ANL-85-17

ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY

ANNUAL REPORT FOR 1984

by

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

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

Operated by THE UNIVERSITY OF CHICAGO
for the U. S. DEPARTMENT OF ENERGY
under Contract W-31-109-Eng-38

Argonne National Laboratory, with facilities in the states of Illinois and Idaho, is owned by the United States government, and operated by The University of Chicago under the provisions of a contract with the Department of Energy.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Printed in the United States of America
Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161

NTIS price codes
Printed copy: A06
Microfiche copy: A01

Distribution Category:
Health and Safety (UC-11)

ANL-85-17

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY ANNUAL REPORT FOR 1984

by

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

Occupational Health and Safety Department

Laboratory Work Performed by

W. J. Dudas	D. M. Ray
T. L. Duffy	J. G. Riha
N. W. Golchert	J. Sedlet
R. B. Kasper	H. C. Svoboda
N. D. Kretz	T. TenKate

March 1985

Approved: D. P. O'Neil
D. P. O'Neil
Department Manager

Preceding Report in This Series: ANL-84-14

TABLE OF CONTENTS

		Page
ABSTRACT		1
I.	INTRODUCTION	1
	A. General	1
	B. Description of Site	3
	C. Population	7
	D. Meteorology	7
	E. Geohydrology	10
	F. Water and Land Use	12
II.	SUMMARY	15
III.	MONITORING RESULTS	21
	A. Radiological	21
	1. Air	23
	2. Surface Water	31
	3. Ground Water	38
	4. Soil, Grass, and Bottom Sediment	41
	5. Milk	50
	6. External Penetrating Radiation	51
	7. Potential Radiation Dose Estimates	55
	a. Radionuclides in Air and Water	55
	b. External Penetrating Radiation Dose	60
	c. Summary	61
	B. Chemical Constituents	61
	1. Surface Water	61
	a. Waste Treatment Plant Effluent Water	63
	b. National Pollutant Discharge Elimination System (NPDES)	65
	c. Sawmill Creek	69
	d. Des Plaines River	73
	2. Ground Water	75

TABLE OF CONTENTS

Page

IV. APPENDIX 91

A. References 91

B. Environmental Permits, Impact Statements, and Activities 93

C. Environmental Quality Standards, Detection Limits, Analytical Errors, and Quality Assurance 95

1. Standards 95

2. Detection Limits 95

3. Quality Assurance Program 97

a. Radiochemical Analysis and Radioactivity Measurements 97

b. Penetrating Radiation 97

c. Chemical Analysis 100

d. Sampling, Sample Storage, Other 102

C. Distribution List 103

D. Acknowledgements 104

LIST OF TABLES

No.	Title	Page
1.	Incremental Population Data in the Vicinity of ANL, 1981	8
2.	ANL Weather Summary, 1984	11
3.	Total Alpha and Beta Activities in Air-Filter Samples, 1984	25
4.	Gamma-Ray Activity in Air-Filter Samples, 1984	26
5.	Strontium, Thorium, Uranium, and Plutonium Concentrations in Air-Filter Samples, 1984	28
6.	Summary of Airborne Radioactive Emissions, 1984	30
7.	Tritiated Water Vapor in Air, 1984	32
8.	Radionuclides in Sawmill Creek Water, 1984	34
9.	Radionuclides in Des Plaines River Water, 1984	36
10.	Radionuclides in Illinois River Water, 1984	37
11.	Radioactivity in ANL Domestic Wells, 1984	39
12.	Tritium Content of Water Near A^2R^2 Excavation, 1984	41
13.	Gamma-Ray Emitting Radionuclides in Soil, 1984	43
14.	Transuranics in Soil, 1984	44
15.	Ground Deposition of Plutonium-239,240	45
16.	Radionuclides in Grass, 1984	48
17.	Radionuclides in Bottom Sediment, 1984	49
18.	Gamma-Ray Emitting Radionuclides in Landfill Sediment Samples, 1984	50
19.	Radionuclides in Milk, 1984	51
20.	Environmental Penetrating Radiation at Off-Site Locations, 1984	52
21.	Environmental Penetrating Radiation at ANL, 1984	53
22.	Concentration-to-Dose Conversion Factors	56
23.	Dose From Airborne Emissions, 1984	58
24.	80 km Population Dose, 1984	58
25.	Radionuclide Concentrations and Dose Estimates for Sawmill Creek Water, 1984	59
26.	Water Quality Standards and Detection Limits	62
27.	Chemical Constituents in Effluents from ANL Treatment Plant, 1984	64

LIST OF TABLES

No.	Title	Page
28.	NPDES Effluent Quality Summary, IL 0034592, Effective through October 28, 1984	67
29.	NPDES Effluent Quality Summary, IL 00334592, (Revised in Effect October 28, 1984)	70
30.	Sawmill Creek - Effect of Sanitary Waste, 1984	72
31.	Chemical Constituents in Sawmill Creek, Location 7M, 1984	74
32.	Chemical Constituents in the Des Plaines River, 1984	75
33.	Well Point and Water Level Elevations of Monitoring Sites at the ANL Landfill	78
34.	Chemical Constituents at ANL Landfill, Well 1	80
35.	Chemical Constituents at ANL Landfill, Well 2	81
36.	Chemical Constituents at ANL Landfill, Well 3	82
37.	Chemical Constituents at ANL Landfill, Well 4	84
38.	Chemical Constituents at ANL Landfill, Drainage Ditch	85
39.	Chemical Constituents at ANL Landfill, Well 5	86
40.	Chemical Constituents at ANL Landfill, Well 6	87
41.	Chemical Constituents at ANL Landfill, Well 7a	88
42.	Environmental Permits in Effect at ANL	94
43.	Concentration Guides and Detection Limits	96
44.	Summary of EPA-QA Samples, 1984	98
45.	Summary of DOE-EML-QAP Samples, 1984	99
46.	Summary of Quality Assurance Studies	101

LIST OF FIGURES

No.	Title	Page
1.	Sampling Locations at Argonne National Laboratory	5
2.	Sampling Locations Near Argonne National Laboratory	6
3.	Monthly and Annual Wind Roses Near Argonne National Laboratory, 1984	9
4.	Plutonium-239,240 Air Concentrations, 1973-1984	29
5.	Plutonium-239,240 Surface Deposition, 1973-1984	46
6.	Penetrating Radiation Measurements at the ANL Site, 1984	54
7.	NPDES Permit Locations	66
8.	Monitoring Well Locations for Landfill	76

ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY ANNUAL REPORT FOR 1984

by

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

ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1984 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, ground water, soil, grass, bottom sediment, and milk; for a variety of chemical constituents in surface water, ground water, and Argonne effluent water; and of the environmental penetrating radiation dose. Sample collections and measurements were made on the site, at the site boundary, and off the Argonne site for comparison purposes. The potential radiation dose to off-site population groups is also estimated. The results of the program are interpreted in terms of the sources and origin of the radioactive and chemical substances (natural, fallout, Argonne, and other) and are compared with applicable environmental quality standards.

I. INTRODUCTION

A. General

This report is prepared to provide the U. S. Department of Energy (DOE) and the public with information on the levels of radioactive and chemical 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 Draft

5484.1A, Chapter III.[1] The Laboratory conducts a continuous environmental monitoring program on and near the Argonne site whose primary purpose is to determine the magnitude, origin, and identity of radioactive or toxic chemical substances in the environment. Of special interest is the detection of any such materials released to the environment by Argonne. One important function of the program is to verify the adequacy of Argonne's pollution controls.

Argonne is a DOE energy research and development laboratory with several principal objectives. It conducts a broad program of research in the basic energy and related sciences (physical, chemical, material, nuclear, biomedical, and environmental), and serves as an important engineering center for the study of nuclear and non-nuclear energy sources. Some of the energy-related research projects conducted during 1984 were safety studies for light water and breeder reactors, development of components and materials for fission and fusion reactors, improvements in the utilization of coal for power production (particularly high sulfur coal), electrochemical energy sources including fuel cells and batteries for vehicles and for energy storage, solar energy utilization, evaluation of heat exchangers for the recovery of waste heat from engines, and operation of the National Battery Test Laboratory. Other areas of research are the use of superconducting magnets for improved nuclear particle accelerators, coal technology, fundamental coal chemistry studies, 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, characterization and monitoring of energy-related pollutants, and the effect of acidic precipitation on vegetation, soil, and surface water quality. A significant portion of these laboratory studies requires the use of radioactive and chemically toxic substances.

The principal nuclear facilities at the Laboratory are a 200 kW light-water cooled and moderated biological research reactor (Janus) fueled with enriched uranium; the Argonne Thermal Source Reactor (ATSR), a 10 kW research reactor fueled with enriched uranium; a superconducting heavy ion

linear accelerator (Argonne Tandem Linac Accelerating System, ATLAS); a 60-inch cyclotron; several other charged particle accelerators (principally of the Van de Graaff and Dynamitron type); a large fast neutron source (Intense Pulsed Neutron Source, IPNS) in which high energy protons strike a heavy metal target to produce the neutrons; cobalt-60 irradiation sources; chemical and metallurgical plutonium laboratories; and several hot cells and laboratories designed for work with multicurie quantities of the actinide elements and with irradiated reactor fuel elements. Two experiments were begun in these facilities in 1984 that have a potential environmental impact: 1) proof-of-breeding in light-water reactors, which involves the dissolution and analysis of irradiated thorium and uranium-233 dioxide fuel elements and 2) recovery of tritium from reactor irradiated ceramic lithium compounds. Two major facilities, a 12.5 GeV proton accelerator (Zero Gradient Synchrotron, ZGS) and a 5 MW heavy water enriched uranium reactor (CP-5) were not in operation during 1984. The CP-5 facility is awaiting decontamination and decommissioning, while this work has been completed at the ZGS. Also located on the ANL site is the DOE New Brunswick Laboratory, a safeguards plutonium and uranium measurements and analytical chemistry laboratory. The principal non-nuclear activities at Argonne that may produce a measurable impact on the environment are a coalfired boiler (No. 5) and the heat exchanger studies for waste heat recovery. 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 heat recovery studies use moderately large quantities of toxic or flammable organic compounds such as toluene, Freon, biphenyl oxides, methyl pyridine, and trifluoroethanol.

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.

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

The largest topographical feature is the Des Plaines River channel, about 1.6 km (1 mi) wide. This channel contains the River, the Chicago Sanitary and Ship Canal, and the Illinois and Michigan Canal. Their presence extends the uninhabited area about 1.6 km (1 mi) south of the site. The elevation of the channel surface is 180 m (590 ft) above sea level. Bluffs, which comprise the south border of the site, rise from the channel at varying slope angles of 15° to 60°, reaching an average elevation of 200 m (650 ft) above sea level at the top. The land then slopes gradually upward reaching the average site elevation of 220 m (725 ft) above sea level at 940 m (3,000 ft) from the bluffs. Several large ravines oriented in a north-south direction are located in the southern portion of the site. The bluffs and ravines generally are forested with mature deciduous trees. The remaining portion of the site changes in elevation by no more 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.[2]

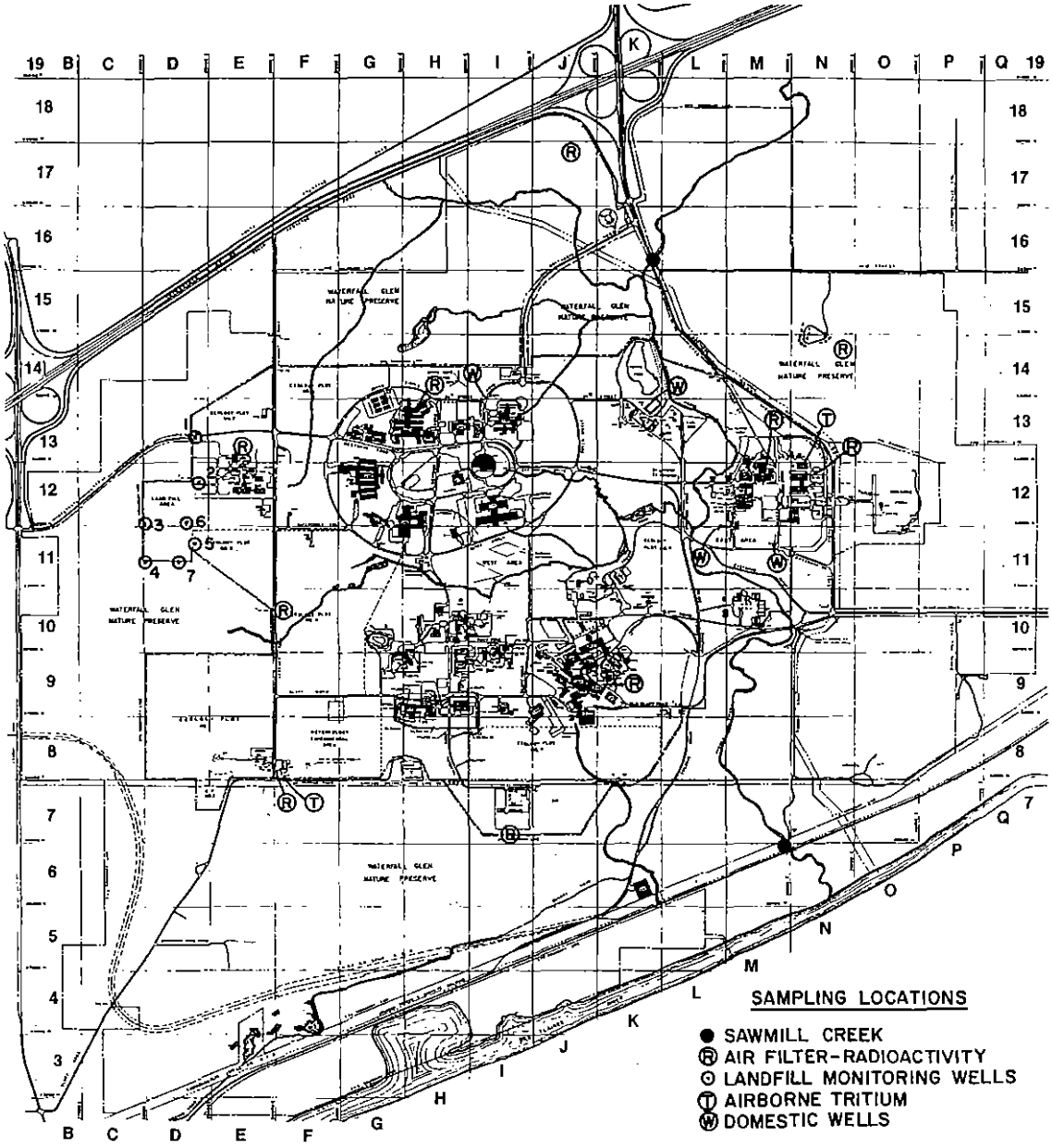

Fig. 1. Sampling Locations at Argonne National Laboratory

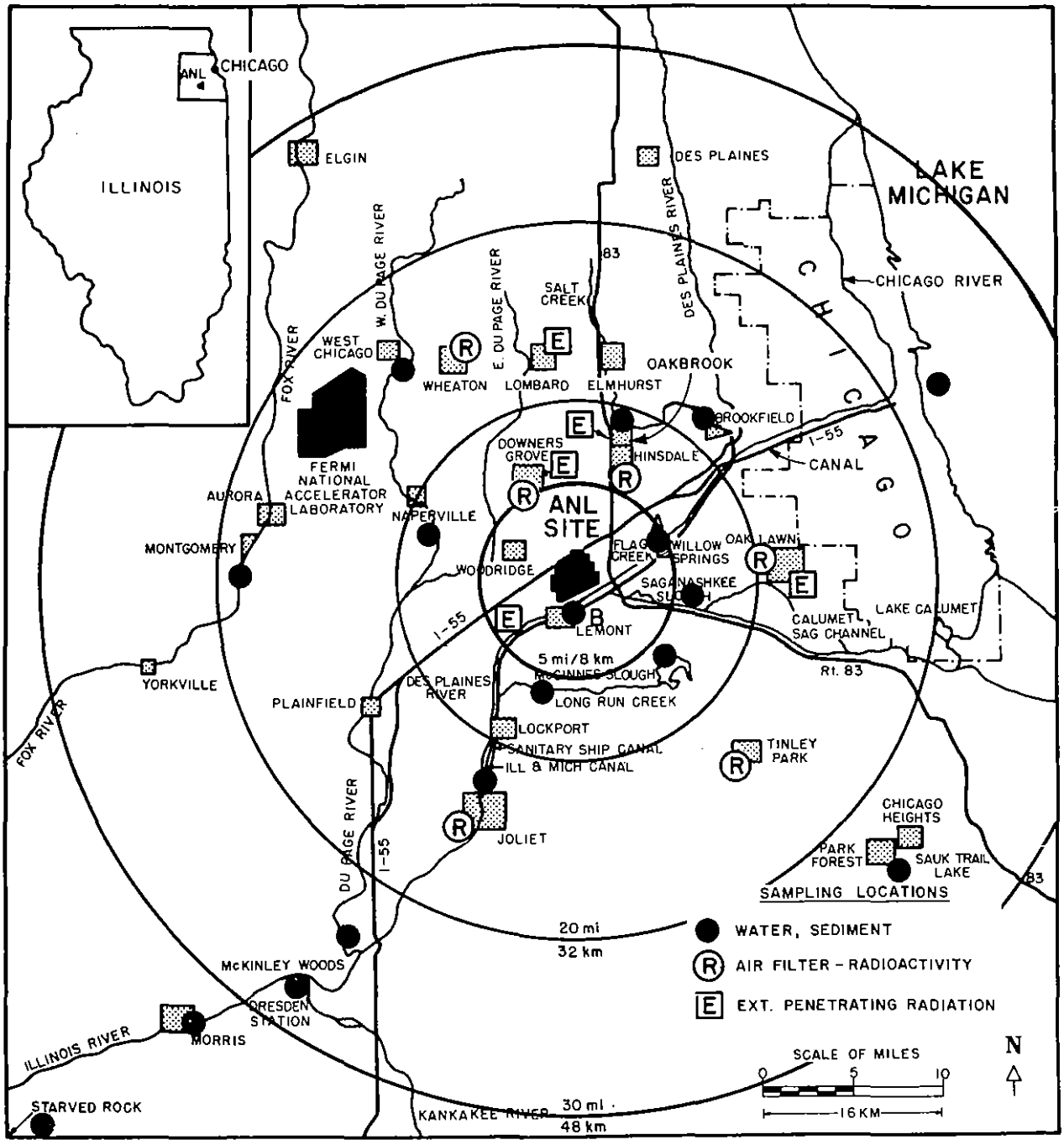

Fig. 2. Sampling Locations Near Argonne National Laboratory

C. Population

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

D. Meteorology

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

The most important meteorological parameters for the purposes of this report are wind direction, wind speed, temperature, and precipitation. The wind data are used to calculate radiation doses from air emissions and in selecting air sampling locations and distances. Temperature and precipitation data are useful in interpreting some of the environmental results. Since on-site meteorological data are no longer available in suitable form, measurements taken in 1984 at the nearest U. S. Weather Bureau station, O'Hare Airport, 31 km (18 mi) north of Argonne, were used. The 1984 average monthly and annual wind roses are shown in Figure 3. The wind roses are polar coordinate plots in which the lengths of the radii represent the percentage frequency of wind speeds in classes of 2.01-6 m/s (4.5-13.4 mph), 6.01-10 m/s (13.4-22.4 mph), and greater than 10.01 m/s (22.4 mph). The number in the center represents the percent of observations of wind speed less than 2 m/s (4.5 mph) in all directions. The direction of the radii from the center 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.

TABLE 1

Incremental Population Data in the Vicinity of ANL, 1981

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

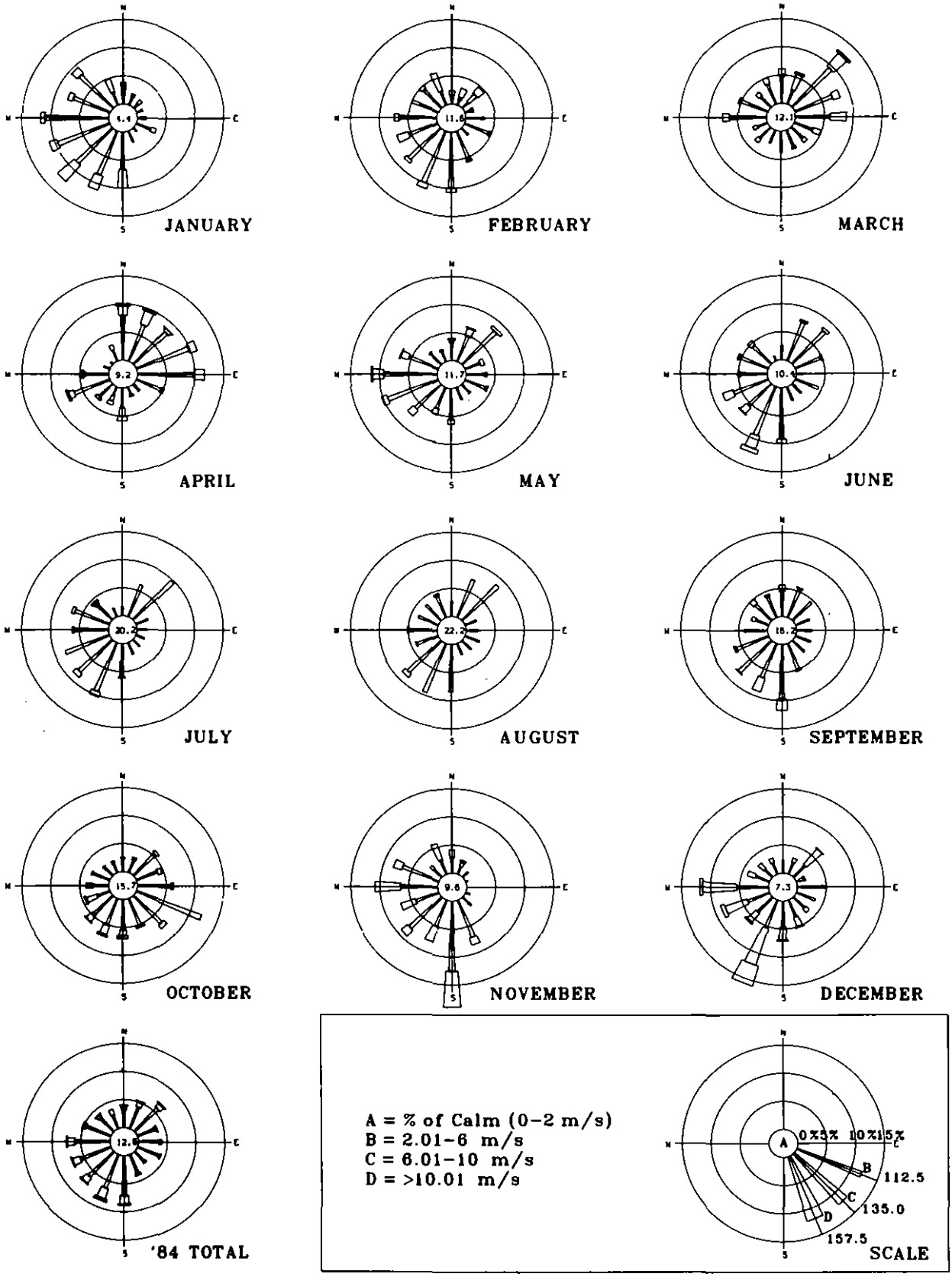

Fig. 3. Monthly and Annual Wind Roses Near Argonne National Laboratory, 1984

A comparison of the monthly wind roses indicates that the winds are sufficiently variable so that monitoring for airborne releases must be carried out in all directions from the site. For example, the dominant wind direction in March is northeast while in November it is south. The annual average wind rose for 1984 is consistent with the long term average wind direction, which is usually from the west to south. Precipitation and temperature data for 1984 are shown in Table 2. Temperatures and precipitation were both normal in 1984 compared to the long term average.

E. Geohydrology

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

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

The four domestic water wells now in use are about 90 m (300 ft) deep in the Niagaran dolomite, about 300 feet from the ground surface. One well, in the Galesville sandstone 490 m (1,600 ft) deep, is not used because the water table has dropped below the pumping level. The water level in the Niagaran dolomite has remained reasonably stable under Laboratory pumping, dropping about 3.7 m (12 ft) between 1960 and 1980. The aquifer appears to be adequate for future Laboratory use, but this ground water source is used throughout the area. There are also several monitoring wells

TABLE 2

Weather Summary, 1984*

Month	Precipitation (cm) Amount	Precipitation (cm) Average	Temperature (°C) Monthly Average	Temperature (°C) Average
January	2.92	4.06	-8.3	-5.9
February	3.53	3.33	1.1	-3.3
March	7.62	6.58	-1.4	2.2
April	10.44	9.30	7.7	9.3
May	11.40	8.00	13.1	15.1
June	5.13	10.36	21.3	20.3
July	8.10	9.22	21.3	22.8
August	5.33	8.97	22.7	22.2
September	9.75	8.51	16.2	18.2
October	8.00	5.79	12.6	11.9
November	6.71	5.23	3.3	4.3
December	7.42	5.33	-0.5	-2.4
Total	86.35	84.68		

*Data obtained from the National Oceanic and Atmospheric Administration for the weather station at O'Hare International Airport. The average is for the years 1951-1980.

and small capacity water wells used for laboratory experiments, fire protection, and sanitary facilities.

F. Water and Land Use

The principal stream that drains the site is Sawmill Creek. It carries effluent water continuously from a sewage treatment plant (Marion Brook Treatment Plant) located a few kilometers north of the site. The plant has an operating capacity of about 11.4 megaliters (3 million gallons) per day. In addition, the residential development in the area has resulted in the collection and channeling of additional runoff water into the Creek. Treated sanitary and laboratory waste water from Argonne are combined and discharged into Sawmill Creek at location 7M in Figure 1. This effluent averaged 2.8 megaliters (0.75 million gallons) per day, while the water flow in the Creek upstream of the waste-water outfall averaged about 34 megaliters (9.1 million gallons) per day during 1984. The combined Argonne effluent consisted of 63% laboratory waste water and 37% sanitary waste water.

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

from Argonne, where water is used to replenish ground water supplies by infiltration. In the vicinity of the Laboratory, only subsurface water (from both shallow and deep aquifers) and Lake Michigan water are used for drinking purposes.

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

I. SUMMARY

This is one in a series of annual reports prepared to provide the U. S. Department of Energy (DOE), environmental agencies, 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-84-14.[4] *Included in this report are the results of measurements obtained in 1984 for a number of radionuclides in air, surface water, ground water, soil, grass, bottom sediment, and milk; for a variety of chemical constituents in surface and subsurface water, and for the external penetrating radiation dose.*

The major airborne radionuclides released from the Laboratory were hydrogen-3, carbon-11, argon-41, krypton-85, and radon-220, all as gases. The maximum dose from these nuclides at the site boundary was 0.09 mrem/yr 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 0.05 mrem/yr, which is 0.01% of the Radiation Protection Standard of 500 mrem/yr. These releases constitute an insignificant addition to the dose received from the natural background radiation, which is about 88 mrem/yr. The total 80-km population dose from these radionuclides was less than 3.2 man-rem for 1984, compared to approximately 700,000 man-rem from the natural background radiation.*

Particulate radioactivity (total alpha and beta, fission and activation products, plutonium, thorium, and uranium) was measured in air-filter samples collected continuously at the site perimeter and off the site. No

---

* *Unless otherwise indicated, the hazard due to a given concentration of a radioactive nuclide is assessed in this report by comparison with the annual dose limits, or Radiation Protection Standards and Concentration Guides (CG) for uncontrolled areas specified by the U. S. Department of Energy Order 5480.1, Chapter XI.*[5] *The pertinent CGs are listed in the Appendix, Section V.C. 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.*

activity attributable to Argonne operations could be detected. These samples contained only radionuclides from natural sources and nuclear test detonations. Almost all of the radioactive materials injected into the air by previous atmospheric nuclear tests have either decayed or have been deposited on the ground.

Argonne waste water is discharged into Sawmill Creek, and this stream was sampled above and below the site to evaluate the effect of Argonne operations on its radioactive content. The nuclides (for which analyses were made) added to the Creek in the waste water, and the percent of the Concentration Guide (CG) of their net average Creek concentrations, were hydrogen-3, 0.003%; strontium-90, 0.042%; neptunium-237, 0.00022%; plutonium-239,240, 0.00040%; americium-241, 0.00040%; and curium-244 and/or californium-249, 0.00004%. The concentrations are all very low compared to the CG standard.

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

Plutonium concentrations in soil showed the same general range and average at the site perimeter and off the site as in the past years. The average plutonium-239,240 content of the top 5 cm (2 in) of soil was 0.7×10^{-3} $\mu Ci/m^2$ at the site perimeter and 0.5×10^{-3} $\mu Ci/m^2$ off the site. The corresponding plutonium-238 averages were 0.03×10^{-3} $\mu Ci/m^2$ and 0.02×10^{-3} $\mu Ci/m^2$, respectively. The plutonium content of grass was similar to that found in previous years and was about a factor of 10^4 less than soil from the same locations. The results were within the range reported by other laboratories for fallout from test detonations, and the plutonium found in soil and grass is attributed to this source. The plutonium content of samples from beds of off-site streams and ponds ranged from 1×10^{-9} $\mu Ci/g$ to 18×10^{-9} $\mu Ci/g$ of plutonium-239,240, a range found in previous years to be normal for fallout plutonium in such materials. Higher concentrations were found in the sediment just

below the Laboratory waste water outfall as a result of their presence in Argonne waste water, but they decreased to normal, or fallout, levels 50 meters downstream of the outfall.

Milk from a dairy farm near the Laboratory was analyzed for several fission products, including hydrogen-3. Hydrogen-3 concentrations averaged < 113 x 10^{-9} µCi/mL. The strontium-90 concentration (2.8 x 10^{-9} µCi/mL), and the cesium-137 concentration (0.7 x 10^{-9} µCi/mL) were similar to 1983 results. These radionuclides resulted from nuclear test detonations, and are not related to Argonne operations.

Measurements of penetrating radiation were made at several locations at the site boundary and off the site. The off-site results averaged 88 ± 11 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 370 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 decreased to 107 ± 29 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 31 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 about 0.06 mrem/yr. Thus, doses to individuals near the site from these sources will not exceed 0.012% of the 500 mrem/yr limit.

Concentrations of chemical constituents and other water quality parameters were measured in Argonne waste and effluent water and in Sawmill Creek. The results were compared to the standards adopted by the State of Illinois as well as National Pollutant 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 or Sawmill Creek could be seen.

All the results obtained for the waste water were below the State standards except for mercury, which exceeded the standard in 31% of the samples measured. The average mercury concentration was 81% of the standard. Efforts are continuing to lower these concentrations by additional treatment of batches of waste water that contain elevated levels.

The results for the NPDES permit effluent locations were in general compliance with the exception of mercury at Location 001 and pH at Location 009 (Fig. 7). The permit was extensively revised and renewed effective October 28, 1984. The mercury concentration limit was changed from 0.5 µg/L to 3.0 µg/L, and subsequent samples were in compliance partially because the limit was changed and partially because of more extensive mercury removal treatments. The pH at Location 009 remains above the limit, due to the basicity of water from the water softening process. This location will be in compliance when this softening process is replaced in 1985. In any event, this effluent has no measurable effect on the receiving stream.

The average values in Sawmill Creek for dissolved oxygen and most chemical constituents were within the State of Illinois Water Quality Standards. The average levels of copper and iron were 181 and 146% of the State standards, and individual values exceeded the standards 53 and 37% of the time. Iron and copper concentrations increase in response to rainfall runoff which increases the amounts of these elements in the stream. The relative concentrations of iron and copper in the Creek and in the waste water, and the dilution of waste water by a factor of ten in the Creek, shows that elevated levels of these elements in the Creek are not due to their presence in waste water. Mercury concentrations exceeded the State standard 24% of the time, but the average concentration was only 61% of the standard. The mercury was released in Argonne waste water, and improved cleaning operations are expected to lower these levels in the future. The concentration of mercury in the Des Plaines River was not affected by the amounts in Argonne effluent water.

Monitoring wells in the ANL sanitary landfill area were examined for selected inorganic and organic constituents. Levels of manganese in two of the wells were elevated as was the pH in one and arsenic in another. Similar results were obtained in the past, and there is no indication of migration. Organic compounds (acetone, benzene, perchloroethylene, toluene, trichloroethylene, and xylene) were not detected.

The average concentrations and total amounts of radioactive and chemical pollutants released by Argonne to the environment did not constitute a health hazard. Any individual discharges from the Laboratory that exceeded acceptable standards were temporary, and when they did occur, investigations were undertaken to identify the source and reduce their discharge.

III. MONITORING RESULTS

A. Radiological

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

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

with a frequency such that the average value is one-half of the detection limit. The averages that are obtained by using these two methods under the conditions indicated are believed to give an adequate picture of the average concentration at locations where the concentrations not only varied greatly, but were at times not detectable. Penetrating radiation measurements are reported in units of millirem (mrem) per year and population dose in man-rem.

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

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

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

1. Air

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

The total alpha and beta activities in the individual weekly samples are summarized in Table 3. 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 gamma-ray emitters, as determined by gamma-ray spectrometry performed on composite weekly samples are given in Table 4. The gamma-ray detector is a shielded germanium diode calibrated for each gamma-ray emitting nuclide measured.

The alpha activities, principally due to naturally occurring nuclides, averaged the same as the past several years and were in their normal range. The average beta activity for the year, 2.4×10^{-14} µCi/mL, was identical to 1982 and 1983 averages. The beta activity was also from natural sources. The fission products from previous nuclear tests found and reported in the past were not detected in 1984 due to decay and/or deposition. The most recent atmospheric nuclear test took place on October 16, 1980, by the People's Republic of China. The beryllium-7 exhibits a spring increase in concentration, indicating its stratospheric origin. The lead-210 in air is due to the radioactive decay of the naturally occurring uranium decay series, principally from the decay of gaseous radon-222 in air.

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

Plutonium and thorium were separated on an anion exchange column and the uranium was extracted from the column effluent. Following the extraction, the aqueous phase was analyzed for radiostrontium by a standard radiochemical procedure. The separated plutonium, thorium, and uranium fractions were electrodeposited and measured by alpha spectrometry. The chemical recoveries were monitored by adding known amounts of plutonium-242, thorium-234, and uranium-232 tracers prior to ignition. Since alpha spectrometry cannot distinguish between plutonium-239 and plutonium-240, it

TABLE 3

TOTAL ALPHA AND BETA ACTIVITIES IN AIR-FILTER SAMPLES, 1984*
(CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

MONTH	LOCATION	NO. OF SAMPLES	ALPHA ACTIVITY AV.	ALPHA ACTIVITY MIN.	ALPHA ACTIVITY MAX.	BETA ACTIVITY AV.	BETA ACTIVITY MIN.	BETA ACTIVITY MAX.
JANUARY	PERIMETER	32	2.1	0.9	3.5	29	14	40
	OFF-SITE	21	2.1	0.4	4.0	30	19	37
FEBRUARY	PERIMETER	28	2.0	0.7	5.3	22	16	29
	OFF-SITE	20	1.5	0.8	2.8	22	14	28
MARCH	PERIMETER	34	2.4	1.2	3.8	21	12	32
	OFF-SITE	20	2.0	0.5	3.6	20	8	29
APRIL	PERIMETER	34	1.9	0.5	3.7	17	9	25
	OFF-SITE	20	1.6	0.1	2.9	18	8	60
MAY	PERIMETER	29	2.3	1.1	4.8	18	9	24
	OFF-SITE	20	2.2	1.2	3.5	18	14	26
JUNE	PERIMETER	31	2.1	0.8	3.6	19	8	34
	OFF-SITE	22	1.8	0.8	3.9	17	5	25
JULY	PERIMETER	35	2.5	0.3	13	22	6	34
	OFF-SITE	19	1.9	0.8	3.1	21	6	30
AUGUST	PERIMETER	31	2.2	1.0	5.4	28	17	43
	OFF-SITE	19	2.1	1.1	3.3	25	17	34
SEPTEMBER	PERIMETER	34	2.2	0.8	4.8	26	14	47
	OFF-SITE	16	1.9	0.4	3.7	25	10	38
OCTOBER	PERIMETER	34	2.0	0.6	4.0	25	3	40
	OFF-SITE	20	2.3	0.1	4.4	26	7	39
NOVEMBER	PERIMETER	28	2.6	0.8	6.3	26	19	44
	OFF-SITE	19	1.8	0.9	2.7	22	14	32
DECEMBER	PERIMETER	23	2.3	1.2	4.1	36	23	54
	OFF-SITE	20	2.8	0.7	6.6	35	19	64
ANNUAL SUMMARY	PERIMETER	373	2.2 ± 0.1	0.3	13	24 ± 1	3	54
	OFF-SITE	236	2.0 ± 0.1	0.1	6.6	23 ± 1	5	64
% CG	PERIMETER	-	(0.002)	(0.0003)	(0.013)	0.024	0.003	0.054
% CG	OFF-SITE	-	(0.002)	(0.0001)	(0.007)	0.023	0.005	0.064

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

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

TABLE 4

GAMMA RAY ACITVITY IN AIR-FILTER SAMPLES, 1984
(CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

MONTH	LOCATION	BE7	PB210
JANUARY	PERIMETER	62	37
	OFF-SITE	71	52
FEBRUARY	PERIMETER	58	37
	OFF-SITE	69	42
MARCH	PERIMETER	86	27
	OFF-SITE	83	33
APRIL	PERIMETER	84	18
	OFF-SITE	103	21
MAY	PERIMETER	102	19
	OFF-SITE	111	24
JUNE	PERIMETER	101	20
	OFF-SITE	99	24
JULY	PERIMETER	100	25
	OFF-SITE	103	34
AUGUST	PERIMETER	98	35
	OFF-SITE	95	42
SEPTEMBER	PERIMETER	91	36
	OFF-SITE	111	42
OCTOBER	PERIMETER	65	37
	OFF-SITE	73	48
NOVEMBER	PERIMETER	81	41
	OFF-SITE	76	41
DECEMBER	PERIMETER	73	58
	OFF-SITE	70	65
ANNUAL SUMMARY	PERIMETER	83 ± 10	33 ± 7
	OFF-SITE	89 ± 10	39 ± 8
% CG	PERIMETER	(0.00021)	(0.407)
	OFF-SITE	(0.00022)	(0.486)

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 5.

The average strontium-90 and plutonium-239 concentrations were similar to 1983 results. Strontium-89 was not observed above the detection limit of 1 x 10^{-16} µCi/mL. The plutonium-239 concentrations at location 7I are slightly elevated in February, March, and April. Since the sampler is located in an area where radioactive waste material is stored before shipment to a disposal site, the elevated concentrations may have resulted from work in that area. The highest monthly concentration was only 4.9 x 10^{-4}% of the CG.

Figure 4 shows the monthly plutonium-239 off-site air concentrations for the past 12 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 thorium and uranium concentrations are in the same range found during the past several years and are considered to be of natural origin. The amounts of thorium and uranium in a sample were proportional to the mass of inorganic material collected on the filter paper, and the bulk of these elements in the air was due to resuspension of soil. In contrast, the amount of plutonium in the air samples contributed by soil, if the resuspended soil has the same plutonium concentration as the first centimeter on the ground, ranged from about 7% in January to 28% in December of the total plutonium in the samples.

The major airborne effluents released during 1984 are listed in Table 6 by location. The increase from Building 200 is due to work on the "proof-of-breeding" program and the hydrogen-3 from Building 212 is from the tritium recovery studies. In addition to the nuclides listed in Table 6, several other fission products were also released in millicurie or smaller amounts.

TABLE 5

STRONTIUM, THORIUM, URANIUM AND PLUTONIUM CONCENTRATIONS IN AIR-FILTER SAMPLES, 1984
(CONCENTRATIONS IN 1E-18 MICROCURIES/ML)

MONTH	LOCATION[1]	SR-90	TH-228	TH-230	TH-232[2]	U-234	U-235	U-238[2]	PU-238	PU-239[3]
JANUARY	7I	40 ± 30	5 ± 2	11 ± 2	6 ± 1	15 ± 3	< 0.3	13 ± 1	< 0.3	2.0 ± 0.7
	12N	20 ± 20	2 ± 2	11 ± 1	5 ± 1	15 ± 2	0.4 ± 0.2	15 ± 1	< 0.3	2.3 ± 1.5
	OFF-SITE	20 ± 30	1 ± 2	-	2 ± 1	11 ± 4	< 0.3	7 ± 2	0.4 ± 1.2	0.9 ± 0.8
FEBRUARY	7I	50 ± 40	9 ± 1	12 ± 1	7 ± 1	11 ± 3	< 0.3	13 ± 1	< 0.3	3.0 ± 1.2
	12N	30 ± 40	9 ± 3	17 ± 3	9 ± 2	28 ± 5	0.4 ± 0.3	21 ± 2	< 0.3	1.1 ± 1.1
	OFF-SITE	30 ± 10	5 ± 1	12 ± 1	7 ± 1	13 ± 2	< 0.3	15 ± 1	< 0.3	1.2 ± 0.8
MARCH	7I	20 ± 10	13 ± 5	14 ± 4	6 ± 4	11 ± 1	0.6 ± 0.6	12 ± 2	< 0.3	4.0 ± 2.9
	12N	30 ± 10	7 ± 1	21 ± 1	7 ± 1	34 ± 3	0.9 ± 0.3	36 ± 2	< 0.3	0.9 ± 0.6
	OFF-SITE	30 ± 10	4 ± 1	6 ± 1	4 ± 1	8 ± 1	< 0.3	7 ± 1	0.5 ± 0.5	2.1 ± 0.6
APRIL	7I	20 ± 20	15 ± 2	33 ± 2	14 ± 2	9 ± 4	0.5 ± 0.3	9 ± 1	< 0.3	4.9 ± 1.3
	12N	20 ± 10	1 ± 1	3 ± 1	1 ± 1	21 ± 5	1.0 ± 0.4	25 ± 2	< 0.3	2.1 ± 0.8
	OFF-SITE	20 ± 10	16 ± 3	16 ± 3	12 ± 3	5 ± 3	< 0.3	5 ± 3	< 0.3	1.8 ± 1.6
MAY	7I	40 ± 20	14 ± 1	18 ± 1	14 ± 1	22 ± 2	0.4 ± 0.2	20 ± 1	< 0.3	1.0 ± 0.4
	12N	40 ± 40	16 ± 1	24 ± 2	11 ± 1	-	-	-	0.5 ± 0.4	1.2 ± 0.4
	OFF-SITE	20 ± 10	3 ± 3	4 ± 2	2 ± 2	3 ± 2	< 0.3	4 ± 1	0.4 ± 0.4	0.7 ± 1.0
JUNE	7I	30 ± 30	10 ± 1	14 ± 1	8 ± 1	14 ± 1	0.4 ± 0.6	13 ± 1	< 0.3	1.3 ± 0.6
	12N	< 10	13 ± 2	15 ± 2	8 ± 1	19 ± 2	0.4 ± 0.6	17 ± 2	< 0.3	1.3 ± 0.6
	OFF-SITE	40 ± 20	8 ± 1	13 ± 1	8 ± 1	15 ± 1	< 0.3	13 ± 1	< 0.3	1.7 ± 0.6
JULY	7I	< 10	5 ± 3	6 ± 3	3 ± 2	8 ± 1	< 0.3	7 ± 1	< 0.3	1.8 ± 1.5
	12N	20 ± 20	3 ± 6	7 ± 6	5 ± 6	17 ± 2	0.7 ± 0.8	17 ± 2	< 0.3	1.7 ± 0.8
	OFF-SITE	20 ± 10	5 ± 7	8 ± 8	7 ± 7	8 ± 1	< 0.3	9 ± 1	< 0.3	0.8 ± 0.7
AUGUST	7I	-	11 ± 2	15 ± 2	5 ± 1	13 ± 2	< 0.3	14 ± 2	< 0.3	1.6 ± 0.9
	12N	< 10	7 ± 2	16 ± 3	4 ± 2	21 ± 2	< 0.3	24 ± 2	< 0.3	2.4 ± 1.2
	OFF-SITE	-	3 ± 2	4 ± 2	3 ± 2	6 ± 1	< 0.3	6 ± 1	< 0.3	1.2 ± 0.8
SEPTEMBER	7I	< 10	7 ± 3	11 ± 3	6 ± 2	9 ± 2	0.6 ± 1.5	9 ± 2	< 0.3	0.9 ± 0.8
	12N	20 ± 10	4 ± 1	8 ± 1	4 ± 1	9 ± 1	0.5 ± 0.6	9 ± 1	< 0.3	0.8 ± 0.3
	OFF-SITE	< 10	5 ± 3	10 ± 3	5 ± 2	11 ± 1	0.5 ± 1.2	10 ± 2	< 0.3	1.5 ± 1.0
OCTOBER	7I	20 ± 10	4 ± 4	7 ± 3	7 ± 3	7 ± 2	< 0.3	7 ± 2	< 0.3	0.5 ± 0.6
	12N	< 10	3 ± 1	5 ± 1	3 ± 1	7 ± 1	0.5 ± 0.5	7 ± 1	< 0.3	0.6 ± 0.6
	OFF-SITE	< 10	2 ± 2	5 ± 2	3 ± 2	4 ± 1	< 0.3	5 ± 1	< 0.3	0.5 ± 0.4
NOVEMBER	7I	< 10	7 ± 3	10 ± 3	7 ± 2	12 ± 2	< 0.3	12 ± 2	< 0.3	0.6 ± 0.6
	12N	< 10	3 ± 2	8 ± 2	3 ± 2	9 ± 2	< 0.3	10 ± 2	< 0.3	0.6 ± 0.6
	OFF-SITE	20 ± 20	5 ± 3	9 ± 3	6 ± 2	10 ± 2	0.6 ± 1.3	10 ± 2	< 0.3	0.7 ± 0.7
DECEMBER	7I	< 10	9 ± 2	11 ± 2	7 ± 2	14 ± 1	< 0.3	14 ± 1	< 0.3	0.9 ± 0.6
	12N	< 10	9 ± 2	17 ± 2	7 ± 1	23 ± 2	1.3 ± 0.7	23 ± 2	< 0.3	0.7 ± 0.5
	OFF-SITE	< 10	3 ± 2	6 ± 2	4 ± 2	8 ± 2	< 0.3	10 ± 2	< 0.3	0.8 ± 0.6
ANNUAL	7I	20 ± 10	9 ± 2	13 ± 4	7 ± 2	12 ± 2	< 0.4	12 ± 2	< 0.3	1.9 ± 0.8
	12N	20 ± 10	6 ± 3	13 ± 4	6 ± 2	18 ± 5	0.6 ± 0.2	19 ± 5	< 0.3	1.3 ± 0.4
SUMMARY	OFF-SITE	20 ± 10	5 ± 2	9 ± 2	5 ± 2	8 ± 2	< 0.3	8 ± 2	< 0.3	1.2 ± 0.3
%CG	7I	0.01	(4.4)	(4.4)	(0.7)	(0.30)	< 0.01	(0.24)	< 0.03	0.2
	12N	0.01	(3.2)	(4.2)	(0.6)	(0.46)	(0.01)	(0.37)	< 0.03	0.1
(X 1E-3)	OFF-SITE	0.01	(2.5)	(2.9)	(0.5)	(0.21)	< 0.01	(0.17)	< 0.03	0.1

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

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

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

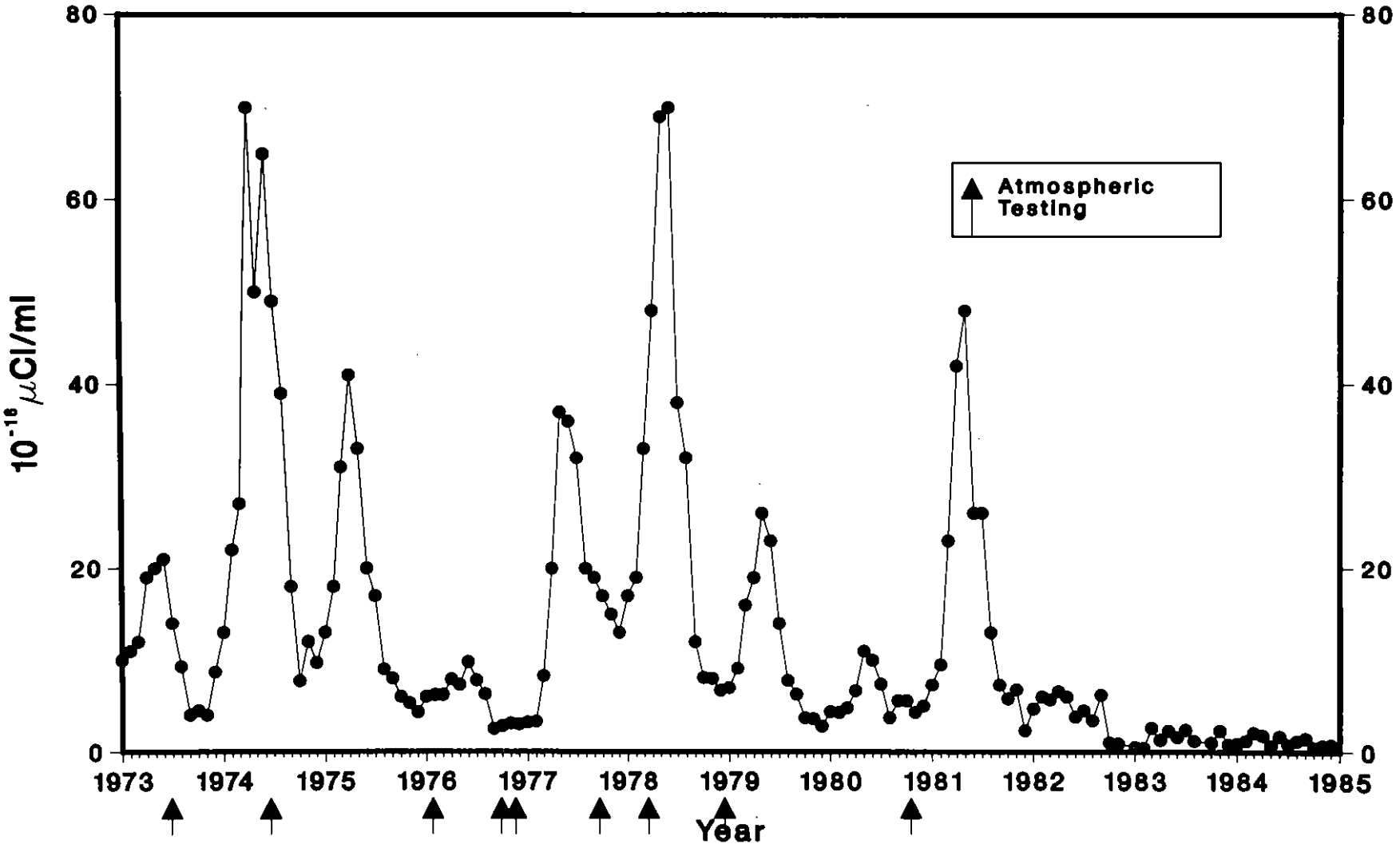

Fig. 4. Plutonium-239,240 Air Concentrations, 1973-1984

TABLE 6

Summary of Airborne Radioactive Emissions, 1984
(Curies/Year)

Building	Nuclide	Concentration
200	Hydrogen-3	4.0
	Krypton-85	69.4
	Radon-220	76.9
202 (Janus)	Argon-41	0.88
212	Hydrogen-3	58.9
	Krypton-85	1.9
375 (IPNS)	Carbon-11	85.6

Because the Laboratory recently began a program that could result in the release of tritiated water vapor into the air, the air sampling program for tritium in air was re-established. Samples were collected at Locations 8F (at the southwest corner of the site) and at 12N (on the east perimeter of the site). The tritiated water vapor was collected by adsorption on silica gel and was measured by counting the desorbed water in a liquid scintillation spectrometer. The results are given in Table 7. Based on the data in Table 6, the principal source of the tritiated water vapor should be from Building 212, Location 12I. Since the winds are usually from the west to south, the tritium concentrations should be higher at Location 12N. However, the concentrations were similar at both perimeter locations, but a factor of two to three less than when these measurements were made in 1979.[7] *Another potential source of tritiated water vapor is the CP-5 reactor, Location 9H, which although not operating, may be exhausting some residual tritium. If the background concentrations measured in 1979 away from the Laboratory are still applicable, and are subtracted from the concentrations in Table 7, the tritium concentrations due to ANL operations are about 25% less than the total measured. The fenceline annual dose at Location 8F would be 0.010 mrem/y and at Location 12N would be 0.008 mrem/y.*

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-liter

TABLE 7

Tritiated Water Vapor in Air, 1984
(Concentrations in 10^{-12} µCi/mL)

Month	Location*	No. of Samples	Avg.	Min.	Max.
March	8F	7	7.5	0.5	22.7
	12N	7	2.6	1.2	8.2
April	8F	8	3.3	0.1	14.8
	12N	8	1.9	0.4	9.4
May	8F	9	3.6	0.9	7.0
	12N	9	4.4	1.1	7.5
June	8F	9	10.2	0.7	29.3
	12N	9	4.5	1.1	8.5
July	8F	8	4.1	0.5	13.7
	12N	8	5.3	1.4	16.1
August	8F	9	5.5	0.6	18.7
	12N	9	4.1	1.6	14.9
September	8F	9	2.1	0.5	5.0
	12N	9	2.0	0.8	4.2
October	8F	9	2.7	0.5	8.5
	12N	9	2.4	0.4	5.6
November	8F	8	0.8	0.2	2.3
	12N	8	2.3	0.5	5.5
December	8F	7	1.6	0.4	4.1
	12N	7	2.2	0.6	4.7
Annual Summary	8F	83	4.1 ± 1.8	0.1	29.3
	12N	83	3.2 ± 0.8	0.4	16.1
Percent CG	8F	-	0.0021	0.00005	0.015
	12N	-	0.0016	0.00020	0.008

*Locations are given in terms of the grid coordinates in Figure 1.

samples by chemical separation methods followed by alpha spectrometry.[8,9] *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 87% 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 8. Comparison of the results, and 95% confidence levels of the averages, for the two sampling locations shows that the nuclides whose presence in the Creek water can be attributed to Argonne operations were hydrogen-3, neptunium-237, plutonium-239, americium-241, and occasionally strontium-90, plutonium-238, curium-242 and/or californium-252, and curium-244 and/or californium-249. The percentage of individual samples containing activity attributable to Argonne was 65% for hydrogen-3, 90% for neptunium-237, 100% for plutonium-239, and 94% for americium-241. The concentrations of all these nuclides were low compared to the CGs. The principal radionuclide added to the Creek by Argonne waste water, in terms of concentration, was hydrogen-3. Its average net concentration was equivalent to 0.003% of the CG. The decrease in the hydrogen-3 over the past few years is due to the cessation of the operation of the CP-5 reactor. The total concentration, regardless of source, must be used in assessing the

TABLE 8

RADIONUCLIDES IN SAWMILL CREEK WATER, 1984

TYPE OF ACTIVITY	* LOCATION	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/ML) AVG.	MIN.	MAX.	PERCENT CG AVG.	MIN.	MAX.
ALPHA (NONVOLATILE)	16K	12	3.1 ± 0.1	0.9	9.1	(0.102)	(0.032)	(0.303)
	7M	249	2.4 ± 0.3	1.0	5.1	(0.081)	(0.034)	(0.171)
BETA (NONVOLATILE)	16K	12	24 ± 1	11	38	(0.81)	(0.36)	(1.26)
	7M	249	19 ± 2	6	30	(0.62)	(0.21)	(1.01)
HYDROGEN-3	16K	12	< 125	< 100	205	< 0.0042	< 0.003	0.0068
	7M	249	207 ± 36	< 100	789	0.0069	< 0.003	0.0263
STRONTIUM-90	16K	12	< 0.27	< 0.25	0.38	< 0.091	< 0.08	0.13
	7M	249	0.40 ± 0.09	< 0.25	1.75	0.133	< 0.08	0.58
RADIUM-226	7M	249	0.69 ± 0.16	0.19	3.13	(2.31)	(0.63)	(10.4)
** URANIUM (NATURAL)	16K	12	1.1 ± 0.1	0.5	2.0	(0.0028)	(0.0013)	(0.0051)
	7M	249	1.1 ± 0.1	0.6	1.8	(0.0028)	(0.0015)	(0.0045)
NEPTUNIUM-237	16K	12	-	-	< 0.001	-	-	< 0.000033
	7M	249	0.0076 ± 0.0023	< 0.001	0.049	0.00025	< 0.00003	0.0016
PLUTONIUM-238	16K	12	-	-	< 0.001	-	-	< 0.00002
	7M	249	< 0.0020	< 0.001	0.025	0.00004	< 0.00002	0.00050
PLUTONIUM-239	16K	12	-	-	< 0.0010	-	-	< 0.000020
	7M	249	0.0208 ± 0.0097	0.0015	0.2463	0.00042	0.000030	0.00493
AMERICIUM-241	16K	12	-	-	< 0.001	-	-	< 0.000025
	7M	244	0.0168 ± 0.0095	< 0.001	0.226	0.00042	< 0.000025	0.0056
CURIUM-242 AND/OR CALIFORNIUM-252	16K	12	-	-	< 0.001	-	-	< 0.000005
	7M	244	< 0.0011	< 0.001	0.0026	< 0.000006	< 0.000005	0.000013
CURIUM-244 AND/OR CALIFORNIUM-249	16K	12	-	-	< 0.001	-	-	< 0.000014
	7M	244	0.0035 ± 0.0019	< 0.001	0.0440	0.000050	< 0.000014	0.00063

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

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

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 1984, the hydrogen-3 content of 21 other lakes and streams ranged from $< 100 \times 10^{-9}$ µCi/mL to 338×10^{-9} µCi/mL and averaged 156×10^{-9} µCi/mL.

The total radioactive effluent discharged to the Creek in Argonne waste water can be estimated from the average net concentrations and the volume of water carried by the Creek. These totals are 1.0 Ci of hydrogen-3, 1.6 mCi of strontium-90, 0.07 mCi of neptunium-237, 0.26 mCi of plutonium-239, 0.20 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 9. 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, 1.4×10^{-9} µCi/mL and 10×10^{-9} µCi/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.

The radioactivity in samples of Illinois River water, shown in Table 10, was similar to those found previously at these same locations. No

TABLE 9

RADIONUCLIDES IN DESPLAINES RIVER WATER, 1984

TYPE OF ACTIVITY	LOCATION *	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/ML) AVG.	MIN.	MAX.	PERCENT CG AVG.	MIN.	MAX.
ALPHA (NONVOLATILE)	A	11	1.8 ± 0.3	1.1	2.4	(0.062)	(0.037)	(0.081)
	B	24	1.8 ± 0.2	0.9	2.5	(0.060)	(0.030)	(0.083)
BETA (NONVOLATILE)	A	11	15 ± 4	7	23	(0.49)	(0.23)	(0.75)
	B	24	14 ± 2	8	25	(0.45)	(0.26)	(0.85)
HYDROGEN-3	A	11	113 ± 35	< 100	195	0.0037	< 0.003	0.0065
	B	24	< 120	< 100	228	< 0.0040	< 0.003	0.008
STRONTIUM-90	A	11	< 0.29	< 0.25	0.38	< 0.096	< 0.08	0.13
	B	24	0.29 ± 0.05	< 0.25	0.66	0.10	< 0.08	0.22
URANIUM ** (NATURAL)	A	11	1.3 ± 0.3	0.8	2.0	(0.0033)	(0.0020)	(0.0050)
	B	24	1.2 ± 0.1	0.5	1.7	(0.0030)	(0.0014)	(0.0043)
NEPTUNIUM-237	A	11	< 0.0010	< 0.001	0.001	< 0.00003	< 0.00003	0.00004
	B	12	-	-	< 0.001	-	-	< 0.00003
PLUTONIUM-238	A	11	-	-	< 0.001	-	-	< 0.00002
	B	12	-	-	< 0.001	-	-	< 0.00002
PLUTONIUM-239	A	11	-	-	< 0.0010	-	-	< 0.000020
	B	12	-	-	< 0.0010	-	-	< 0.000020
AMERICIUM-241	A	11	-	-	< 0.001	-	-	< 0.000025
	B	12	-	-	< 0.001	-	-	< 0.000025
CURIUM-242 AND/OR CALIFORNIUM-252	A	11	-	-	< 0.001	-	-	< 0.000005
	B	12	-	-	< 0.001	-	-	< 0.000005
CURIUM-244 AND/OR CALIFORNIUM-249	A	11	-	-	< 0.001	-	-	< 0.000014
	B	12	-	-	< 0.001	-	-	< 0.000014

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

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

TABLE 10

Radionuclides in Illinois River Water, 1984

(Concentrations in 10^{-9} µCi/mL)

Date Collected	Location	Alpha*	Beta*	Hydrogen-3	Uranium** (natural)	Plutonium-239
June 5	McKinley Woods State Park	1.0 ± 0.2	7.5 ± 0.3	< 100	1.2 ± 0.2	< 0.001
June 5	Below Dresden Power Station	1.2 ± 0.2	1.7 ± 0.3	175 ± 93	1.2 ± 0.2	< 0.001
June 5	Morris	1.4 ± 0.2	5.1 ± 0.3	118 ± 92	1.2 ± 0.2	-
June 5	Starved Rock State Park	0.8 ± 0.2	2.8 ± 0.3	163 ± 92	1.2 ± 0.2	-
October 9	McKinley Woods State Park	0.6 ± 0.2	9.2 ± 0.3	338 ± 100	0.3 ± 0.1	< 0.001
October 9	Below Dresden Power Station	1.0 ± 0.2	7.6 ± 0.3	269 ± 99	0.5 ± 0.1	< 0.001
October 9	Morris	0.6 ± 0.2	8.3 ± 0.3	255 ± 99	0.6 ± 0.1	-
October 9	Starved Rock State Park	1.3 ± 0.2	8.0 ± 0.3	295 ± 100	0.6 ± 0.1	-

*Nonvolatile activity.

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

radioactivity originating at Argonne could be detected in the Des Plaines or Illinois Rivers.

Special samples were collected in May 1984 around the suspect landfill located at 7J in Figure 1. Material was buried in this area between January 1965 and October 1967 and consisted of items that did not show any radioactivity by survey, but could be contaminated in inaccessible areas, such as inside pipes. Water samples were collected at two drain points in the landfill area and at the point where the surface drainage leaves the site. The samples were analyzed for tritiated water and by gamma-ray spectrometry. No gamma-ray activities above ambient levels were detected. The tritium concentrations at the burial site were 1.5×10^{-5} μCi/mL and 9.4×10^{-5} μCi/mL, while the concentration at the site boundary was 3.6×10^{-5} μCi/mL. Although these concentrations are several hundred times ambient levels, they are all well below the DOE standard for tritium in water of 3×10^{-3} μCi/mL. This area will be monitored regularly to establish trends. Sediment samples collected from the drainage bed will be discussed in Section III.A.4.

3. Ground Water

The Laboratory domestic water is provided by four wells which are described in Section I.E. and the locations are illustrated in Figure 1. Samples from each well were collected quarterly at the well head and analyzed for several types of radioactivity. The 1984 results are in Table 11. In addition to the well water samples, one tap water sample was collected and the results are also in Table 11.

Since the Laboratory is a "non-community water system",[10] the EPA standards for this type of system apply. For the nuclides measured in Table 11, the following limits are established:

Gross alpha particle activity	15 pCi/L (10^{-9} μCi/mL)
Gross beta particle activity	15 pCi/L (10^{-9} μCi/mL)
Hydrogen-3	2×10^{4} pCi/L (10^{-9} μCi/mL)
Strontium-90	8 pCi/L (10^{-9} μCi/mL)
Radium-226	5 pCi/l (10^{-9} μCi/mL)

TABLE 11

Radioactivity in ANL Domestic Wells, 1984

(Concentrations in 10^{-9} μCi/mL)

Type of Activity	Location	No. of Samples	Avg.	Min.	Max.
Alpha (nonvolatile)	Well #1	4	3.6 ± 1.2	1.8	4.5
	Well #2	4	4.8 ± 1.1	3.4	6.0
	Well #3	4	2.8 ± 0.9	1.6	3.6
	Well #4	4	2.3 ± 1.3	0.6	3.4
	Tap	1	±	-	0.44
Beta (nonvolatile)	Well #1	4	7.0 ± 1.3	8.4	5.4
	Well #2	4	8.0 ± 0.5	6.7	8.7
	Well #3	4	6.4 ± 0.7	5.8	7.2
	Well #4	4	6.4 ± 0.8	5.2	6.9
	Tap	1	±	-	5.8
Hydrogen-3	Well #1	4	264 ± 59	184	322
	Well #2	4	126 ± 36	< 100	177
	Well #3	4	108 ± 11	< 100	123
	Well #4	4	±	-	< 100
	Tap	1	-	-	190
Strontium-90	Well #1	1	-	-	< 0.25
	Well #2	1	-	-	< 0.25
	Well #3	1	-	-	< 0.25
	Well #4	1	-	-	< 0.25
	Tap	1	-	-	< 0.25
Radium-226	Well #1	1	-	-	1.16
	Well #2	1	-	-	1.29
	Well #3	1	-	-	0.83
	Well #4	1	-	-	0.78
	Tap	1	-	-	0.06
Uranium-234	Well #1	1	-	-	0.22
	Well #2	1	-	-	0.16
	Well #3	1	-	-	0.17
	Well #4	1	-	-	0.11
	Tap	1	-	-	0.06
Uranium-238	Well #1	1	-	-	0.14
	Well #2	1	-	-	0.09
	Well #3	1	-	-	0.11
	Well #4	1	-	-	0.05
	Tap	1	-	-	0.04

The uranium results would be covered by the gross alpha standard. Inspection of Table 11 indicates that all measurements are well within the EPA drinking water standards. This program is being conducted to demonstrate the Laboratory's compliance with the EPA drinking water regulations.

Wells No. 1 and No. 2 had measurable levels of hydrogen-3 at various times during the year, although the average concentration was only 1% of the EPA Standard. It is speculated that the source of the hydrogen-3 was from liquid wastes that were placed in holding ponds in the sewage treatment area (Location 10M in Figure 1) in the 1950's. The tritiated water migrated down through the soil to the dolomite, and was drawn into the wells. Well No. 1, which is about 200 m north of the treatment area, had higher hydrogen-3 concentrations than Well No. 2, which is about 300 m from the treatment area. Although the normal subsurface water flow gradient is in the south direction, the cone of depression created by the pumping on these wells would overpower the normal pattern. The holding ponds have not been used for a number of years. Two hydrogen-3 results from Well No. 3 were very slightly above the detection limit, but these are considered to be within the normal fluctuation range for the measurement of hydrogen-3 in water.

In the 1982 monitoring report,[11] an unusual occurrence was reported which consisted of the loss from the A^2R^2 excavation of about 7.5 megaliters (2 million gallons) of ponded water containing about 26 mCi of tritiated water. Quarterly monitoring of wells and a seep likely to be in the subsurface flow path of this water continued during 1984 and the results are summarized in Table 12. The concentrations of tritiated water were in the normal range and none was detected that could be attributed to this release. In 1982, elevated concentrations, up to 1200 x 10^{-9} µCi/mL, were measured in Well No. 9 from this release. The concentration in the tritiated water that remains in the A^2R^2 excavation continued to decrease from the 1.2 x 10^{-6} µCi/mL in 1983 to 0.9 x 10^{-6} µCi/L in 1984 and the excavation is being filled with building rubble as it becomes available. Monitoring will continue in the wells and the excavation. The wells are used to measure water levels, and are not a source of drinking water.

TABLE 12

Tritium Content of Water Near A^2R^2 Excavation, 1984
(Concentrations in 10^{-9} μCi/mL)

Sample	Location*	No. of Samples	Avg.	Min.	Max.
Well #9	9F	4	110 ± 12	< 100	128
Well #10	11J	4	129 ± 58	< 100	217
Well D181	8F	4	-	-	< 100
Well D185	8E	4	-	-	< 100
Seep	4EF	4	117 ± 22	< 100	147
A^2R^2	10G	4	910 ± 31	864	932

*See Figure 1.

4. Soil, Grass, and Bottom Sediment

The radioactive content of soil, grass, and bottom sediment was measured at the site perimeter and off the site. The purpose of the off-site sampling was to measure deposition for comparison with perimeter samples, and with results obtained by other organizations for samples collected at large distances from nuclear installations. Such comparisons are useful in determining if the soil activity near Argonne is normal. For this purpose, the ASTM site selection criteria, sample collection, and sample preparation techniques were used.[12] Sites were selected in several directions and at various distances from the Laboratory. Each site was selected on the basis that the soil appeared, or was known to have been, undisturbed for a number of years. Attempts were made to select open, level, grassy areas that were mowed at reasonable intervals. Public parks were selected when available.

Each soil sample consisted of ten cores totaling 864 cm^2 in area by 5 cm deep. Through 1976, samples had been collected down to 30 cm to measure total deposition and as a result of five years of sample collection

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

The results for the gamma-ray emitting nuclides in soil are presented in Table 13. The antimony-125 is a residual from past atmospheric nuclear tests by the People's Republic of China. Other intermediate half-life fission products reported previously have decayed to below their detection limits. The cesium-137 levels are similar to those found over the past several years, and represent an accumulation from nuclear tests over a period of many years. The annual average concentrations for the perimeter and off-site samples are similar. The plutonium and americium concentrations are given in Table 14. The ranges and concentrations of plutonium and americium in soil are similar at both perimeter and off-site locations. For fallout americium-241 in soil, about 10% is due to direct deposition, while about 90% 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.[13]

Composite monthly precipitation samples were analyzed for plutonium-239. The results are given in Table 15, along with results since 1973 for comparison. The 1984 deposition by precipitation was equivalent to 0.1% of the total plutonium deposited through 1976, which is 2.2×10^{-3} $\mu Ci/m^2$.[14] Deposition since 1976 has not changed this value significantly. The data in Table 15 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 twelve years in Figure 4 shows excellent correlation.

TABLE 13

Gamma-Ray Emitting Radionuclides in Soil, 1984

(Concentrations in 10^{-6} μCi/g)

Date Collected	Location	Potassium-40	Antimony-125	Cesium-137	Radium-226 (Bismuth-214)	Thorium-228 (Lead-212)	Thorium-232 (Actinium-228)
	Perimeter*						
June 8	7M	15.31 ± 0.53	0.31 ± 0.10	0.48 ± 0.03	1.10 ± 0.06	0.73 ± 0.03	0.63 ± 0.07
June 8	13N	16.82 ± 0.36	0.50 ± 0.08	1.03 ± 0.02	1.17 ± 0.04	1.01 ± 0.02	0.89 ± 0.05
June 8	15H	16.86 ± 0.58	0.37 ± 0.10	0.26 ± 0.02	1.09 ± 0.06	0.98 ± 0.04	0.89 ± 0.08
June 8	13C/D	21.74 ± 0.77	0.62 ± 0.13	0.88 ± 0.04	1.66 ± 0.07	1.30 ± 0.05	0.93 ± 0.10
June 8	8G	18.75 ± 0.73	0.58 ± 0.13	1.23 ± 0.04	1.55 ± 0.07	1.22 ± 0.05	0.90 ± 0.09
October 23	12C	18.63 ± 0.74	0.46 ± 0.11	0.99 ± 0.04	1.75 ± 0.07	1.26 ± 0.05	0.92 ± 0.10
October 23	14L	18.73 ± 0.90	0.38 ± 0.13	0.35 ± 0.03	1.17 ± 0.08	1.04 ± 0.05	0.82 ± 0.12
October 23	12-0	15.35 ± 0.55	0.34 ± 0.09	0.94 ± 0.04	1.13 ± 0.06	0.98 ± 0.04	0.79 ± 0.08
October 23	8N	10.59 ± 0.46	0.33 ± 0.09	0.29 ± 0.02	0.69 ± 0.05	0.48 ± 0.03	0.39 ± 0.06
October 23	5D	15.60 ± 0.62	0.30 ± 0.12	0.85 ± 0.04	1.06 ± 0.06	0.94 ± 0.04	0.88 ± 0.09
	Average	16.84 ± 1.88	0.42 ± 0.07	0.73 ± 0.22	1.24 ± 0.20	0.99 ± 0.16	0.80 ± 0.11
	Off-Site						
June 5	Dresdan Lock & Dam, IL	17.19 ± 0.75	0.48 ± 0.12	1.15 ± 0.05	1.22 ± 0.08	0.86 ± 0.04	0.79 ± 0.10
June 5	McKinley Woods State Park, IL	21.86 ± 0.67	0.52 ± 0.10	< 0.1	1.39 ± 0.07	0.94 ± 0.03	0.97 ± 0.09
June 12	Saganashkee Slough, IL	20.98 ± 0.93	0.68 ± 0.16	0.56 ± 0.04	1.52 ± 0.09	1.07 ± 0.05	0.89 ± 0.11
June 12	McGinnis Slough, IL	22.63 ± 0.77	0.54 ± 0.13	0.83 ± 0.04	1.75 ± 0.07	1.15 ± 0.04	0.83 ± 0.09
June 12	Willow Springs, IL	20.92 ± 0.68	0.44 ± 0.11	0.29 ± 0.02	1.76 ± 0.07	1.12 ± 0.04	0.96 ± 0.09
October 9	Channahon, IL	14.02 ± 0.54	0.39 ± 0.10	0.30 ± 0.02	0.93 ± 0.05	0.66 ± 0.03	0.56 ± 0.07
October 9	Morris, IL	15.59 ± 0.55	0.45 ± 0.10	0.24 ± 0.02	1.52 ± 0.07	0.86 ± 0.03	0.78 ± 0.08
October 9	Starved Rock State Park, IL	16.94 ± 0.72	0.59 ± 0.13	0.90 ± 0.04	1.29 ± 0.07	1.15 ± 0.04	0.94 ± 0.10
October 11	Romeoville, IL	17.14 ± 0.75	0.40 ± 0.12	1.43 ± 0.07	1.46 ± 0.07	1.16 ± 0.05	0.92 ± 0.10
October 11	Naperville, IL	13.92 ± 0.84	0.40 ± 0.13	1.25 ± 0.05	1.11 ± 0.08	1.07 ± 0.05	0.84 ± 0.12
	Average	18.12 ± 2.05	0.49 ± 0.06	0.71 ± 0.30	1.40 ± 0.17	1.00 ± 0.11	0.85 ± 0.08

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

TABLE 14

Transuranics in Soil, 1984

Date Collected	Location	Plutonium-238		Plutonium-239		$^{238}Pu/^{239}Pu$	Americium-241		$^{241}Am/^{239}Pu$
		10^{-9} µCi/g	10^{-3} µCi/m^2	10^{-9} µCi/g	10^{-3} µCi/m^2		10^{-9} µCi/g	10^{-3} µCi/m^2	
	Perimeter*								
June 8	7M	0.4 ± 0.2	0.022 ± 0.010	9.6 ± 0.8	0.499 ± 0.039	0.043	1.8 ± 0.4	0.095 ± 0.019	0.191
June 8	13N	1.2 ± 0.3	0.052 ± 0.012	21.9 ± 1.3	0.970 ± 0.058	0.054	1.9 ± 0.3	0.083 ± 0.013	0.086
June 8	15H	0.3 ± 0.1	0.016 ± 0.006	4.7 ± 0.4	0.225 ± 0.017	0.072	0.8 ± 1.0	0.037 ± 0.046	0.170
June 8	13C/D	0.6 ± 0.2	0.026 ± 0.009	15.9 ± 1.1	0.698 ± 0.047	0.038	3.1 ± 2.9	0.137 ± 0.126	0.200
June 8	8G	0.8 ± 0.2	0.046 ± 0.013	21.2 ± 1.2	1.176 ± 0.067	0.039	3.4 ± 1.7	0.191 ± 0.093	0.162
October 23	12C	0.8 ± 0.2	0.039 ± 0.012	18.0 ± 1.3	0.873 ± 0.061	0.044	1.2 ± 0.4	0.061 ± 0.022	0.070
October 23	14L	0.4 ± 0.2	0.022 ± 0.010	6.8 ± 0.6	0.426 ± 0.037	0.053	1.4 ± 0.3	0.086 ± 0.020	0.204
October 23	12-O	1.1 ± 0.4	0.046 ± 0.014	20.6 ± 1.5	0.892 ± 0.064	0.052	2.8 ± 0.4	0.124 ± 0.018	0.138
October 23	8N	0.5 ± 0.2	0.026 ± 0.009	9.1 ± 0.7	0.480 ± 0.036	0.055	1.8 ± 0.8	0.092 ± 0.046	0.192
October 23	5D	0.8 ± 0.2	0.033 ± 0.008	17.4 ± 1.0	0.753 ± 0.046	0.043	2.3 ± 0.4	0.099 ± 0.017	0.131
	Average	0.7 ± 0.2	0.033 ± 0.008	14.5 ± 4.0	0.699 ± 0.184	0.049	2.1 ± 0.5	0.101 ± 0.026	0.154
	Off-Site								
June 5	Dresden Lock and Dam, IL	1.2 ± 0.6	0.045 ± 0.025	21.4 ± 2.7	0.839 ± 0.108	0.054	2.7 ± 2.8	0.107 ± 0.111	0.127
June 5	McKinley Woods State Park, IL	0.6 ± 0.2	0.030 ± 0.011	2.3 ± 0.4	0.124 ± 0.019	0.244	1.8 ± 0.3	0.095 ± 0.016	0.760
June 12	Saganashkee Slough, IL	0.4 ± 0.2	0.015 ± 0.007	10.5 ± 0.8	0.446 ± 0.032	0.034	2.2 ± 0.7	0.095 ± 0.030	0.213
June 12	McGinnis Slough, IL	0.8 ± 0.3	0.035 ± 0.011	15.2 ± 1.2	0.693 ± 0.054	0.050	2.5 ± 0.5	0.113 ± 0.024	0.163
June 12	Flagg Creek, Hinsdale, IL	0.3 ± 0.2	0.014 ± 0.008	5.3 ± 0.5	0.250 ± 0.025	0.057	1.8 ± 0.5	0.086 ± 0.023	0.343
October 9	DuPage River, Channahon, IL	0.1 ± 0.2	0.005 ± 0.008	5.7 ± 0.6	0.256 ± 0.029	0.021	2.7 ± 0.5	0.122 ± 0.022	0.476
October 9	Illinois River, Morris, IL	0.3 ± 0.2	0.015 ± 0.008	6.0 ± 0.6	0.306 ± 0.030	0.050	0.9 ± 0.2	0.045 ± 0.011	0.145
October 9	Starved Rock State Park, IL	0.5 ± 0.4	0.027 ± 0.020	14.0 ± 1.5	0.723 ± 0.079	0.037	3.3 ± 0.5	0.171 ± 0.024	0.236
October 11	Romeoville, IL	1.0 ± 0.3	0.033 ± 0.010	28.3 ± 1.8	0.976 ± 0.060	0.034	5.9 ± 0.8	0.204 ± 0.029	0.209
October 11	Naperville, IL	0.7 ± 0.5	0.024 ± 0.017	23.4 ± 2.3	0.817 ± 0.081	0.029	3.6 ± 0.5	0.127 ± 0.018	0.155
	Average	0.6 ± 0.2	0.024 ± 0.008	13.2 ± 5.6	0.543 ± 0.190	0.061	2.7 ± 0.9	0.117 ± 0.028	0.283

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

TABLE 15

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

Month	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984
January	-	0.60	1.1	0.25	0.17	0.37	0.57	0.15	0.06	0.23	0.53	< 0.1
February	0.14	1.1	1.7	0.38	0.15	0.12	0.43	0.21	0.43	< 0.1	0.23	0.20
March	0.27	2.0	1.5	0.58	0.74	4.7	-	0.50	0.48	< 0.1	0.27	0.16
April	0.40	5.2	4.0	0.21	1.0	3.3	1.78	0.56	3.70	< 0.1	0.10	0.14
May	0.50	4.6	1.8	0.58	1.8	4.7	0.58	0.81	1.43	1.10	0.26	0.28
June	0.16	3.5	1.9	0.65	1.2	3.0	0.73	0.54	1.11	0.69	0.11	0.28
July	0.32	5.6	0.48	0.26	0.71	1.7	0.45	0.67	0.51	-	0.27	0.10
August	0.34	1.0	0.38	0.15	1.6	0.54	0.54	0.62	0.40	0.55	0.11	0.10
September	0.27	0.25	0.10	0.06	1.5	0.69	0.21	0.65	0.29	0.37	0.13	< 0.1
October	0.30	0.45	0.12	0.30	0.85	0.34	0.53	0.53	0.36	0.16	0.18	< 0.1
November	0.22	0.73	0.08	0.05	0.72	0.74	0.09	0.13	0.24	0.13	0.33	< 0.1
December	0.46	0.71	0.21	0.12	0.43	0.40	0.13	0.12	0.07	0.13	0.19	< 0.1
Average Monthly Deposition	0.31 ± 0.07	2.1 ± 1.2	1.1 ± 0.7	0.30 ± 0.13	0.91 ± 0.31	1.7 ± 1.0	0.55 ± 0.27	0.46 ± 0.14	0.76 ± 0.26	0.33 ± 0.13	0.23 ± 0.12	0.15 ± 0.04
Annual Deposition	3.38	25.74	13.37	3.59	10.87	20.60	6.04	5.49	9.08	3.66	2.71	1.76
Percent Added to Existing	0.2	1.2	0.6	0.2	0.5	0.9	0.3	0.3	0.4	0.2	0.1	0.1

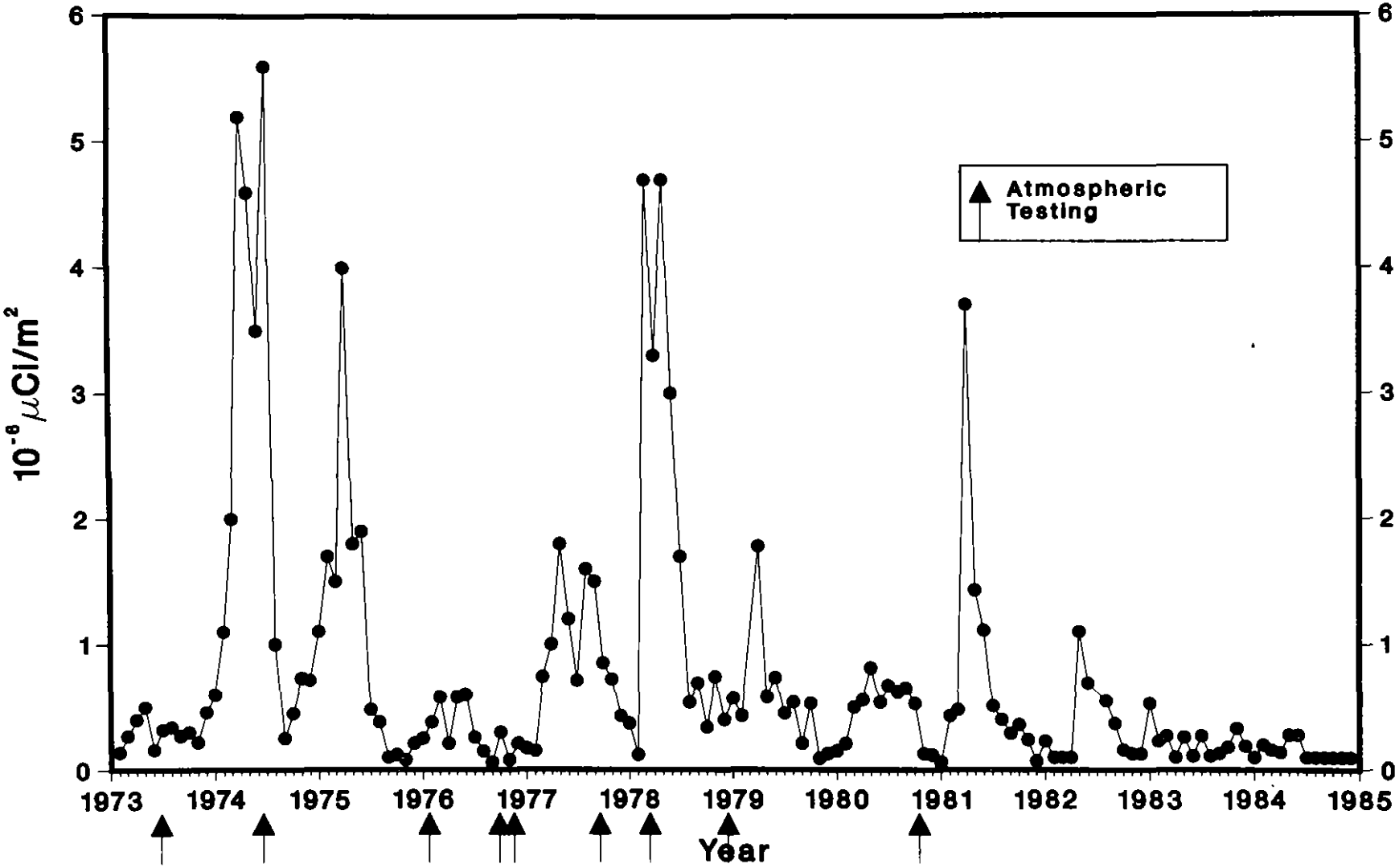

Fig. 5. Plutonium-239,240 Surface Deposition, 1973-1984

The results of radionuclide concentrations measured in grass are given in Table 16. The annual averages and concentration ranges were similar at the perimeter and off-site locations as well as similar to previous years, indicating no contribution from ANL operations. In terms of deposition, the plutonium-239 concentration was a factor of about 10^4 less in the grass than in the soil from the same location.

Results of analysis of bottom sediment samples for gamma-ray emitters and transuranics are given in Table 17. The annual off-site averages are in the same range found in off-site samples collected in previous years. The cesium-137 and plutonium content of the pond at Location 15H is similar to concentrations in off-site ponds of the same approximate size and composition. Plutonium results vary widely between locations and are strongly dependent on the retentiveness of the bottom material.

A set of samples was collected on July 27, 1984, 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 17 and indicate that the sample above the 7M outfall is similar to the off-site samples. The plutonium and americium concentrations are the highest at the outfall and decrease rapidly with distance, indicating their origin in Argonne waste water. Similar sets of samples were collected on August 11, 1983, September 15, 1982, and September 24, 1980. Comparison of plutonium concentrations indicates that the 1982 results were an order of magnitude higher at the outfall, but similar farther downstream, while the 1980 and 1983 results were more like the 1984 set. The changes in concentrations of the various nuclides with time indicates the dynamic nature of the sediment material in this area.

Three sediment samples were collected at the same locations water samples were obtained at the D-319 suspect landfill, see Section III.A.2. The samples were prepared as previously described and the radioactivity determined by gamma-ray spectrometry. The results are in Table 18. Very small amounts of cesium-137 and possibly cobalt-60 were measured in the sediment at the base of the buried material. However, sediment at the boundary has concentrations of these nuclides that are at ambient levels. Monitoring will continue to determine the status of this area.

TABLE 16

Radionuclides in Grass, 1984

Date Collected	Location	(Concentrations in 10^{-9} µCi/g) Cesium-137	Plutonium-239	(10^{-6} µCi/m^2) Deposition of Plutonium-239
	Perimeter*			
June 8	7M	25 ± 25	< 0.1	< 0.01
June 8	13N	10 ± 20	< 0.1	< 0.01
June 8	15H	< 10	1.8 ± 0.5	0.391 ± 0.102
June 8	13C/D	20 ± 20	0.1 ± 0.3	0.022 ± 0.050
June 8	8G	20 ± 30	< 0.1	< 0.01
October 23	12C	< 10	0.1 ± 0.1	0.011 ± 0.011
October 23	14L	< 10	0.6 ± 0.2	0.059 ± 0.023
October 23	12-O	< 10	0.2 ± 0.2	0.018 ± 0.014
October 23	8N	20 ± 30	< 0.1	< 0.01
October 23	5D	10 ± 20	0.1 ± 0.1	0.012 ± 0.011
	Average	13 ± 5	0.3 ± 0.3	0.055 ± 0.075
	Off-Site			
June 5	Dresden Lock and Dam, IL	10 ± 20	1.9 ± 0.5	0.159 ± 0.0401
June 5	McKinley Woods State Park, IL	20 ± 30	< 0.1	< 0.01
June 12	Saganashkee Slough, IL	< 10	< 0.1	< 0.01
June 12	McGinnis Slough, IL	25 ± 25	0.2 ± 0.1	0.057 ± 0.033
June 12	Hinsdale, IL	30 ± 25	< 0.1	< 0.01
October 9	Channahon, IL	10 ± 20	0.5 ± 0.2	0.075 ± 0.023
October 9	Morris, IL	< 10	0.4 ± 0.2	0.036 ± 0.023
October 9	Starved Rock State Park, IL	< 10	0.4 ± 0.2	0.028 ± 0.015
October 11	Romeoville, IL	< 10	2.2 ± 0.3	0.609 ± 0.087
October 11	Naperville, IL	10 ± 20	0.2 ± 0.1	0.054 ± 0.026
	Average	12 ± 6	0.6 ± 0.5	0.105 ± 0.116

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

TABLE 17

Radionuclides in Bottom Sediment, 1984

Date Collected	Location	Concentrations in 10^{-6} µCi/g Antimony-125	Cesium-137	Radium-226	Thorium-228	Thorium-232	Concentrations in 10^{-9} µCi/g Plutonium-238	Plutonium-239	Americium-241
	Perimeter*								
June 8	Pond - 15H	0.32 ± 0.11	1.73 ± 0.05	0.85 ± 0.05	0.57 ± 0.03	0.45 ± 0.06	1.9 ± 0.4	38.0 ± 2.3	3.3 ± 0.4
July 27	50 m Above ANL Outfall	0.27 ± 0.11	0.09 ± 0.02	0.96 ± 0.07	0.42 ± 0.03	0.48 ± 0.08	< 0.1	3.6 ± 0.1	< 0.1
July 27	7M Outfall	0.19 ± 0.08	0.78 ± 0.03	0.75 ± 0.05	0.42 ± 0.03	0.42 ± 0.06	8.4 ± 0.6	89.6 ± 3.3	9.9 ± 0.3
July 27	50 m Below ANL Outfall	0.42 ± 0.13	0.07 ± 0.02	0.90 ± 0.07	0.38 ± 0.03	0.50 ± 0.08	1.3 ± 0.4	11.5 ± 1.2	1.9 ± 0.8
July 27	100 m Below ANL Outfall	0.29 ± 0.11	0.11 ± 0.02	0.96 ± 0.07	0.40 ± 0.03	0.59 ± 0.09	1.1 ± 0.6	9.9 ± 1.6	0.9 ± 1.6
July 27	500 m Below ANL Outfall	0.24 ± 0.09	0.21 ± 0.02	0.82 ± 0.06	0.40 ± 0.03	0.47 ± 0.07	1.4 ± 0.3	16.1 ± 1.1	3.0 ± 1.2
	Off-Site								
June 5	DuPage River, Channahon, IL	0.63 ± 0.12	0.26 ± 0.02	1.42 ± 0.06	1.03 ± 0.03	1.04 ± 0.09	0.6 ± 0.3	7.1 ± 0.8	3.1 ± 0.2
June 12	Saganashkee Slough, IL	0.38 ± 0.10	0.16 ± 0.02	1.49 ± 0.07	0.98 ± 0.03	0.87 ± 0.08	1.0 ± 0.3	17.7 ± 1.3	5.6 ± 0.5
June 12	McGinnis Slough, IL	0.34 ± 0.10	0.78 ± 0.04	1.16 ± 0.07	0.70 ± 0.04	0.67 ± 0.09	0.5 ± 0.2	4.2 ± 0.6	6.6 ± 0.4
October 9	Illinois River, Morris, IL	0.27 ± 0.08	0.02 ± 0.01	0.46 ± 0.04	0.49 ± 0.03	0.45 ± 0.07	< 0.1	0.4 ± 0.2	< 0.1
October 9	Illinois River, Starved Rock State Park, IL	0.31 ± 0.09	< 0.1	0.60 ± 0.06	0.32 ± 0.03	0.22 ± 0.07	< 0.1	0.2 ± 0.3	< 0.1
October 11	Long Run Creek, Lemont, IL	0.44 ± 0.09	0.25 ± 0.02	0.97 ± 0.06	0.68 ± 0.03	0.58 ± 0.07	0.5 ± 0.2	7.5 ± 0.6	1.5 ± 0.7
October 11	Des Plaines River, Romeoville, IL	0.59 ± 0.16	0.46 ± 0.04	1.82 ± 0.09	1.14 ± 0.05	1.08 ± 0.12	0.5 ± 0.3	7.4 ± 0.9	1.1 ± 0.8
October 11	DuPage River, Naperville, IL	0.83 ± 0.19	0.27 ± 0.04	1.40 ± 0.09	1.88 ± 0.06	1.51 ± 0.12	1.2 ± 0.4	18.0 ± 1.6	2.6 ± 0.4
	Average	0.48 ± 0.14	0.28 ± 0.18	1.17 ± 0.33	0.90 ± 0.34	0.80 ± 0.29	0.6 ± 0.3	7.8 ± 4.8	2.6 ± 1.7

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

TABLE 18

Gamma-Ray Emitting Radionuclides in Landfill Sediment Samples, 1984
(Concentrations in 10^{-6} μCi/g)

	319 Drain	317 Drain	Drain at Boundary
Potassium-40	19.4 ± 0.9	17.5 ± 0.9	20.1 ± 0.9
Cobalt-60	0.13 ± 0.07	< 0.1	0.10 ± 0.07
Cesium-137	1.57 ± 0.06	0.22 ± 0.03	0.29 ± 0.03
Radium-226 (Bismuth-214)	1.04 ± 0.08	1.26 ± 0.08	1.32 ± 0.08
Thorium-228 (Lead-212)	0.89 ± 0.05	0.97 ± 0.05	1.06 ± 0.05
Thorium-232 (Actinium-228)	0.88 ± 0.12	0.80 ± 0.11	0.89 ± 0.12

5. Milk

Raw milk was collected monthly from a local dairy farm south of Lemont and analyzed for several radioactive nuclides. Cesium-137 was analyzed with a detection limit of 5×10^{-10} μCi/mL by an ion-exchange separation followed by gamma-ray spectrometry. The other nuclides were analyzed by the same methods used for water and with the same detection limits. The results are given in Table 19. The average strontium-90 and cesium-137 concentrations were similar to the 1983 results. These nuclides are fission products from nuclear tests and their presence in milk is not related to Argonne operations.

The concentrations given in Table 19 may be compared to the CGs for drinking water given in the Appendix, Part C. The drinking water CGs are based on an intake of 2.2 liters per day. The consumption of one liter of milk per day would result in an average intake of 0.5% of the strontium-90 and 0.003% of the cesium-137 and hydrogen-3 Concentration Guides.

TABLE 19

Radionuclides in Milk, 1984
(Concentrations in 10^{-9} µCi/ml)

Date Collected	Hydrogen-3	Strontium-90	Cesium-137
January 4	155 ± 89	3.1 ± 0.4	0.5 ± 0.4
February 1	120 ± 89	2.7 ± 0.4	0.8 ± 0.3
March 7	124 ± 89	2.8 ± 0.4	0.8 ± 0.3
April 4	< 100	2.0 ± 0.4	0.6 ± 0.3
May 2	< 100	0.8 ± 0.4	1.0 ± 0.3
June 6	106 ± 175	4.2 ± 0.6	0.9 ± 0.3
July 11	< 100	3.3 ± 0.3	0.6 ± 0.5
August 1	< 100	3.8 ± 1.4	1.3 ± 0.2
September 5	154 ± 177	3.3 ± 1.5	0.4 ± 0.4
October 10	< 100	2.4 ± 2.1	0.5 ± 0.2
November 7	< 100	3.0 ± 1.4	0.6 ± 0.2
December 5	< 100	2.4 ± 0.7	0.2 ± 0.2
Average	< 113	2.8 ± 0.5	0.7 ± 0.2

6. External Penetrating Radiation

Measurements were made with calcium fluoride and lithium fluoride thermoluminescent dosimeter (TLD) chips. Each measurement was the average of four chips exposed in the same packet. The response of the chips was determined with a National Bureau of Standards standard radium-226 source, and the results calculated in terms of air dose. Dosimeters were exposed at a number of locations at the site boundary and on the site. 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 20 and 21, and the site boundary and on-site readings are also shown in Figure 6. Measurements were made

for the four successive exposure periods shown in the tables and the results were calculated in terms of annual dose for ease in comparing measurements made for different elapsed times. The uncertainty given in the tables for an average is the 95% confidence limit calculated from the standard deviation of the average.

TABLE 20

Environmental Penetrating Radiation at Off-Site Locations, 1984

	Dose Rate (mrem/year)				
	Period of Measurement				
Location	1/24-4/17	4/17-7/18	7/18-10/16	10/16-1/8	Average
Downers Grove	76	84	88	95	86 ± 15
Lemont	85	82	102	92	90 ± 16
Lombard	82	93	96	90	90 ± 11
Oak Brook	89	97	103	97	96 ± 11
Oak Lawn	74	75	83	82	79 ± 9
Average	81 ± 9	86 ± 12	94 ± 12	91 ± 8	88 ± 11

The off-site results averaged 88 ± 11 mrem/y, within the range found previously at the same locations. If the off-site locations are an accurate sample of the radiation background in the area, then annual averages at the site in the range of 88 ± 11 mrem/y may be considered normal with a 95% probability. To compare boundary results for individual sampling periods, the standard deviation of the 20 individual off-site results is useful. This value is 8.6 mrem/y, so that individual results in the range of 88 ± 17 mrem/y 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

TABLE 21

Environmental Penetrating Radiation at ANL, 1984

Location	Dose Rate (mrem/year)				
	Period of Measurement				
	1/24-4/17	4/17-7/18	7/18-10/16	10/16-1/8	Average
14L - Boundary	75	79	82	79	79 ± 5
14I - Boundary	118	122	123	114	119 ± 6
14G - Boundary	80	85	89	87	85 ± 7
9/10EF - Boundary	77	85	89	-	84 ± 19
8H - Boundary	97	105	110	101	103 ± 10
8H - Boundary, Center, St. Patrick's Cemetery	94	105	110	101	102 ± 12
7I - Boundary	296	474	593	463	457 ± 221
6I - 200 m N of Quarry Road	86	121	117	102	107 ± 29
9L - Boundary	64	69	75	72	70 ± 9
9H - 50 m SE of CP-5	539	582	569	556	561 ± 34
8H - 65 m S of Building 316	86	112	94	100	98 ± 20
8H/I - 200 m NW of Waste Storage Area	108	127	134	119	122 ± 21
7I - Center, Waste Storage Area	5,750	5,140	5,190	5,700	5,450 ± 590
10/11K - Lodging Facilities	68	66	75	75	71 ± 9
9J - Between ZGS Condenser and Building 370	56	49	66	62	58 ± 14
13J - 135 m E of Building 202	159	166	177	155	164 ± 18
12M - 30 m W of Building 55	75	65	72	72	71 ± 8
9I - 65 m NE of Building 350, 230 m NE of Building 316	80	132	103	88	101 ± 42

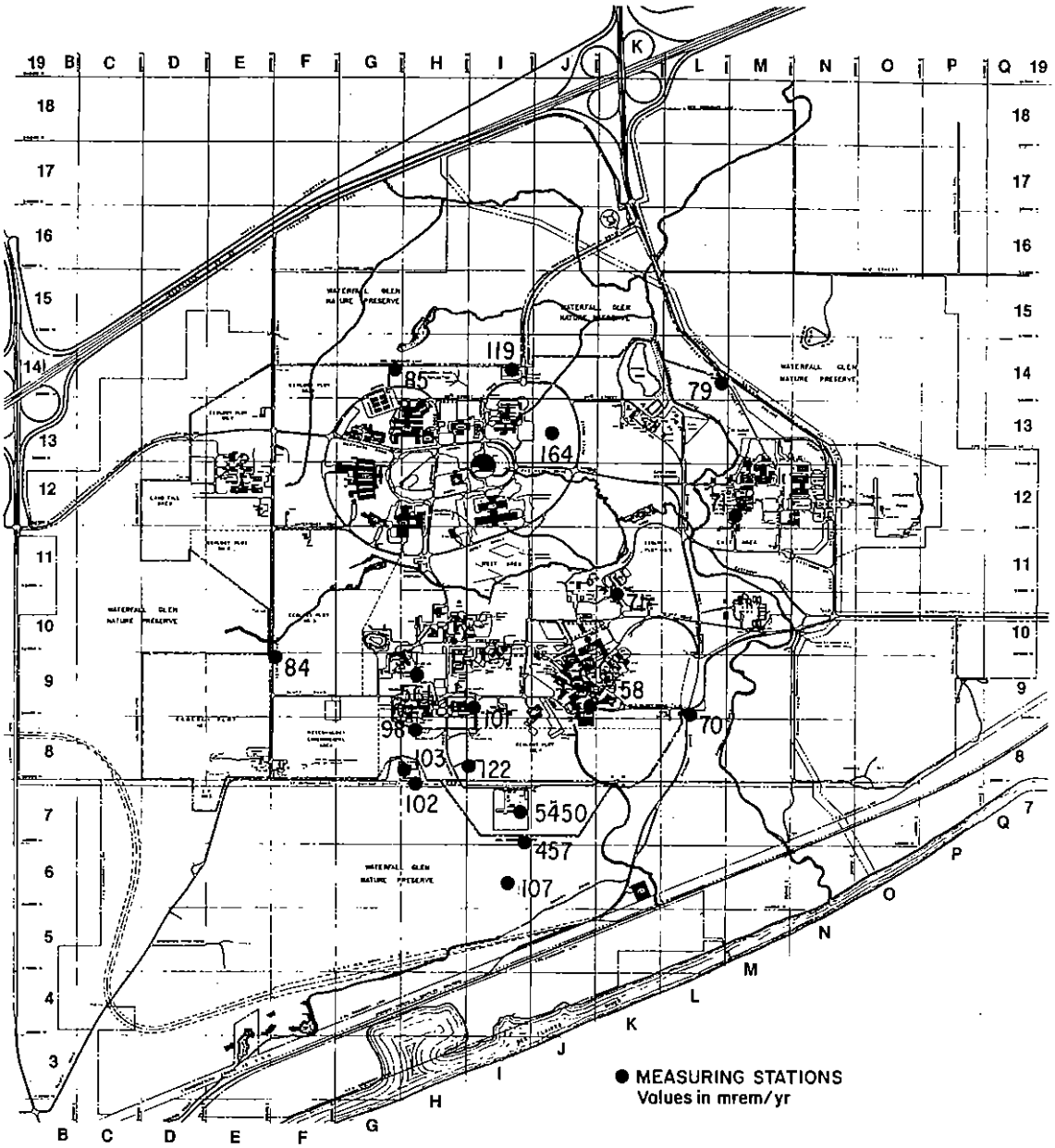

Fig. 6. Penetrating Radiation Measurements at the ANL Site, 1984

grid 7I. Waste is packaged and temporarily kept in this area prior to removal for permanent storage elsewhere. The net above-normal dose at this location was about 370 mrem/y, about twice the 1983 average. This increase resulted from an unusually large amount of repackaging of waste in this area. In previous years, this value has ranged from 560 mrem/y in 1980 to 114 mrem/y in 1977. About 300 m (0.2 mi) south of the fence in grid 6I, the measured dose dropped to 107 ± 29 mrem/y, close to normal levels. The above-normal dose at the 8H/I Location, 20 m NW of the Waste Storage Area, about 25 mrem/y, is also due to the Storage Area, as discussed in the 1982 report.[11] At Location 14I, at the north boundary, the dose rate was 31 mrem/y above normal, about 50% higher than in 1983. This dose is due to the use of cobalt-60 irradiation sources in Building 202.

The dose in the south portion of grid 8H is of interest since this area includes St. Patrick's Cemetery, which was in use before Argonne was constructed and is open to visitors. In 1984, as in previous years, this dose is estimated to be 5 to 10 mrem/y above the off-site average. Possible explanations are that the 8H dose rates are natural (since the differences between the off-site and 8H averages are not statistically significant at the 95% confidence level) or that the monument stones in the Cemetery produce the elevated rates (since above normal dose rates were obtained from one of the large red granite stones, and granite is known to contain above average levels of natural thorium and its decay products).

7. Potential Radiation Dose Estimates

a. Radionuclides in Air and Water

The radiation doses at the site boundary and off the site that could have been received by the public from radioactive materials leaving the site were calculated by two methods. Where measured radionuclide concentrations in water were available, conversion of concentration to dose was based on the ratio of environmental concentrations to the Concentrations Guides given in Table 22. This table gives the annual radiation doses (after 70 years of intake for long-lived nuclides in bone) that would result from continuous exposure at the specified concentrations for those nuclides whose presence in the environment are attributable to Argonne.

TABLE 22

Concentration-to-Dose Conversion Factors

Nuclide	Medium	Concentration* (μCi/mL)	Dose (rem)	Critical Organ
Americium-241	Water	4×10^{-6}	1.5	Kidney
	Water	5×10^{-6}	3	Bone
Argon-41	Air	4×10^{-8}	0.5	Whole Body
Californium-249	Water	4×10^{-6}	3	Bone
Californium-252	Water	7×10^{-6}	1.5	GI (LLI)
	Water	2×10^{-5}	3	Bone
Carbon-11	Air	6×10^{-8}	0.5	Whole Body
Curium-242	Water	2×10^{-5}	1.5	GI (LLI)
	Water	2×10^{-4}	3	Bone
Curium-244	Water	7×10^{-6}	3	Bone
Hydrogen-3 (H_2O)	Air	2×10^{-7}	0.5	Whole Body
	Water	3×10^{-3}	0.5	Whole Body
Krypton-85	Air	3×10^{-7}	0.5	Whole Body
Neptunium-237	Water	3×10^{-6}	3	Bone
Plutonium-238	Water	5×10^{-6}	3	Bone
Plutonium-239	Water	5×10^{-6}	3	Bone
Radon-220	Air	1×10^{-8}	0.5	Whole Body
Strontium-90	Water	3×10^{-7}	3	Bone

*The concentrations, except as noted below, are the Concentration Guides specified in DOE Order 5480.1, Chapter XI for individuals in uncontrolled areas. For americium-241 (bone), californium-252 (GI and bone), and curium-242 (bone) the concentrations listed are one-tenth of the 168 hour occupational values specified by the ICRP.[17] For carbon-11, the concentration is taken from Reference 18. The concentration-to-dose conversion factors are based on ICRP Publication 2 methodology.[17]

Hydrogen-3, carbon-11, argon-41, krypton-85, and radon-220 are the major airborne radioactive effluents released from the Laboratory. The hydrogen-3 (as water vapor) exposure is by inhalation. The exposure pathway for the other nuclides is directly from air to man by submersion. The concentrations and dose rates were calculated for these nuclides by a computer program based on an atmospheric dispersion model.[15,16] *The following parameters were used in the calculations:*

a) *annual release rates: hydrogen-3, 62.9 Ci; carbon-11, 85.6 Ci; argon-41, 0.88 Ci; krypton-85, 71.3 Ci; and radon-220, 76.9 Ci.*
b) *meteorological data: the wind speed and direction data shown in Figure 3.*
c) *the usual parameters for stack height, building, wake, plume, momentum, temperature, etc.*

The calculations were carried out to 80 km (50 mi) using the population distribution of 16 segments and ten distance increments given in Table 1. 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 23. The dose to the nearest resident is 0.01% of the 500 mrem/y Radiation Protection Standard.[5]

The population data in Table 1 was used to calculate the cumulative population dose from gaseous radioactive effluents. The results are given in Table 24, 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. Although this water is not used for drinking purposes, the

TABLE 23

Dose From Airborne Emissions, 1984
(Millirems)

Nuclide	Quantity Released (Ci)	Maximum Perimeter		Maximum Individual	
		Annual	50-Year	Annual	50-Year
Hydrogen-3	62.9	0.06	0.06	0.04	0.04
Carbon-11	85.6	0.02	0.02	0.009	0.009
Argon-41	0.88	0.0004	0.0004	0.0002	0.0002
Krypton-85	71.3	0.005	0.005	0.003	0.003
Radon-220	76.9	0.006	0.006	0.004	0.004

TABLE 24

80 km Population Dose, 1984

Source	Man-Rems	
	Annual	50-Year
Hydrogen-3	2.94	2.94
Carbon-11	0.21	0.21
Argon-41	0.013	0.013
Krypton-85	0.30	0.30
Radon-220	0.0016	0.0016
Natural	6.99×10^5	-

dose that would be received by an individual ingesting water at the concentrations found in the Creek was calculated. A minor exposure route is from water to man by ingestion 710 km (370 mi) downstream from Argonne (Section I.E.), but the dilution of Sawmill Creek water at this point is so great that the dose calculation is meaningless. No other exposure pathways are

significant. Those nuclides added to Sawmill Creek by Argonne waste water, their net concentrations in the Creek, and the corresponding dose rates, if water at these concentrations were used as the sole water supply by an individual, are given in Table 25. The dose rates were all well below the standards for individuals in uncontrolled areas. It should be emphasized that Sawmill Creek is not used for drinking, swimming, or boating. Inspection of the area shows there are few fish in the stream, and they do not constitute a significant source of food for any individual.

TABLE 25

Radionuclide Concentrations and Dose Estimates for Sawmill Creek Water, 1984

Nuclide	*Conc. (10^{-9} μCi/mL (net avg.)*	*Dose mrem/year*	*Percent of Standard**
Hydrogen-3	*82*	*0.014*	*0.0027*
Strontium-90	*0.13*	*1.3*	*0.042*
Neptunium-237	*0.0066*	*0.0066*	*0.00022*
Plutonium-239	*0.0198*	*0.011*	*0.00040*
Americium-241	*0.0158*	*0.0059 (kidney)* *0.0095 (bone)*	*0.00040* *0.00032*
Curium-244	*0.0025*	*0.0011 (bone)*	*0.00004*
Californium-249	*0.0025*	*0.0019*	*0.00006*

**DOE Order 5480.1 Standard.*[5]

As indicated in Table 8, occasional Sawmill Creek samples (less than 10) contained traces of plutonium-238 and curium-242 and/or californium-252 nuclides, but the averages were only slightly greater than the detection limit. The annual dose due to an individual consuming water at these concentrations can be calculated as was done for those nuclides more commonly found in Creek water, but the method of averaging probably overestimates the true concentration. These annual doses range from 1×10^{-3} to 2×10^{-5} mrem/y for these nuclides.

The Environmental Protection Agency (EPA) has established drinking water standards based on a dose of 4 mrem/y for man-made beta particle and

photon emitting radionuclides.[10] *For hydrogen-3, the EPA standard is 2 x* 10^{-5} *µCi/mL and for strontium-90, it is 8 x* 10^{-9} *µCi/mL. The concentrations in Table 26 correspond to 0.4% for hydrogen-3 and 1.6% for strontium-90 of the EPA standards. No specific EPA standards exist for the transuranic nuclides.*

b. External Penetrating Radiation Dose

Above normal fenceline 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.6.

At Location 7I, the fenceline dose from Argonne was about 370 mrem/y. Approximately 300 m (0.3 mi) south of the fenceline (grid 6I), the measured dose averaged 107 $\pm$ *29 mrem/y, close to the upper end of the normal range, 99 mrem/y. There are no individuals living in this area. The closest residents are about 1.6 km (1 mi) south of the fenceline. At this distance, the calculated dose rate from the Waste Storage Area is about 0.01 mrem/y, if the energy of the radiation were 0.66 MeV (the cesium-137 gamma-ray energy) and about 0.05 mrem/y (if the energy were 1.3 MeV, the cobalt-60 energy). In the area north of the site, where the fenceline radiation dose from the cobalt-60 sources in Building 202 was measured at 31 mrem/y, the nearest residents are 750 m (0.47 mi) to the north-northwest. The calculated dose at that location was about 0.06 mrem/y.*

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

c. Summary

The total dose received by off-site residents was a combination of the separate pathways that contribute to this total: carbon-11, argon-41, krypton-85, and radon-220 submersion dose, hydrogen-3 inhalation dose, and cobalt-60 external radiation dose. The highest dose was about 0.1 mrem/y (0.02% of the 500 mrem/y standard) 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 3.2 man-rem.

B. Chemical Constituents

1. Surface Water

The environmental water data contained in this section were collected to ascertain the effectiveness of Argonne's effluent pollution controls and determine compliance with State regulations and National Pollutant Discharge Elimination System (NPDES) permits. The appropriate stream and effluent standards are listed in the ILLINOIS POLLUTION CONTROL BOARD RULES AND REGULATIONS, Chapter 3.[19] *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 26. 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.*

TABLE 26

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	0.003
Copper	0.02	1.0	0.0008
Cyanide	0.025	0.025	0.020
Fluoride	1.4	15	0.02
Iron	1.0	2.0	0.05
Lead	0.1	0.1	0.002
Manganese	1.0	1.0	0.001
Mercury	0.0005	0.0005	0.0001
Nickel	1.0	1.0	0.003
pH	6.5-9.0	5.0-10.0	-
Selenium	1.0	1.0	0.005
Silver	0.005	0.1	0.0002
Sulfate	500	-	1.0
Temperature	-	-	0.1°C
Total Dissolved Solids	1000	-	-
Zinc	1.0	1.0	0.01

The analytical methods have been described in previous reports in this series. The results of measurements for chemical constituents are expressed as milligrams (mg) or micrograms (µg) per liter (L). Averages were calculated as described in Section III.A. Yearly averages are reported with a (±) limit value. This value is the standard error at the 95% confidence limit and is calculated from the standard deviation of the yearly average. Only when sample concentrations are random does this value represent the actual distribution occurring at the sampling location. As in the past, emphasis has been placed on Sawmill Creek, since this is the principal route for waste water leaving the Argonne site, and on the NPDES permit locations.

a. Waste Treatment Plant Effluent Water

The major discharge of waste water from Argonne operations is by way of the waste treatment plant. The water volume from this source was approximately 2.8 megaliters (0.75 million gallons) per day in 1984 and was comprised of 37% sanitary waste water and 63% water from laboratory operations. Laboratory waste is directed to a large holding pond with an overflow which controls the flow to about 1300 liters/minute (350 gallons/minute). The discharge occurs over a 24-hour period. Since the sanitary system is fairly constant, the overall output is constant.

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

The results obtained for chemical constituents in the effluent water from the treatment plant are shown in Table 27. All of the average concentrations were below the State standards, except for mercury. The average value for mercury was 88% of the State standard and 31% of the samples exceeded this value. The corresponding values for 1983 were 116% and 39%. Efforts are continuing to lower mercury 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.

TABLE 27
CHEMICAL CONSTITUENTS IN EFFLUENTS FROM ANL TREATMENT PLANT, 1984
(CONCENTRATIONS IN MICROGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	CONCENTRATION MIN.	CONCENTRATION MAX.	PERCENT OF STANDARD (AVG.)	PERCENT EXCEEDING STATE STANDARD
ARSENIC	12	-	-	< 5	< 2.0	0
BARIUM	51	23 ± 3	8	67	1.2	0
BERYLLIUM	12	0.04 ± 0.02	0.01	0.11	-	-
CADMIUM	51	0.6 ± 0.1	< 0.2	2.4	0.4	0
COPPER	51	35 ± 13	10	314	3.5	0
FLUORIDE	51	478 ± 77	240	1430	3.2	0
IRON	51	199 ± 16	100	290	10	0
LEAD	51	3.2 ± 0.4	< 2.0	7.5	3.2	0
MANGANESE	51	64 ± 29	9	484	6.4	0
MERCURY	51	0.44 ± 0.09	0.05	1.40	88	31
NICKEL	51	8 ± 1	3	29	0.8	0
PH	249	-	6.9	8.5	-	0
SELENIUM	12	-	-	< 5	< 0.5	0
SILVER	51	2.2 ± 0.3	0.4	5.5	2.2	0
ZINC	51	55 ± 4	29	94	5.5	0

Concentration of all the other constituents are similar to last year's values and are not due to Argonne operations.

b. National Pollutant Discharge Elimination System (NPDES)

The Laboratory discharges are regulated by NPDES Permit #IL 0034592. This permit is subject to renewal about every three to five years. The period of January 1, 1984, through October 28, 1984, is covered by the permit in effect since May 19, 1981. Effective October 1984, this permit was renewed to March 1, 1989, and several revisions of requirements have occurred. The locations previously described and shown in Figure 7 * *remain the same, except for Location 010 which has been eliminated, and did not contain any water in 1984. The location formerly denoted as 001 (waste treatment plant) has been divided into 001A (sanitary waste) and 001B (laboratory waste). Requirements for the combined effluent are still listed under Location 001.*

Requirements for total dissolved solids and residual chlorine have been eliminated. Measurement of fats, oil, and greases have been added to all locations except 001, 007, and 008. Chemical oxygen demand has been added to Location 001B (laboratory waste) and zinc has been added to Location 006. The limit for mercury has been changed from an overall limit of 0.5 µg/L to a 30-day average of 3 µg/L and a daily maximum of 6 µg/L.

All samples collected at Locations 001A, 001B, and 001 were collected once/week. All other samples were collected once/month. This applied to both versions of the permit. Results from the period January-October appear in Table 28. Total suspended solids exceeded the permit levels occasionally at several locations and in all cases, excessive rainfall had occurred. One sample exceeded the limit for fecal coliform and sample contamination during collection was suspected. The water which is discharged at Location 009 has a pH greater than the limit of 9.0 because it contains water treatment chemicals, and this value was always exceeded. The radium-226 limit was exceeded on four occasions due to the natural radium content

---

* *For purposes of clarity, the location numbers in the figure are given without the zeros. Thus, permit location 001 is shown in the figure as 1.*

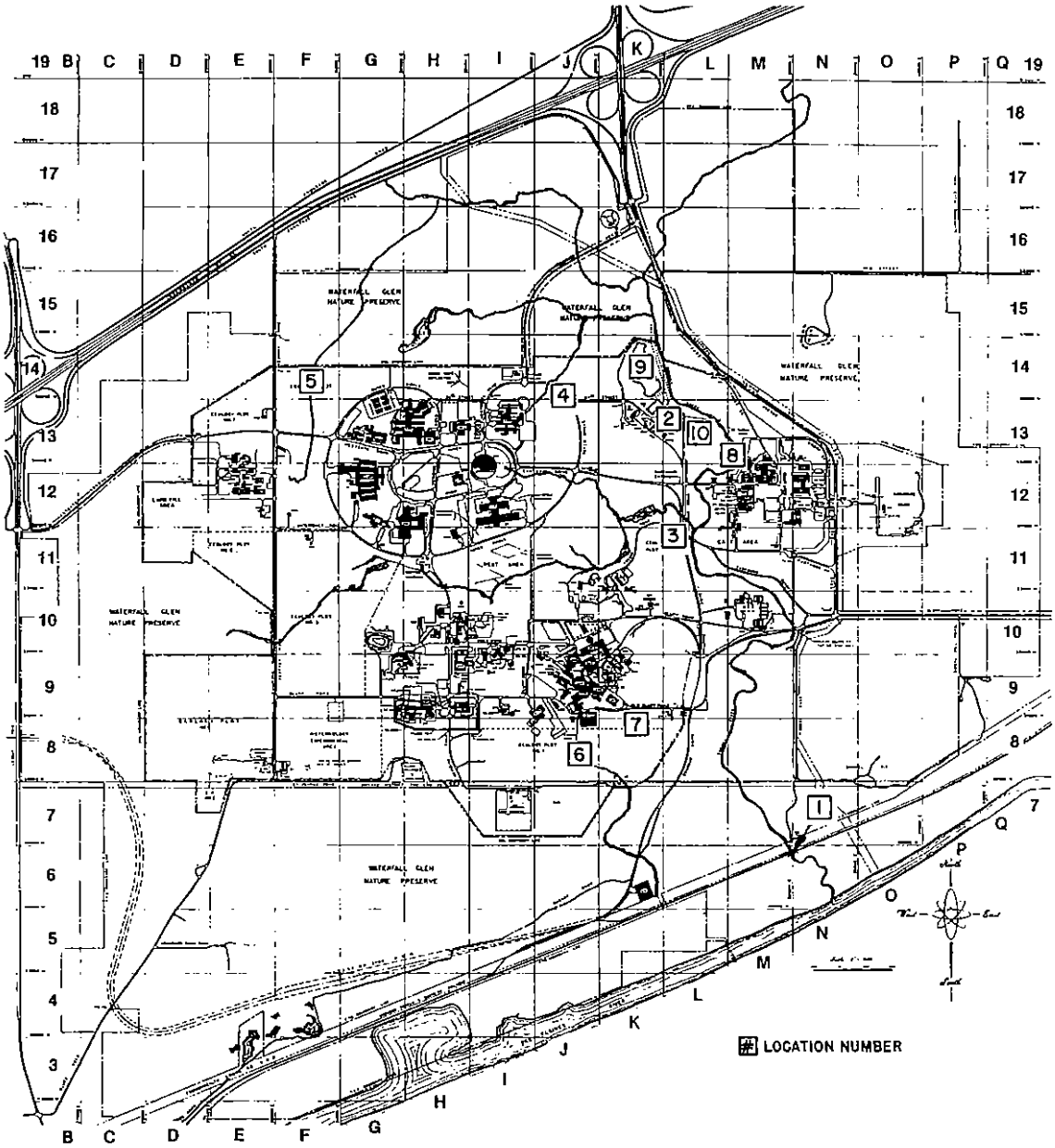

Fig. 7. NPDES Permit Locations

TABLE 28

NPDES Effluent Quality Summary
IL 0034592, Effective through October 28, 1984

Discharge Location	Permit Constituent	Concentration Limits mg/L 30 Day Average	Daily Max.	Number Exceeding Limit	*R Measured / Permit
001	Flow	None		0	-
	pH	6-9		0	-
	Biochemical Oxygen Demand (BOD)	30	75	0	-
	Total Suspended Solids (TSS)	30	75	0	-
	Fecal Coliform	-	400 organisms / 100 mL	1	6
	Total Residual Chlorine (TRCL)	-	0.75	0	-
	Mercury	-	0.0005	34	1.1-20
	Radioactivity:				
	Gross Beta		100 pCi/L	0	-
	Radium-226		1 pCi/L	4	1.6-3.1
	Strontium-90		2 pCi/L	0	-
002	Flow	None		0	-
	pH	6-9		0	-
	Temperature	< 2.8°C Rise		0	-
	TSS	-	15	2	1.1-2.0
003	Flow	None		0	-
	pH	6-9		0	-
	Temperature	< 2.8°C Rise		0	-
	TSS	-	15	1	2.5
	Total Dissolved Solids (TDS)	-	3500	0	-
	TRCL	-	0.75	0	-

TABLE 28 (contd)

Discharge Location	Permit Constituent	Concentration Limits mg/L 30 Day Average	Daily Max.	Number Exceeding Limit	*R Measured / Permit
004	Flow	None		0	-
	pH	6-9		0	-
	TSS	-	15	1	1.5
	TDS	-	3500	0	-
	Temperature	< 2.8°C Rise		0	-
005	Flow	None		0	-
	pH	6-9		0	-
	Fats, Oil, and Grease (FOG)	15	30	0	-
	TSS	-	15	1	7
	Temperature	< 2.8°C Rise		0	-
006	Flow	None		0	-
	pH	6-9		0	-
	TSS	-	15	2	1.1-3.0
007	Flow	None		0	-
	pH	6-9		2	10.7-11.7
008	Flow	None		0	-
	pH	6-9		0	-
009	Flow	None		0	-
	pH	6-9		10	All > 10
	TSS	-	15	2	1.4-2.6

*R is the range of the ratio of the values of the measurements exceeding the concentration limit to the concentration limit (except for pH, for which the actual values are given).

of the water supply. The limit of 0.5 µg/L for mercury was exceeded in most cases despite the steps taken to reduce levels. Only four of these samples exceeded the new one-time limit of 6.0 µg/L and these occurred during a period when the Laboratory portion of the treatment plant was being extensively cleaned.

Results for November and December are shown in Table 29. All results for mercury were in compliance with the discharge limit. One fecal coliform result exceeded the limit due to low chlorine residual caused by a chlorine monitor malfunction. Finally, one oil and grease sample at Location 002 exceeded the limit, but resampling the next day as well as subsequent sampling indicates that the result was due to contamination on the discharge pipe. The pH at 009 was exceeded for reasons previously discussed. This situation should be alleviated in summer 1985, when an ion-exchange treatment will replace the high pH treatment.

On November 21, 1984, approximately 190 liters (50 gallons) of No. 2 fuel oil was released from the 800 Area (12E) into the stream which is defined as NPDES Location 005. The oil was collected by using absorbents and booms. Subsequent NPDES samples for oil, fats, and grease at this location have contained less than the detection limit of 5 mg/L. The NPDES permit limit is 30 mg/L.

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 100 m (310 ft) downstream of the outfall [7M (down)].

The data from these studies are in Table 30. Average levels for ammonia nitrogen were below the State limit at both locations for the second year after having always exceeded this limit in the past. The dissolved

TABLE 29

NPDES Effluent Quality Summary

IL 00334592 (Revised in Effect October 28, 1985)

Discharge Location	Permit Constituent	Concentration Limits mg/L 30 Day Average	Daily Max.	Number Exceeding Limit	*R Measured / Permit
001A	Flow	None		0	-
	BOD	30	60	0	-
	TSS	30	60	0	-
001B	Flow	None		0	-
	Chemical Oxygen Demand	-	-	0	-
	TSS	15	30	0	-
	Mercury	0.003	0.006	0	-
001	pH	6-9		0	-
	Fecal Coliform	-	400 organisms / 100 mL	1	48
	BOD	30	60	0	-
	TSS	30	60	0	-
002	Flow	None		0	-
	pH	6-9		0	-
	TSS	15	30	0	-
	Temperature	< 2.8°C Rise		0	-
	FOG	-		0	-
003	Flow	None		0	-
	pH	6-9		0	-
	TSS	15	30	0	-
	Temperature	< 2.8°C Rise		0	-
	FOG	-		0	-

TABLE 29 (contd)

Discharge Location	Permit Constituent	Concentration Limits mg/L 30 Day Average	Daily Max.	Number Exceeding Limit	*R Measured / Permit
004	Flow	None		0	-
	pH	6-9		0	-
	TSS	15	30	1	1.2
	Temperature	< 2.8°C Rise		0	-
	FOG	-	-	0	-
005	Flow	None		0	-
	pH	6-9		0	-
	Temperature	< 2.8°C Rise		0	-
	FOG	15	30	0	-
006	Flow	None		0	-
	pH	6-9		0	-
	TSS	15	30	0	-
	Zinc	1.0	2.0	0	-
	FOG	-	-	0	-
007	Flow	None		0	-
	pH	6-9		0	-
	Temperature	< 2.8°C Rise		0	-
008	Flow	None		0	-
	pH	6-9		0	-
	FOG	-	-	0	-
009	Flow	None		0	-
	pH	6-9		2	All > 9
	TSS	15	30	0	-
	FOG	-	-	0	-

*R is the range of the ratio of the values of the measurements exceeding the concentration limit to the concentration limit (except for pH, for which the actual values are given).

TABLE 30

SAWMILL CREEK - EFFECT OF SANITARY WASTE, 1984

CONSTITUENT	* LOCATION	NO. OF SAMPLES	CONCENTRATION (MG/L) AVG.	MIN.	MAX.	AVG. PERCENT OF STANDARD	PERCENT EXCEEDING STATE STANDARD
AMMONIA NITROGEN	7M (UP)	51	1.2 ± 0.5	0.1	8.2	83	27
	7M (DOWN)	51	1.0 ± 0.3	0.1	4.5	67	22
CHLORIDE	7M (UP)	27	424 ± 54	209	713	-	-
	7M (DOWN)	27	386 ± 43	209	586	-	-
CYANIDE	7M (DOWN)	51	-	-	< 0.01	40	0
DISSOLVED OXYGEN	7M (UP)	51	11.2 ± 0.4	8.1	14.4	-	0
	7M (DOWN)	51	11.1 ± 0.4	8.1	13.8	-	0
DISSOLVED SOLIDS	7M (UP)	51	1200 ± 110	310	1910	120	71
	7M (DOWN)	51	1120 ± 90	380	1670	112	67
PH **	7M (UP)	51	-	7.5	8.7	-	0
	7M (DOWN)	51	-	7.5	8.7	-	0
SULFATE	7M (UP)	27	169 ± 14	73	243	34	0
	7M (DOWN)	27	162 ± 11	74	207	32	0
TEMPERATURE ***	7M (UP)	51	13.7 ± 2.3	1.8	27.3	-	-
	7M (DOWN)	51	13.8 ± 2.2	2.2	27.1	-	-

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

** UNIT

*** DEGREES CENTIGRADE

oxygen levels obtained during 1984 were all above the State minimum of 5 mg/L. The total dissolved solids above the outfall exceeded the State standard in about 71% of the samples, and increased the solids content of the downstream sample.

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 31.

None of the individual samples for arsenic, barium, cadmium, chromium, fluoride, lead, nickel, selenium, and zinc exceeded the State standards. The levels of mercury averaged 61% of the State limit and exceeded this limit 24% of the time, compared to 24% and 2%, respectively, in 1983. This increase was due primarily to cleaning operations in the treatment plant to remove residual mercury contamination. Inadvertent losses to the effluent system occurred during a short time span. As in the past, high iron levels reflect high turbidity conditions and are not related to Argonne releases. The levels of copper and silver exceeded State limits in 49% and 8% of the samples, respectively, but they are not due to ANL operations since the average stream and effluent copper and silver concentrations are essentially the same, but the effluent water is diluted by about a factor of 10 with stream water.

d. Des Plaines River

The effect of Sawmill Creek on the Des Plaines River was evaluated by collecting samples at Willow Springs (upstream of Argonne) and at Lemont (downstream of Argonne). These samples were analyzed for total mercury, hexavalent chromium, total iron, and total zinc. The results are in Table 32. In no case was there any indication that hexavalent chromium, mercury, or zinc levels were affected by the Argonne effluent.

TABLE 31

CHEMICAL CONSTITUENTS IN SAWMILL CREEK LOCATION 7M,* 1984
(CONCENTRATIONS IN MICROGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	CONCENTRATION MIN.	CONCENTRATION MAX.	PERCENT OF STANDARD (AVG.)	PERCENT EXCEEDING STATE STANDARD
ARSENIC	12	-	-	< 5	< 0.5	0
BARIUM	51	76 ± 9	22	206	1.5	0
BERYLLIUM	12	0.06 ± 0.03	0.01	0.17	-	-
CADMIUM	51	1.2 ± 0.2	0.6	3.1	2.4	0
CHROMIUM(III)	51	6 ± 1	3	23	0.6	0
CHROMIUM(VI)	51	-	-	< 3	< 6.0	0
COPPER	51	34 ± 13	8	247	172	49
FLUORIDE	51	537 ± 44	240	890	38	0
IRON	51	1460 ± 410	320	8070	146	45
LEAD	51	9 ± 2	2	44	9.1	0
MANGANESE	51	222 ± 69	29	1150	22	4
MERCURY	51	0.30 ± 0.08	< 0.05	1.29	61	24
NICKEL	51	12 ± 3	3	42	1.2	0
PH	249	-	7.0	8.7	-	0
SELENIUM	12	-	-	< 5	< 0.5	0
SILVER	51	2.0 ± 0.4	0.4	6.0	41	8
ZINC	51	64 ± 10	23	194	6.4	0

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

TABLE 32

CHEMICAL CONSTITUTENTS IN THE DES PLAINES RIVER, 1984

CONSTITUENT	LOCATION *	NO. OF SAMPLES	CONCENTRATION(MICROGRAM/LITER) AVG.	MIN.	MAX.
CHROMIUM(VI)	A	12	-	-	< 6.0
	B	22	-	-	< 6.0
IRON	A	12	941 ± 333	224	1720
	B	23	902 ± 225	174	2440
MERCURY	A	12	-	-	< 0.05
	B	23	-	-	< 0.05
ZINC	A	12	41 ± 12	10	82
	B	23	41 ± 10	20	119

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

2. Ground Water

A sanitary landfill for non-radioactive waste is located on the western edge of the Argonne site (Figures 1 and 8). This landfill operates under Illinois EPA permit No. 1981-29-OP and is further described in Reference 11. Operation of the ANL landfill began in July, 1966. During the period 1969-1978, substantial quantities of liquid organic and inorganic waste were disposed onsite by adding them to an open "French drain" located in the northeast sector of the landfill.[2] This practice is no longer allowed.

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

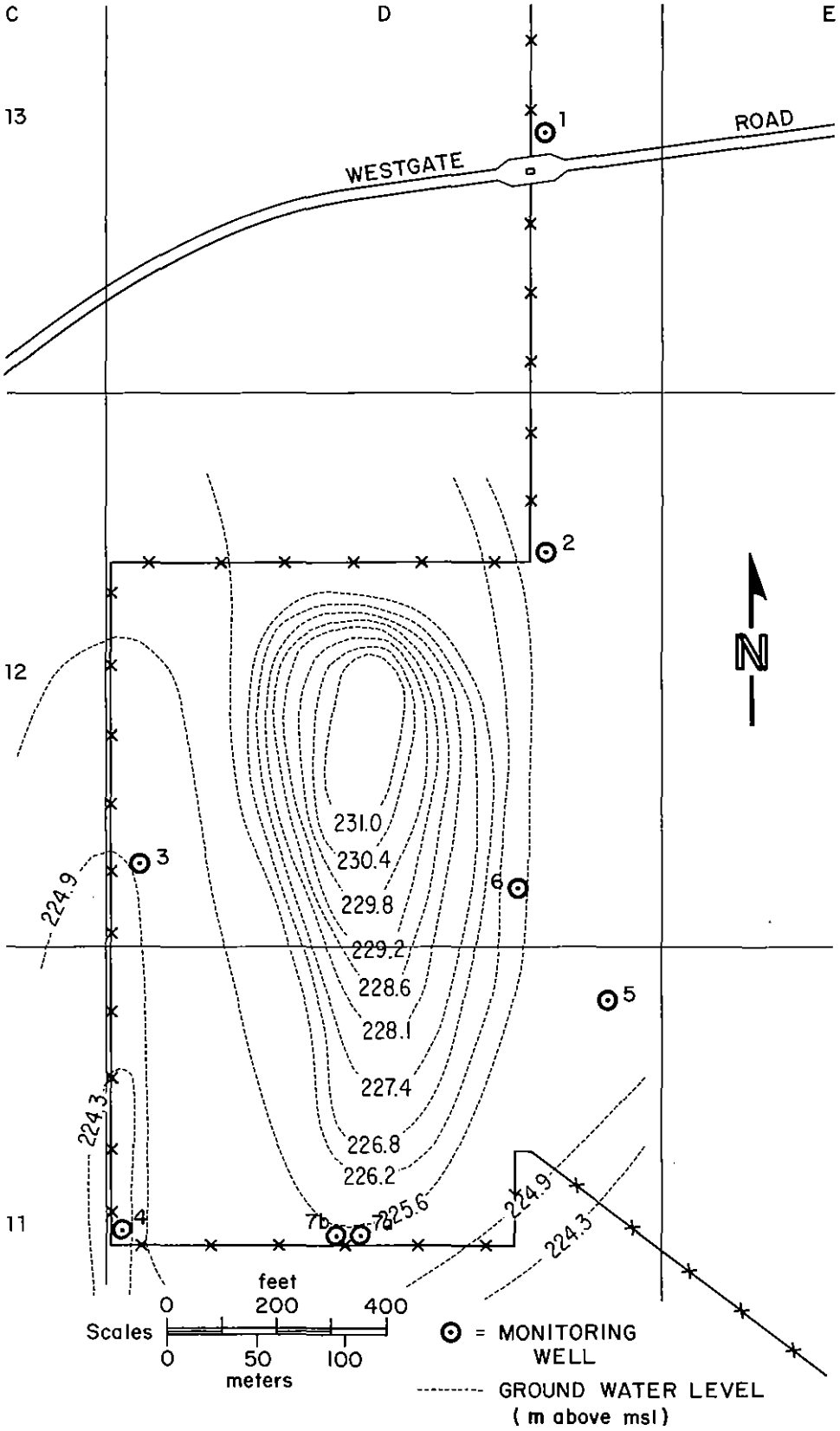

Fig. 8. Monitoring Well Locations for Landfill

Wells No. 2, No. 3, and No. 4 were placed in the landfill itself. In April, 1980,[20] a more comprehensive study was initiated. Three additional test wells were placed in the landfill at previously untested locations. Well No. 6 was placed in the east section to sample any water flowing out of the landfill in the southeasterly direction. Wells No. 7a and No. 7b were nested and were installed to measure vertical water movement as well as to provide monitoring of water from two depths. Test borings were also made in the fill area to determine soil conditions where refuse had been added.

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

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

All of the wells have been studied from 1979 until the present to determine constancy of water levels, recharge rates, and chemical characteristics. The water levels, measured quarterly, for 1984 are shown in Table 33. In addition, the precipitation for a 30-day period prior to sampling and the average levels for 1983 are shown. The average water levels for Wells Nos. 1-4 were similar in 1984 and 1983. The average water levels in Wells No. 6 and No. 7a were much higher in 1984 than in 1983. Wells No. 6 and No. 7a are believed to be better prepared and sealed than the other wells and are more accurate measures of the condition of the ground water.

TABLE 33

Well Point and Water Level Elevations[1] of Monitoring Sites at the ANL Landfill

Well No.	Ground Surface Elevation	Well Point Elevation	Quarterly Measurements 1st	2nd	3rd	4th	Average 1984	1983[2]
1	227.53	218.23	227.38	226.28	225.28	226.47	226.35	226.83
2	230.58	220.83	226.16	225.70	224.70	225.22	225.44	226.04
3	226.77	218.08	224.82	224.67	223.54	224.03	224.27	224.85
4	227.23	221.13	225.43	225.09	222.63	225.43	224.64	225.16
5	227.53	215.34	*	221.86	216.71	*	219.28	222.17
6	229.91	215.13	223.69	224.18	222.96	*	223.61	221.68
7a	227.81	220.19	225.55	224.91	224.73	225.86	225.26	223.54
7b	227.81	214.09	**	**	**	**	**	**
(3)	-	-	87.9	111.8	56.1	96.0	88.0	94.6

*Not measured.

**Dry.

Notes: (1) All measurements are in meters above mean sea level.

(2) Based on three measurements at each well.

(3) Precipitation in millimeters for 30 day period prior to measurements.

Samples collected in the third quarter, which was preceded by a low precipitation level of 56.1 mm, showed a large drop in water levels in Wells Nos. 1-5. Well No. 6 was effected somewhat less and the effect on Well No. 7a was small.

The recharge rates of the wells parallel this. Wells No. 1 and No. 4 recharge almost immediately upon emptying. Wells No. 2, No. 3, and No. 5 have intermediate recharge rates while No. 6 and No. 7a recharge within 24 hours. It appears that the water level surrounding Well No. 7a is gradually increasing which could indicate a flow change in the southerly direction. Well No. 7b was dry in 1984 as in past years confirming the fact that water is not permeating down to this level.

On the basis of past data, selected elements were studied in all wells in 1984. Similar studies were carried out in a drainage ditch surrounding the landfill. Well No. 1 (Table 34) has persistently high chloride and total dissolved solid levels. The proximity of this well to Westgate Road and its rapid recharge rate indicates that surface runoff has free access to this well and that the chloride comes from salt used for deicing.

Well No. 2 (Table 35) has had a pH of 11 or greater since the beginning of sampling. This was presumably due to the nearby disposal of carbonate-type water softening materials. The high pH water was confined to this area since no other wells have had elevated, or even changing pH values. The pH of samples obtained in 1984 were successively 10, 8.4, 9.8, and 8.4. This indicates the depletion of the source of basicity and the value of 9.8 in the third quarter could be due to the low precipitation level. In any event, it appears that the water in this area was once contaminated with alkaline chemicals which is no longer occurring.

Well No. 3 (Table 36) is characterized by the presence of arsenic at the same levels as in previous years. The levels of iron are extremely high and manganese levels are elevated. Chloride and sulfate levels are very low. The source of the arsenic is unknown but the constancy of the levels suggest it is naturally occurring. Arsenic has not been found in any other well.

TABLE 34

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

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	MIN.	MAX.
ARSENIC	4	-	-	< 5
BARIUM	4	212 ± 71	158	260
CHLORIDE*	4	844 ± 151	730	983
DISSOLVED SOLIDS*	4	2240 ± 690	1770	2930
FLUORIDE	4	163 ± 24	142	184
IRON	4	663 ± 625	228	1260
MANGANESE	4	227 ± 85	174	312
MERCURY	4	-	-	< 0.05
PH	4	-	7.0	7.1
SULFATE*	4	126 ± 38	87	152

* CONCENTRATION IN MILLIGRAM/LITER.

TABLE 35

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

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	MIN.	MAX.
ARSENIC	4	-	-	< 5
BARIUM	4	177 ± 12	169	185
CHLORIDE*	4	25 ± 8	17	29
DISSOLVED SOLIDS*	4	265 ± 65	195	291
FLUORIDE	4	146 ± 14	132	154
IRON	4	-	-	< 100
MANGANESE	4	7 ± 9	2	15
MERCURY	4	-	-	< 0.05
PH	4	-	8.4	10
SULFATE*	4	69 ± 10	60	77

* CONCENTRATION IN MILLIGRAM/LITER.

TABLE 36

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

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	MIN.	MAX.
ARSENIC	4	15 ± 6	12	21
BARIUM	4	656 ± 759	275	1470
CHLORIDE*	4	12 ± 7	5	16
DISSOLVED SOLIDS*	4	795 ± 17	784	811
FLUORIDE	4	135 ± 33	100	152
IRON	4	4930 ± 4810	1110	8800
MANGANESE	4	365 ± 200	249	576
MERCURY	4	-	-	< 0.05
PH	4	-	6.7	6.8
SULFATE*	4	33 ± 52	2	81

* CONCENTRATION IN MILLIGRAM/LITER.

Well No. 4 (Table 37) has very high levels of manganese but relatively low iron levels. In addition, very high levels of sulfate were found. This well has a fast recharge rate and is located close to a ditch which drains a marshy area. This ditch was sampled in 1984 and the results are shown in Table 38. No elevated values are seen with the exception of a moderately elevated chloride level. Both managanese and sulfate levels are low. This would indicate little or no connection between the ditch and Well No. 4.

Well No. 5 (Table 39) serves as a reference downstream of the landfill. The pH was the same as in 1983 (7.5) and contained very low manganese and iron levels. Well No. 6 (Table 40) contains very high levels of iron and manganese and moderate to high levels of chloride and sulfate. There is a possibility that the manganese resulted from manganese sulfate added to the area, but the high levels could be indicative of natural levels. Results from Well No. 7a (Table 41) are about the same as in past years with no outstanding characteristics.

The most characteristic factor in the results for both 1983 and 1984 is the constancy of the pH values at each well. With the exception of Well No. 2, which was previously discussed, the pH values of each well tested in 1984 are the same as 1983 values within 0.1 pH unit, although different from each other. It appears that some buffering system is available in each which maintains the pH. In addition, arsenic is found in only Well No. 3 at about the same level, and manganese in Wells No. 4 and No. 6 is high and fairly constant. It does not appear that water is moving from one well site to another and some type of equilibrium exists in each well.

All of the wells were examined for organic content. The first quarter samples were examined using a carbonyl specific test previously developed. All wells contained less than the detection limit for acetone and formaldehyde of 10 µg/L. Samples collected in the other quarters were examined for benzene, toluene, xylene, trichloroethylene, and perchloroethylene using a newly instituted microextraction procedure. Results for benzene, toluene, and xylene were all less than 5 µg/L. Results for tri- and perchloroethylene were all less than 20 µg/L. To date no evidence exists

TABLE 37

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

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	MIN.	MAX.
ARSENIC	4	-	-	< 5
BARIUM	4	160 ± 87	108	235
CADMIUM	2	0.95 ± 0.22	0.90	1.00
CHLORIDE*	4	180 ± 137	86	319
CHROMIUM(VI)	2	-	-	< 3
COPPER	2	7 ± 10	5	10
DISSOLVED SOLIDS*	4	1260 ± 360	940	1500
FLUORIDE	4	187 ± 43	144	218
IRON	4	436 ± 317	100	596
LEAD	2	1	1	1
MANGANESE	4	1340 ± 640	780	1900
MERCURY	4	-	-	< 0.05
NICKEL	2	7 ± 8	5	9
PH	4	-	6.8	6.9
SELENIUM	2	-	-	< 5
SILVER	2	0.65 ± 0.22	0.60	0.70
SULFATE*	4	364 ± 150	270	490
ZINC	2	17 ± 30	10	24

* CONCENTRATION IN MILLIGRAM/LITER.

TABLE 38

CHEMICAL CONSTITUENTS AT ANL LANDFILL
DRAINAGE DITCH
(CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	MIN.	MAX.
ARSENIC	2	-	-	< 5
BARIUM	2	136 ± 140	104	169
CADMIUM	2	1.45 ± 3.66	0.60	2.30
CHLORIDE*	2	260 ± 303	189	330
CHROMIUM(VI)	2	-	-	< 3
COPPER	2	8 ± 12	5	11
DISSOLVED SOLIDS*	2	892 ± 1160	622	1160
FLUORIDE	2	180 ± 69	164	196
IRON	2	115 ± 65	100	130
LEAD	2	1	1	1
MANGANESE	2	100 ± 230	47	154
MERCURY	2	-	-	< 0.05
NICKEL	2	8 ± 14	5	11
PH	2	-	7.4	7.5
SELENIUM	2	-	-	< 5
SILVER	2	0.60 ± 1.29	0.30	0.90
SULFATE*	2	91 ± 37	82	99
ZINC	2	22	22	22

* CONCENTRATION IN MILLIGRAM/LITER.

TABLE 39

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

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	MIN.	MAX.
ARSENIC	1	-	-	< 5
BARIUM	1	-	-	155
CHLORIDE*	1	-	-	13
DISSOLVED SOLIDS*	1	-	-	362
FLUORIDE	1	-	-	154
IRON	1	-	-	< 100
MANGANESE	1	-	-	37
MERCURY	1	-	-	< 0.05
PH	1	-	7.5	7.5
SULFATE*	1	-	-	115

* CONCENTRATION IN MILLIGRAM/LITER.

TABLE 40

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

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	MIN.	MAX.
ARSENIC	3	-	-	< 5
BARIUM	3	184 ± 71	159	228
CHLORIDE*	3	147 ± 75	100	172
DISSOLVED SOLIDS*	3	1290 ± 51	1260	1310
FLUORIDE	3	117 ± 61	82	148
IRON	3	12200 ± 2760	10700	13700
MANGANESE	3	3920 ± 540	3720	4250
MERCURY	3	-	-	< 0.05
PH	3	-	6.5	6.6
SULFATE*	3	124 ± 21	112	135

* CONCENTRATION IN MILLIGRAM/LITER.

TABLE 41

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

CONSTITUENT	NO. OF SAMPLES	CONCENTRATION AVG.	MIN.	MAX.
ARSENIC	4	-	-	< 5
BARIUM	4	150 ± 79	97	231
CHLORIDE*	4	19 ± 7	13	25
DISSOLVED SOLIDS*	4	483 ± 82	417	544
FLUORIDE	4	136 ± 22	116	154
IRON	4	-	-	< 100
MANGANESE	4	372 ± 86	287	430
MERCURY	4	-	-	< 0.05
PH	4	-	7.1	7.3
SULFATE*	4	98 ± 31	76	129

* CONCENTRATION IN MILLIGRAM/LITER.

for movement of organic materials into the test wells, although studies are continuing to improve sampling and analytical methods.

V. APPENDIX

A. References

1. U. S. Department of Energy, EFFLUENT AND ENVIRONMENTAL MONITORING PROGRAM REQUIREMENTS, DOE DRAFT ORDER 5484.1A, CHAPTER III (1984).

2. ENVIRONMENTAL ASSESSMENT RELATED TO THE OPERATION OF ARGONNE NATIONAL LABORATORY, Argonne National Laboratory, Argonne, Illinois, DOE/EA-0181, August 1982.

3. H. Moses and M. A. Bogner, "Fifteen-Year Climatological Summary, Jan. 1, 1950-Dec. 31, 1964." U.S.A.E.C. Report ANL-7084 (September, 1967); H. Moses and J. H. Willett, "Five-Year Climatological Summary, July 1949-June 1954," U.S.A.E.C. Report ANL-5592.

4. N. W. Golchert, T. L. Duffy, and J. Sedlet, "Environmental Monitoring at Argonne National Laboratory, Annual Report for 1983," U.S.D.O.E. Report ANL-84-14 (March 1984).

5. U. S. Department of Energy, REQUIREMENTS FOR RADIATION PROTECTION, DOE Order 5480.1, Chapter XI (1981).

6. ICRP Publication 26, "Recommendations of the International Commission on Radiological Protection," ANNALS OF THE ICRP, Vol. 1, No. 3, Pergamon Press (1977); ICRP Publication 30, "Limits for Intakes of Radionuclides by Workers," Ibid (1978, et seq).

7. N. W. Golchert, T. L. Duffy, and J. Sedlet, "Environmental Monitoring at Argonne National Laboratory, Annual Report for 1979," U.S.D.O.E. Report ANL-80-29 (March 1980).

8. N. W. Golchert and J. Sedlet, "Radiochemical Determination of Plutonium in Environmental Water Samples," RADIOCHEM. RADIOANAL. LETTERS, 12, 215 (1972).

9. N. W. Golchert and J. Sedlet, "Determination of Transuranics in Environmental Water," in M. H. Campbell (ed.), HIGH-LEVEL RADIOACTIVE WASTE MANAGEMENT (Proceedings of a Symposium, American Chemical Society, April 1-2, 1974), American Chemical Society, Washington, DC, 1976, p. 152.

10. "National Interim Primary Drinking Water Regulations," Report EPA-570/9-76-003, Environmental Protection Agency, Office of Water Supply, U. S. Government Printing Office, Washington, DC.

11. N. W. Golchert, T. L.Duffy, and J.Sedlet, "Environmental Monitoring at Argonne National Laboratory, Annual Report for 1982," U.S.D.O.E. Report ANL-83-26 (March 1983).

12. "Sampling Surface Soil for Radionuclides, C-998-83," 1983 ANNUAL BOOK OF ASTM STANDARDS, Vol. 12.01, p. 792. "Soil Sample Preparation for the Determination of Radionuclides, C-999-83," IBID, p. 796.

13. B. G. Bennett, "Environmental Aspects of Americium," U.S.D.O.E. Report EML-348, December, 1978.

14. N. W. Golchert, T. L. Duffy, and J. Sedlet, "Environmental Monitoring at Argonne National Laboratory, Annual Report for 1976," U.S.E.R.D.A Report ANL-77-13 (March 1977).

15. J. F. Sagendorf and J. T. Goll, "XOQDOQ Program for the Meteorological Evaluation of Routine Effluent Releases at Nuclear Power Stations," U. S. Nuclear Regulatory Commission, NUREG-0324 (September 1977).

16. K. F. Eckerman, F. J. Congel, A. K. Roecklein, and W. J. Pasciak, "User's Guide to GASPAR Code," U. S. Nuclear Regulatory Commission, NUREG-0597 (June 1980).

17. International Commission on Radiological Protection, REPORT OF COMMITTEE II ON PERMISSIBLE DOSE FOR INTERNAL RADIATION, ICRP

Publication 2 (1959), and ICRP Publication 6 (1962), Pergamon Press, New York.

18. *C. Yamaguchi, "MPC Calculations for the Radionuclides Produced During Accelerator Operation," HEALTH PHYSICS, 29, 393-397 (1975).*

19. *ILLINOIS POLLUTION CONTROL BOARD, RULES AND REGULATIONS, Chapter 3, 1972.*

20. *APPLICATION FOR PERMIT TO DEVELOP AND/OR OPERATE A SOLID WASTE MANAGEMENT SITE, Soil Testing Services, Inc., Northbrook, IL, for Argonne National Laboratory, STS Job No. 14236-P, June 30, 1980.*

B. Environmental Permits, Impact Statements, and Activities

The environmental permits in effect at ANL are shown in Table 42. As shown, all the permits have been issued by the Illinois Environmental Protection Agency. The compliance status of the NPDES permit is discussed in Section III.B.1. The monitoring data obtained for the landfill are discussed in Section III.B.2. The limits or standards are not specified in the permit to operate the landfill, but the number of monitoring wells and the types of measurements and results are within the IEPA guidelines.

For Boiler No. 5 operations, sulfur dioxide and particulates are measured in the exhaust stack and the results reported to the IEPA. The Boiler is operating in compliance with the permit. The vapor recovery system for gas dispensing facilities was constructed, and is operating, as designed and satisfies its permit. The permit to operate the hazardous waste disposal facility is in interim status and a final permit application is expected in October, 1985.

The environmental effects of new construction projects, modifications to facilities, and other activities that can have an environmental impact are evaluated before work is begun. During 1984, no draft or final Environmental Impact Statements or Environmental Assessments were completed, and

TABLE 42

Environmental Permits in Effect at ANL

Permit Type	Facility	Application/ Permit No.	Agency	Expires
Fossil Energy Users Laboratory (FEUL) Facility Air Emissions Permit	FEUL-MHD Building 145	Per. C8012024	IEPA	9/13/86
Operating	ANL Steam Plant-ANL Boiler No. 5	043802AAA D/02	IEPA	7/23/86
Construct and Own	Water Pollution Control Facility Coal-Pile Runoff Containment Area	1980-EB-1568	IEPA	Covered by NPDES Permit 3/1/89
Construction Operation	Emco Wheaton coaxial vapor recovery system for gas dispensing facilities	HG490	IEPA	6/24/85 Subject to special conditions
NPDES	Laboratory Water Effluents	IL0034592	IEPA	3/1/89
Sludge Drying Bed Replacement	Sanitary Plant Laboratory Plant	1981-EB-1776	IEPA	Covered by NPDES Permit 3/1/89
Lift Station Construction	For Boiler wastewater, coal-pile runoff overflow lime pond and water pond overflow	1982-HB-0402	IEPA	Operation covered by NPDES Permit 3/1/89
Operating	Landfill	1981-20-OP	IEPA	Special Conditions
Hazardous Waste Facility-Operation RCRA	Buildings 325C and 306	IL3-890-008946	IEPA	Interim

no significant environmental activities or pollution abatement projects were undertaken.

C. 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,[5] and are used in this report to assess the hazard of a measured concentration of a radioactive nuclide. The Order distinguishes two CGs, one for occupational exposure in controlled areas and one for uncontrolled areas, beyond the site boundary where individuals can be exposed nonoccupationally, for 168 hours per week. The CGs in the table are for uncontrolled areas. For water, the standard selected was for the soluble form of the radionuclide; for air, the standard for the insoluble form was selected.

2. Detection Limits

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

TABLE 43

Concentration Guides and Detection Limits
(μCi/mL)

Nuclide or Activity	Concentration Guide Water	Concentration Guide Air	Detection Limit Water	Detection Limit Air
Americium-241	4×10^{-6}	4×10^{-10}	1×10^{-12}	-
Argon-41	-	4×10^{-8}	-	1.5×10^{-8}
Barium-140	3×10^{-5}	-	2×10^{-9}	-
Beryllium-7	-	4×10^{-8}	-	5×10^{-15}
Californium-249	4×10^{-6}	-	1×10^{-12}	-
Californium-252	7×10^{-6}	-	1×10^{-12}	-
Cesium-137	2×10^{-5}	5×10^{-10}	-	1×10^{-16}
Curium-242	2×10^{-5}	-	1×10^{-12}	-
Curium-244	7×10^{-6}	-	1×10^{-12}	-
Hydrogen-3	3×10^{-3}	-	1×10^{-7}	-
Krypton-85	-	3×10^{-7}	-	-
Neptunium-237	3×10^{-6}	-	1×10^{-12}	-
Plutonium-238	5×10^{-6}	1×10^{-12}	1×10^{-12}	3×10^{-19}
Plutonium-239	5×10^{-6}	1×10^{-12}	1×10^{-12}	3×10^{-19}
Radium-226	3×10^{-8}	-	1×10^{-10}	-
Radon-220	-	1×10^{-8}	-	-
Strontium-89	3×10^{-6}	1×10^{-9}	2×10^{-9}	1×10^{-16}
Strontium-90	3×10^{-7}	2×10^{-10}	2.5×10^{-10}	1×10^{-17}
Thorium-228	-	2×10^{-13}	-	1×10^{-18}
Thorium-230	-	3×10^{-13}	-	1×10^{-18}
Thorium-232	-	1×10^{-12}	-	1×10^{-18}
Uranium-234	-	4×10^{-12}	-	1×10^{-19}
Uranium-235	-	4×10^{-12}	-	1×10^{-19}
Uranium-238	-	5×10^{-12}	-	1×10^{-19}
Uranium - natural*	4×10^{-5}	4×10^{-12}	2×10^{-10}	2×10^{-17}
Alpha**	3×10^{-6}	1×10^{-10}	2×10^{-10}	2×10^{-16}
Beta**	to 1×10^{-7}	to 1×10^{-13}	1×10^{-9}	5×10^{-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.

3. Quality Assurance Program

a. Radiochemical Analysis and Radioactivity Measurements

All nuclear instrumentation is calibrated with standard sources obtained from the U. S. National Bureau of Standards (NBS), if possible. If NBS standards were not available for particular nuclides, standards from the Amersham Corporation were used. The equipment is usually checked on a daily basis with secondary counting standards to insure proper operation. Samples were periodically analyzed in duplicate or with the addition of known amounts of a radionuclide to check precision and accuracy. In addition, standard and intercomparison samples distributed by the Quality Assurance Branch of the U. S. Environmental Protection Agency (EPA-QA) are analyzed regularly. Results of our participation in the EPA-QA program during 1984 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).

In addition, participation continued in the DOE Environmental Measurements Laboratory Quality Assurance Program (DOE-EML-QAP), a semi-annual distribution of four or five different sample matrices containing various combinations of radionuclides. Results for 1984 are collected in Table 45. In the table, the comparison is made between the EML value, which is the result of replicate determinations by that Laboratory, and the value obtained in our laboratory. To assist in judging the quality of the results, typical errors for our analyses are 2-50% and the error in the EML results is 1-30% (depending on the nuclide and the amount present). For most analyses for which the differences are large (> 20%), the concentrations were quite low and the differences were within the measurement uncertainties.

b. Penetrating Radiation

Our laboratory participated in the Sixth International Environmental Dosimeter Intercomparison Project conducted by the DOE Radiological and

TABLE 44

Summary of EPA-QA Samples, 1984

Type of Sample	Analysis	Number Analyzed	Average Difference from Added
Air Filter	Total Alpha	2	43%
	Total Beta	2	3%
	Strontium-90	2	13%
	Cesium-137	2	47%
Milk	Potassium-40	2	5%
	Strontium-89	2	21%
	Strontium-90	2	14%
	Iodine-131	3	5%
	Cesium-137	2	5%
Water	Total Alpha	1	47%
	Total Beta	1	22%
	Hydrogen-3	2	4%
	Chromium-51	3	2%
	Cobalt-60	4	2%
	Zinc-65	3	3%
	Strontium-89	3	11%
	Strontium-90	3	7%
	Ruthenium-106	3	7%
	Iodine-131	1	21%
	Cesium-134	4	3%
	Cesium-137	4	5%
	Total Uranium	3	11%
	Plutonium-239	2	16%

TABLE 45

Summary of DOE-EML-QAP Samples, 1984

	Average Difference From EML Value				
	Air Filters	Water	Soil	Tissue	Vegetation
Hydrogen-3	-	10% (1)	-	-	-
Beryllium-7	4% (2)	-	-	-	-
Potassium-40	-	-	8% (2)	20% (1)	10% (2)
Manganese-54	4% (1)	4% (1)	-	-	-
Cobalt-60	2% (1)	3% (1)	8% (1)	-	9% (2)
Strontium-90	10% (2)	17% (2)	14% (2)	35% (1)	4% (2)
Cesium-137	5% (2)	1% (2)	12% (2)	3% (1)	10% (2)
Radium-226	-	-	11% (2)	9% (1)	-
Uranium-234	9% (2)	5% (2)	17% (1)	-	-
Uranium-238	13% (2)	5% (2)	22% (1)	-	-
Plutonium-238	-	-	16% (1)	-	-
Plutonium-239	1% (2)	32% (2)	23% (2)	_	21% (2)
Americium-241	10% (2)	12% (2)	27% (1)	_	-

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

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

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

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

c. *Chemical Analysis*

With each set of atomic absorption analyses, standard amounts of trace metals were analyzed in blank solutions at concentrations corresponding to

50% and 100% of the current State standards. Recoveries were determined by comparing these results to results obtained by analyzing stream and effluent samples to which identical concentrations were added. Average recoveries ranged from about 80-120%, with a standard deviation of 10-30%. 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, zinc, and organic materials. The results (Table 46) were in good agreement with the reference values.

TABLE 46

Summary of Quality Assurance Studies

Constituent	*Average Difference From Reference Value*
Cadmium	*2% (16)*
Lead	*5% (16)*
Zinc	*4% (16)*
Benzene	*4% (4)*
Carbon Tetrachloride	*3% (4)*
Chloroform	*4% (4)*
Methyl Chloroform	*5% (4)*
Methylethyl Ketone	*6% (4)*
Toluene	*4% (4)*
Trichloroethylene	*3% (4)*
O-xylene	*5% (4)*

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

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.

C. Distribution List

Argonne National Laboratory	No. of Copies
ANL Contract Copy	1
ANL Libraries	2
ANL Patent Dept.	1
C. L. Cheever	1
W. J. Dudas	1
T. L. Duffy	5
A. J. Dvorak	1
N. W. Golchert	5
P. F. Gustafson	1
R. L. Herriford	1
R. B. Kasper	1
N. D. Kretz	1
A. H. Kuljian, Jr.	1
F. P. Marchetti	1
D. Nelson	1
D. P. O'Neil	1
OTD File #30.6	1
E. G. Pewitt	1
D. M. Ray	1
J. G. Riha	1
J. Sedlet	25
A. F. Stehney	1
H. C. Svoboda	1
T. TenKate	1
TIS Files	6
J. P. Unik	1
M. A. Wahlgren	1
R. A. Wynveen	1

External	No. of Copies
DOE Technical Information Center for distribution per UC-11	207
DOE-CH Division of Operational and Environmental Safety	5
DOE-HQ Division of Operational Safety	10
Manager, DOE-CH	1
Samuel Baker, Fermilab	1
Robert Buddemeier, Lawrence Livermore Laboratory	1
DuPage County Forest Preserve District	1
DuPage County (IL) Health Department	1
David Edgington, University of Wisconsin at Milwaukee	1
Don Etchison, Illinois Department of Nuclear Safety, Springfield, IL	1
Billy M. Farmer, Mound Laboratory	1
Thomas C. Gunderson, Los Alamos National Laboratory	1
Andrew P. Hull, Brookhaven National Laboratory	1
Illinois Department of Public Health, Springfield, IL	1
Illinois Environmental Protection Agency, Springfield, IL	1
J. B. McCaslin, Lawrence Berkeley Laboratory	1
Tom Oakes, Oak Ridge National Laboratory	1
U. S. Environmental Protection Agency, Region 5, Chicago, IL	1
H. L. Volchok, DOE-EML, New York	1

D. Acknowledgements

We are indebted to C. L. Cheever of the ANL Plant Facilities and Services Division, who provided some of the NPDES data in Section III.B.2.b. and the environmental permit information in Section V.B., and to the ANL-OHS Health Physics Section, who provided some of the radioactive gaseous effluent data. J. P. Unik (ANL), J. J. Nelsen (DOE-CH), and T. W. Joseph (DOE-CH) reviewed the draft manuscript, and made a number of constructive comments for which we are grateful. The dedicated effort of Rita M. Beaver, who typed and prepared the manuscript and performed numerous other tasks needed to complete the report, is greatly appreciated.