landfill

In April, 1980,[19] a much more comprehensive study was initiated. Three additional test wells were placed in the landfill at previously untested locations. Well No. 6 was placed in the east section in the line of flow with Well No. 5. Wells No. 7a and No. 7b were nested and were installed to measure vertical water movement as well as to provide monitoring of water from two depths. Test borings were also made in the fill area to determine soil conditions where refuse had been added.

The study established the following important characteristics. There is a lateral perched water condition at a depth varying from about 6 m (20 ft) on the north to about 7.6 m (25 ft) on the south. This is caused by the relatively impermeable condition of the underlying clay which restricts downward water flow. There are several granular seams in the soil above the clay which permit lateral flow. Additionally, regional groundwater flow patterns indicate that there is a southerly flow of water with a slight westerly flow.

These facts indicate that water in the test wells is from surface infiltration and that it moves horizontally approximately 7.6 m (25 ft) below the surface. Penetration to the dolomite aquifer used for water supplies which lies 15-30 m (50-100 ft) below the surface is very slow. Indeed, core samples obtained at 11-12 m (35-40 ft) below the surface were only moist indicating that the aquifer is below this level and little downward movement is occurring. If this movement were occurring, this stratum would be saturated.

All of the wells have been studied from 1979 through 1983 to determine constancy of water levels, recharge rates, and chemical characteristics. Table 36 shows the annual average water levels during this period. The water levels have been fairly constant over this period, except for Wells No. 6 and No. 7a. In September, 1981, the surface above Well No. 6 was raised about 1.8 m (6 ft) and in all probability the subsurface soil permeability was disturbed. In November and December of 1982, the precipitation was about three times normal and the higher levels in these two wells reflect this.

TABLE 36

Water Levels and Depth of Monitoring Wells in ANL Landfill

Well No.	Depth From Surface (m)	1979	1980	1981	1982	1983
1	9.30	2.29 (1)	10.67 (3)	1.01 (4)	0.91 (5)	0.70 (3)
2	9.45	5.85 (1)	50.60 (5)	4.88 (1)	4.30 (1)	4.54 (3)
3	8.23	2.62 (1)	2.38 (3)	2.47 (4)	2.35 (5)	1.92 (3)
4	6.10	2.04 (1)	2.23 (5)	1.83 (4)	1.98 (5)	2.07 (3)
5	13.56	10.36 (1)	6.71 (3)	*	*	5.36 (3)
6	13.41		12.47 (1)	1.21 (4)	7.74 (5)	8.23 (3)
7a	7.62		6.46 (1)	6.25 (6)	6.22 (9)	4.27 (3)
7b	13.72		**	**	**	**

*Not measured.

**Dry.

Note: The water level is measured from the surface.
The number in parentheses is the number of measurements made each year.

The recharge rate of Wells No. 1 and No. 4 is so rapid that it is difficult to measure. It appears that both are poorly sealed or that the surrounding soil has a high rate of permeability and surface water is entering them. The recharge rate of the other wells is about 24 hours and the water appears to be characteristic of the perched layer. The fact that no water has been found in Well No. 7b is confirmation that the impermeable clay layer is indeed at about 14 m (45 ft). Well No. 7a, which is above the clay layer 7.6 m (25 ft), recharges in about 24 hours and appears representative of the ground water.

In 1982,[4] extensive results were reported for Wells No. 1, No. 3, No. 4, No. 6, and No. 7a. Results of the same scope for Wells No. 2 and No. 5 are reported here. The main focus of the 1983 studies was to determine if the results obtained earlier were persistent and reproducible, particularly those that appeared abnormal. Well No. 1 (Table 37) is characterized by high levels of chloride and results obtained in 1983 were essentially the same as in 1982. The proximity of this well to Westgate Road and its high recharge rate indicates that the chloride is coming from surface runoff and is due to road salting. A positive mercury result was obtained in 1982, but not in 1983. The pH, 7.1, is essentially the same as last year. Well No. 2 (Table 38) has had a high pH (11) since sampling began and is probably caused by the disposal of alkaline water softening compounds. This high pH water seems to be confined to this area because the landfilled material prevents water flow in a southerly (normal) direction.[19] Well No. 3 (Table 39) is characterized by the presence of arsenic. This element has been found in all samples tested in 1982 and 1983. The 1983 value is about 50% of the 1982 value.

Well No. 4 (Table 40) had elevated levels of manganese in 1982, and the results in 1983 were high but about half of the 1982 results. Chloride levels were also elevated. This well has a high recharge rate and is located close to a ditch which drains a marshy area adjacent to Westgate Road. Well No. 5 (Table 41) has a pH of about 7.6 and very low iron and manganese levels. Well No. 6 (Table 42) had a high level of manganese in 1982 and a higher level in 1983. The cause is uncertain, except that a substantial amount of manganous sulfate was added to the

TABLE 37
CHEMICAL CONSTITUENTS AT ANL LANDFILL
WELL 1
(CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES		CONCENTRATION	
		AVG.	MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	155 ± 133	124	186
CADMIUM	1	-	-	1.30
CHLORIDE*	2	780 ± 172	740	820
DISSOLVED SOLIDS*	2	1780 ± 140	1750	1810
LEAD	1	-	-	3
MANGANESE	2	160 ± 114	134	187
MERCURY	2	-	-	< 0.05
PH	2	-	7.1	7.1
SELENIUM	2	-	-	< 5
SILVER	1	-	-	1.30

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* CONCENTRATION IN MILLIGRAM/LITER.

TABLE 38
CHEMICAL CONSTITUENTS AT ANL LANDFILL
WELL 2
(CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	177 ± 4	176	178
CADMIUM	2	-	-	< 0.20
CHLORIDE*	2	51 ± 24	45	56
CHROMIUM(III)	2	-	-	< 3
COPPER	2	1	1	1
CYANIDE	2	-	-	< 10
DISSOLVED* SOLIDS	2	270 ± 60	256	284
FLUORIDE	2	163 ± 65	148	178
IRON	2	-	-	< 100
LEAD	2	1	1	1
MANGANESE	2	2 ± 4	1	3
MERCURY	2	-	-	< 0.05
NICKEL	2	3	3	3
PH	2	-	11	11
SELENIUM	2	-	-	< 5
SILVER	2	-	-	< 0.20
ZINC	2	10	10	10

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* CONCENTRATION IN MILLIGRAM/LITER.

TABLE 39
CHEMICAL CONSTITUENTS AT ANL LANDFILL
WELL 3
(CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	MIN.	MAX.
ARSENIC	2	18 ± 13	15	20
BARIUM	2	269 ± 136	237	300
CADMIUM	1	-	-	0.80
CHLORIDE[*]	2	20 ± 4	19	21
DISSOLVED SOLIDS[*]	2	762 ± 2	761	762
LEAD	1	-	-	1
MANGANESE	2	303 ± 54	291	316
MERCURY	1	-	-	< 0.05
PH	2	-	6.8	6.9
SELENIUM	2	-	-	< 5
SILVER	1	-	-	< 0.20

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[*] CONCENTRATION IN MILLIGRAM/LITER.

TABLE 40
CHEMICAL CONSTITUENTS AT ANL LANDFILL
WELL 4
(CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

	NO. OF		CONCENTRATION	
CONSTITUENT	SAMPLES	AVG.	MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	109 ± 110	84	135
BENZENE	1	-	-	< 30
CADMIUM	1	-	-	0.60
CHLORIDE*	2	130 ± 13	127	133
DISSOLVED SOLIDS*	2	982 ± 194	937	1030
LEAD	1	-	-	1
MANGANESE	2	982 ± 695	821	1140
MERCURY	2	-	-	< 0.05
PH	2	-	6.9	6.9
SELENIUM	2	-	-	< 5
SILVER	1	-	-	< 0.20
TOLUENE	1	-	-	< 20
XYLENE	1	-	-	< 20

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* CONCENTRATION IN MILLIGRAM/LITER.

TABLE 41
CHEMICAL CONSTITUENTS AT ANL LANDFILL
WELL 5
(CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	56 ± 86	36	76
CADMIUM	2	< 0.20	< 0.20	0.30
CHLORIDE*	2	21 ± 11	18	23
CHROMIUM(III)	2	-	-	< 3
CHROMIUM(VI)	2	-		< 3
COPPER	2	2 ± 6	1	4
CYANIDE	2	-	-	< 10
DISSOLVED SOLIDS*	2	354 ± 108	329	379
FLUORIDE	2	162 ± 26	156	168
IRON	2	-	-	< 100
LEAD	2	1	1	1
MANGANESE	2	29 ± 22	24	34
MERCURY	2	-	-	< 0.05
NICKEL	2	5 ± 8	3	6
PH	2	-	7.5	7.6
SELENIUM	2	-	-	< 5
SILVER	2	-	-	< 0.20
ZINC	2	10	10	10

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* CONCENTRATION IN MILLIGRAM/LITER.

TABLE 42
CHEMICAL CONSTITUENTS AT ANL LANDFILL
WELL 6
(CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

	NO. OF		CONCENTRATION	
CONSTITUENT	SAMPLES	AVG.	MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	109 ± 114	83	136
CADMIUM	1	-	-	0.30
CHLORIDE*	2	114 ± 49	102	125
DISSOLVED SOLIDS*	2	1180 ± 220	1130	1230
LEAD	1	-	-	1
MANGANESE	2	5430 ± 2380	4880	5990
MERCURY	1	-	-	< 0.05
PH	2	-	6.5	6.6
SELENIUM	2	-	-	< 5
SILVER	1	-	-	0.10

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* CONCENTRATION IN MILLIGRAM/LITER.

"French drain" in 1974. The dissolved solids level is about the same as last year, which could indicate that manganese is moving into this well on a preferential basis. Mercury at trace levels was found in 1982 in Wells No. 6 and No. 7a (Table 43) but the 1983 data did not show positive results.

Wells No. 4 and No. 7a , which had the only positive organic compound results in 1982, were reexamined and benzene, toluene, and xylene were not detected as shown in the tables. Studies will be continued using techniques that are capable of detecting a broad spectrum of organic materials at the nanogram/Liter level.

In summary, study of all test wells has indicated that the perched water is unaffected by the landfill materials, with the possible exceptions of basicity in Well No. 2, arsenic in Well No. 3, and manganese in Wells No. 4 and No. 6. Organic infiltration has not been established, based on past studies, but the focus in 1984 will be on selected materials from this group.

TABLE 43
CHEMICAL CONSTITUENTS AT ANL LANDFILL
WELL 7A
(CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

	NO. OF		CONCENTRATION	
CONSTITUENT	SAMPLES	AVG.	MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	102 ± 60	88	116
BENZENE	1	-	-	< 30
CADMIUM	1	-	-	0.70
CHLORIDE*	2	22 ± 9	20	24
DISSOLVED SOLIDS*	2	443 ± 41	433	452
LEAD	1	-	-	1
MANGANESE	2	289 ± 189	245	333
MERCURY	2	-	-	< 0.05
PH	2	-	7.3	7.3
SELENIUM	2	-	-	< 5
SILVER	1	-	-	< 0.20
TOLUENE	1	-	-	< 20
XYLENE	1	-	-	< 20

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* CONCENTRATION IN MILLIGRAM/LITER.

IV. UNUSUAL OCCURRENCE

A. 1982 Loss of Pond Water

In the 1982 monitoring report,[4] an unusual occurrence was reported which consisted of the loss of about 7.5 megaliters (2 million gallons) of ponded water containing about 26 mCi of tritiated water. Monitoring continued during 1983 for tritium at four locations to determine any pattern or trends in the subsurface movement of the tritium. In addition, samples of the residual water remaining in the excavation were periodically sampled and analyzed for tritium. Monitoring of the water in the pond began in 1973 and annual average results through 1983 are given in Table 44.

TABLE 44

Average Tritiated Water Content of Water From the Excavation (Concentrations in 10^{-6} µCi/mL)

Year	No. of Samples	Results
1973	2	8.1
1974	3	6.8
1975	3	9.1
1976	2	11.1
1978	1	11.4
1979	1	8.7
1980	1	8.4
1981	4	4.3
1982	11	2.9
1983	9	1.2

The decrease is due to the elimination of the source of the tritium (the CP-5 reactor) and dilution by rainwater.

Sampling of the three wells and one seep south of the excavation continued in 1983 and the results are listed in Table 45. Samples from wells in all other directions of the pond gave no evidence of migration in these directions. As discussed in last year's report, the water from the excavation appears to have moved in a southwest direction and was detected in Well No. 9. The concentrations have decreased gradually with time, until by summer of 1983, tritium levels were below the detection limit and stayed low for the remainder of the year. It would seem that the water from the excavation is slowly moving horizontally in the dolomite as a pulse. Quarterly subsurface sampling will continue to determine if the tritium pulse appears at any other location.

TABLE 45

Tritiated Water Content of Wells Near A^2R^2 Excavation, 1982-1983 (Concentrations in 10^{-6} µCi/mL)

Date Collected	Well No. 9	Well D181	Well D185	4EF Seep
March 11, 1982	1.2 ± 0.2	< 0.1	< 0.1	< 0.1
March 19, 1982	1.7 ± 0.1	-	-	-
March 26, 1982	1.4 ± 0.1	-	-	-
April 9, 1982	1.0 ± 0.1	-	-	-
April 16, 1982	1.0 ± 0.1	-	-	-
April 26, 1982	0.6 ± 0.1	-	-	-
May 14, 1982	0.7 ± 0.1	-	-	-
May 25, 1982	0.7 ± 0.1	< 0.1	< 0.1	0.69 ± 0.11
June 25, 1982	0.7 ± 0.1	< 0.1	< 0.1	0.10 ± 0.10
July 9, 1982	0.5 ± 0.1	-	-	-
July 26, 1982	0.8 ± 0.1	< 0.1	< 0.1	< 0.1
August 30, 1982	0.5 ± 0.1	< 0.1	< 0.1	< 0.1
September 17, 1982	0.6 ± 0.1	< 0.1	< 0.1	< 0.1
October 25, 1982	0.6 ± 0.1	< 0.1	< 0.1	< 0.1
November 29, 1982	0.7 ± 0.1	< 0.1	< 0.1	0.13 ± 0.10
January 4, 1983	-	< 0.1	< 0.1	-
January 24, 1983	0.7 ± 0.1	< 0.1	< 0.1	< 0.1
February 23, 1983	0.7 ± 0.1	< 0.1	0.2 ± 0.1	< 0.1
March 31, 1983	0.6 ± 0.1	< 0.1	0.1 ± 0.1	0.2 ± 0.1
April 26, 1983	0.6 ± 0.1	< 0.1	< 0.1	< 0.1
May 24, 1983	0.5 ± 0.1	< 0.1	-	< 0.1
July 25, 1983	< 0.1	< 0.1	< 0.1	0.1 ± 0.1
September 26, 1983	< 0.1	< 0.1	< 0.1	< 0.1
December 2, 1983	< 0.1	< 0.1	< 0.1	< 0.1

V. APPENDIX

A. References

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B. Environmental Quality Standards, Detection Limits, Analytical Errors, and Quality Assurance

1. Standards

All of the standards and detection limits for chemical constituents, and some of those for radionuclides and external radiation, are given in the main body of the report. In addition, in Table 46 are collected air and water environmental quality standards and detection limits (minimum detectable amounts) for all radionuclides, and for those materials, for which measurements were made. These standards are the Concentration Guides given in DOE Order 5480.1, Chapter XI,[5] and are used in this report to assess the hazard of a measured concentration of a radioactive nuclide. The Order distinguishes two CGs, one for occupational exposure in controlled areas and one for uncontrolled areas, beyond the site boundary where individuals can be exposed nonoccupationally, for 168 hours per week. The CGs in the table are for uncontrolled areas. For water, the standard selected was for the soluble form of the radionuclide; for air, the standard for the insoluble form was selected.

2. Detection Limits

The detection limits were chosen so that the measurement uncertainty at the 95% confidence level is equal to the measured value. The relative

TABLE 46

Concentration Guides and Detection Limits
(μCi/mL)

Nuclide or Activity	Concentration Guide		Detection Limit	
	Water	Air	Water	Air
Americium-241	4×10^{-6}	4×10^{-10}	1×10^{-12}	-
Antimony-125	-	9×10^{-10}	-	1×10^{-16}
Argon-41	-	4×120^{-8}	-	1.5×10^{-8}
Barium-140	3×10^{-5}	-	2×10^{-9}	-
Beryllium-7	-	4×10^{-8}	-	5×10^{-15}
Californium-249	4×10^{-6}	-	1×10^{-12}	-
Californium-252	7×10^{-6}	-	1×10^{-12}	-
Cesium-137	2×10^{-5}	$5 \times 1-^{-10}$	-	1×10^{-16}
Curium-242	2×10^{-5}	-	1×10^{-12}	-
Curium-244	7×10^{-6}	-	1×10^{-12}	-
Hydrogen-3	3×10^{-3}	-	1×10^{-7}	-
Krypton-85	-	3×10^{-7}	-	-
Neptunium-237	3×10^{-6}	-	1×10^{-12}	-
Plutonium-238	5×10^{-6}	1×10^{-12}	1×10^{-12}	3×10^{-19}
Plutonium-239	5×10^{-6}	1×10^{-12}	5×10^{-13}	3×10^{-19}
Radium-226	3×10^{-8}	-	1×10^{-13}	-
Strontium-89	3×10^{-6}	1×10^{-9}	2×10^{-9}	1×10^{-16}
Strontium-90	3×10^{-7}	2×10^{-10}	2.5×10^{-10}	1×10^{-17}
Thorium-228	-	2×10^{-13}	-	1×10^{-18}
Thorium-230	-	3×10^{-13}	-	1×10^{-18}
Thorium-232	-	1×10^{-12}	-	1×10^{-18}
Uranium-234	-	4×10^{-12}	-	1×10^{-19}
Uranium-235	-	4×10^{-12}	-	1×10^{-19}
Uranium-238	-	5×10^{-12}	-	1×10^{-19}
Uranium - natural*	4×10^{-5}	4×10^{-12}	2×10^{-10}	2×10^{-17}
Alpha**	3×10^{-6} to	1×10^{-10} to	2×10^{-10}	2×10^{-16}
Beta**	1×10^{-7}	1×10^{-13}	1×10^{-9}	5×10^{-16}

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*Concentration Guides converted from the "special curie" used in DOE Order 5480.1 Chapter XI to the standard curie.

**The Concentration Guides for unknown mixtures depend, within the range given, on whether certain radionuclides are known to be present in concentrations less than 0.1 of their CGs, and the sum of the fraction of the CGs for all such nuclides is less than 0.25. For most total alpha and beta results given in this report, the largest uncontrolled CG is applicable.

error in a result decreases with increasing concentration. At a concentration equal to twice the detection limit, the error is about 50% of the measured value, and at 10 times the detection limit, the error is about 10%.

3. Quality Assurance Program

a. Radiochemical Analysis and Radioactivity Measurements

All nuclear instrumentation is calibrated with standard sources obtained from the U. S. National Bureau of Standards (NBS), if possible. If NBS standards were not available for particular nuclides, standards from the Amersham Corporation were used. The equipment is usually checked on a daily basis with secondary counting standards to insure proper operation. Samples were periodically analyzed in duplicate or with the addition of known amounts of a radionuclide to check precision and accuracy. In addition, standard and intercomparison samples distributed by the Quality Assurance Branch of the U. S. Environmental Protection Agency (EPA-QA) are analyzed regularly. Results of our participation in the EPA-QA program during 1982 are given in Table 47. In the table, the comparison is made between the EPA-QA value, which is the quantity added to the sample by that Laboratory, and the value obtained in our laboratory. To assist in judging the quality of the results, typical errors for our analyses are 2-50% and the error in the EPA-QA results is 2-5% (our estimate).

In addition, participation in the re-established DOE Environmental Measurements Laboratory Quality Assurance Program (DOE-EML-QAP), a semi-annual distribution of four or five different sample matrices containing various combinations of radionuclides, was resumed. Results for 1983 are collected in Table 48. In the table, the comparison is made between the EML value, which is the result of replicate determinations by that Laboratory, and the value obtained in our laboratory. More than 95% of all the intercomparison samples received were analyzed for the radionuclides for which results were requested. To assist in judging the quality of the results, typical errors for our analyses are 2-50% and the error in the EML results is 1-30% (depending on the nuclide and the amount present). For most analyses for which the differences are large (> 20%), the

TABLE 47

Summary of EPA-QA Samples, 1983

Type of Sample	Analysis	Number Analyzed	Average Difference from Added
Air Filter	Total Alpha	4	48%
	Total Beta	4	12%
	Strontium-90	3	7%
	Cesium-137	4	28%
Milk	Potassium-40	3	6%
	Strontium-89	3	11%
	Strontium-90	3	12%
	Iodine-131	3	3%
	Cesium-137	3	5%
Water	Total Alpha	2	52%
	Total Beta	2	52%
	Hydrogen-3	2	3%
	Chromium-51	3	9%
	Cobalt-60	5	5%
	Zinc-65	3	5%
	Strontium-89	4	5%
	Strontium-90	4	10%
	Ruthenium-106	3	4%
	Iodine-131	3	11%
	Cesium-134	5	3%
	Cesium-137	5	5%
	Radium-226	3	35%
	Radium-228	2	32%
	Total Uranium	3	12%
	Plutonium-239	2	5%

TABLE 48

Summary of DOE-EML-QAP Samples, 1933

	Average Difference From EML Value				
	Air Filters	Water	Soil	Tissue	Vegetation
Hydrogen-3	-	3% (2)	-	-	-
Beryllium-7	10% (2)	-	-	-	-
Potassium-40	-	-	14% (2)	-	1% (1)
Chromium-51	21% (1)	4% (1)	-	-	-
Manganese-54	-	2% (2)	-	-	-
Cobalt-57	16% (1)	-	-	-	-
Cobalt-60	8% (2)	3% (1)	-	22% (1)	-
Strontium-90	17% (2)	8% (2)	22% (2)	17% (1)	9% (1)
Cesium-137	10% (2)	2% (2)	3% (2)	24% (2)	28% (2)
Cerium-144	-	1% (1)	-	-	-
Radium-226	-	-	6% (2)	-	-
Uranium-234	8% (2)	6% (2)	-	-	-
Uranium-238	9% (2)	11% (2)	-	-	-
Plutonium-238	-	-	32% (1)	11% (2)	9% (2)
Plutonium-239	11% (2)	27% (2)	35% (1)	3% (2)	14% (2)
Americium-241	15% (2)	13% (2)	24% (1)	21% (1)	4% (1)

Note: The figure in parentheses is the number of samples.

concentrations were quite low and the differences were within the measurement uncertainties.

b. Penetrating Radiation

Our laboratory participated in the Sixth International Environmental Dosimeter Intercomparison Project conducted by the DOE Radiological and Environmental Sciences Laboratory in Idaho Falls, Idaho, and the DOE Environmental Measurements Laboratory (EML) in New York, NY. Participants supplied four sets of each type of dosimeter they wished to evaluate. One set was exposed in the field to a mixture of natural and man-made gamma radiation at the Idaho National Engineering Laboratory; one set each was exposed in the laboratory to a known (but unrevealed) quantity of radiation at the beginning of the field exposure (then placed in the field); and one set at the end of the field exposure period. The fourth set served as a control. The laboratory exposures were to a cesium-137 source, and were done for a fading experiment. After exposure, the dosimeters were returned to the participant for measurement. The field exposure was measured by an EML high-pressure ion chamber for comparison. The results, in milliroentgens (mR), were:

		Laboratory Exposure	
	Field Exposure	Begin	End
Delivered Dose	43.5 ± 2.2	202 ± 10	158 ± 8
ANL Value			
calcium fluoride	40.0 ± 6.0	209 ± 23	172 ± 23
lithium fluoride	40.0 ± 9.0	204 ± 43	140 ± 18

The uncertainties listed are the standard deviations as estimated by the sponsors for their values and by us for our results. All results agree with the delivered dose within the measurement error at the 95% confidence level. The most important comparison is for the field exposures. Here the differences between the ion chamber result and our results were 3.5 mR ± 6.4 mR (8.1% ± 14.7%) for calcium fluoride and 3.5 mR ± 9.3 mR (8.1% ± 21.4%) for lithium fluoride. The standard deviations for our results are relatively large because the transit and storage dose, as measured with the control dosimeters, is significant and must be subtracted from the total dose.

c. Chemical Analysis

With each set of atomic absorption analyses, standard amounts of trace metals were analyzed in blank solutions at concentrations corresponding to 50% and 100% of the current State standards. Recoveries were determined by comparing these results to results obtained by analyzing stream and effluent samples to which identical concentrations were added. Average recoveries ranged from about 85-100%, with a standard deviation of about 10%. Recovery studies were also performed for ammonia nitrogen, fluoride, and hexavalent chromium analyses, and similar results were obtained.

Intercomparison air-filter samples furnished by the National Institute of Occupational Safety and Health (NIOSH) were analyzed for cadmium, lead, and zinc. The results (Table 49) were in good agreement with the reference values.

TABLE 49

Summary of Quality Assurance Studies

Constituent	Average Difference From Reference Value
Cadmium	4% (16)
Lead	4% (16)
Zinc	5% (16)
High Volume Air Flow	0.4% (2)

Note: The figures in parentheses are the number of samples analyzed.

The air flows of the high volume air samplers used for TSP measurements were compared to air flow standards furnished by the USEPA. Results at both locations were within 0.4% of the reference values.

d. Sampling, Sample Storage, Other

Many factors enter into an overall quality assurance program other than the analytical quality control discussed above. Representative sampling is of prime importance. The continuous water sampler in Sawmill Creek provides a representative sample for a critical sampling location since the concentration of pollutants in the waste water may vary appreciably during each 24-hour period.

The accuracy of the flowmeters in the air sampling equipment is verified periodically with a calibrated rotameter, as well as by comparison to a Roots meter primary standard.

Samples are pre-treated in a manner designed to maintain the integrity of the constituent sought. For example, samples for trace radionuclide analysis are acidified immediately after collection to prevent hydrolytic loss of metal ions.

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