

**ENVIRONMENTAL MONITORING AT  
ARGONNE NATIONAL LABORATORY  
ANNUAL REPORT FOR 1982**

by

**N. W. Golchert, T. L. Duffy,  
and J. Sedlet**



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**ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS**

**Prepared for the U. S. DEPARTMENT OF ENERGY**

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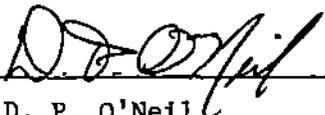
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ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1982 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 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. The potential radiation dose to off-site population groups is also estimated.

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.<sup>1</sup> 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 1982 were safety studies for light water and breeder reactors, development of components and materials for fission and fusion reactors and for improved coal combustion, 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 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 fully-enriched uranium; one critical assembly or zero power reactor (ZPR-9), that is fueled at various times with plutonium, uranium, or a combination of the two; the Argonne Thermal Source Reactor (ATSR), a 10 kW research reactor fueled with enriched uranium; a prototype superconducting

heavy ion linear accelerator; a 60-inch cyclotron; several other charged particle accelerators (principally of the Van de Graaff and Dynamitron type); a large fast neutron source (IPNS, Intense Pulsed Neutron Source) 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 (ZGS, Zero Gradient Synchrotron) and a 5 MW heavy-water enriched-uranium reactor (CP-5) were not in operation during 1982. The CP-5 facility is awaiting decontamination and decommissioning, while this work is essentially complete 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 nonnuclear facilities at Argonne that may produce a measurable impact on the environment are the coal-fired boiler #5 and the ocean thermal energy conversion (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 District in 1973 for their use as a public recreational area, nature preserve, and demonstration forest.

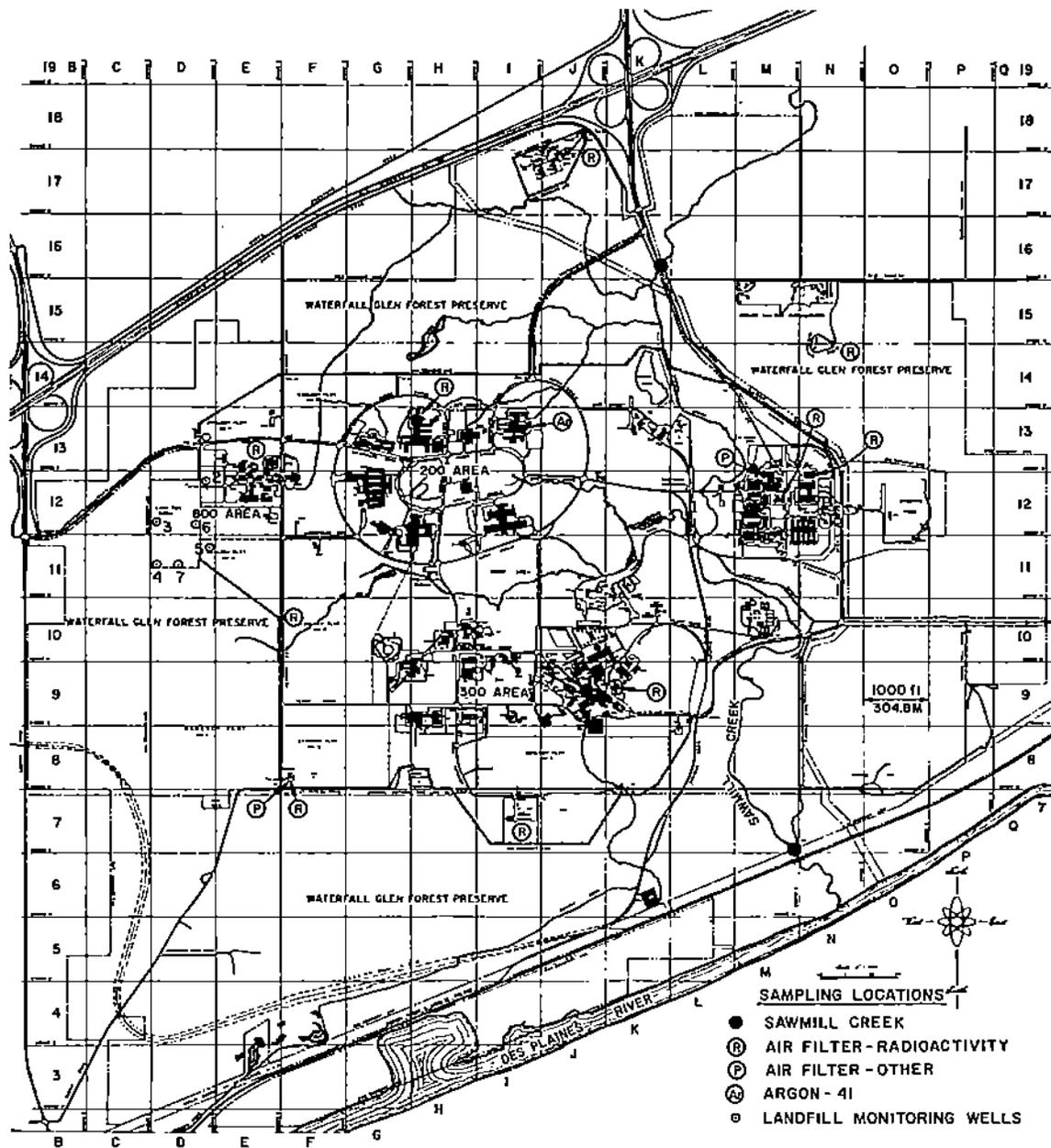


Fig. 1. Sampling Locations at Argonne National Laboratory

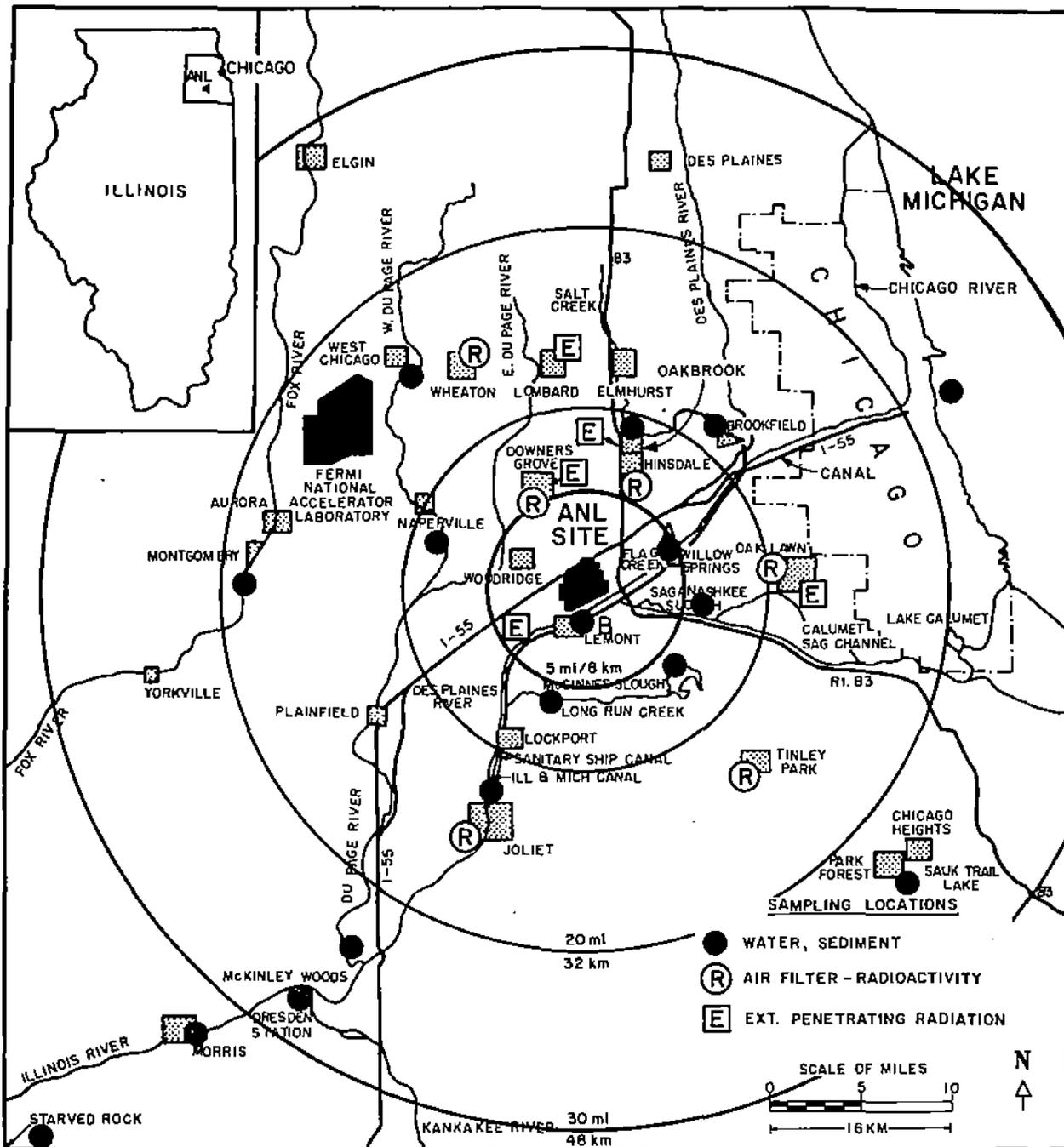


Fig. 2. Sampling Locations Near Argonne National Laboratory

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 deciduous trees of an average height of 15-18 m (50-60 ft). The remaining portion of the site changes in elevation by no more than 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.<sup>2</sup>

### 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<sup>3</sup> and provides an adequate sample of the climate. The data collected since 1964 have not been published.

The most important meteorological parameters for the purposes of this report are wind direction, wind speed, temperature, and precipitation. The 1982 average monthly and annual wind roses on the Argonne site 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 radius represents the average wind speed for the direction covering 11.25° on either side of the radius. For example, in the plot labeled January, the west component is dominant. The length and direction of this vector shows that, of the total wind observations, about 9% were in the 2.01-6 m/s range, about 10% were in the 6.01-10 m/s range, about 1% were greater than 10.01 m/s; and about 20% of the observations were from the direction between 258.75° and 281.25°.

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

TABLE 1

## Incremental Population Data in the Vicinity of ANL, 1981

Distance, miles Distance, km	In Thousands									
	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		36347	86265	151270	538.0	3679.4	6638.6	7396.0	7942.8

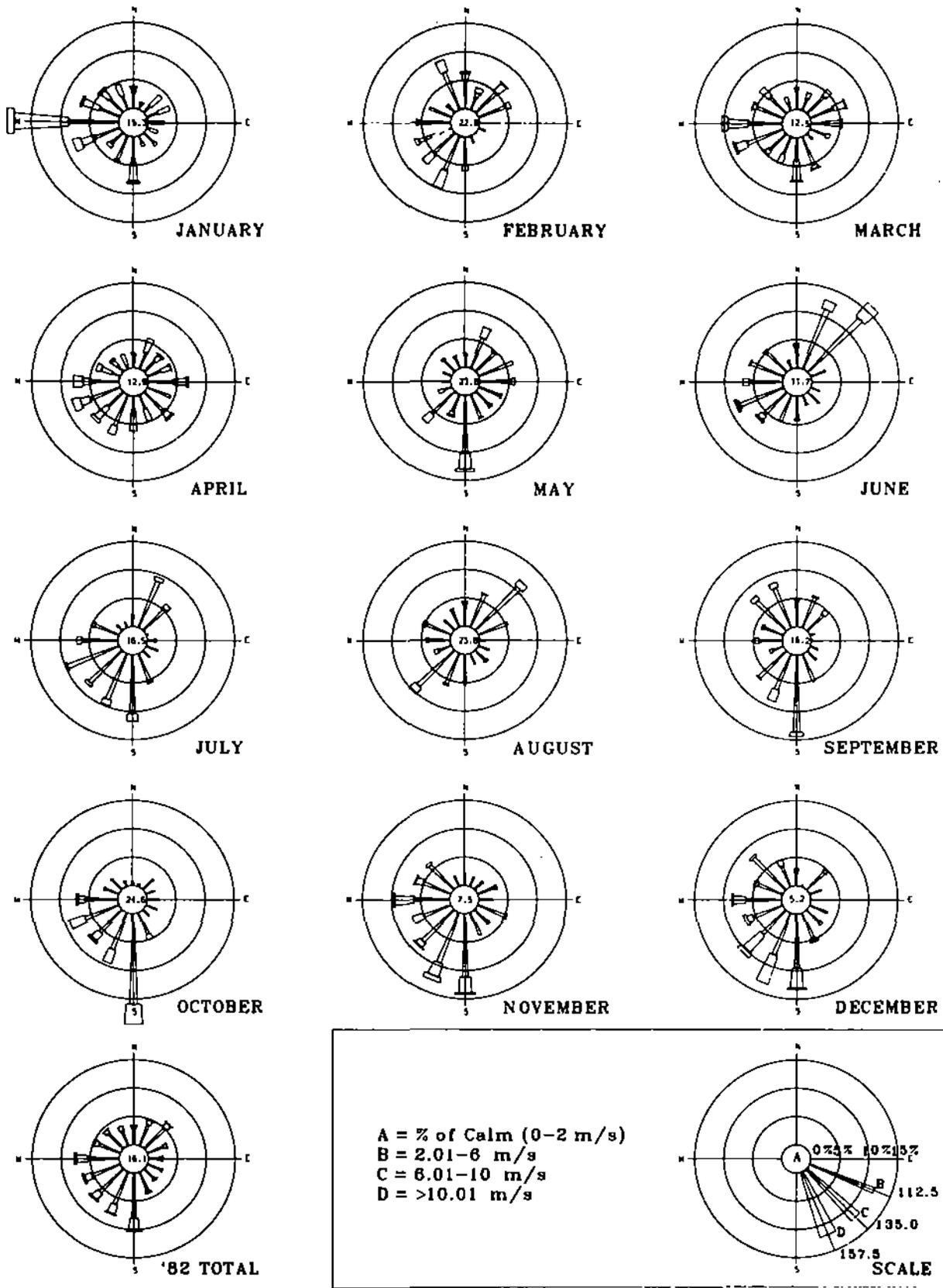


Fig. 3. Monthly and Annual Wind Roses at Argonne National Laboratory, 1982.

wind direction in May is south, while in June it is northeast. Compared to the past several years, the annual average wind rose for 1982 displayed a more southerly dominance. The long term average wind direction is usually from the west to southwest.

The precipitation and temperature data for 1982 are shown in Table 2. Temperatures averaged about 1.5°C lower in 1982 compared to the long term average. Significantly above normal amounts of precipitation in July, November, and December resulted in a total precipitation in 1982 of 109.3 cm (43.0 in), about 25% higher than the long term annual average of 85.2 cm (35.5 in).

#### E. Geohydrology

The geology of the Laboratory area consists of about 30 m (100 ft) of glacial till on top of 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 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 shales 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 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

TABLE 2

## ANL Weather Summary, 1982

Month	Precipitation (cm)		Temperature (°C)	
	Amount	Average <sup>*</sup>	Monthly Average	Average <sup>**</sup>
January	7.37	4.88	-11.0	-4.2
February	1.04	4.57	-5.8	-2.7
March	10.54	6.93	1.7	2.5
April	7.06	7.98	6.9	8.8
May	5.28	8.81	17.9	14.6
June	3.96	9.47	16.7	20.2
July	21.16	8.66	23.4	23.2
August	9.98	8.10	20.4	22.5
September	2.92	8.08	16.7	18.7
October	4.78	6.55	11.8	12.5
November	17.65	5.88	3.9	4.7
December	17.58	5.30	2.2	-1.5

<sup>\*</sup> Average precipitation, 1873-1977, U. S. Weather Bureau, Chicago, Illinois.

<sup>\*\*</sup> Average temperature 1871-1977, U. S. Weather Bureau, Chicago, Illinois.

appears to be adequate for future Laboratory use, but this ground water source is widely used throughout DuPage County. 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, which 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 2.8 megaliters (0.75 million gallons) per day, while the water flow in the Creek upstream of the waste-water outfall averaged about 36 megaliters (10 million gallons) per day during 1982. 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 for some 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. Very approximate estimates of usage are 600 individuals per day on weekends and 500 individuals for group educational purposes. The average stay is about two hours. Sawmill Creek flows through the south portion of the Preserve on its way to the Des Plaines River. This region of the Preserve (formerly named Rocky Glen) was used principally for picnicking, hiking, and overnight camping by youth groups, but has been closed for rehabilitation since June 6, 1976. East and southeast of Argonne and the Des Plaines River are located several large forest preserves of the Cook County Forest Preserve District. 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 corn and soybeans, the data are for 1981; for milk, wheat, and oats, the data are for 1982.

TABLE 3

## Agricultural Production Near ANL, 1981, 1982

County	Milk		Corn	Soybeans	Wheat	Oats
	No. of Cows	Million Pounds				
DuPage	200	2.2	2.1	0.60	0.08	0.24
Cook	200	2.2	2.1	0.74	0.041	0.045
Will	3,000	33.1	18.8	5.2	0.29	0.24
Kane	5,800	63.9	15.7	2.8	0.197	0.21
Kendall	600	6.6	12.0	2.6	0.077	0.079
Grundy	700	7.7	15.4	3.4	0.034	0.053
Lake	1,400	15.4	2.5	1.0	0.344	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-82-12.<sup>4</sup> Included in this report are the results of measurements obtained in 1982 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. Some intermediate half-life fission products were detected at all sampling locations and are the result of fallout from the Chinese atmospheric nuclear test of October 16, 1980. No activity attributable to Argonne operations could be detected.

The plutonium-239,240 concentrations in air averaged  $4 \times 10^{-18}$   $\mu\text{Ci/ml}$ ,\* about five times less than the 1981 value, and were nearly identical at all sampling locations. The monthly variations showed a "spring maximum" in stratospheric fallout of plutonium similar to that observed for beryllium-7 (a naturally-occurring nuclide produced in the stratosphere by cosmic-ray interactions) and several fission products. The results indicated that the airborne plutonium was from atmospheric nuclear test detonations and there was no evidence that any of the plutonium originated at Argonne. The average plutonium concentration was equivalent

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\*The radioactivity units are described in Section III.

to 0.0004% of the Concentration Guide (CG).\*

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 0.02 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 0.01 mrem/yr. These releases constitute an insignificant addition to the dose received from the natural background radiation, which is about 96 mrem/yr. The total 80-km population dose from these radionuclides was less than 0.3 man-rem for 1982.

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 CG of their net average Creek concentrations, were hydrogen-3, 0.003%; strontium-90, < 0.02%; neptunium-237, 0.00016%; plutonium-239,240, 0.00010%; americium-241, 0.00058%; 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.

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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. 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.

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  $0.9 \times 10^{-3} \mu\text{Ci}/\text{m}^2$  at the site perimeter and  $0.8 \times 10^{-3} \mu\text{Ci}/\text{m}^2$  off the site. The corresponding plutonium-238 averages were  $0.04 \times 10^{-3} \mu\text{Ci}/\text{m}^2$  and  $0.03 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ , 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  $2 \times 10^{-9} \mu\text{Ci}/\text{g}$  to  $10 \times 10^{-9} \mu\text{Ci}/\text{g}$  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 Laboratory waste water outfall.

Milk from a dairy farm near the Laboratory was analyzed for several fission products, including hydrogen-3. Hydrogen-3 concentrations averaged  $< 117 \times 10^{-9} \mu\text{Ci}/\text{ml}$ . The strontium-90 concentration ( $3.5 \times 10^{-9} \mu\text{Ci}/\text{ml}$ ), and the cesium-137 concentration ( $1.1 \times 10^{-9} \mu\text{Ci}/\text{ml}$ ) decreased by 50% compared to 1981. These radio-nuclides 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 96 mrem/yr with a standard deviation of 10 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 410 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  $111 \pm 19$  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 25 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 about 0.05 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 also about 0.05 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 total suspended particulates (TSP) were determined upwind and downwind of the coal burning steam plant. Average values of  $46 \mu\text{g}/\text{m}^3$  and  $48 \mu\text{g}/\text{m}^3$  were obtained. These are essentially the same as obtained in previous years and were below the primary Federal standard of  $60 \mu\text{g}/\text{m}^3$ .

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 007 (9K) and 009 (14K), and suspended solids at 005 (14F). The average mercury level of  $1 \mu\text{g}/\text{l}$  (twice the permit level) resulted from releases in Argonne waste water which occurred even though reasonable care is taken to prevent them. The pH values that exceeded the limit ( $\text{pH} > 9.0$ ) 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, except ammonia nitrogen and dissolved solids, were within the State of Illinois Water Quality Standards. The average ammonia nitrogen

level above the Argonne waste-water outfall was 2 times the State standard, and exceeded this value in 46% of the samples, while levels below the outfall were 1.7 times the State standard and exceeded this value in 46% of the samples. The average concentration of dissolved solids above the waste-water outfall exceeded the State standard by 26%, while below the outfall it was 15% above the standard. The elevated levels of ammonia nitrogen and total dissolved solids were due to effluents from the Marion Brook Sewage Treatment Plant and other upstream sources. The average levels of copper and iron were 136 and 160% of the State standards, and individual values exceeded the standards 40 and 45% of the time. Mercury concentrations exceeded the State standard 4% of the time and averaged 33% 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. Test 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.

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.

## 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 ( $\mu\text{Ci/ml}$ ) for water, air, and milk and microcuries per gram (g) and/or square meter ( $\text{m}^2$ ) for soil, bottom sediment, and vegetation. When a nuclide was not detected, the result is given as less than ( $\langle$ ) 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 ( $\langle$ ) 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.<sup>5</sup> 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,<sup>6</sup> 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 by DOE and all regulatory agents 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 asbestos-cellulose filter paper 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 listed in Table 5.

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,  $2.5 \times 10^{-14}$   $\mu\text{Ci/ml}$ , was about five times less than the 1981 average and similar to the 1980 average. The decrease is due to the decay of intermediate half-life radionuclides injected into the stratosphere by the October 16, 1980, atmospheric nuclear test by the People's Republic of China. The nuclides measured in 1981 but which are now no longer detectable from this test are: manganese-54, yttrium-88, ruthenium-103, and cerium-141. The stratospherically produced beryllium-7 and the test-produced radionuclides in Table 5 exhibit the same spring increase in concentration, indicating their stratospheric origin.

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 a polystyrene filter medium. The total air volume filtered for the monthly samples was about  $25,000 \text{ m}^3$ . Samples were ignited at  $600^\circ\text{C}$  to remove organic matter and prepared for analysis by vigorous treatment with hot hydrochloric, hydrofluoric, and nitric acids. This treatment has been found in our laboratory to solubilize plutonium that has been ignited at  $1000^\circ\text{C}$ .

TABLE 4  
 TOTAL ALPHA AND BETA ACTIVITIES IN AIR-FILTER SAMPLES, 1982<sup>\*</sup>  
 (CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

MONTH	LOCATION	NO. OF SAMPLES	ALPHA ACTIVITY			BETA ACTIVITY		
			AV.	MIN.	MAX.	AV.	MIN.	MAX.
JANUARY	OFF-SITE PERIMETER	19	2.4	1.3	3.8	38	15	59
		30	2.8	1.7	4.4	42	30	57
FEBRUARY	OFF-SITE PERIMETER	16	1.8	0.9	2.6	35	19	52
		32	1.7	0.7	2.6	34	16	52
MARCH	OFF-SITE PERIMETER	21	1.7	0.3	3.3	22	5	34
		33	1.5	0.5	2.6	25	9	40
APRIL	OFF-SITE PERIMETER	21	2.2	0.9	4.5	27	16	39
		31	2.1	0.7	3.5	23	18	40
MAY	OFF-SITE PERIMETER	19	2.1	0.5	4.5	23	12	41
		35	2.0	0.5	4.3	25	11	40
JUNE	OFF-SITE PERIMETER	23	1.3	0.6	2.5	17	8	26
		33	1.4	0.7	2.3	16	8	24
JULY	OFF-SITE PERIMETER	20	1.6	0.8	3.0	21	10	29
		34	1.3	0.7	1.8	20	11	29
AUGUST	OFF-SITE PERIMETER	22	1.8	0.4	3.5	22	4	34
		37	1.7	0.8	3.4	23	18	36
SEPTEMBER	OFF-SITE PERIMETER	18	1.5	0.7	3.3	19	9	33
		32	1.2	0.5	2.3	18	6	23
OCTOBER	OFF-SITE PERIMETER	21	2.0	0.6	3.1	25	13	41
		38	1.7	0.9	3.8	24	11	46
NOVEMBER	OFF-SITE PERIMETER	19	1.9	1.1	4.0	24	11	34
		33	1.3	0.7	3.6	25	16	35
DECEMBER	OFF-SITE PERIMETER	19	1.3	0.5	2.2	24	13	35
		25	1.5	0.6	2.6	26	16	41
ANNUAL SUMMARY	OFF-SITE PERIMETER	238	1.8 ± 0.1	0.3	4.5	25 ± 1	4	59
		393	1.7 ± 0.1	0.5	4.4	26 ± 1	6	57
% CG	OFF-SITE	-	(0.002)	(0.0003)	(0.005)	0.025	0.004	0.059
% CG	PERIMETER	-	(0.002)	(0.0005)	(0.004)	0.026	0.006	0.057

\* 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.

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NOTE: (1E-15 = 10<sup>-15</sup>)

TABLE 5  
 GAMMA-RAY ACTIVITY IN AIR-FILTER SAMPLES, 1982  
 (CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

MONTH	LOCATION	BE7	ZR95	NB95	RU106-RH106	SB125	CS137	CE144
JANUARY	OFF-SITE PERIMETER	59	0.2	0.1	1.1	0.1	0.2	1.2
		67	0.2	0.2	0.6	0.1	0.2	1.0
FEBRUARY	OFF-SITE PERIMETER	79	0.1	0.2	1.0	< 0.1	0.2	1.3
		79	0.2	0.2	0.7	0.1	0.2	1.1
MARCH	OFF-SITE PERIMETER	65	< 0.1	0.1	0.5	0.1	0.3	1.1
		76	0.1	0.1	0.6	0.1	0.2	1.1
APRIL	OFF-SITE PERIMETER	124	0.1	0.1	0.8	0.1	0.5	1.4
		132	0.1	0.1	0.7	0.2	0.5	1.9
MAY	OFF-SITE PERIMETER	105	< 0.1	< 0.1	1.0	0.1	0.4	1.6
		121	< 0.1	0.1	0.8	0.1	0.4	1.4
JUNE	OFF-SITE PERIMETER	85	< 0.1	< 0.1	0.5	< 0.1	0.4	0.8
		81	< 0.1	< 0.1	0.5	< 0.1	0.3	0.7
JULY	OFF-SITE PERIMETER	79	< 0.1	< 0.1	0.5	< 0.1	0.3	0.5
		86	< 0.1	< 0.1	0.5	0.1	0.2	0.6
AUGUST	OFF-SITE PERIMETER	76	< 0.1	< 0.1	0.5	< 0.1	0.2	0.6
		82	< 0.1	< 0.1	0.5	< 0.1	0.2	0.6
SEPTEMBER	OFF-SITE PERIMETER	52	< 0.1	< 0.1	0.5	< 0.1	0.1	0.5
		53	< 0.1	< 0.1	0.5	< 0.1	0.1	0.5
OCTOBER	OFF-SITE PERIMETER	68	< 0.1	< 0.1	0.5	< 0.1	0.2	0.5
		66	< 0.1	< 0.1	0.5	< 0.1	0.1	0.5
NOVEMBER	OFF-SITE PERIMETER	62	< 0.1	< 0.1	0.5	< 0.1	0.1	0.5
		65	< 0.1	< 0.1	0.5	< 0.1	0.1	0.5
DECEMBER	OFF-SITE PERIMETER	47	< 0.1	< 0.1	0.5	< 0.1	0.1	0.5
		57	< 0.1	< 0.1	0.5	< 0.1	0.2	0.5
ANNUAL SUMMARY	OFF-SITE PERIMETER	75±13 80±14	< 0.1 < 0.1	< 0.1 < 0.1	0.7±0.2 0.6±0.1	< 0.10 < 0.11	0.3±0.1 0.2±0.1	0.9±0.3 0.9±0.3
% CG (X 1E-3)	OFF-SITE PERIMETER	(0.19) (0.20)	< .01 < .01	< .04 < .04	0.3 0.3	< .010 < .011	0.05 0.04	0.4 0.4

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, 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 five compared to 1981 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  $1 \times 10^{-16}$   $\mu\text{Ci/ml}$ . Figure 4 shows the monthly plutonium-239 air concentrations for the past ten 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

TABLE 6  
STRONTIUM, THORIUM, URANIUM AND PLUTONIUM CONCENTRATIONS IN AIR-FILTER SAMPLES, 1982  
(CONCENTRATIONS IN  $10^{-18}$  MICROCURIES/ML)

MONTH	LOCATION <sup>1</sup>	2								3	
		SR-90	TH-228	TH-230	TH-232	U-234	U-235	U-238	PU-238	PU-239	
JANUARY	7I	170 ± 30	20 ± 3	30 ± 3	24 ± 3	38 ± 3	1.3 ± 0.6	30 ± 3	< 0.3	4.5 ± 0.8	
	12H	140 ± 10	22 ± 3	42 ± 4	27 ± 3	56 ± 3	2.1 ± 1.0	63 ± 4	< 0.3	5.2 ± 0.7	
	OFF-SITE	160 ± 10	13 ± 2	10 ± 1	10 ± 1	19 ± 2	0.5 ± 0.3	21 ± 2	< 0.3	4.5 ± 0.7	
FEBRUARY	7I	160 ± 20	12 ± 1	13 ± 1	11 ± 2	11 ± 2	0.6 ± 0.3	13 ± 1	0.9 ± 0.8	6.1 ± 1.1	
	12H	160 ± 10	6 ± 1	12 ± 1	8 ± 1	27 ± 2	0.6 ± 0.3	25 ± 1	0.6 ± 0.6	7.0 ± 0.9	
	OFF-SITE	130 ± 10	7 ± 2	11 ± 1	7 ± 1	13 ± 1	1.3 ± 0.3	8 ± 1	0.6 ± 0.8	5.0 ± 1.0	
MARCH	7I	150 ± 10	28 ± 4	33 ± 3	30 ± 3	20 ± 2	0.9 ± 0.3	15 ± 1	< 0.3	6.8 ± 0.8	
	12H	140 ± 10	13 ± 3	22 ± 2	15 ± 2	49 ± 2	2.1 ± 0.4	29 ± 2	0.7 ± 0.6	5.3 ± 0.8	
	OFF-SITE	130 ± 10	6 ± 2	9 ± 1	6 ± 1	6 ± 1	0.2 ± 0.3	6 ± 1	0.4 ± 0.5	5.1 ± 0.7	
APRIL	7I	240 ± 10	22 ± 3	24 ± 2	19 ± 2	18 ± 1	0.7 ± 0.2	16 ± 1	< 0.3	7.7 ± 1.0	
	12H	280 ± 10	14 ± 4	24 ± 3	15 ± 2	24 ± 2	0.6 ± 0.6	19 ± 1	0.6 ± 0.6	6.4 ± 1.1	
	OFF-SITE	260 ± 10	17 ± 3	22 ± 2	13 ± 2	20 ± 1	0.4 ± 0.2	17 ± 1	0.4 ± 0.4	5.8 ± 0.8	
MAY	7I	190 ± 10	19 ± 3	25 ± 2	18 ± 2	24 ± 2	1.0 ± 0.4	22 ± 2	1.0 ± 0.4	6.3 ± 0.9	
	12H	190 ± 20	12 ± 3	25 ± 2	13 ± 2	23 ± 1	0.8 ± 0.3	19 ± 1	< 0.3	5.4 ± 0.7	
	OFF-SITE	200 ± 50	14 ± 4	23 ± 2	16 ± 2	10 ± 2	0.6 ± 0.2	13 ± 1	< 0.3	6.3 ± 1.1	
JUNE	7I	110 ± 10	8 ± 1	12 ± 1	8 ± 1	20 ± 2	0.7 ± 0.7	24 ± 2	0.6 ± 0.6	3.7 ± 0.7	
	12H	110 ± 10	7 ± 3	11 ± 1	7 ± 1	24 ± 2	0.5 ± 0.3	21 ± 2	0.5 ± 0.6	4.5 ± 0.9	
	OFF-SITE	140 ± 10	9 ± 2	10 ± 1	9 ± 1	10 ± 1	0.7 ± 0.4	11 ± 1	0.4 ± 0.4	3.3 ± 0.5	
JULY	7I	120 ± 10	14 ± 3	15 ± 2	10 ± 1	18 ± 1	0.6 ± 0.4	13 ± 1	0.7 ± 0.4	5.0 ± 0.8	
	12H	80 ± 10	13 ± 3	18 ± 2	13 ± 2	20 ± 2	0.5 ± 0.4	19 ± 1	< 0.3	4.8 ± 0.8	
	OFF-SITE	60 ± 50	3 ± 2	4 ± 1	2 ± 1	4 ± 1	0.5 ± 0.2	4 ± 1	0.6 ± 0.5	3.6 ± 0.9	
AUGUST	7I	90 ± 10	7 ± 4	11 ± 1	7 ± 1	10 ± 3	0.3 ± 0.7	13 ± 2	0.8 ± 0.4	3.1 ± 0.7	
	12H	70 ± 10	8 ± 3	15 ± 2	8 ± 1	26 ± 2	0.6 ± 0.9	23 ± 2	< 0.3	3.6 ± 0.7	
	OFF-SITE	60 ± 10	4 ± 4	8 ± 1	5 ± 1	16 ± 1	0.4 ± 0.7	13 ± 2	0.9 ± 0.5	3.4 ± 0.8	
SEPTEMBER	7I	50 ± 10	2 ± 1	6 ± 2	2 ± 1	13 ± 1	0.2 ± 0.4	8 ± 1	0.6 ± 0.8	6.5 ± 1.4	
	12H	30 ± 20	9 ± 2	17 ± 2	7 ± 1	15 ± 1	0.3 ± 0.4	10 ± 1	0.4 ± 0.4	4.6 ± 4.1	
	OFF-SITE	30 ± 20	1 ± 1	1 ± 1	1 ± 1	6 ± 1	0.2 ± 0.6	9 ± 1	0.5 ± 0.6	7.6 ± 1.3	
OCTOBER	7I	50 ± 10	7 ± 2	19 ± 2	8 ± 1	13 ± 2	0.8 ± 0.6	13 ± 2	0.5 ± 0.6	1.2 ± 0.6	
	12H	40 ± 20	9 ± 1	15 ± 2	9 ± 1	8 ± 2	0.3 ± 0.3	8 ± 2	< 0.3	1.2 ± 0.4	
	OFF-SITE	50 ± 10	3 ± 2	6 ± 2	3 ± 1	11 ± 1	0.9 ± 0.3	7 ± 1	0.6 ± 0.4	0.7 ± 0.4	
NOVEMBER	7I	20 ± 10	11 ± 2	14 ± 2	5 ± 1	13 ± 1	0.4 ± 0.2	8 ± 1	< 0.3	0.7 ± 0.3	
	12H	30 ± 10	5 ± 1	8 ± 1	5 ± 1	7 ± 1	0.3 ± 0.4	8 ± 1	< 0.3	1.1 ± 0.4	
	OFF-SITE	40 ± 20	3 ± 1	7 ± 2	3 ± 1	8 ± 2	0.6 ± 0.5	7 ± 1	0.4 ± 0.5	1.0 ± 0.4	
DECEMBER	7I	30 ± 10	6 ± 1	11 ± 2	6 ± 1	16 ± 2	0.6 ± 0.4	9 ± 1	0.8 ± 0.4	0.9 ± 0.4	
	12H	30 ± 10	13 ± 7	16 ± 4	10 ± 4	23 ± 2	0.7 ± 0.6	23 ± 2	0.4 ± 0.5	1.6 ± 0.6	
	OFF-SITE	30 ± 30	9 ± 2	11 ± 2	9 ± 1	15 ± 2	0.3 ± 0.8	10 ± 2	0.8 ± 0.9	0.6 ± 0.8	
ANNUAL	7I	110 ± 40	13 ± 5	18 ± 5	12 ± 5	18 ± 5	0.7 ± 0.2	16 ± 5	0.5 ± 0.2	4 ± 1	
	12H	110 ± 50	11 ± 3	19 ± 5	11 ± 4	25 ± 9	0.8 ± 0.4	22 ± 9	< 0.3	4 ± 1	
SUMMARY	OFF-SITE	110 ± 40	7 ± 3	10 ± 4	7 ± 3	11 ± 3	0.5 ± 0.2	10 ± 3	0.5 ± 0.1	4 ± 1	
XCB (X $10^{-3}$ )	7I	0.06	(6.5)	(5.9)	(1.2)	(0.44)	(0.02)	(0.32)	0.05	0.4	
	12H	0.06	(5.4)	(6.2)	(1.1)	(0.63)	(0.02)	(0.44)	< 0.03	0.4	
	OFF-SITE	0.06	(3.7)	(3.4)	(0.7)	(0.29)	(0.01)	(0.21)	0.05	0.4	

<sup>1</sup> PERIMETER LOCATIONS ARE GIVEN IN TERMS OF THE GRID COORDINATES IN FIGURE 1.

<sup>2</sup> THE CONCENTRATIONS IN UNITS OF MICROGRAMS/CUBIC METER CAN BE OBTAINED BY MULTIPLYING THE VALUE IN MICROCURIES/ML BY  $2.96 \times 10^{12}$  FOR URANIUM-238 AND BY  $9 \times 10^{12}$  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.

<sup>3</sup> PLUTONIUM-240 IS INCLUDED (SEE TEXT).

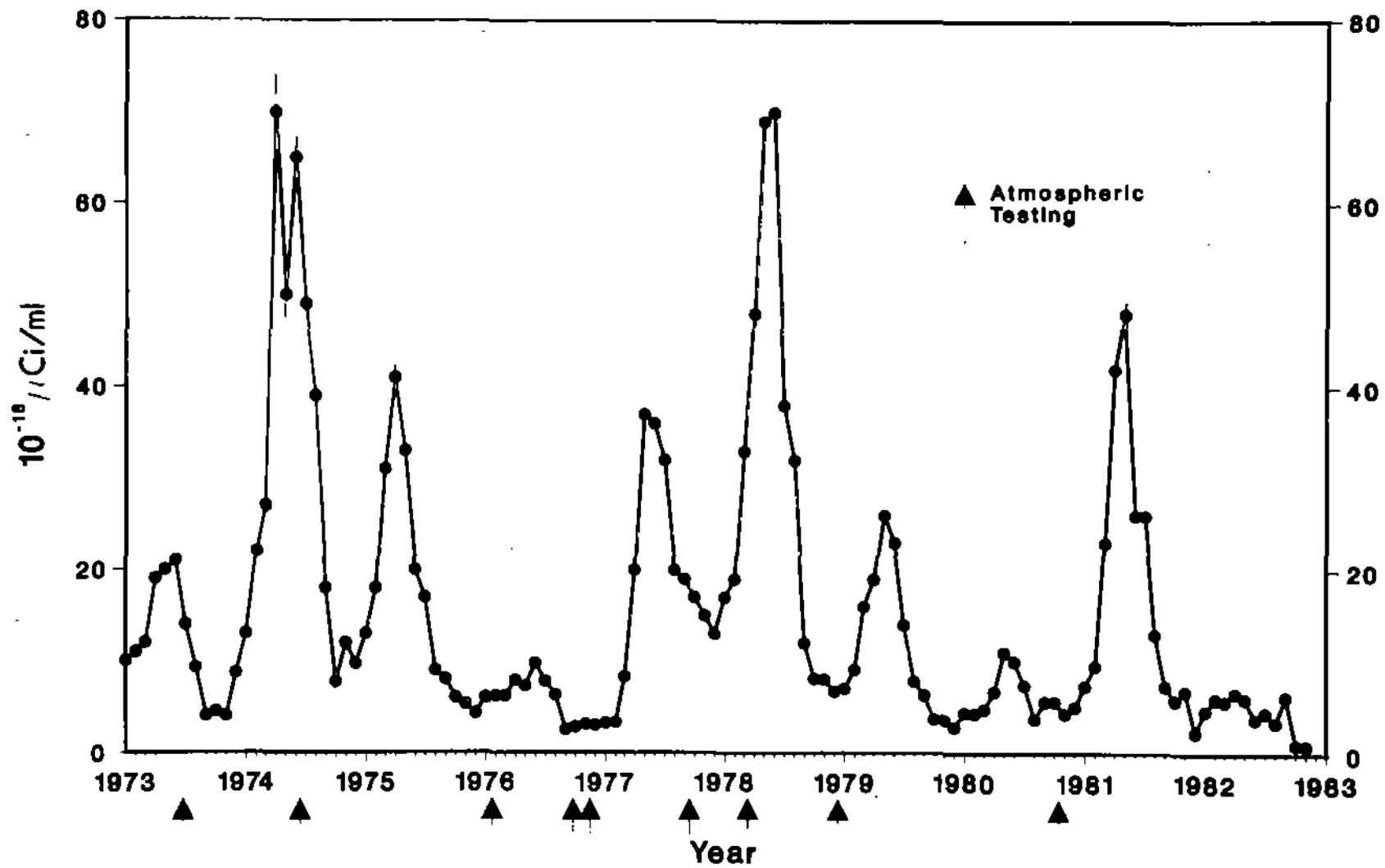


Fig. 4. Plutonium-239,240 in Air Concentrations, 1973-1982.

resuspended soil has the same plutonium concentration as the first centimeter on the ground, ranged from about 0.9% in September to 17% in October 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 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 1982 was about 0.56 Ci. The major airborne effluents released were 100 Ci of carbon-11 from the IPNS facility (location 9J) and 8.4 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.<sup>7,8</sup> 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 94% 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 43% for hydrogen-3, 86% for neptunium-237, 100% for plutonium-239, and 84% 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 (equivalent to 0.003% of the CG) was the lowest since such measurements were initiated. The decrease in the hydrogen-3 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 1982, the hydrogen-3

TABLE 7  
 RADIONUCLIDES IN SAWHILL CREEK, 1982

TYPE OF ACTIVITY	LOCATION <sup>*</sup>	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/ML)			AVG.	PERCENT CG	
			AVG.	MIN.	MAX.		MIN.	MAX.
ALPHA (NONVOLATILE)	16K	12	1.7 ± 0.1	0.7	3.7	(0.056)	(0.023)	(0.123)
	7H	248	1.7 ± 0.3	0.4	5.5	(0.057)	(0.013)	(0.184)
BETA (NONVOLATILE)	16K	12	19 ± 1	9	34	(0.63)	(0.30)	(1.14)
	7H	248	18 ± 2	8	30	(0.59)	(0.27)	(1.01)
HYDROGEN-3	16K	12	< 110	< 100	179	< 0.0037	< 0.003	0.0060
	7H	248	196 ± 58	< 100	1270	0.0065	< 0.003	0.0423
STRONTIUM-90	16K	12	< 0.30	< 0.25	0.58	< 0.102	< 0.08	0.19
	7H	248	0.28 ± 0.05	< 0.25	1.11	0.095	< 0.08	0.37
IODINE-131	16K	12	< 1.9	< 1	11.0	< 0.63	< 0.33	3.66
	7H	245	< 1.0	< 1	1.6	< 0.35	< 0.33	0.54
BARIUM-140	16K	12	-	-	< 2	-	-	< 0.007
	7H	118	-	-	< 2	-	-	< 0.007
RADIUM-226	7H	248	0.54 ± 0.11	0.15	2.30	(1.78)	(0.50)	(7.66)
URANIUM (NATURAL) <sup>**</sup>	16K	12	1.3 ± 0.1	0.2	1.8	(0.0031)	(0.0005)	(0.0046)
	7H	248	1.4 ± 0.1	0.7	2.9	(0.0036)	(0.0017)	(0.0073)
NEPTUNIUM-237	16K	11	< 0.001	< 0.001	0.001	< 0.00003	< 0.00003	0.00004
	7H	243	0.0057 ± 0.0016	< 0.001	0.026	0.00019	< 0.00003	0.0009
PLUTONIUM-238	16K	11	-	-	< 0.001	-	-	< 0.00002
	7H	243	< 0.0022	< 0.001	0.015	0.00004	< 0.00002	0.00030
PLUTONIUM-239	16K	11	< 0.00056	< 0.0005	0.0009	< 0.00001	< 0.00001	0.00002
	7H	243	0.0056 ± 0.0015	0.0008	0.0255	0.00011	0.000016	0.00051
AMERICIUM-241	16K	12	< 0.001	< 0.001	0.001	< 0.000025	< 0.000025	0.00003
	7H	248	0.0233 ± 0.0353	< 0.001	0.906	0.00058	< 0.000025	0.0227
CURIUM-242 AND/OR CALIFORNIUM-252	16K	12	-	-	< 0.001	-	-	< 0.000005
	7H	248	< 0.0012	< 0.001	0.0026	< 0.000005	< 0.000005	0.000013
CURIUM-244 AND/OR CALIFORNIUM-249	16K	12	-	-	< 0.001	-	-	< 0.000014
	7H	248	0.0022 ± 0.0005	< 0.001	0.0065	0.000031	< 0.000014	0.00009

<sup>\*</sup> LOCATION 16K IS UPSTREAM FROM THE ARGONNE SITE AND LOCATION 7H IS DOWNSTREAM FROM THE ARGONNE WASTE-WATER OUTFALL.  
<sup>\*\*</sup> URANIUM CONCENTRATIONS IN UNITS OF MICROGRAMS/L CAN BE OBTAINED BY MULTIPLYING THE CONCENTRATION GIVEN BY 1.48 X 10<sup>9</sup>.

content of 21 other lakes and streams ranged from  $< 100 \times 10^{-9}$   $\mu\text{Ci/ml}$  to  $200 \times 10^{-9}$   $\mu\text{Ci/ml}$  and averaged  $< 113 \times 10^{-9}$   $\mu\text{Ci/ml}$ .

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 1.2 Ci of hydrogen-3,  $< 1.0$  mCi of strontium-90, 0.07 mCi of neptunium-237, 0.07 mCi of plutonium-239, 0.3 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,  $1.2 \times 10^{-9}$   $\mu\text{Ci/ml}$  and  $12 \times 10^{-9}$   $\mu\text{Ci/ml}$ , respectively, of 21 off-site surface water samples collected this year (excluding the Des Plaines River) were similar to the levels found in previous years.

Comparison of the results of the water samples presented in Tables 7 and 8 indicate that the total alpha and beta activities were slightly higher above the site, which indicates that at times Argonne waste water contained less of these materials than Creek water. The higher activities above the site were probably due to the water added to the Creek by a sewage treatment plant. The large amount of dissolved solids present in the sewage is naturally accompanied by a small amount of radioactive materials, and evidently increases the radioactivity in the Creek water. Iodine-131 is still occasionally found in the River and Creek. It is

TABLE 3  
 RADIONUCLIDES IN DESPLAINES RIVER WATER, 1982

TYPE OF ACTIVITY	LOCATION <sup>#</sup>	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/ML)			PERCENT CG		
			AVG.	MIN.	MAX.	AVG.	MIN.	MAX.
ALPHA (NONVOLATILE)	A	12	1.5 ± 0.1	0.9	2.0	(0.050)	(0.030)	(0.065)
	B	24	1.5 ± 0.2	0.4	3.2	(0.051)	(0.015)	(0.105)
BETA (NONVOLATILE)	A	12	14 ± 1	8	27	(0.46)	(0.26)	(0.90)
	B	24	14 ± 2	8	27	(0.46)	(0.26)	(0.89)
HYDROGEN-3	A	12	< 121	< 100	250	< 0.0040	< 0.003	0.008
	B	24	< 118	< 100	205	< 0.0039	< 0.003	0.007
STRONTIUM-90	A	12	0.27 ± 0.01	< 0.25	0.43	0.09	< 0.08	0.14
	B	24	0.30 ± 0.04	< 0.25	0.48	0.10	< 0.08	0.16
IODINE-131	A	12	< 1.2	< 1	3.0	< 0.41	< 0.33	1.00
	B	24	< 1.2	< 1	2.3	< 0.39	< 0.33	0.76
BARIUM-140	A	12	-	-	< 2	-	-	< 0.007
	B	12	-	-	< 2	-	-	< 0.007
URANIUM <sup>**</sup> (NATURAL)	A	12	1.3 ± 0.1	0.6	2.0	(0.0033)	(0.0015)	(0.0050)
	B	24	1.3 ± 0.2	0.8	2.6	(0.0033)	(0.0019)	(0.0066)
NEPTUNIUM-237	A	11	-	-	< 0.001	-	-	< 0.00003
	B	11	< 0.0013	< 0.001	0.004	< 0.00004	< 0.00003	0.00013
PLUTONIUM-238	A	11	-	-	< 0.001	-	-	< 0.00002
	B	11	-	-	< 0.001	-	-	< 0.00002
PLUTONIUM-239	A	11	< 0.00054	< 0.0005	0.0009	< 0.000011	< 0.00001	0.000018
	B	10	< 0.00054	< 0.0005	0.0007	< 0.000011	< 0.00001	0.000014
AMERICIUM-241	A	12	-	-	< 0.001	-	-	< 0.000025
	B	12	< 0.0011	< 0.001	0.001	< 0.000027	< 0.000025	0.000035
CURIUM-242 AND/OR CALIFORNIUM-252	A	12	-	-	< 0.001	-	-	< 0.000005
	B	11	-	-	< 0.001	-	-	< 0.000005
CURIUM-244 AND/OR CALIFORNIUM-249	A	12	-	-	< 0.001	-	-	< 0.000014
	B	12	< 0.0011	< 0.001	0.0019	< 0.000015	< 0.000014	0.000027

<sup>#</sup> LOCATION A, NEAR HILLON SPRINGS, IS UPSTREAM AND LOCATION B, NEAR LEMONT, IS DOWNSTREAM FROM THE MOUTH OF SAKMILL CREEK. SEE FIGURE 2.

<sup>\*\*</sup> URANIUM CONCENTRATIONS IN UNITS OF MICROGRAMS/L CAN BE OBTAINED BY MULTIPLYING THE CONCENTRATION GIVEN BY  $1.48 \times 10^{-9}$ .

assumed that the source of the iodine-131 is a medical facility or a sewage treatment plant that has processed excretions from an individual who has received a medical injection of iodine-131.<sup>9</sup>

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 River.

### 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 from weapons testing 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 purpose, the site selection criteria and sample preparation techniques of the DOE Environmental Measurements Laboratory<sup>10</sup> were used. 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<sup>2</sup> 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<sup>2</sup> 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 method described in Section

TABLE 9

Radionuclides in Illinois River Water, 1982  
(Concentrations in  $10^{-9}$   $\mu\text{Ci/ml}$ )

Date Collected	Location	Alpha*	Beta*	Hydrogen-3	Uranium** (natural)	Plutonium-239
May 18	McKinley Woods State Park	1.5 $\pm$ 0.3	13.5 $\pm$ 0.4	114 $\pm$ 101	0.7 $\pm$ 0.1	0.00008 $\pm$ 0.00011
May 18	Below Dresden Power Station	1.3 $\pm$ 0.3	7.1 $\pm$ 0.3	122 $\pm$ 101	1.1 $\pm$ 0.1	0.00013 $\pm$ 0.00009
May 18	Morris	1.7 $\pm$ 0.3	7.9 $\pm$ 0.3	133 $\pm$ 101	1.5 $\pm$ 0.1	-
May 18	Starved Rock State Park	1.4 $\pm$ 9.3	9.2 $\pm$ 0.3	< 100	1.0 $\pm$ 0.1	-
October 12	McKinley Woods State Park	0.5 $\pm$ 0.2	8.4 $\pm$ 0.3	< 100	0.6 $\pm$ 0.1	0.00027 $\pm$ 0.00015
October 12	Below Dresden Power Station	0.6 $\pm$ 0.2	7.7 $\pm$ 0.3	< 100	0.9 $\pm$ 0.1	0.00017 $\pm$ 0.00005
October 12	Morris	0.7 $\pm$ 0.2	9.9 $\pm$ 0.3	< 100	1.0 $\pm$ 0.1	-
October 12	Starved Rock State Park	0.8 $\pm$ 0.2	7.3 $\pm$ 0.3	< 100	0.9 $\pm$ 0.1	-

\* Nonvolatile activity.

\*\* Uranium concentrations in units of  $\mu\text{g/l}$  can be obtained by multiplying the concentration by  $1.48 \times 10^9$ .

III.A for air-filter residue. Results are given in terms of the oven-dried sample weight.

Soil samples were analyzed by gamma-ray spectrometry and radiochemically for plutonium and americium. The plutonium and americium were separated from the same 100 gram aliquot of soil. The results for the gamma-ray nuclides in soil are presented in Table 10. The antimony-125 and cerium-144 are residuals from the October 16, 1980, atmospheric nuclear test by the People's Republic of China. The other intermediate half-life fission products reported last year have decayed to below their detection limits. The cesium-137 levels are similar to those found over the past several 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. About 10% of the americium-241 in soil is due to direct deposition from fallout, while about 90% of the americium-241 is from the 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.<sup>11</sup>

Composite monthly precipitation samples were analyzed for plutonium-239. The results are given in Table 12, along with results since 1973 for comparison. The 1982 deposition by precipitation was equivalent to 0.2% of the total plutonium deposited through 1976, which is  $2.2 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ .<sup>12</sup> 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 ten years in Figure 4 shows excellent correlation.

The results of radionuclides measured in grass are given in Table 13. The annual averages were similar between perimeter and off-site locations. 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.

TABLE 10

Gamma-Ray Emitting Radionuclides in Soil, 1982  
(Concentrations in  $10^{-6}$   $\mu\text{Ci/g}$ )

Date Collected	Location	Antimony-125	Cesium-1347	Cerium-144
	<u>Perimeter*</u>			
May 25	8G	< 0.05	1.26 $\pm$ 0.04	< 0.01
May 25	13D	< 0.05	0.94 $\pm$ 0.03	< 0.01
May 25	15H	< 0.05	1.70 $\pm$ 0.03	0.16 $\pm$ 0.04
May 25	13N	< 0.05	0.59 $\pm$ 0.02	< 0.01
May 25	7M	< 0.05	0.69 $\pm$ 0.02	< 0.01
October 13	8N	0.07 $\pm$ 0.03	0.41 $\pm$ 0.02	< 0.01
October 13	5D	< 0.05	0.90 $\pm$ 0.03	< 0.01
October 13	12-0	< 0.05	1.12 $\pm$ 0.03	< 0.01
October 13	14L	< 0.05	0.73 $\pm$ 0.03	< 0.01
October 13	12C	< 0.05	1.13 $\pm$ 0.03	< 0.01
	Average	< 0.06	0.95 $\pm$ 0.24	< 0.03
	<u>Off-Site</u>			
May 18	Morris, IL	< 0.05	0.34 $\pm$ 0.02	0.34 $\pm$ 0.07
May 18	Starved Rock State Park, IL	< 0.05	0.79 $\pm$ 0.03	< 0.01
May 18	Channahon, IL	< 0.05	1.08 $\pm$ 0.04	< 0.01
June 15	Brookfield, IL	< 0.05	0.45 $\pm$ 0.02	< 0.01
June 15	Western Springs, IL	< 0.05	0.55 $\pm$ 0.02	< 0.01
October 12	McKinely Woods State Park, IL	< 0.05	0.43 $\pm$ 0.03	< 0.01
October 12	Dresden Lock and Dam, IL	< 0.05	1.34 $\pm$ 0.04	< 0.01
October 14	Willow Springs, IL	0.10 $\pm$ 0.03	0.62 $\pm$ 0.02	0.43 $\pm$ 0.05
October 14	Saganashkee Slough, IL	< 0.05	0.82 $\pm$ 0.03	< 0.01
October 14	McGinnis Slough, IL	< 0.05	1.22 $\pm$ 0.03	< 0.01
	Average	< 0.06	0.76 $\pm$ 0.22	< 0.09

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

TABLE 11

## Transuranics in Soil, 1982

Date Collected	Location	Plutonium-238		Plutonium-239		$^{238}\text{Pu}/^{239}\text{Pu}$	Americium-241		$^{241}\text{Am}/^{239}\text{Pu}$	
		$10^{-9}$ $\mu\text{Ci/g}$	$10^{-3}$ $\mu\text{Ci/m}^2$	$10^{-9}$ $\mu\text{Ci/g}$	$10^{-3}$ $\mu\text{Ci/m}^2$		$10^{-9}$ $\mu\text{Ci/g}$	$10^{-3}$ $\mu\text{Ci/m}^2$		
<u>Perimeter*</u>										
May 25	8G	$0.8 \pm 0.2$	$0.050 \pm 0.011$	$24.0 \pm 0.9$	$1.61 \pm 0.06$	0.031	$4.7 \pm 0.4$	$0.318 \pm 0.028$	0.20	
May 25	13D	$0.7 \pm 0.2$	$0.038 \pm 0.012$	$17.0 \pm 1.0$	$0.92 \pm 0.06$	0.041	$3.3 \pm 0.4$	$0.179 \pm 0.023$	0.19	
May 25	15H	$1.3 \pm 0.3$	$0.069 \pm 0.014$	$31.3 \pm 1.5$	$1.69 \pm 0.08$	0.041	$8.0 \pm 0.5$	$0.433 \pm 0.027$	0.26	
May 25	13N	$0.4 \pm 0.2$	$0.020 \pm 0.008$	$10.0 \pm 0.7$	$0.54 \pm 0.04$	0.036	$2.4 \pm 0.4$	$0.132 \pm 0.024$	0.24	
May 25	7M	$0.8 \pm 0.2$	$0.044 \pm 0.010$	$15.6 \pm 0.9$	$0.86 \pm 0.05$	0.051	$3.7 \pm 0.3$	$0.206 \pm 0.018$	0.24	
October 13	8N	$0.3 \pm 0.2$	$0.015 \pm 0.008$	$7.7 \pm 0.6$	$0.39 \pm 0.03$	0.037	$1.6 \pm 0.3$	$0.080 \pm 0.015$	0.20	
October 13	5D	$0.6 \pm 0.2$	$0.028 \pm 0.008$	$15.8 \pm 0.9$	$0.78 \pm 0.05$	0.036	$2.8 \pm 0.5$	$0.138 \pm 0.026$	0.18	
October 13	12-0	$0.7 \pm 0.3$	$0.038 \pm 0.016$	$20.0 \pm 1.5$	$1.02 \pm 0.08$	0.037	$5.1 \pm 0.8$	$0.260 \pm 0.042$	0.25	
October 13	14L	$0.6 \pm 0.2$	$0.028 \pm 0.012$	$13.4 \pm 1.0$	$0.65 \pm 0.05$	0.044	$4.2 \pm 0.8$	$0.204 \pm 0.039$	0.32	
October 13	12C	$1.0 \pm 0.2$	$0.040 \pm 0.009$	$20.0 \pm 1.2$	$0.82 \pm 0.05$	0.049	$4.0 \pm 0.5$	$0.164 \pm 0.022$	0.20	
	Average	$0.7 \pm 0.2$	$0.037 \pm 0.010$	$17.5 \pm 4.3$	$0.93 \pm 0.27$	0.040	$4.0 \pm 1.1$	$0.217 \pm 0.065$	0.23	
<u>Off-Site</u>										
May 18	Morris, IL	$0.3 \pm 0.2$	$0.016 \pm 0.009$	$8.5 \pm 0.7$	$0.50 \pm 0.04$	0.032	$1.6 \pm 0.4$	$0.091 \pm 0.023$	0.18	
May 18	Starved Rock State Park, IL	$0.7 \pm 0.2$	$0.051 \pm 0.014$	$17.4 \pm 1.0$	$1.20 \pm 0.07$	0.042	$2.4 \pm 0.5$	$0.168 \pm 0.033$	0.14	
May 18	Channahon, IL	$1.0 \pm 0.3$	$0.056 \pm 0.016$	$23.4 \pm 1.5$	$1.25 \pm 0.08$	0.044	$5.1 \pm 0.7$	$0.272 \pm 0.037$	0.22	
June 15	Brookfield, IL	$0.6 \pm 0.2$	$0.035 \pm 0.010$	$10.4 \pm 0.6$	$0.67 \pm 0.04$	0.052	$2.5 \pm 0.4$	$0.157 \pm 0.024$	0.24	
June 15	Western Springs, IL	$0.5 \pm 0.2$	$0.027 \pm 0.008$	$9.4 \pm 0.6$	$0.50 \pm 0.03$	0.054	-	-	-	
October 12	McKinley Woods State Park, IL	$0.3 \pm 0.2$	$0.016 \pm 0.013$	$5.7 \pm 0.8$	$0.29 \pm 0.04$	0.055	$1.7 \pm 0.2$	$0.088 \pm 0.012$	0.30	
October 12	Dresden Lock & Dam, IL	$0.9 \pm 0.2$	$0.042 \pm 0.018$	$26.7 \pm 1.4$	$1.24 \pm 0.07$	0.034	$4.3 \pm 0.4$	$0.199 \pm 0.018$	0.16	
October 14	Willow Springs, IL	$0.5 \pm 0.2$	$0.030 \pm 0.014$	$9.9 \pm 0.9$	$0.65 \pm 0.06$	0.046	$1.4 \pm 0.3$	$0.093 \pm 0.020$	0.14	
October 14	Saganashkee Slough, IL	$0.4 \pm 0.2$	$0.020 \pm 0.011$	$14.8 \pm 1.2$	$0.70 \pm 0.06$	0.029	$3.6 \pm 1.0$	$0.173 \pm 0.046$	0.25	
October 14	McGinnis Slough, IL	$0.6 \pm 0.2$	$0.024 \pm 0.008$	$21.7 \pm 1.0$	$0.96 \pm 0.04$	0.025	$2.4 \pm 0.7$	$0.106 \pm 0.030$	0.11	
	Average	$0.6 \pm 0.2$	$0.032 \pm 0.009$	$14.8 \pm 4.5$	$0.80 \pm 0.22$	0.041	$2.8 \pm 0.9$	$0.150 \pm 0.041$	0.19	

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

TABLE 12

Ground Deposition of Plutonium-239,240  
(Units of  $10^{-6}$   $\mu\text{Ci}/\text{m}^2$ )

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

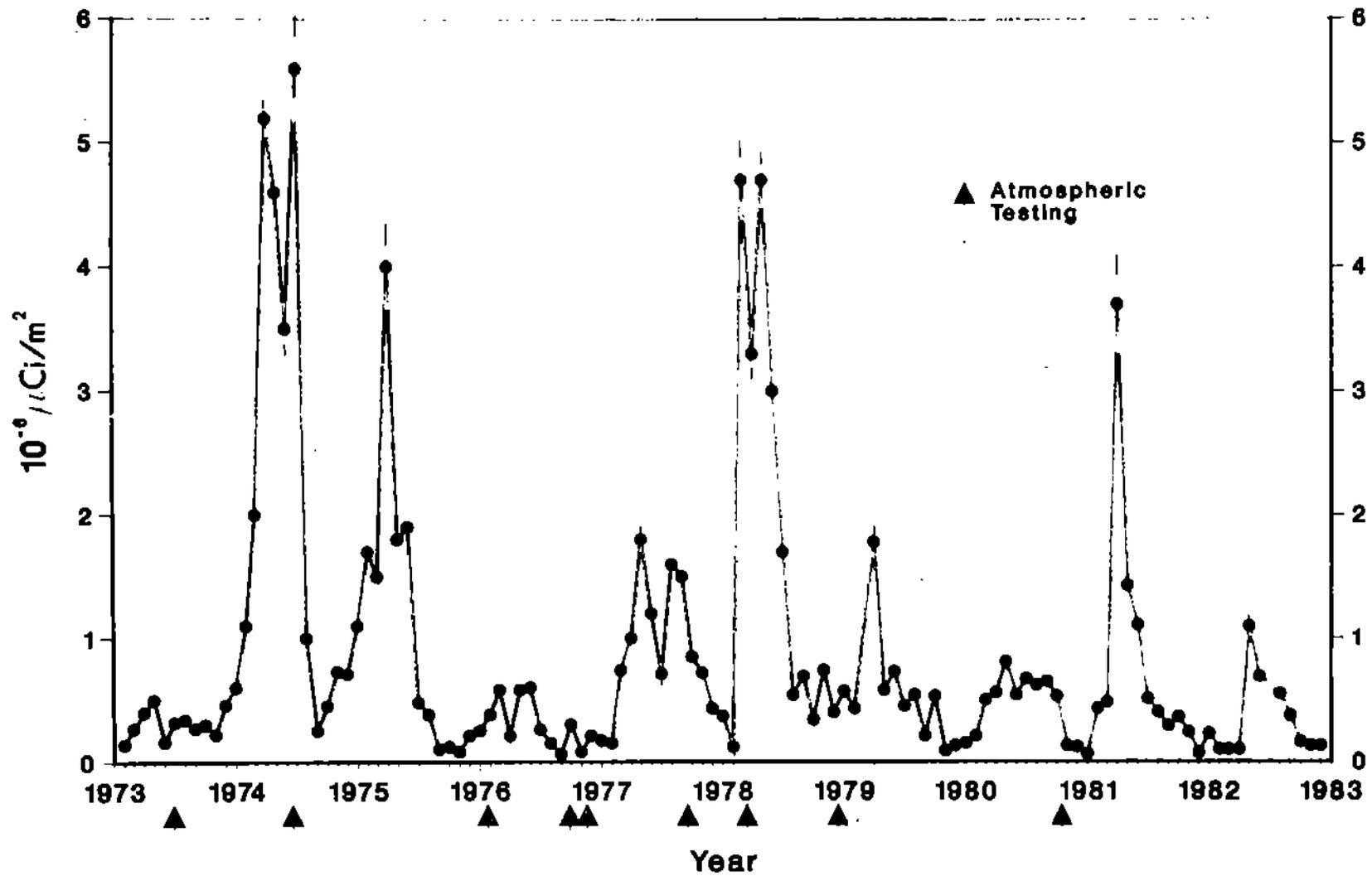


Fig. 5. Plutonium-239,240 in Surface Deposition, 1973-1982.

TABLE 13

## Radionuclides in Grass Samples, 1982

Date Collected	Location	Concentrations in $10^{-9}$ $\mu\text{Ci/g}$					$10^{-6}$ $\mu\text{Ci/m}^2$	
		Cesium-137	Thorium-228	Thorium-230	Thorium-232	Uranium (natural)	Plutonium-239	Deposition of Plutonium-239
	<u>Perimeter*</u>							
May 25	8G	< 50	$3.8 \pm 1.2$	$4.9 \pm 1.0$	$1.5 \pm 0.4$	$44 \pm 5$	$0.1 \pm 0.1$	$0.024 \pm 0.015$
May 25	13D	< 50	$2.2 \pm 0.6$	$2.6 \pm 0.4$	$3.0 \pm 0.5$	$31 \pm 3$	< 0.1	< 0.01
May 25	15H	< 50	$1.5 \pm 0.5$	$2.1 \pm 0.3$	$2.0 \pm 0.3$	$67 \pm 5$	$0.2 \pm 0.1$	$0.018 \pm 0.011$
May 25	13N	< 50	$0.7 \pm 0.1$	$1.5 \pm 0.2$	$1.0 \pm 0.1$	$77 \pm 10$	$0.4 \pm 0.1$	$0.047 \pm 0.013$
May 25	7H	< 50	$0.8 \pm 0.4$	$1.6 \pm 0.2$	$0.8 \pm 0.3$	$51 \pm 4$	$0.1 \pm 0.1$	$0.024 \pm 0.014$
October 13	8N	< 50	$0.9 \pm 0.5$	$1.2 \pm 0.6$	$0.6 \pm 0.4$	$37 \pm 17$	$0.2 \pm 0.1$	$0.051 \pm 0.024$
October 13	5D	< 50	$0.5 \pm 0.5$	$1.2 \pm 0.2$	$0.5 \pm 0.3$	$24 \pm 2$	$0.1 \pm 0.1$	$0.040 \pm 0.030$
October 13	12-0	< 50	$0.4 \pm 0.3$	$1.0 \pm 0.2$	$0.4 \pm 0.4$	$18 \pm 10$	$1.2 \pm 0.2$	$0.380 \pm 0.056$
October 13	14L	< 50	$0.8 \pm 0.2$	$1.9 \pm 0.4$	$0.9 \pm 0.3$	$43 \pm 4$	$0.1 \pm 0.1$	$0.067 \pm 0.039$
October 13	12C	< 50	$0.8 \pm 0.2$	$1.8 \pm 0.5$	$0.7 \pm 0.4$	$8 \pm 3$	$0.2 \pm 0.1$	$0.081 \pm 0.038$
	Average	< 50	$1.2 \pm 0.7$	$2.0 \pm 0.7$	$1.1 \pm 0.5$	$40 \pm 13$	$0.3 \pm 0.2$	$0.074 \pm 0.070$
	<u>Off-Site</u>							
May 18	Morris, IL	< 50	$2.6 \pm 0.6$	$4.0 \pm 0.4$	$3.9 \pm 0.6$	$52 \pm 6$	$0.1 \pm 0.1$	$0.012 \pm 0.009$
May 18	Starved Rock State Park, IL	$60 \pm 30$	$1.5 \pm 0.3$	$2.1 \pm 0.3$	$1.9 \pm 0.3$	$47 \pm 5$	$0.6 \pm 0.1$	$0.073 \pm 0.014$
May 18	Channahon, IL	< 50	$0.3 \pm 0.4$	$0.9 \pm 0.2$	$0.4 \pm 0.2$	$70 \pm 6$	$0.3 \pm 0.1$	$0.030 \pm 0.007$
June 15	Brookfield, IL	< 50	$1.8 \pm 0.4$	$2.2 \pm 0.3$	$1.3 \pm 0.2$	$57 \pm 7$	$0.1 \pm 0.1$	$0.023 \pm 0.010$
June 15	Western Springs, IL	< 50	$2.9 \pm 0.6$	$4.7 \pm 0.6$	$1.8 \pm 0.3$	$64 \pm 6$	< 0.1	< 0.01
October 12	McKinley Woods State Park, IL	< 50	$2.1 \pm 0.8$	$1.9 \pm 0.7$	$1.1 \pm 0.5$	$54 \pm 6$	$0.3 \pm 0.1$	$0.096 \pm 0.025$
October 12	Dresden Lock and Dam, IL	< 50	$1.4 \pm 0.7$	$1.7 \pm 0.8$	$0.7 \pm 0.5$	$26 \pm 2$	$0.3 \pm 0.1$	$0.065 \pm 0.006$
October 14	Willow Springs, IL	< 50	$0.6 \pm 0.3$	$1.0 \pm 0.5$	$0.5 \pm 0.3$	$38 \pm 9$	$0.2 \pm 0.1$	$0.022 \pm 0.011$
October 14	Saganashkee Slough, IL	< 50	$0.9 \pm 0.4$	$1.4 \pm 0.4$	$0.7 \pm 0.3$	$69 \pm 7$	$0.2 \pm 0.1$	$0.019 \pm 0.011$
October 14	McGinnis Slough, IL	< 50	$0.6 \pm 0.2$	$1.1 \pm 0.6$	$0.6 \pm 0.4$	$27 \pm 2$	$0.2 \pm 0.2$	$0.036 \pm 0.035$
	Average	< 51	$1.5 \pm 0.6$	$2.1 \pm 0.8$	$1.3 \pm 0.7$	$50 \pm 10$	$0.2 \pm 0.1$	$0.038 \pm 0.019$

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

Concentration ranges were the same at the perimeter and off-site, and similar to previous years, indicating no contribution from ANL operations.

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 of concentrations 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 September 15, 1982, 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 concentrations are the highest at the outfall and fall off rapidly with distance, approximating the off-site results at 500 m below the 7M outfall. The presence of the 45 day half-life iron-59 implies a relatively recent release. The ability to monitor for the transplutonium nuclides by analysis of 100 g soil or sediment sample is a recently acquired technique. In addition to those transplutonium nuclides listed in Table 14, americium-243 ( $36 \times 10^{-9}$   $\mu\text{Ci/g}$ ) and californium-252 ( $1.2 \times 10^{-9}$   $\mu\text{Ci/g}$ ) were also determined in the sample collected at the outfall. A similar set of samples was collected on 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. This difference also implies a relatively recent release, or may reflect the inhomogeneous nature of the bottom sediment.

#### 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  $1 \times 10^{-10}$   $\mu\text{Ci/ml}$  by an ion-exchange separation followed by beta counting. Cesium-137 was analyzed with a detection limit of  $5 \times 10^{-10}$   $\mu\text{Ci/ml}$  by an ion-exchange separation followed by gamma-ray spectrometry. The other nuclides were analyzed by the same

TABLE 14

## Radionuclides in Bottom Sediment, 1982

Date Collected	Location	Concentrations in $10^{-6}$ $\mu\text{Ci/g}$					Concentrations in $10^{-9}$ $\mu\text{Ci/g}$			
		$^{59}\text{Fe}$	$^{60}\text{Co}$	$^{125}\text{Sb}$	$^{137}\text{Cs}$	$^{144}\text{Ce}$	$^{238}\text{Pu}$	$^{239}\text{Pu}$	$^{241}\text{Am}$	$^{244}\text{Cm}$
	<u>Perimeter*</u>									
May 25	15H	< 0.01	< 0.01	$0.08 \pm 0.04$	$1.16 \pm 0.04$	$0.17 \pm 0.07$	$1.6 \pm 0.4$	$17.6 \pm 1.4$	$1.4 \pm 0.3$	< 0.1
September 15	50 m above 7M Outfall	< 0.01	< 0.01	< 0.05	$0.02 \pm 0.01$	< 0.05	$0.1 \pm 0.1$	$1.5 \pm 0.3$	$0.4 \pm 0.1$	< 0.1
September 15	7M Outfall	$3.2 \pm 0.1$	$0.46 \pm 0.02$	< 0.05	$0.57 \pm 0.02$	$0.19 \pm 0.07$	$112 \pm 7$	$739 \pm 19$	$213 \pm 2$	$46 \pm 1$
September 15	50 m below 7M Outfall	$0.28 \pm 0.04$	$0.03 \pm 0.01$	< 0.05	$0.18 \pm 0.01$	< 0.05	$4.5 \pm 0.4$	$24.4 \pm 1.2$	$10.6 \pm 0.5$	$2.3 \pm 0.2$
September 15	100 m below 7M Outfall	$0.17 \pm 0.03$	$0.02 \pm 0.01$	< 0.05	$0.10 \pm 0.01$	< 0.05	$2.4 \pm 0.3$	$15.1 \pm 0.7$	$2.9 \pm 0.4$	< 0.1
September 15	500 m below 7M Outfall	< 0.01	< 0.01	< 0.05	$0.09 \pm 0.01$	< 0.05	$0.8 \pm 0.2$	$6.1 \pm 0.5$	$0.7 \pm 0.1$	< 0.1
	<u>Off-Site</u>									
May 18	Illinois River, Morris, IL	< 0.01	< 0.01	< 0.05	$0.03 \pm 0.01$	< 0.05	$0.2 \pm 0.1$	$4.2 \pm 0.5$	$0.2 \pm 0.1$	< 0.1
May 18	Illinois River, Starved Rock State Park, IL	< 0.01	< 0.01	< 0.05	$0.03 \pm 0.01$	< 0.05	$0.2 \pm 0.2$	$7.6 \pm 0.8$	$0.2 \pm 0.1$	< 0.1
June 15	Des Plaines River, Brookfield, IL	< 0.01	< 0.01	$0.12 \pm 0.03$	$0.56 \pm 0.03$	$0.46 \pm 0.07$	$0.4 \pm 0.2$	$9.8 \pm 0.7$	$1.0 \pm 0.2$	< 0.1
June 15	Salt Creek, Western Springs, IL	< 0.01	< 0.01	< 0.05	$0.11 \pm 0.02$	< 0.05	$0.1 \pm 0.1$	$1.9 \pm 0.3$	$0.3 \pm 0.2$	< 0.1
June 15	Long Run Creek, Lemont, IL	< 0.01	< 0.01	< 0.05	$0.30 \pm 0.02$	< 0.05	$0.2 \pm 0.1$	$8.3 \pm 0.6$	$1.4 \pm 0.3$	< 0.1
October 12	DuPage River, Channahon, IL	< 0.01	< 0.01	< 0.05	$0.14 \pm 0.01$	< 0.05	$0.1 \pm 0.1$	$3.8 \pm 0.4$	$0.9 \pm 0.5$	< 0.1
October 12	Illinois River, McKinley Woods State Park, IL	< 0.01	< 0.01	< 0.05	$0.05 \pm 0.01$	< 0.05	$0.2 \pm 0.1$	$1.4 \pm 0.3$	$0.3 \pm 0.1$	< 0.1
October 12	Illinois River, Dresden Lock & Dam, IL	< 0.01	< 0.01	< 0.05	$0.10 \pm 0.01$	< 0.05	$0.2 \pm 0.1$	$1.6 \pm 0.3$	$0.6 \pm 0.2$	< 0.1
October 14	Saganashkee Slough, IL	< 0.01	< 0.01	< 0.05	$0.07 \pm 0.01$	< 0.05	$0.4 \pm 0.1$	$1.7 \pm 0.6$	$0.8 \pm 0.3$	< 0.1
October 14	McGinnis Slough, IL	< 0.01	< 0.01	< 0.05	$0.35 \pm 0.02$	< 0.05	$0.3 \pm 0.1$	$7.0 \pm 0.6$	-	< 0.1
	Average	< 0.01	< 0.01	< 0.06	$0.17 \pm 0.11$	< 0.09	$0.2 \pm 0.1$	$4.7 \pm 2.0$	$0.6 \pm 0.3$	< 0.1

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 decreased by about 50% compared to 1981 and were similar to the 1980 results. 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 Concentration Guides.

TABLE 15

Radionuclides in Milk, 1982  
(Concentrations in  $10^{-9}$   $\mu$ Ci/ml)

Date Collected	Hydrogen-3	Strontium-90	Cesium-137
January 6	< 100	3.1 $\pm$ 0.4	1.5 $\pm$ 0.3
February 3	< 100	3.2 $\pm$ 0.5	1.2 $\pm$ 0.2
March 3	163 $\pm$ 104	2.6 $\pm$ 0.1	1.8 $\pm$ 0.4
April 7	< 100	3.1 $\pm$ 0.2	1.4 $\pm$ 0.4
May 5	< 100	3.4 $\pm$ 0.4	1.4 $\pm$ 0.4
June 2	< 100	3.9 $\pm$ 0.6	0.9 $\pm$ 0.4
July 7	145 $\pm$ 101	5.4 $\pm$ 0.2	0.6 $\pm$ 0.3
August 4	164 $\pm$ 102	5.5 $\pm$ 0.4	0.8 $\pm$ 0.3
September 8	< 100	3.7 $\pm$ 0.4	0.7 $\pm$ 0.2
October 6	130 $\pm$ 100	3.3 $\pm$ 0.2	0.8 $\pm$ 0.2
November 3	< 100	2.3 $\pm$ 0.2	0.9 $\pm$ 0.3
December 1	< 100	3.0 $\pm$ 0.5	0.8 $\pm$ 0.4
Average	< 117	3.5 $\pm$ 0.6	1.1 $\pm$ 0.2

## 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 calibrated with a National Bureau of Standards 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 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, and 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 89 to 98 days, and in total covered the period from January 12, 1982, to January 17, 1983. 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  $96 \pm 10$  mrem/yr. From 1973 to 1981, the corresponding values varied from  $89 \pm 7$  mrem/yr (1979) to  $100 \pm 23$  mrem/yr (1973); the nine-year average was  $93 \pm 3$  mrem/yr. Thus, the background radiation has been quite consistent and the 1982 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 (and averaged

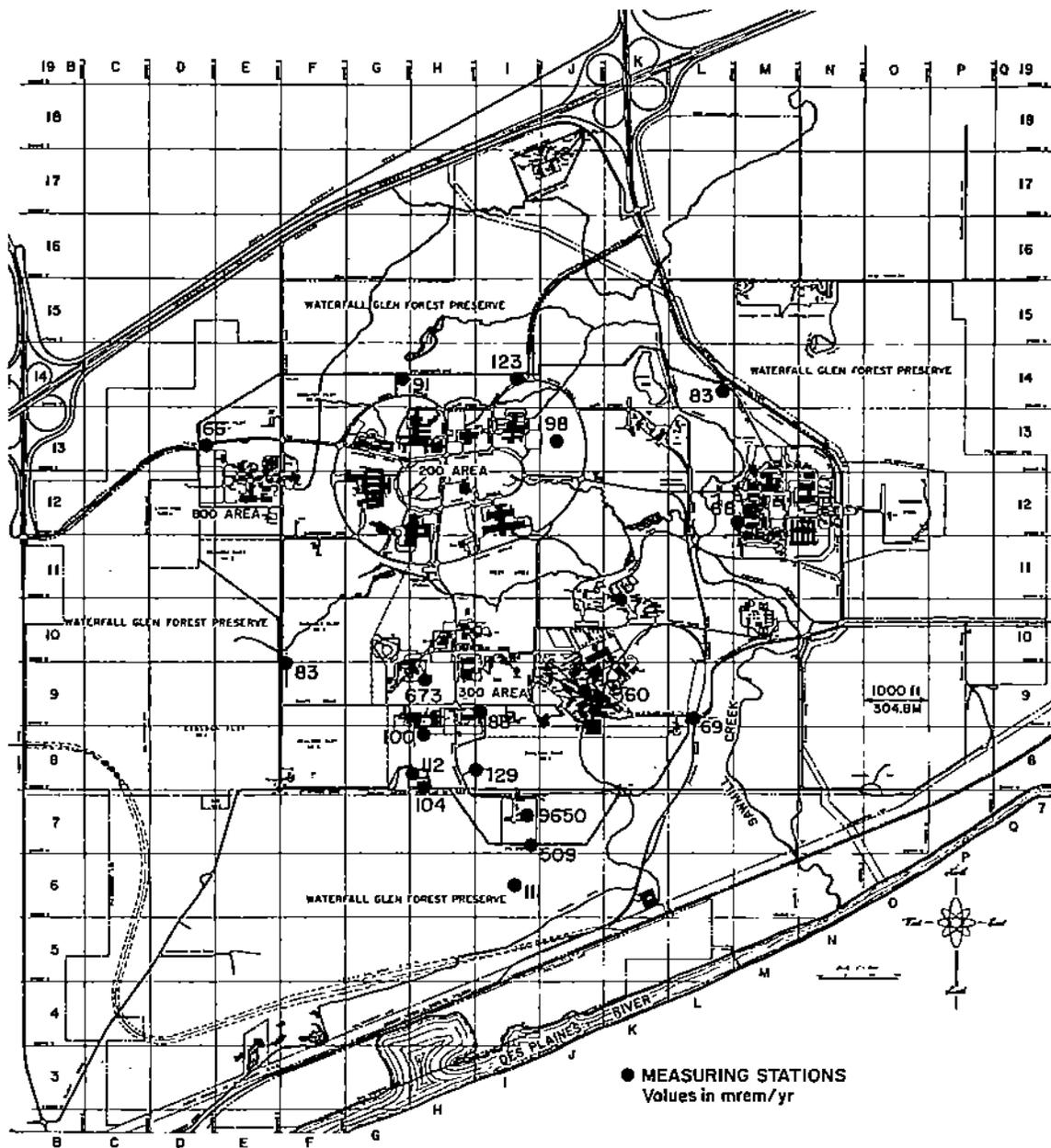


Fig. 6. Penetrating Radiation Measurements at the ANL Site, 1982

10 mrem/yr) below and at Oakbrook, 3 to 11 mrem/yr (average, 9 mrem/yr) above, the overall average.

TABLE 16

## Environmental Penetrating Radiation at Off-Site Locations, 1982

Location	Dose Rate (mrem/year)				Average
	Period of Measurement				
	1/12-4/13	4/13-7/20	7/20-10/20	10/20-1/17	
Downers Grove	83	98	88	99	92 ± 11
Lemont	88	96	101	109	99 ± 12
Lombard	90	98	102	103	98 ± 8
Oakbrook	94	-	112	112	106 ± 26
Oak Lawn	76	80	89	87	83 ± 8
Average	86 ± 8	93 ± 12	98 ± 12	102 ± 12	96 ± 10

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  $96 \pm 10$  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 value is 10 mrem/yr, so that individual results in the range of  $96 \pm 21$  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

TABLE 17

## Environmental Penetrating Radiation at ANL, 1982

Location	Dose Rate (mrem/year)				Average
	Period of Measurement				
	1/12-4/13	4/13-7/20	7/20-10/20	10/20-1/17	
14L - Boundary	75	84	84	89	83 ± 8
14I - Boundary	123	126	122	122	123 ± 3
14G - Boundary	84	97	90	91	91 ± 7
13D - Boundary	61	67	66	71	66 ± 6
9/10EF - Boundary	77	86	85	83	83 ± 6
8H - Boundary	102	112	110	93	104 ± 12
8H - Boundary, Center, St. Patrick's Cemetery	109	-	112	115	112 ± 4
7I - Boundary	753	593	413	278	509 ± 288
6I - 200 m N of Quarry Road	-	106	115	-	111 ± 19
9L - Boundary	67	70	72	68	69 ± 3
9H - 50 m SE of CP-5	697	683	674	638	673 ± 35
8H - 65 m S of Building 316	101	103	99	96	100 ± 4
8H - 200 m NW of Waste Storage Area	145	135	123	111	129 ± 20
7I - Center, Waste Storage Area	18,600	7,630	6,440	5,940	9,650 ± 8,340
10/11K - Lodging Facilities	69	73	74	68	71 ± 4
9J - Between ZGS Condenser and Building 370	65	64	59	52	60 ± 8
13J - 135 m E of Building 202	194	208	192	199	198 ± 10
12M - 30 m W of Building 55	67	65	66	73	68 ± 5
9I - 65 m NE of Building 350, 230 m NE of Building 316	97	90	86	77	88 ± 12

location was about 410 mrem/yr, 16% less than in 1981; 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  $111 \pm 19$  mrem/yr, which is close to normal levels. As shown in Table 18, the dose rate in the Waste Storage Area and at the 7I boundary decreased during the year, by a factor of about three, as a result of the shipment of a substantial portion of the stored waste to disposal sites. The above-normal dose at the 7I boundary was about 4.5% of the dose at the center of the area. The above-normal dose at the 8H (Heliport) location 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 8H.

The dose at 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 1982, as in previous years, this dose is estimated to be 5 to 10 mrem/yr above normal for this location, and was formerly attributed to radiation from the Storage Area at location 7I.<sup>4</sup> This conclusion appeared reasonable, in view of the off-site averages, the topography of the area, and other considerations. However, the dose at 8H did not decrease as did the dose rates at other locations where the 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 (F-test and t-test), and differences between the averages were found not to be significant at the 95% confidence level. However, the consistency of the apparent 5-10 mrem/yr elevated dose in the Cemetery indicates that the difference may be real. If the difference were due to statistical variation only, it would be expected that the dose in the Cemetery would fluctuate above and below the off-site average. It is possible that the granite stones in the Cemetery produce the elevated readings, since granite contains higher concentrations of natural radioactivities than soil or sandstone.<sup>13</sup> A survey of the area with a portable gamma-ray detector showed that the granite gravestones did emit more radiation than those made of sandstone. Additional tests will be conducted to determine if the general gamma-ray dose in the Cemetery is actually due to the relatively large amount of granite. A similar survey in the 7I Storage Area confirmed the presence of

sufficient gamma-ray emitters to produce the dose observed by the TL dosimeters.

## 6. Potential Radiation Dose Estimates

### a. Air and Waterborne Radionuclides

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 that would result from continuous exposure at the specified concentrations for those nuclides whose presence in the environment are attributable to Argonne. For carbon-11, argon-41, and krypton-85 released from stacks, doses were calculated from an atmospheric dispersion model which made use of a source term and meteorological data.

The principal exposure pathway for radioactive substances released from Argonne is directly from air to man by submersion. Although Sawmill Creek 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 is 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 the the dose calculation is meaningless. No other exposure pathways are significant.

Carbon-11, argon-41, and krypton-85 represent the major gaseous radioactive effluents released from the Laboratory. The concentrations and dose rates were calculated for those three nuclides by a computer program based on an atmospheric dispersion model.<sup>14,15</sup> The following parameters were used in the calculations:

- a) annual release rates: carbon-11, 100 Ci; argon-41, 0.56 Ci; and krypton-85, 8.4 Ci.

TABLE 18

## Concentration-to-Dose Conversion Factors

Nuclide	Medium	Concentration* ( $\mu\text{Ci/ml}$ )	Dose* (rem)	Critical Organ
Americium-241	Water	$4 \times 10^{-6}$	1.5	Kidney
	Water	$5 \times 10^{-6}$	3	Bone
Argon-41	Air	$4 \times 10^{-8}$	0.5	Whole Body
Californium-249	Water	$4 \times 10^{-6}$	3	Bone
Californium-252	Water	$7 \times 10^{-6}$	1.5	GI (LLI)
	Water	$2 \times 10^{-5}$	3	Bone
Carbon-11	Air	$6 \times 10^{-8}$	0.5	Whole Body
Curium-242	Water	$2 \times 10^{-5}$	1.5	GI (LLI)
	Water	$2 \times 10^{-4}$	3	Bone
Curium-244	Water	$7 \times 10^{-6}$	3	Bone
Hydrogen-3 ( $\text{H}_2\text{O}$ )	Air	$2 \times 10^{-7}$	0.5	Whole Body
	Water	$3 \times 10^{-3}$	0.5	Whole Body
Iodine-131	Air	$1 \times 10^{-10}$	1.5	Thyroid
Krypton-85	Air	$3 \times 10^{-7}$	0.5	Whole Body
Neptunium-237	Water	$3 \times 10^{-6}$	3	Bone
Plutonium-238	Water	$5 \times 10^{-6}$	3	Bone
Plutonium-239	Water	$5 \times 10^{-6}$	3	Bone
Strontium-90	Water	$3 \times 10^{-7}$	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.<sup>13</sup> For carbon-11, the values are taken from reference 17.

- 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.

TABLE 19

Dose From Airborne Emissions, 1982  
(Millirems)

Nuclide	Pathway	Maximum Perimeter		Maximum Individual	
		Annual	50-Year	Annual	50-Year
Carbon-11	Submersion	0.02	0.02	0.01	0.01
Argon-41	Submersion	0.0002	0.0002	0.0001	0.0001
Krypton-85	Submersion	0.0005	0.0005	0.0002	0.0002

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 representative of the entire area within an 80 km (50 mi) radius.

The only location where radionuclides attributable to Argonne operations could be found in off-site water was Sawmill Creek below the wastewater outfall. The 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.

As indicated in Table 7, occasional 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  $1 \times 10^{-3}$  to  $2 \times 10^{-5}$  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.<sup>18</sup> For hydrogen-3, the EPA standard is  $2 \times 10^{-5}$   $\mu\text{Ci/ml}$  and for strontium-90, it is  $8 \times 10^{-9}$   $\mu\text{Ci/ml}$ . The concentrations in Table 22 correspond to 0.4% for hydrogen-3 and < 0.6% for strontium-90 of the EPA standards. No specific EPA standards exist for the transuranic nuclides.

TABLE 20

## 80 km Population Dose, 1982

Source	Man-Rems	
	Annual	50-Year
Carbon-11	0.246	0.246
Argon-41	0.009	0.009
Krypton-85	0.042	0.042
Natural	$7.63 \times 10^5$	-

TABLE 21

Radionuclide Concentrations and Dose Estimates  
for Sawmill Creek Water, 1982

Nuclide	Conc. <sub>10<sup>-9</sup></sub> (net avg.) μCi/ml	Dose mrem/year	Percent of Standard*
Hydrogen-3	86	0.014	0.0028
Strontium-90	< 0.05	< 0.5	< 0.017
Neptunium-237	0.0047	0.0047	0.00016
Plutonium-239	0.0050	0.0030	0.00010
Americium-241	0.0233	0.0087 (kidney) 0.014 (bone)	0.00058 0.00047
Curium-244	0.0022	0.0009 (bone)	0.00003
Californium-249	0.0012	0.0009	0.00003

\* DOE Order 5480.1 Standard.<sup>5</sup>

## 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 410 mrem/yr. Approximately 300 m (0.3 mi) south of the fence line (grid 6I), the measured dose averaged  $111 \pm 19$  mrem/yr, slightly above 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 0.010 mrem/yr, if the energy of the radiation were 0.66 MeV, and 0.051 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 27 mrem/yr, the nearest residents are 750 m (0.47 mi) to the north-northwest. The calculated dose at that location was about 0.05 mrem/yr.

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.<sup>5</sup> 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 combined from 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) and trace metals 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<sup>19</sup> as well as continuing previous studies. 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. Sampling for trace metals was discontinued, but the TSP studies were continued to measure general air quality. These samples were retained and will be examined for additional parameters in the future.

Twenty-four-hour samples were obtained using samplers equipped with constant flow regulation and time programming. Air flow rates were maintained at  $1.2 \text{ m}^3/\text{minute}$  and this flow was verified by comparison with a Roots meter which served as a primary air flow standard. State and Federal standards for TSP require that the annual geometric mean not exceed  $75 \text{ } \mu\text{g}/\text{m}^3$  for the primary standard or  $60 \text{ } \mu\text{g}/\text{m}^3$  for the secondary standard. The 24-hour standards are  $260 \text{ } \mu\text{g}/\text{m}^3$  and  $150 \text{ } \mu\text{g}/\text{m}^3$ , respectively. The results at 12M averaged  $48 \text{ } \mu\text{g}/\text{m}^3$  (with a geometric standard deviation of  $1.6 \text{ } \mu\text{g}/\text{m}^3$ ) while those at 8F averaged  $46 \text{ } \mu\text{g}/\text{m}^3$  (with a geometric standard deviation of  $1.7 \text{ } \mu\text{g}/\text{m}^3$ ). These results are about 10% lower than last year and very similar to previous years. The only sample that exceeded the 24-hour secondary standard was obtained on January 11, 1982, at 8F and contained  $216 \text{ } \mu\text{g}/\text{m}^3$ . The sample at 12M on this date was  $81 \text{ } \mu\text{g}/\text{m}^3$ . It appears that the source of the elevated TSP was upwind of the Laboratory. Samples obtained at both locations during the first two weeks of May were elevated, but did not exceed the 24-hour standards.

## 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 regulations. The appropriate standards are listed in the ILLINOIS POLLUTION CONTROL BOARD RULES AND REGULATIONS, Chapter 3.<sup>20</sup> 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 ( $\mu\text{g}$ ) per liter (l). Averages were calculated as described in Section III.A. Yearly averages are reported

TABLE 22

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

Constituent	State Standard		Detection Limit
	Stream	Effluent	
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	-
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

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 2.8 megaliters (0.75 million gallons) per day in 1982 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 are shown in Table 23. All of the average concentrations were below the State standards. The average value for mercury was 84% of the State standard and 27% of the samples exceeded this value. 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 and are probably a measurement of ambient levels, not due to Argonne operations.

TABLE 23  
 CHEMICAL CONSTITUENTS IN EFFLUENTS FROM ANL TREATMENT PLANT, 1982  
 (CONCENTRATIONS IN MICROGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.	PERCENT OF STANDARD (AVG.)	PERCENT EXCEEDING STATE STANDARD
ARSENIC	51	-	-	< 5	< 2.0	0
BARIUM	51	22 ± 9	4	166	1.1	0
BERYLLIUM	12	0.02 ± 0.01	0.01	0.03	-	-
CADMIUM	51	0.6 ± 0.1	0.3	2.2	0.4	0
CHROMIUM(III)	47	3.3 ± 0.2	< 3.0	6.0	0.3	0
CHROMIUM(VI)	47	3.3 ± 0.2	< 3.0	6.0	1.1	0
COPPER	51	26 ± 2	16	47	2.6	0
FLUORIDE	51	306 ± 34	196	960	2.0	0
IRON	51	258 ± 15	134	377	13	0
LEAD	51	2.5 ± 0.4	< 2.0	8.0	2.5	0
MANGANESE	51	28 ± 4	9	88	2.8	0
MERCURY	83	0.42 ± 0.06	0.05	1.84	84	27
NICKEL	51	9 ± 2	2	25	0.9	0
PH	245	-	6.7	8.6	-	0
SELENIUM	51	-	-	< 5	< 0.5	0
SILVER	51	2.6 ± 0.3	1.1	4.8	2.6	0
ZINC	51	106 ± 30	43	799	11	0

b. National Pollution Discharge Elimination System (NPDES)

A new and expanded NPDES permit program was begun in March, 1981. The permit provides for 10 sampling locations, which are shown in Figure 7. 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 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 1981 or 1982. 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  $\mu\text{g}/\text{l}$  for mercury was exceeded 80% of the time by various amounts. In December, 1981, new effluent regulations were adopted by the Illinois Pollution Control Board which provides a monthly limit, by exemption, of 3.0  $\mu\text{g}/\text{l}$  for mercury discharges from research facilities. If this limit were applied to the NPDES parameters, all the results would be in compliance. Occasional 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. However, at location 005, the suspended solids were usually above the limit of 15  $\text{mg}/\text{l}$ . The source of this problem is being investigated. At locations 007 and 009, the pH was usually above the upper limit of 9.0.

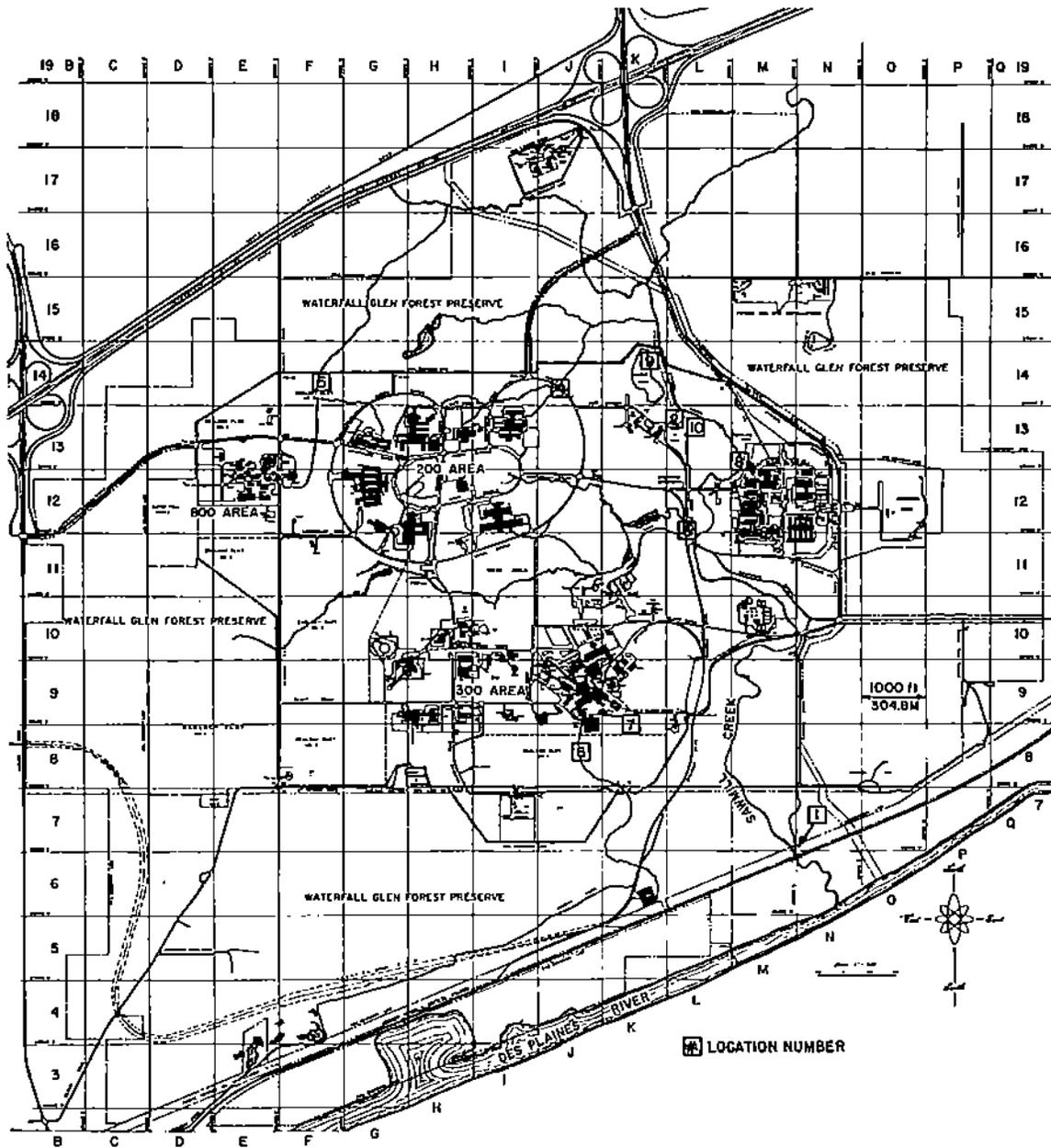


Fig. 7. NPDES Permit Locations

TABLE 24

 ANL National Pollution Discharge Elimination  
 System Data Summary, Location 001 (7M), 1982

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.75	-	-	MGD	0	Cont.	N/A
	Permit Condition	0.84	-	-			Cont.	N/A
B.O.D.	Reported	4.6	-	72	mg/l	0	1/7	24 Hr.
	Permit Condition	30	-	75		-	1/7	24 Hr.
Suspended Solids	Reported	5	-	20	mg/l	0	2/7	24 Hr.
	Permit Condition	30	-	75		-	1/7	24 Hr.
Fecal Coliform Bacteria	Reported	-	-	6800	Number/ 100 ml	1	1/7	Grab
	Permit Condition	200	-	400		-	1/7	Grab
Residual Chlorine	Reported	0.20	-	-	mg/l	0	1/7	Grab
	Permit Condition	0.75	-	-		-	1/7	Grab
Mercury	Reported	0.001	-	0.0026	mg/l	41	1/7	Grab
	Permit Condition	-	-	0.0005		-	1/7	Grab
Nitrate (N)	Reported	5.6	-	11.1	mg/l	-	1/7	24 Hr.
	Permit Condition	-	-	-		-	-	Grab
pH	Reported	-	7.0	9.2	Units	1	1/7	24 Hr.
	Permit Condition	-	6.0	9.0		-	1/7	Grab
Gross Beta	Reported	17.6	-	30.2	pCi/l	0	1/7	Grab
	Permit Condition	-	-	100		-	1/7	Grab
Radium-226	Reported	0.53	-	2.30	pCi/l	3	1/7	Grab
	Permit Condition	-	-	1.0		-	1/7	Grab
Strontium-90	Reported	0.30	-	1.11	pCi/l	0	1/7	Grab
	Permit Condition	-	-	2.0		-	1/7	Grab

TABLE 25

 ANL National Pollution Discharge Elimination  
 System Data Summary, Location 002 (13L), 1982

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.13	-	-	MGD	0	1/30	Single Approximation
	Permit Condition	-	-	-		-	1/30	
Total Suspended Solids	Reported			1110	mg/l	1	1/30	Grab
	Permit Condition	-	-	15		-	1/30	Grab
Temperature	Reported	All Within Range			°F	0	1/30	Grab
	Permit Condition	Natural Temp. +5°				-	1/30	Grab
pH	Reported	-	6.0	10.1	Units	1	1/30	Grab
	Permit Condition	-	6.5	9.0		-	1/30	Grab

TABLE 26

ANL National Pollution Discharge Elimination  
System Data Summary, Location 003 (12L), 1982

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.83	0.2	3.3	MGD	0	Single Reading	N/A
	Permit Condition	-	-	-				
Total Suspended Solids	Reported	9.2	0.9	26.8	mg/l	1	1/30	Grab
	Permit Condition	-	-	15				
Total Chlorine Residual	Reported	-	-	0.10	mg/l	0	1/30	Grab
	Permit Condition	-	-	0.75				
Total Dissolved Solids	Reported	665	432	1360	mg/l	0	1/30	Grab
	Permit Condition	-	-	3500				
Temperature	Reported	All Results Within Limits			°F	-	1/30	Grab
	Permit Condition	Natural Temp. +5°						
pH	Reported	-	7.9	8.5	Units	0	1/30	Grab
	Permit Condition	-	6.0	9.0				

TABLE 27

ANL National Pollution Discharge Elimination  
System Data Summary, Location 004 (14J), 1982

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.1	0.03	0.4	MGD	0	Single Approximation	
	Permit Condition	-	-	-		-	Single Apoxroximation	
Suspended Solids	Reported	5.6	1.4	9.0	mg/l	0	1/30	Grab
	Permit Condition	-	-	15		-	1/30	Grab
Total Dissolved Solids	Reported	625	378	2090	mg/l	0	1/30	Grab
	Permit Condition	-	-	3500		-	1/30	Grab
Temperature	Reported	All Results Within Limits				0	1/30	Grab
	Permit Condition	Natural Temp. +5°			°F	-	1/30	Grab
pH	Reported	-	7.5	8.1	Units	0	1/30	Grab
	Permit Condition	-	6.0	9.0		-	1/30	Grab

TABLE 28

 ANL National Pollution Discharge Elimination  
 System Data Summary, Location 005 (14F), 1982

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.12	0.04	0.27	MGD	0	Single Approximation	
	Permit Condition	-	-	-		-	Single Approximation	
Suspended Solids	Reported	27.2	7.4	50.4	mg/l	8	1/30	Grab
	Permit Condition	-	-	15		-	1/30	Grab
Fat, Oil, and Grease	Reported	-	-	< 5	mg/l	0	1/30	Grab
	Permit Condition	-	-	30		-	1/30	Grab
Temperature	Reported	All Results Within Limits				-	1/30	Grab
	Permit Condition	Natural Temp. +5°			°F	-	1/30	Grab
pH	Reported	-	7.4	7.9	Units	0	1/30	Grab
	Permit Condition	-	6.0	9.0		-	1/30	Grab

TABLE 29

ANL National Pollution Discharge Elimination  
System Data Summary, Location 006 (8J), 1982

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.13	0.02	0.65	MGD	0	Single Approximation	Single Approximation
	Permit Condition	-	-	-				
Suspended Solids	Reported	9.8	1.7	26.6	mg/l	3	1/30	Grab
	Permit Condition	-	-	15				
pH	Reported	-	6.7	8.4	Units	0	1/30	Grab
	Permit Condition	-	6.0	9.0				

TABLE 30

ANL National Pollution Discharge Elimination  
System Data Summary, Location 007 (9K), 1982

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.02	0.01	0.06	MGD	0	Single Approximation	Single Approximation
	Permit Condition	-	-	-				
pH	Reported	-	8.2	10.5	Units	7	1/30	Grab
	Permit Condition	-	6.0	9.0				

TABLE 31

ANL National Pollution Discharge Elimination  
System Data Summary, Location 008 (13M), 1982

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.02	-	0.02	MGD	0	Single Approximation	Single Approximation
	Permit Condition	-	-	-				
pH	Reported	-	7.8	9.1	Units	1	1/30	Grab
	Permit Condition	-	6.0	9.0				

TABLE 32

ANL National Pollution Discharge Elimination  
System Data Summary, Location 009 (14K), 1982

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.03	0.01	0.06	MGD	0	Single Approximation	Single Approximation
	Permit Condition	-	-	-				
Suspended Solids	Reported	2.6	0.10	6.0	mg/l	0	1/30	Grab
	Permit Condition	-	-	15				
pH	Reported	-	8.8	9.6	Units	7	1/30	Grab
	Permit Condition	-	6.0	9.0				

This high pH is probably the result of the use of alkaline water softening compounds and/or stagnant water conditions. The high pH water has no impact on the receiving stream 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. The average level for ammonia nitrogen upstream is 2.0 times the State of Illinois standard and individual samples exceeded the standard 46% of the time. The downstream sample averaged 1.7 times the State standard and exceeded this standard 46% of the time. All of the samples obtained downstream that violated the standard were due to contamination from the upstream sewage treatment plant. The dissolved oxygen levels obtained during 1982 were all above the State minimum of 5 mg/l. The total dissolved solids above the outfall exceeded the State standard frequently, and increased the solids content of the downstream sample. Similar results for these constituents were obtained in 1981.

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.

Individual samples for arsenic, barium, cadmium, chromium, fluoride, lead, nickel, selenium, silver, and zinc did not exceed the State standards. The levels of mercury averaged 33% of the State limit and ex-

TABLE 33

## Sawmill Creek - Effect of Sanitary Waste, 1982

Constituent	Location*	No. of Samples	Concentration (mg/l)			Avg. Percent of Standard	Percent Exceeding State Standard
			Avg.	Min.	Max.		
Ammonia	7M (up)	50	3.0 $\pm$ 0.9	0.1	13.6	200	46
Nitrogen	7M (down)	50	2.6 $\pm$ 0.8	0.1	12.8	170	46
Dissolved Oxygen	7M (up)	50	10.2 $\pm$ 0.7	5.4	15.1	-	-
	7M (down)	50	10.1 $\pm$ 0.6	5.9	15.6	-	-
pH	7M (up)	50	-	7.6	8.9	-	-
	7M (down)	50	-	7.6	8.9	-	-
Sulfate	7M (up)	12	150 $\pm$ 22	89	194	30	0
	7M (down)	12	152 $\pm$ 21	87	195	30	0
Temperature	7M (up)	50	15.0 $\pm$ 2.3	0.3	27.5	-	-
	7M (down)	50	15.0 $\pm$ 2.2	1.6	27	-	-
Total Dissolved Solids	7M (up)	49	1256 $\pm$ 132	536	2022	126	59
	7M (down)	49	1149 $\pm$ 108	505	1826	115	57

\* 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.

TABLE 34  
 CHEMICAL CONSTITUENTS IN SAKHILL CREEK LOCATION 7H, \* 1982  
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.	PERCENT OF STANDARD (AVG.)	PERCENT EXCEEDING STATE STANDARD
ARSENIC	51	6 ± 1	< 5	17	0.6	0
BARIUM	51	63 ± 11	10	163	1.3	0
BERYLLIUM	12	0.09 ± 0.05	0.01	0.27	-	-
CADMIUM	51	1.1 ± 0.1	0.3	3.6	2.1	0
CHLORIDE **	21	394 ± 60	180	680	-	-
CHROMIUM(III)	51	20 ± 10	3	204	2.0	0
CHROMIUM(VI)	51	4 ± 1	< 3	37	7.5	0
COPPER	50	27 ± 8	6	138	136	40
FLUORIDE	51	391 ± 32	178	644	28	0
IRON	51	1600 ± 600	170	12100	160	45
LEAD	51	12 ± 3	1	54	12	0
MANGANESE	51	231 ± 54	40	891	23	0
MERCURY	83	0.16 ± 0.03	< 0.05	0.88	33	4
NICKEL	51	14 ± 3	1	53	1.4	0
PH	247	-	7.2	8.5	-	0
SELENIUM	51	-	-	< 5	< 0.5	0
SILVER	51	1.3 ± 0.2	0.3	4.1	26	0
ZINC	51	77 ± 18	10	326	7.7	0

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

\*\* CONCENTRATION IN MILLIGRAM/LITER.

ceeded this limit 4% of the time. 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 40% of the time, but they are not due to ANL operations since the average stream and effluent levels are the same and 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. Two of the samples had very high levels of suspended material, as evidenced by iron levels of 20.3 and 5.4 mg/l. In no case was there any indication that hexavalent chromium, mercury, or zinc levels were affected by the Argonne effluent.

TABLE 35

Chemical Constituents in the Des Plaines River, 1982

Constituent	Location *	No. of Samples	Concentration (mg/l)		
			Avg.	Min.	Max.
Chromium(VI)	A	12	-	-	< 0.01
	B	24	-	-	< 0.01
Iron	A	12	3.4 + 3.6	0.4	20.3
	B	24	1.3 + 0.4	0.4	5.4
Mercury	A	12	-	-	< 0.0001
	B	24	-	-	< 0.0001
Zinc	A	12	0.11 + 0.10	0.02	0.60
	B	24	0.05 + 0.01	0.02	0.17

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

### 3. Solid Waste

Hazardous solid waste, both radioactive and nonradioactive, is not permanently stored at ANL. Such wastes are packaged in accordance with DOE and U. S. Department of Transportation criteria and shipped to approved repositories. Non-hazardous waste is buried in an on-site sanitary landfill located on the western edge of the site (Figures 1 and 8). This site operates under Illinois EPA permit No. 1981-29-OP. The site is further described in Reference 2.

The effect of this site on adjacent ground water can be estimated by analyses of water obtained from a series of monitoring wells, also shown in Figures 1 and 8. Four water samples were obtained from Wells No. 1, 3, 4, and 7a. Because of reconstruction and resulting sampling problems, only two samples were obtained from Well No. 6. Wells No. 2, 5, and 7b were not sampled during 1982.

The wells were analyzed for many of the water quality parameters listed in Table 22. Results appear in Tables 36 to 40. In general, results appear to be typical for subsurface water. Wells No. 4, 6, and 7a had elevated levels of manganese. Manganese levels in wells 4 and 6 were fairly constant during the sampling period while both manganese and iron levels in well 7a decreased during the course of the year. This could indicate infiltration of surface water into this relatively shallow well. The levels of chloride in Well No. 1 are substantially higher than the other wells. Well No. 1 is located near a major road and the salt used for snow and ice removal is a likely source of the high chloride concentrations. The concentrations of the other constituents are believed to be normal.

From 1969 through 1978, substantial quantities of liquid organic waste were disposed of into an open "French drain" located in the northeast corner of the landfill. Among the many organic waste materials discarded were acetone, benzene, and xylene. Acetone was considered to be a useful tracer of movement since it is miscible with water and large amounts (about 2,600 liters) had been added. Benzene and xylene were taken as representatives

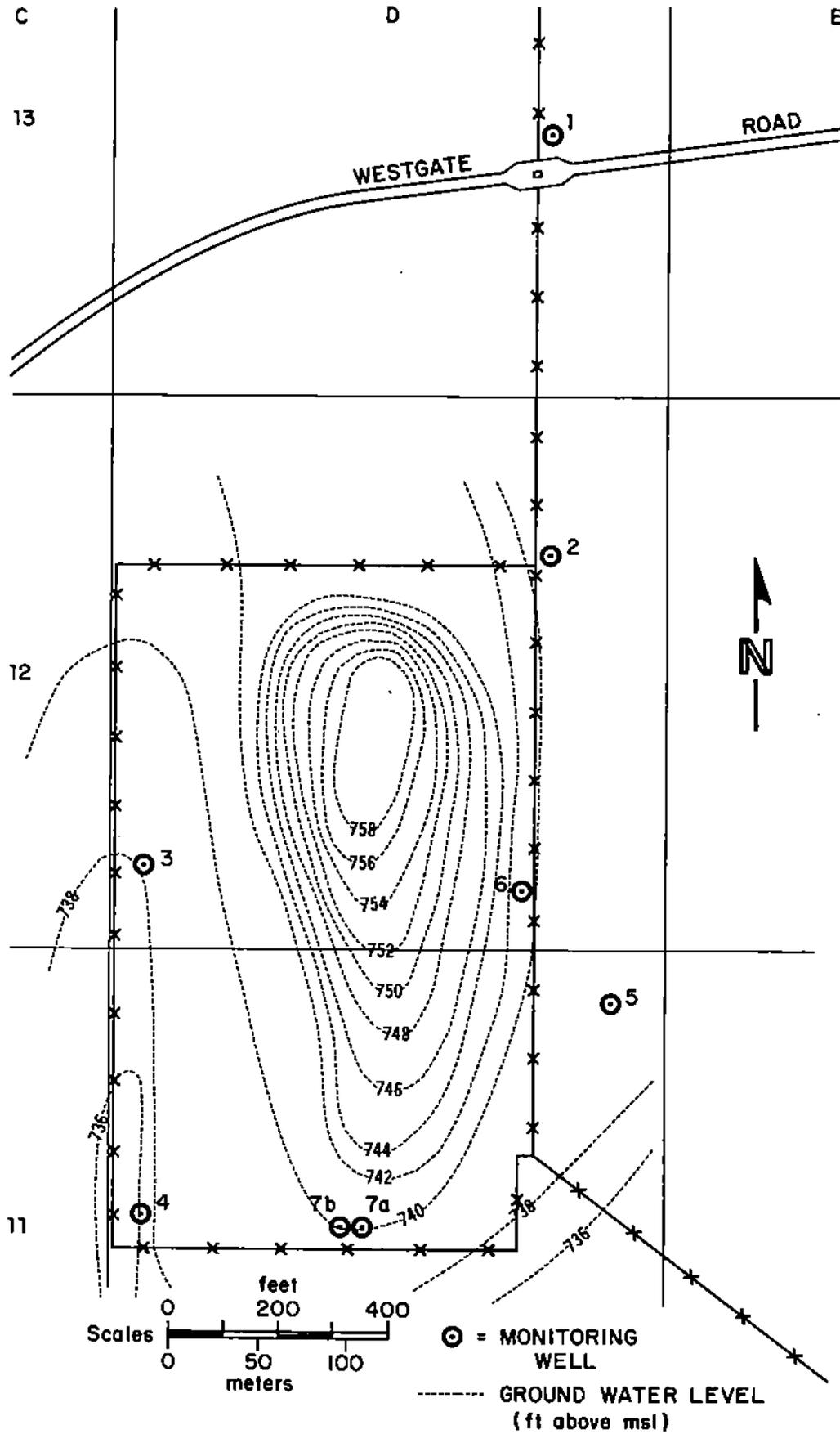


Fig. 8. Monitoring Well Locations for Landfill

TABLE 36  
 CHEMICAL CONSTITUENTS AT ANL LANDFILL  
 HELL 1  
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	3	-	-	< 5
BARIUM	3	193 ± 95	144	247
BENZENE	2	-	-	< 30
CADMIUM	3	1.10 ± 1.29	0.30	1.60
CHLORIDE **	2	790 ± 344	710	870
CHROMIUM(III)	3	-	-	< 3
CHROMIUM(VI)	3	-	-	< 3
COPPER	3	3 ± 2	1	4
CYANIDE	3	-	-	< 10
DISSOLVED SOLID **	3	1940 ± 310	1810	2130
FLUORIDE	3	139 ± 38	122	162
IRON	3	424 ± 578	100	728
LEAD	3	3 ± 3	1	5
MANGANESE	3	224 ± 136	148	296
MERCURY	3	0.16 ± 0.34	< 0.05	0.37
NICKEL	3	15 ± 18	8	27
PH	3	-	7.1	7.3
SELENIUM	3	-	-	< 5
SILVER	3	1.13 ± 0.56	0.80	1.40
XYLENE	2	-	-	< 20
ZINC	3	15 ± 16	10	25

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 \*\*  
 CONCENTRATION IN MILLIGRAM/LITER.

TABLE 37  
 CHEMICAL CONSTITUENTS AT ANL LANDFILL  
 WELL 3  
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ARSENIC	2	34 ± 4	33	35
BARIUM	3	280 ± 212	205	413
BENZENE	2	-	-	< 30
CADMIUM	3	0.47 ± 0.38	0.30	0.70
CHLORIDE**	2	26 ± 19	21	30
CHROMIUM(III)	3	-	-	< 3
CHROMIUM(VI)	3	-	-	< 3
COPPER	3	3 ± 1	3	3
CYANIDE	3	-	-	< 10
DISSOLVED SOLID**	3	762 ± 110	704	824
FLUORIDE	3	127 ± 21	114	136
IRON	3	2540 ± 1300	2260	3670
LEAD	3	2 ± 2	1	3
MANGANESE	3	254 ± 79	205	284
MERCURY	3	-	-	< 0.05
NICKEL	3	10 ± 17	3	20
PH	3	-	6.8	7.0
SELENIUM	3	-	-	< 5
SILVER	3	< 0.20	< 0.20	0.30
XYLENE	2	-	-	< 20
ZINC	3	17 ± 22	10	31

\*\*  
 CONCENTRATION IN MILLIGRAM/LITER.

TABLE 38  
 CHEMICAL CONSTITUENTS AT ANL LANDFILL  
 WELL 4  
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ACETONE	1	-	-	< 20
ARSENIC	1	-	-	< 5
BARIIUM	3	243 ± 186	170	359
BENZENE	2	53 ± 97	< 30	75
CADMIUM	3	0.37 ± 0.70	< 0.20	0.80
CHLORIDE **	2	210 ± 293	142	278
CHROMIUM(III)	3	-	-	< 3
CHROMIUM(VI)	3	-	-	< 3
COPPER	3	3 ± 4	2	5
CYANIDE	3	-	-	< 10
DISSOLVED SOLID **	3	1400 ± 430	1180	1650
FLUORIDE	3	174 ± 26	158	184
IRON	3	737 ± 729	280	986
LEAD	3	3 ± 4	2	5
MANGANESE	3	1920 ± 1810	1320	3050
MERCURY	3	-	-	< 0.05
NICKEL	3	19 ± 13	13	27
PH	3	-	6.9	7.0
SELENIUM	3	-	-	< 5
SILVER	3	0.43 ± 0.38	< 0.20	0.60
XYLENE	2	-	-	< 20
ZINC	3	16 ± 20	10	29

\*\*  
 CONCENTRATION IN MILLIGRAM/LITER.

TABLE 39  
 CHEMICAL CONSTITUENTS AT ANL LANDFILL  
 HELL 6  
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ACETONE	1	-	-	< 20
ARSENIC	1	-	-	< 5
BARIUM	2	155 ± 108	130	180
BENZENE	2	-	-	< 30
CADMIUM	2	-	-	< 0.20
CHLORIDE**	2	91 ± 65	76	106
CHROMIUM(III)	2	-	-	< 3
CHROMIUM(VI)	2	-	-	< 3
COPPER	2	1 ± 2	1	2
CYANIDE	2	-	-	< 10
DISSOLVED SOLID**	2	1270 ± 300	1200	1340
FLUORIDE	2	96 ± 34	88	104
IRON	2	15300 ± 4020	14400	16300
LEAD	2	2 ± 2	2	3
MANGANESE	2	3640 ± 1140	3380	3910
MERCURY	2	0.16 ± 0.26	0.10	0.22
NICKEL	2	16 ± 11	13	18
PH	2	-	6.5	6.6
SELENIUM	2	-	-	< 5
SILVER	1	-	-	0.60
XYLENE	2	-	-	< 20
ZINC	2	31 ± 90	10	52

\*\*  
 CONCENTRATION IN MILLIGRAM/LITER.

TABLE 40  
 CHEMICAL CONSTITUENTS AT ANL LANDFILL  
 WELL 7A  
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.
ACETONE	1	-	-	< 20
ARSENIC	2	-	-	< 5
BARIIUM	3	174 ± 37	151	187
BENZENE	2	47 ± 71	< 30	63
CADMIUM	3	0.37 ± 0.28	< 0.20	0.50
CHLORIDE **	2	23 ± 24	17	28
CHROMIUM(III)	2	-	-	< 3
CHROMIUM(VI)	3	-	-	< 3
COPPER	3	5 ± 2	4	5
CYANIDE	3	-	-	< 10
DISSOLVED SOLID **	2	498 ± 75	480	515
FLUORIDE	3	123 ± 11	116	128
IRON	3	-	-	< 100
LEAD	3	1 ± 1	1	2
MANGANESE	3	528 ± 940	225	1120
MERCURY	3	0.29 ± 0.25	0.17	0.44
NICKEL	3	5 ± 3	3	6
PH	3	-	7.3	7.4
SELENIUM	2	-	-	< 5
SILVER	3	< 0.20	< 0.20	0.30
XYLENE	2	-	-	< 20
ZINC	3	20 ± 32	10	40

\*\*  
 CONCENTRATION IN MILLIGRAM/LITER.

of less soluble materials and substantial quantities were disposed of at this site. Samples from Wells Nos. 1, 3, 4, 6, and 7a were analyzed for benzene and xylene. One sample each from Wells Nos. 4, 6, and 7a was analyzed for acetone. One sample each from Well No. 4 and Well No. 7a indicated benzene levels about twice background. All other results were below detection limits. In 1983, quarterly samples will be obtained from all wells and they will be analyzed for these and other constituents using more sensitive techniques. These preliminary results indicate, however, that there has not been a significant movement from the disposal site to the test wells.

## IV. UNUSUAL OCCURRENCES

A. Loss of Pond Water

In 1967, an excavation was dug (location 10G in Figure 1) for the foundation of the proposed Argonne Advanced Research Reactor (A<sup>2</sup>R<sup>2</sup>). The project was cancelled and the approximately 40 meters (130 foot) diameter by 13 meter (40 foot) deep hole eventually filled with water, creating a pond. The excavation is located about 100 meters Northwest of the CP-5 reactor (location 9H in Figure 1). During operation, the CP-5 reactor exhausted about 1 Ci/day of tritiated water vapor. As a result of rainout and exchange, some of the tritium accumulated in the pond. Monitoring of the water and fish in the pond began in 1973, and the results are given in Table 41 and illustrated in Figure 9. The tritiated water content increased until 1978 and then decreased as the tritium released from CP-5 decreased. Operation of the CP-5 reactor was terminated on September 28, 1979, and the heavy water, which contained the tritium, was removed in January 1980.

Over a period of a few days early in March 1982, the water level fell from about 8 meters (26 feet) above the excavation floor to 2 meters (6 feet) above the floor. It is estimated that about 7.5 megaliters (2 million gallons) of water disappeared by a subsurface route. The tritium concentration of this water was  $3.4 \times 10^{-6}$   $\mu\text{Ci/ml}$ , so the total amount of tritium lost was about 26 mCi.

A reasonable explanation for the water loss can be obtained from a knowledge of the geohydrology of the area.<sup>21</sup> The excavation is in generally low permeability glacial despoits which extend from about 18 to 27 meters (60 to 90 feet) below the ground surface at the Laboratory site. Under this till is the Niagara Dolomite, which ranges in thickness from 60 to 69 meters (200 to 225 feet) in this area. The bottom layer of the glacial deposit probably contains permeable gravel layers in hydrological contact with the dolomite. The existence of solutionally-enlarged openings in the joints and bedding planes of the Niagara Dolomite is well documented. As a result of the hydrostatic pressure from the water in the

excavation, hydrologic piping of the till could occur, providing a conduit for increasingly rapid drainage of water from the excavation into a cavity in the Niagara Dolomite. Bottom sediment and debris probably plugged the channel before all the water drained from the excavation. The piezometric surface of the dolomite aquifer indicates that after the water reached the dolomite, it could flow south toward the springs and seep along the Des Plaines River or toward the drawdown cone around the four domestic water supply wells northeast of the excavation.

TABLE 41

Average Tritiated Water Content of  
Water and Fish from A<sup>2</sup>R<sup>2</sup> Excavation  
(Concentrations in 10<sup>-6</sup>  $\mu$ Ci/ml)

Year	No. of Samples	Water	No. of Samples	Fish <sup>*</sup>
1973	2	8.1	1	6.0
1974	3	6.8	3	5.9
1975	3	9.1	1	8.8
1976	2	11.1	2	8.9
1978	1	11.4	1	11.0
1979	1	8.7	1	9.0
1980	1	8.4	2	7.8
1981	4	4.3	-	-
1982			-	-
Jan.-June	6	3.7	-	-
July-Dec.	5	2.1	-	-

\* Concentration in tissue water.

Samples from wells northeast of the pond gave no evidence of migration in that direction. These wells were the four ANL domestic water wells located in grids 12L, 11M, 13K, and 14I and a water-level monitoring well in grid 10J (Figure 1). South of the excavation, samples were collected from Well No.9 near Kearney and Bluff Roads (location 9F), Well 181 and Well 185 at the ANL Meteorological Station (location 8EF), and a surface seep in the Des Plaines River valley (location 4EF). The results of the tritium

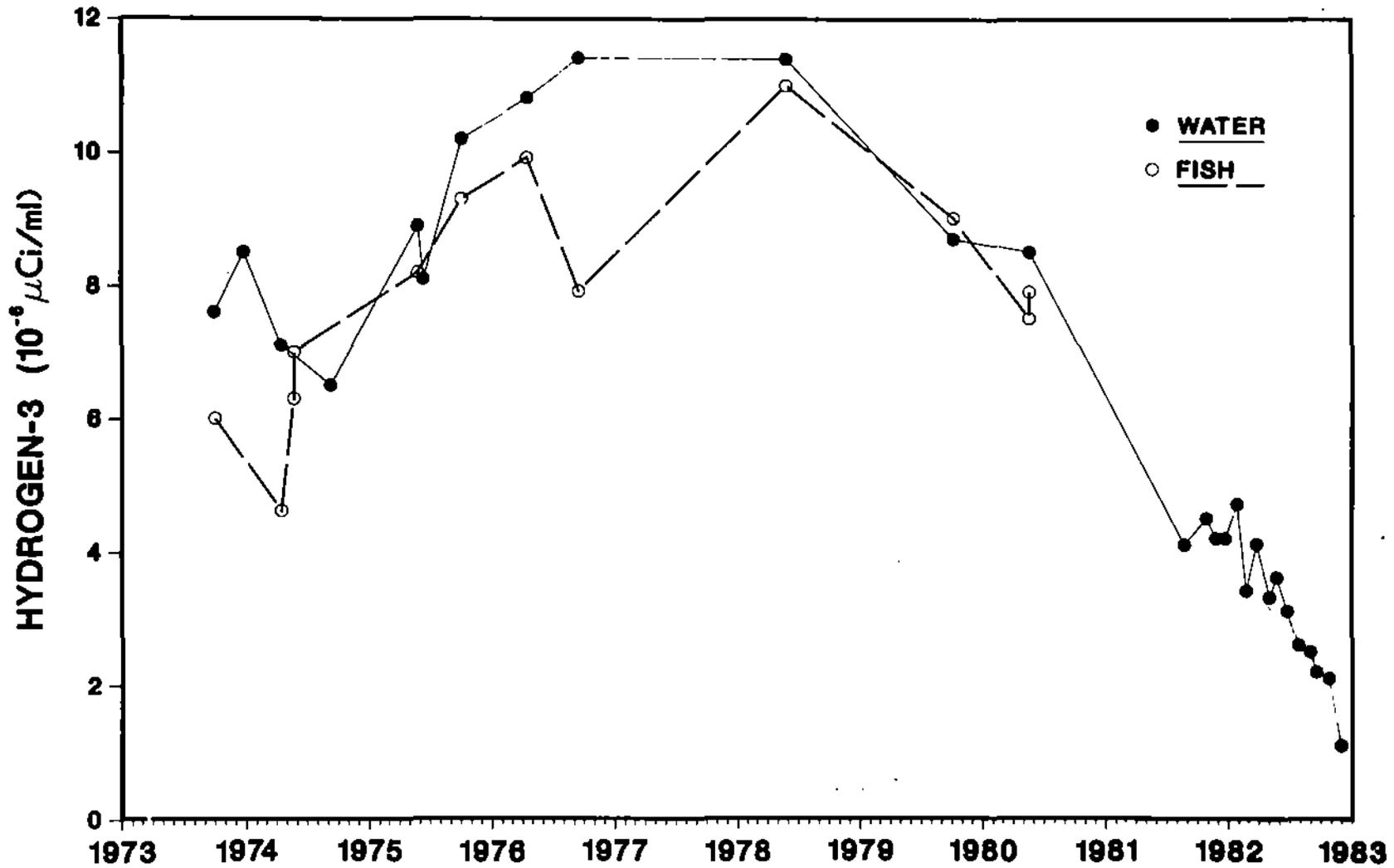


Fig. 9. Tritium Content of Water and Fish from A<sup>2</sup>R<sup>2</sup> Excavation, 1973-1982

analyses of water samples collected at these locations after the event are collected in Table 42. No previous data is available from Well No.9, while prior analyses of the wells at 181 and 185 and the 4EF seep were less than the detection limit of  $0.1 \times 10^{-6}$   $\mu\text{g/ml}$  for tritiated water. All the wells collect water from the Niagara Dolomite. Well No. 9 is a water-level monitoring well; Wells 181 and 185 are used for fire protection and general laboratory purposes in Buildings 181 and 185 and are not part of the principal ANL domestic water supply system.

Based on the data in Table 42, the water from the excavation appears to have moved in a southwest direction. The concentrations at Well #9, about 450 meters (1,500 feet) southwest of the excavation, were significantly elevated a few days after the event and reached a maximum about ten days after the water loss. The maximum concentration found was about 8% of the EPA drinking water standard for tritium (although water from this well is not consumed). The data do not allow a conclusion as to whether the pond water is moving as a subsurface pulse, whether the tritiated water is being diluted by other water, or both.

In the fall of 1982, the excavation began to be filled with concrete and asphalt rubble from building demolition operations. This should prevent the future occurrence of this type of event.

TABLE 42

Tritiated Water Content of Wells  
Near A<sup>2</sup>R<sup>2</sup> Excavation, 1982  
(Concentrations in 10<sup>-6</sup>  $\mu$ Ci/ml)

Date Collected	Well #9	Well D181	Well D185	4EF Seep
March 11	1.2 $\pm$ 0.2	< 0.1	< 0.1	< 0.1
March 19	1.7 $\pm$ 0.1	-	-	-
March 26	1.4 $\pm$ 0.1	-	-	-
April 9	1.0 $\pm$ 0.1	-	-	-
April 16	1.0 $\pm$ 0.1	-	-	-
April 26	0.6 $\pm$ 0.1	-	-	-
May 14	0.7 $\pm$ 0.1	-	-	-
May 25	0.7 $\pm$ 0.1	< 0.1	< 0.1	0.69 $\pm$ 0.11
June 25	0.7 $\pm$ 0.1	< 0.1	< 0.1	0.10 $\pm$ 0.10
July 9	0.5 $\pm$ 0.1	-	-	-
July 26	0.8 $\pm$ 0.1	< 0.1	< 0.1	< 0.1
August 30	0.5 $\pm$ 0.1	< 0.1	< 0.1	< 0.1
September 17	0.6 $\pm$ 0.1	< 0.1	< 0.1	< 0.1
October 25	0.6 $\pm$ 0.1	< 0.1	< 0.1	< 0.1
November 29	0.7 $\pm$ 0.1	< 0.1	< 0.1	0.13 $\pm$ 0.10

## 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 43 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,<sup>5</sup> 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 (except for iodine-131, for which the soluble form was chosen as a more conservative standard).

TABLE 43  
 Concentration Guides and Detection Limits  
 ( $\mu\text{Ci}/\text{ml}$ )

Nuclide or Activity	Concentration Guide		Detection Limit	
	Water	Air	Water	Air
Americium-241	$4 \times 10^{-6}$	$4 \times 10^{-12}$	$1 \times 10^{-12}$	$5 \times 10^{-19}$
Antimony-125	-	$9 \times 10^{-10}$	-	$5 \times 10^{-16}$
Argon-41	-	$4 \times 10^{-8}$	-	$1.5 \times 10^{-8}$
Barium-140	$3 \times 10^{-5}$	$1 \times 10^{-9}$	$2 \times 10^{-9}$	$5 \times 10^{-16}$
Beryllium-7	-	$4 \times 10^{-8}$	-	$5 \times 10^{-15}$
Californium-249	$4 \times 10^{-6}$	-	$1 \times 10^{-12}$	-
Californium-252	$7 \times 10^{-6}$	-	$1 \times 10^{-12}$	-
Cerium-141	-	$5 \times 10^{-9}$	-	$5 \times 10^{-16}$
Cerium-144	-	$2 \times 10^{-10}$	-	$1 \times 10^{-15}$
Cesium-137	$2 \times 10^{-5}$	$5 \times 10^{-10}$	-	$5 \times 10^{-16}$
Cobalt-60	-	$3 \times 10^{-10}$	-	$1 \times 10^{-16}$
Curium-242	$2 \times 10^{-5}$	-	$1 \times 10^{-12}$	-
Curium-244	$7 \times 10^{-6}$	-	$1 \times 10^{-12}$	-
Hydrogen-3	$3 \times 10^{-3}$	$2 \times 10^{-7}$	$1 \times 10^{-7}$	$1 \times 10^{-13}$
Iodine-131	$3 \times 10^{-7}$	$1 \times 10^{-10}$	$3 \times 10^{-9}$	$5 \times 10^{-15}$
Krypton-85	-	$3 \times 10^{-7}$	-	-
Neptunium-237	$3 \times 10^{-6}$	-	$1 \times 10^{-12}$	-
Plutonium-238	$5 \times 10^{-6}$	$1 \times 10^{-12}$	$1 \times 10^{-12}$	$1 \times 10^{-19}$
Plutonium-239	$5 \times 10^{-6}$	$1 \times 10^{-12}$	$5 \times 10^{-13}$	$1 \times 10^{-19}$
Radium-226	$3 \times 10^{-8}$	-	$1 \times 10^{-13}$	-
Ruthenium-103	-	$3 \times 10^{-9}$	-	$5 \times 10^{-16}$
Ruthenium-106	-	$2 \times 10^{-10}$	-	$1 \times 10^{-15}$
Strontium-89	$3 \times 10^{-6}$	$1 \times 10^{-9}$	$2 \times 10^{-9}$	$1 \times 10^{-16}$
Strontium-90	$3 \times 10^{-7}$	$2 \times 10^{-10}$	$2.5 \times 10^{-10}$	$1 \times 10^{-17}$
Thorium-228	-	$2 \times 10^{-13}$	-	$1 \times 10^{-18}$
Thorium-230	-	$3 \times 10^{-13}$	-	$1 \times 10^{-18}$
Thorium-232	-	$1 \times 10^{-12}$	-	$1 \times 10^{-18}$
Uranium-234	-	$4 \times 10^{-12}$	-	$1 \times 10^{-19}$
Uranium-235	-	$4 \times 10^{-12}$	-	$1 \times 10^{-19}$
Uranium-238	-	$5 \times 10^{-12}$	-	$1 \times 10^{-19}$
Uranium - natural*	$4 \times 10^{-5}$	$4 \times 10^{-12}$	$2 \times 10^{-10}$	$2 \times 10^{-17}$
Zirconium-95	-	$1 \times 10^{-9}$	-	$5 \times 10^{-16}$
Alpha**	$3 \times 10^{-6}$	$1 \times 10^{-10}$	$2 \times 10^{-10}$	$2 \times 10^{-16}$
Beta**	$1 \times 10^{-7}$	$1 \times 10^{-13}$	$1 \times 10^{-9}$	$5 \times 10^{-16}$

\* 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.

## 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 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. The DOE Environmental Measurements Quality Assurance Program (DOE-EML-QAP) was not operative during 1982. Results of our participation in the EPA-QA program during 1982 are given in Table 44. 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). For most analyses for which the differences are large (> 20%), the concentrations were quite low and the differences were within the measurement uncertainties.

TABLE 44

## Summary of EPA-QA Samples, 1982

Type of Sample	Analysis	Number Analyzed	Average Difference from Added
Air Filter	Total Alpha	1	12%
	Total Beta	1	10%
	Cesium-137	1	16%
Milk	Potassium-40	2	9%
	Cobalt-60	1	7%
	Strontium-89	1	31%
	Strontium-90	2	35%
	Iodine-131	2	32%
	Cesium-137	2	5%
Soil	Lead-210	1	31%
	Radium-226	1	9%
	Uranium-238	1	27%
Water	Total Alpha	1	42%
	Total Beta	1	8%
	Hydrogen-3	1	1%
	Chromium-51	2	5%
	Cobalt-60	2	1%
	Zinc-65	2	5%
	Strontium-89	2	10%
	Strontium-90	2	12%
	Ruthenium-106	1	10%
	Iodine-131	3	9%
	Cesium-134	3	8%
	Cesium-137	3	5%
	Radium-226	2	37%
	Radium-228	2	11%
	Total Uranium	1	21%
	Plutonium-239	1	10%

## b. Penetrating Radiation

Our laboratory participated in the Sixth International Environmental Dosimeter Intercomparison Project conducted by the School of Public Health, University of Texas (Houston) and the DOE Environmental Measurements Laboratory (EML). The results are not yet available. Our results for the Fifth Intercomparison, conducted in 1980, are reproduced below from our 1981 report.<sup>4</sup>

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 and one set at the end of the field exposure period, and one 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 mR, were:

	<u>Field Exposure</u>	<u>Laboratory Exposure</u>	
		<u>Begin</u>	<u>End</u>
School of Public Health	30.0 ± 3.2	75.2 ± 3.8	88.4 ± 4.4
ANL Value			
calcium fluoride	26.9 ± 1.3	67.5 ± 2.5	83.8 ± 2.5
lithium fluoride	31.0 ± 1.0	79.2 ± 2.6	93.8 ± 3.0

The uncertainties listed are the standard deviations as estimated by the School of Public Health for their values and by us for our results. All results agree with the standard or accepted School of Public Health values 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 3.1 mR ± 3.5 mR (10.3% ± 11.7%) for calcium fluoride and 1.0 mR ± 3.4 mR (3.3% ± 11.3%) for lithium fluoride.

## 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 45) were in good agreement with the reference values.

TABLE 45  
Summary of Quality Assurance Studies

Constituent	Average Difference From Reference Value
Cadmium	4% (16)
Lead	2% (16)
Zinc	2% (16)
High Volume Air Flow	2.5% (2)

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

The air flow 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 about 2.5% higher than 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, but aliquots for radioiodine analyses are withdrawn first, since trace iodine is unstable in acid solution.

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