

Distribution Category:
Health and Safety (UC-41)

ANL--82-12

DE82 013714

ANL-82-12

NOTICE

PORTIONS OF THIS REPORT ARE UNCLASSIFIED
It has been reproduced from the best
available copy to permit the broadest
possible availability.

ARGONNE NATIONAL LABORATORY

9700 South Cass Avenue
Argonne, Illinois 60439

ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY

ANNUAL REPORT FOR 1981

by

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

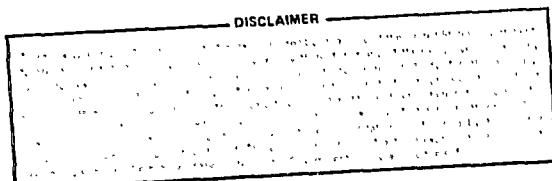
Occupational Health and Safety Division

Laboratory Work Performed by

T. L. Duffy	S. Lewey
N. W. Golchert	D. M. Ray
K. A. Hayes	J. G. Riha
F. S. Iwami	J. Sedlet
R. B. Kasper	H. C. Svoboda

T. TenKate

March 1982



Approved:

D. P. O'Neil
Division Director

Preceding Report in This Series: ANL-81-23

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	9
I. INTRODUCTION	9
A. General	9
B. Description of Site	11
C. Population	14
D. Meteorology	14
E. Geohydrology	17
F. Water and Land Use	19
II. SUMMARY	21
III. MONITORING RESULTS	25
A. Radiological	25
1. Air	27
2. Surface Water	33
3. Soil, Grass, and Bottom Sediment	36
4. Milk	43
5. External Penetrating Radiation	47
6. Potential Radiation Dose Estimates	52
a. Air and Waterborne Radionuclides	52
b. External Penetrating Radiation Dose	57
c. Summary	58
B. Chemical and Biological Pollutants	58
1. Air	58
2. Water	61
a. Waste Treatment Plant Effluent Water	63
b. National Pollution Discharge Elimination System (NPDES)	63
c. Sawmill Creek	66
d. Des Plaines River	79

TABLE OF CONTENTS

	<u>Page</u>
IV. APPENDIX	80
A. References	80
B. Environmental Quality Standards, Detection Limits, Analytical Errors, and Quality Assurance	81
1. Standards	81
2. Detection Limits	83
3. Quality Assurance Program	83
a. Radiochemical Analysis and Radioactivity Measurements	83
b. Penetrating Radiation	85
c. Chemical Analysis	85
d. Sampling, Sample Storage, Other	86
C. Distribution List	88
D. Acknowledgements	89

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Incremental Population Data in the Vicinity of ANL, 1981	15
2.	ANL Weather Summary, 1981	18
3.	Agricultural Production Near ANL, 1980, 1981	20
4.	Total Alpha and Beta Activities in Air-Filter Samples, 1981	28
5.	Gamma-Ray Activity in Air-Filter Samples, 1981	29
6.	Strontium, Thorium, Uranium, and Plutonium Concentrations in Air-Filter Samples, 1981	31
7.	Radionuclides in Sawmill Creek, 1981	35
8.	Radionuclides in Des Plaines River Water, 1981	37
9.	Radionuclides in Illinois River Water, 1981	38
10.	Gamma-Ray Emitting Radionuclides in Soil, 1981	40
11.	Plutonium in Soil, 1981	41
12.	Ground Deposition of Plutonium-239,240	42
13.	Gamma-Ray Emitting Radionuclides in Grass Samples, 1981	44
14.	Radionuclides in Grass Samples, 1981	45
15.	Radionuclides in Bottom Sediment, 1981	46
16.	Radionuclides in Milk, 1981	47
17.	Environmental Penetrating Radiation at Off-Site Locations, 1981 .	48
18.	Environmental Penetrating Radiation at ANL, 1981	50
19.	Concentration-to-Dose Conversion Factors	54
20.	Whole-Body Dose From Airborne Emissions, 1981	55
21.	80 km Whole-Body Population Dose	55
22.	Radionuclide Concentrations and Dose Estimates for Sawmill Creek Water, 1981	56
23.	Summary of National and Illinois Ambient Air Quality Standards .	59
24.	Environmental Air Data, 1981	60
25.	Water Quality Standards and Detection Limits	62
26.	Chemical Constituents in Effluents from ANL Treatment Plant, 1981	64
27.	ANL National Pollution Discharge Elimination System Data Summary, Location 001 (7M), 1981	67
28.	ANL National Pollution Discharge Elimination System Data Summary, Location 002 (13L), 1981	68
29.	ANL National Pollution Discharge Elimination System Data Summary, Location 003 (12L), 1981	69

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
30.	ANL National Pollution Discharge Elimination System Data Summary, Location 004 (14J), 1981	70
31.	ANL National Pollution Discharge Elimination System Data Summary, Location 005 (14F), 1981	71
32.	ANL National Pollution Discharge Elimination System Data Summary, Location 006 (8J), 1981	72
33.	ANL National Pollution Discharge Elimination System Data Summary, Location 007 (9K), 1981	73
34.	ANL National Pollution Discharge Elimination System Data Summary, Location 008 (13M), 1981	74
35.	ANL National Pollution Discharge Elimination System Data Summary, Location 009 (14K), 1981	75
36.	Sawmill Creek - Effect of Sanitary Waste, 1981	77
37.	Chemical Constituents in Sawmill Creek, Location 7M, 1981	78
38.	Chemical Constituents in the Des Plaines River, 1981	79
39.	Concentration Guides and Detection Limits	82
40.	Summary of DOE-EML-QAP Samples, 1981	84
41.	Summary of Quality Assurance Studies	87

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Sampling Locations at Argonne National Laboratory	12
2.	Sampling Locations Near Argonne National Laboratory	13
3.	Monthly and Annual Wind Roses at Argonne National Laboratory, 1981	16
4.	Plutonium-239,240 in Air Concentrations, 1973-1981	32
5.	Plutonium-239,240 Surface Deposition, 1973-1981	39
6.	Penetrating Radiation Measurements at the ANL Site, 1981	51
7.	NPDES Permit Locations	65

ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY

ANNUAL REPORT FOR 1981

by

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

ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1981 are presented and discussed. To evaluate the effect of Argonne operations on the environment, measurements were made for a variety of radionuclides in air, surface water, soil, grass, bottom sediment, and milk; for a variety of chemical constituents in air, surface water, and Argonne effluent water; and of the environmental penetrating radiation dose. Sample collections and measurements were made at the site boundary and off the Argonne site for comparison purposes. Some on-site measurements were made to aid in the interpretation of the boundary and off-site data. The results of the program are interpreted in terms of the sources and origin of the radioactive and chemical substances (natural, fallout, Argonne, and other) and are compared with applicable environmental quality standards. The potential radiation dose to off-site population groups is also estimated.

I. INTRODUCTION

A. General

This report is prepared to provide the U. S. Department of Energy (DOE) and the public with information on the levels of radioactive, chemical, and biological pollutants in the environment of Argonne National Laboratory (ANL) and on the amounts, if any, added to the environment as a result of Argonne operations. The report follows the guidelines given in DOE Order 5484.1, Chapter III.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 potentially toxic chemical and biological 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 an 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 1981 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), electrochemistry of energy storage, solar energy utilization, ocean thermal energy conversion using ammonia as the working fluid, evaluation of heat exchangers, and biomass conversion to alcohol. Other areas of research are the use of superconducting magnets for improved nuclear particle accelerators and magnetohydrodynamics coal technology, the immobilization of radioactive waste products for safe disposal, and the biological effects of small amounts of radiation. Environmental research studies include a Great Lakes radioecology program, toxic trace element ecology, the effect of sulfur dioxide on crop growth, and reclamation of strip mined land. A significant portion of these laboratory studies requires the use of radioactive and chemically-toxic substances.

The principal nuclear facilities at the Laboratory are a 200 kW light-water cooled and moderated biological research reactor (Janus) fueled with fully-enriched uranium; one critical assembly or zero power reactor (ZPR-9), that is fueled at various times with plutonium, uranium, or a combination of the two; the Argonne Thermal Source Reactor (ATSR), a 10 kW research reactor fueled with enriched uranium; a prototype superconducting heavy ion linear accelerator; a 60-inch cyclotron; several other charged particle accelerators (principally of the Van de Graaff and Dynamitron type); a large fast neutron source (IPNS, Intense Pulsed Neutron Source) in which high energy protons strike a heavy metal target to produce the neutrons; cobalt-60 irradiation sources; chemical and metallurgical plutonium laboratories; and several hot cells and laboratories designed for work with multicurie quantities of the actinide elements. Two major facilities, a 12.5 GeV proton accelerator (ZGS, the Zero Gradient Synchrotron) and a 5 MW heavy water-enriched uranium reactor (CP-5) were not in operation during 1981 and are awaiting decontamination and decommissioning. The principal non-nuclear facilities at Argonne that may produce a measurable impact on the environment are the coal-fired

atmospheric fluidized bed boiler (FBB) and the ocean thermal energy conversion (OTEC) studies.

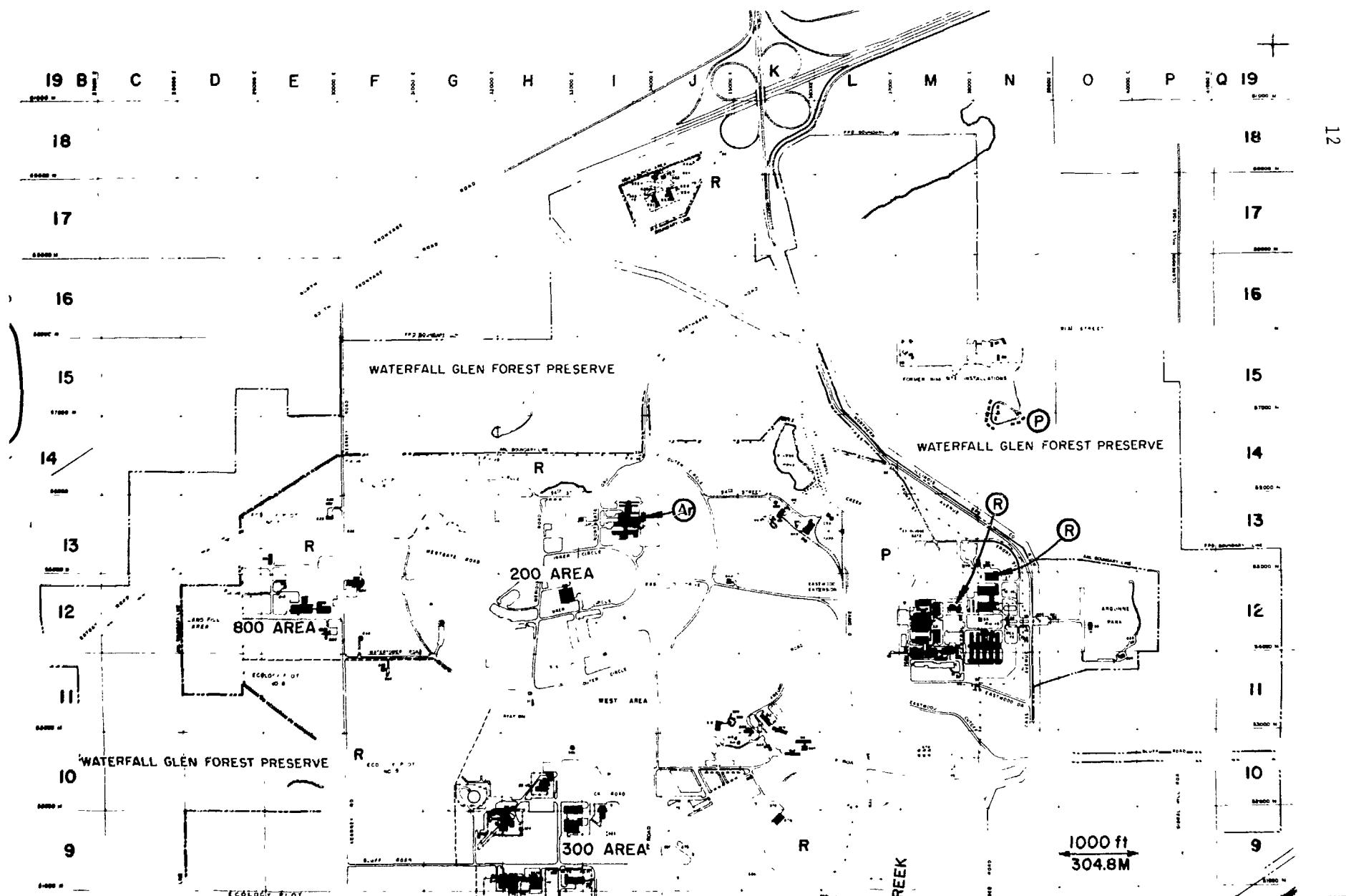
The fluidized bed boiler is designed to burn high sulfur (3.75%) Illinois coal and to produce export steam for Laboratory use. Low sulfur coal was used in the first part of 1981 with no controls to test the system and high sulfur coal was burned in the latter part of the year with controls in place. The OTEC system has the potential for water pollution since it uses large amounts of ammonia.

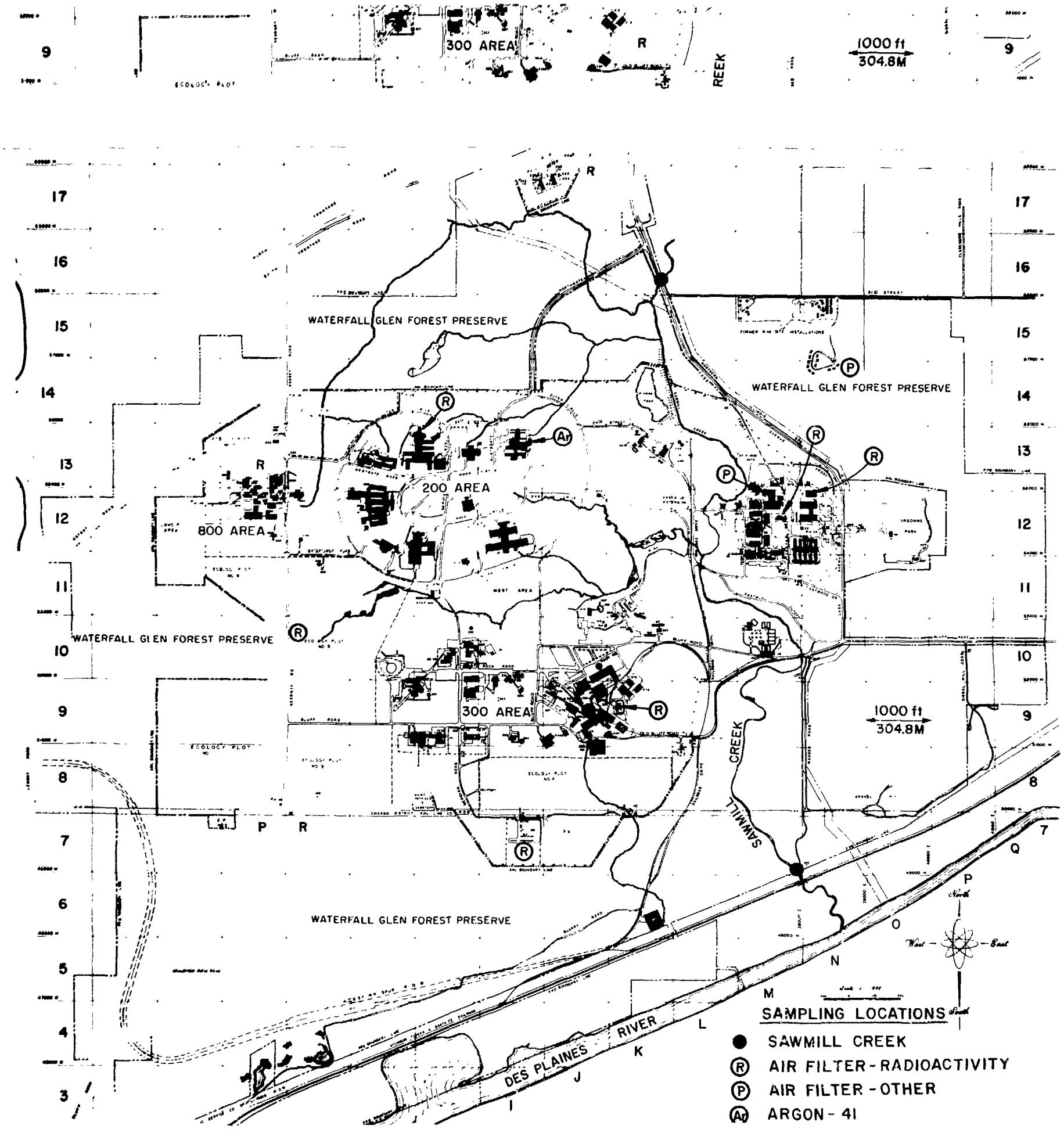
B. Description of Site

Argonne National Laboratory (Illinois site) occupies the central 6.88 sq km (1,700 acres) of a 15.14-sq km (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 8.26-km (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





SAMPLING LOCATIONS

- SAWMILL CREEK
- (R) AIR FILTER - RADIOACTIVITY
- (P) AIR FILTER - OTHER
- (A) ARGON - 41

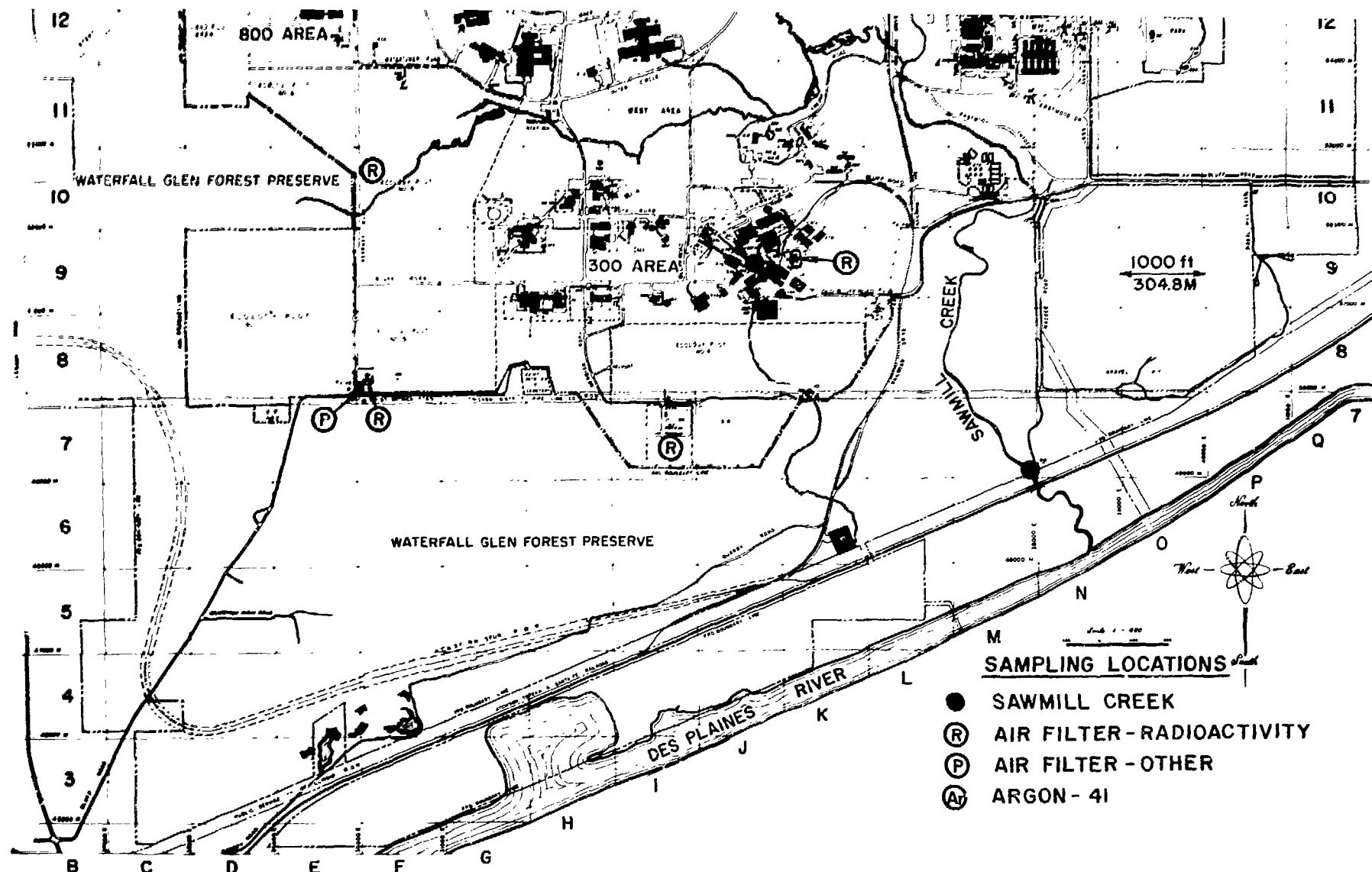


FIG. 1. Sampling Locations at Argonne National Laboratory

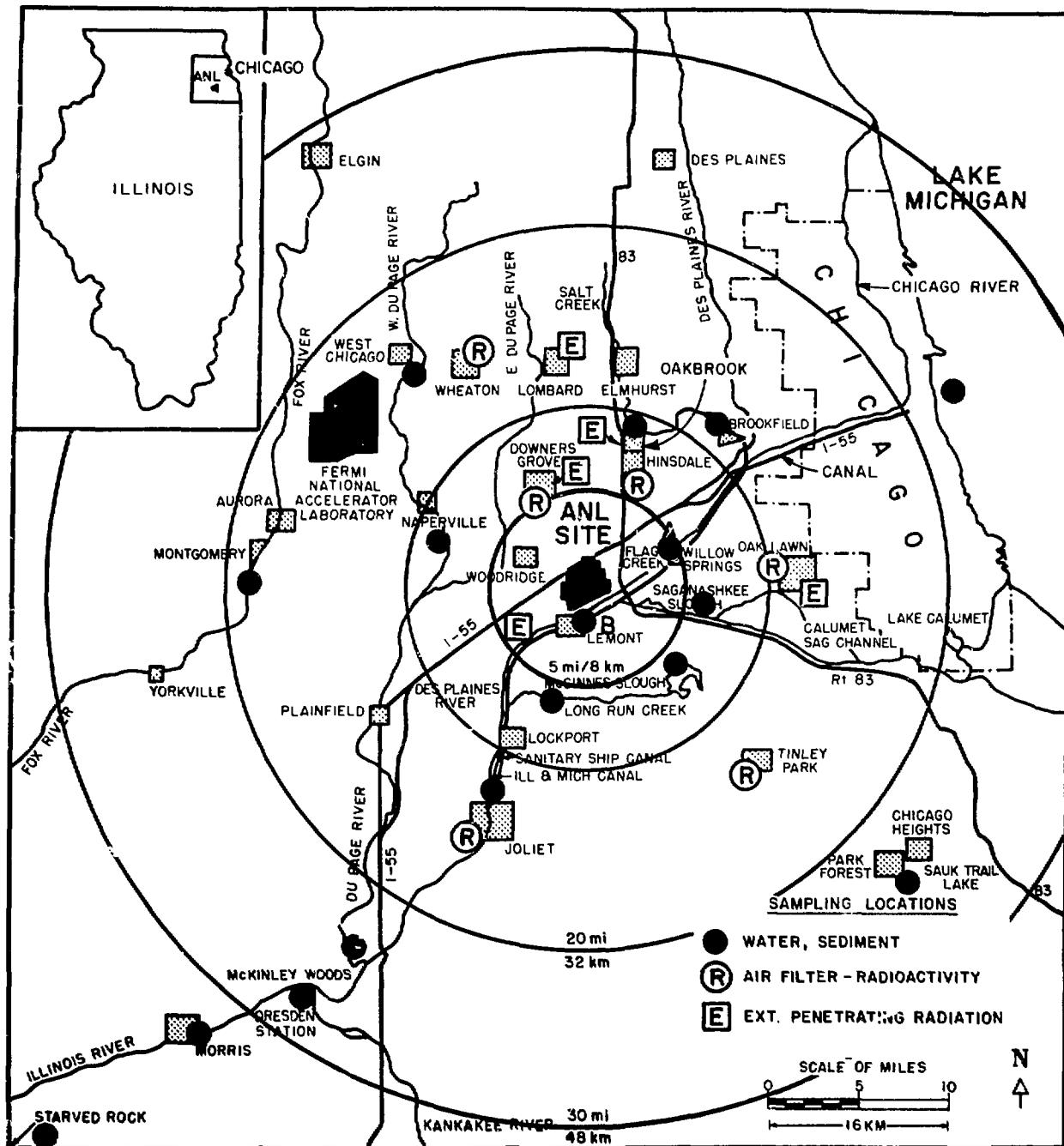


FIG. 2. Sampling Locations Near Argonne National Laboratory

level at the top. The land then slopes gradually upward reaching the average site elevation of 220 m (725 ft) above sea level at 940 m (3,000 ft) from the bluffs. Several large ravines oriented in a north-south direction are located in the southern portion of the site. The bluffs and ravines generally are forested with deciduous trees of an average height of 15-18 m (50-60 ft). The remaining portion of the site changes in elevation by no more than 7.6 m (25 ft) in a distance of 150 horizontal m (500 ft). In the southern portion of the Forest Preserve, the Chicago District Pipe Line Co. and the Atchison, Topeka, and Santa Fe Railroad have rights-of-way. Additional information about the site is given in the Draft Environmental Assessment.²

C. Population

The area around Argonne has exhibited a large population growth in the past 20 years. Large areas of farmland have been converted into housing. A directional and annular 80-km (50-mi) population distribution for the area, which is used for the population dose calculations later in this report, is shown in Table 1. The population distribution, centered on the CP-5 reactor, 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

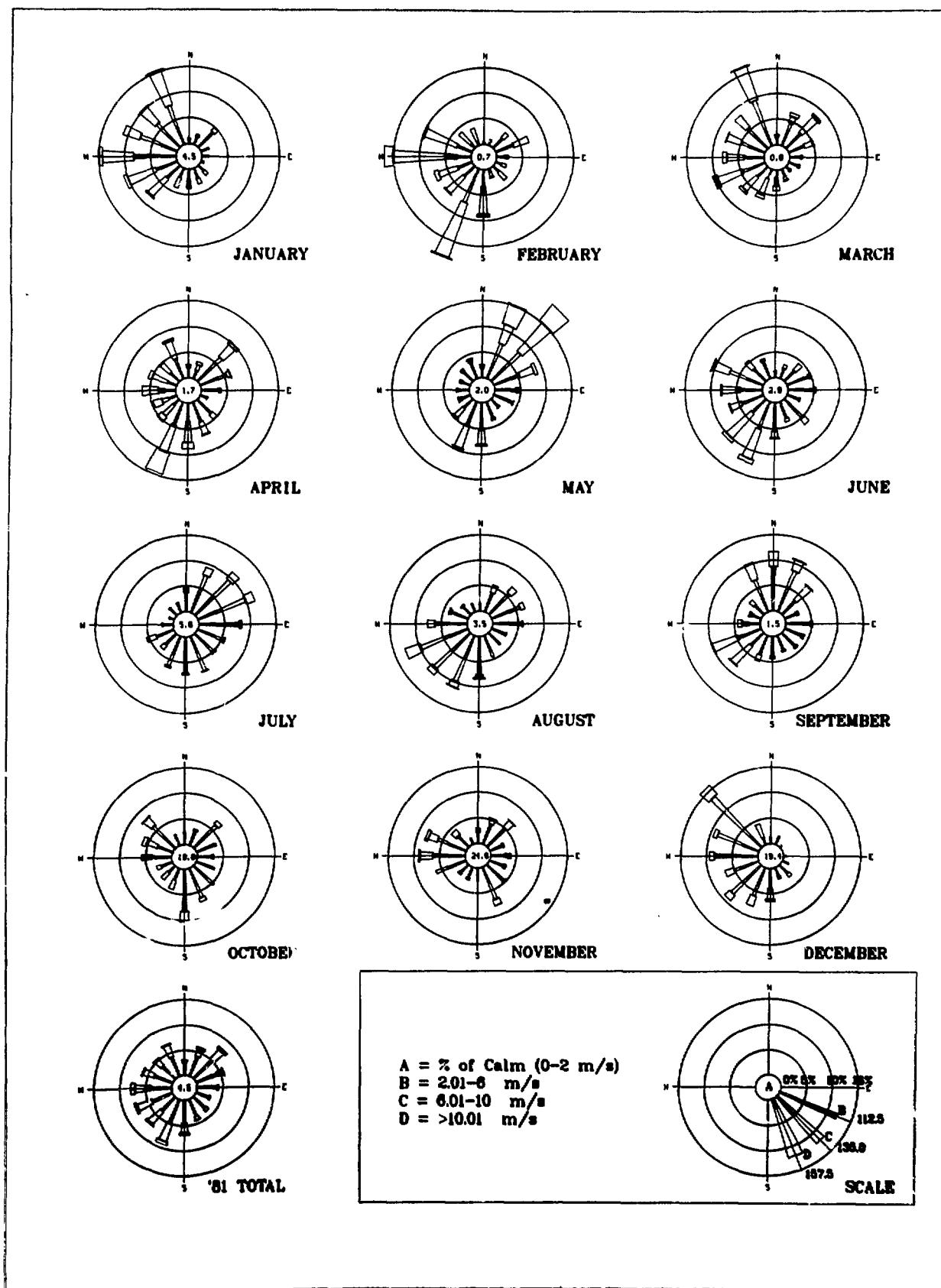
The climate of the area is that of the upper Mississippi Valley, as moderated by Lake Michigan. A summary of the meteorological data collected on the site from 1950 to 1964 is available³ and provides an adequate sample of the climate. The data collected since 1964 has not been published.

The most important meteorological parameters for the purposes of this report are wind direction, wind speed, temperature, and precipitation. The 1981 average monthly and annual wind roses on the Argonne site are shown in Figure 3. The wind roses are polar coordinate plots in which the lengths of the radii represent the percentage frequency of wind speeds in classes of 2.01-6 m/s (4.5-13.4 mph), 6.01-10 m/s (13.4-22.4 mph), and greater than 10.01 m/s (22.4 mph). The number in the center represents the percent of

TABLE 1

Incremental Population Data in the Vicinity of ANL, 1981

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



observations of wind speed less than 2 m/s (4.5 mph) in all directions. The direction of the radii represents the direction from which the wind blows. Sixteen radii are shown on each plot at 22.5° intervals; each radius represents the average wind speed for the direction covering 11.25° on either side of the radius. For example, in the plot labeled May, the northeasterly component is predominant. The length and direction of this vector shows that, of the total wind observations, about 8% were in the 2.01-6 m/s range, about 7% were in the 6.01-10 m/s range, about 5% were greater than 10.01 m/s; and about 20% of the observations were from the direction between 33.75° and 56.25° (northeast).

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 April is south-southwest, while in May it is northeast. This type of wind pattern is typical of this area.

The precipitation and temperature data at Argonne for 1981 are shown in Table 2. Temperatures were fairly close to normal most of the year. Precipitation deviated significantly from the long-term monthly averages, with below normal precipitation in January and March and above normal precipitation in April, June, and August. The 1981 total precipitation (93.1 cm, 36.7 in) was about 10% higher than the annual average (85.2 cm, 33.5 in).

E. Geohydrology

The geology of the Laboratory area consists of about a 30 m (98 ft) thick deposit of glacial till on top of dolomite bedrock. The bedrock at the Laboratory 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.

There are two principal aquifers that are used as water supplies in the Laboratory area. The upper aquifer is the Niagaran-Alexandrian dolomite, which is about 60 m (197 ft) thick in the Laboratory area, and has a piezometric surface between 15 and 30 m (49 and 98 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

TABLE 2

ANL Weather Summary, 1981

Month	Amount	Precipitation (cm)	Temperature (°C)	
		Average *	Monthly Average	Average **
January	0.43	4.88	-5.8	-4.2
February	5.56	4.57	-2.9	-2.7
March	1.50	6.93	3.6	2.5
April	14.35	7.98	11.6	8.8
May	8.58	8.81	13.4	14.6
June	15.44	9.47	21.3	20.2
July	9.52	8.66	21.9	23.2
August	17.68	8.10	21.3	22.5
September	6.60	8.08	16.8	18.7
October	4.57	6.55	9.5	12.5
November	6.25	5.88	4.9	4.7
December	2.67	5.30	-3.9	-1.5

* Average precipitation, 1873-1977, U. S. Weather Bureau, Chicago, Illinois.

** Average temperature 1871-1977, U. S. Weather Bureau, Chicago, Illinois.

shale separates the upper dolomite aquifer from the underlying sandstone aquifer. This shale retards hydraulic connection between the upper and lower aquifers.

The four Laboratory wells now in use are about 90 m (300 ft) deep in the Niagaran dolomite. 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 widely used throughout DuPage County.

F. Water and Land Use

The principal stream that drains the site is Sawmill Creek. This Creek was formerly an intermittent stream, responding in flow rate largely to precipitation runoff. It now carries effluent water continuously from a county sewage treatment plant (Marion Brook Treatment Plant) located a few kilometers north of the site, which has an operating capacity of about 11.4 megaliters (3 million gallons) per day. In addition, the residential development in the area has resulted in the collection and channeling of additional runoff water into the Creek. Treated sanitary and laboratory waste water from Argonne are combined and discharged into Sawmill Creek at location 7M in Figure 1. This effluent averaged 2.7 megaliters (0.72 million gallons) per day, while the water flow in the Creek upstream of the waste-water outfall averaged about 36 megaliters (10 million gallons) per day during 1981. The combined Argonne effluent consisted of 70% laboratory waste water and 30% sanitary waste water.

Sawmill Creek and the Des Plaines River above Joliet, about 21 km (13 mi) southwest of Argonne, receive very little recreational or industrial use. A few people fish in these waters downstream from Argonne and some duck hunting takes place on the Des Plaines River. Water from the Chicago Sanitary and Ship Canal is used for some industrial purposes, such as hydroelectric generators and condensers, and for irrigation at the State prison near Joliet. It is also used as secondary cooling water by Argonne. 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 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 groundwater supplies by infiltration. In the vicinity of the Laboratory, only subsurface water (from both shallow and deep aquifers) and Lake Michigan water are used for drinking purposes.

The principal recreational area near Argonne is Waterfall Glen Forest

Preserve, which surrounds the site as described in Section I.B. and is shown in Figure 1. The area is available for hiking, skiing, and equestrian sports. Very approximate estimates of usage are 600 individuals per day on weekends and 500 individuals for group educational purposes during 1981. The average stay is about two hours. Sawmill Creek flows through the south portion of the Preserve on its way to the Des Plaines River. This region of the Preserve (formerly named Rocky Glen) was used principally for picnicking, hiking, and overnight camping by youth groups, but has been closed for rehabilitation since June 6, 1976. East and southeast of Argonne and the Des Plaines River are located several large forest preserves of the Cook County Forest Preserve District. The preserves include the two sloughs shown in Figure 2, McGinnis and Saganashkee, as well as other smaller lakes. These areas are used for picnicking, boating, fishing, and hiking. A small park located in the eastern portion of the Argonne site (12-0 in Figure 1) is for the use of Argonne and Department of Energy employees only.

The most recently available information on milk and principal agricultural production in a 10,600 sq km (4,100 sq mi) area around Argonne is shown by County in Table 3. These estimates were obtained from the Illinois Crop Reporting Service of the State Department of Agriculture. For corn and soybeans, the data are for 1980; for milk, wheat, and oats, the data are for 1981.

TABLE 3

Agricultural Production Near ANL, 1980, 1981

County	Milk		Corn	Soybeans	Wheat	Oats
	No. of Cows	Million Pounds				
DuPage	200	2.2	2.2	0.61	0.13	0.045
Cook	200	2.2	2.2	0.74	0.13	0.048
Will	3,000	32.4	18.9	5.2	0.37	0.28
Kane	6,300	68.1	15.8	2.8	0.31	0.30
Kendall	700	7.6	12.0	2.6	0.16	0.10
Grundy	700	7.6	15.4	3.4	0.082	0.062
Lake	1,600	17.3	2.5	1.0	0.48	0.12

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

II. SUMMARY

This is one in a series of annual reports prepared to provide the U. S. Department of Energy (DOE) and the public with information on the level of radioactive and chemical pollutants in the environment and on the amounts of such substances, if any, added to the environment as a result of Argonne operations. The previous report in this series is ANL-81-23.⁴ Included in this report are the results of measurements obtained in 1981 for a number of radionuclides in air, surface water, soil, grass, bottom sediment, and milk; for a variety of chemical constituents in air and water, and for the external penetrating radiation dose.

Total alpha and beta activities, fission and activation products, plutonium, thorium, and uranium were measured in air-filter samples collected continuously at the site perimeter and off the site. All of the off-site and perimeter samples contained only radionuclides from natural sources and nuclear test detonations. Intermediate half-life fission products were detected at all sampling locations and are the result of fallout from the Chinese atmospheric nuclear test of October 16, 1980. No activity attributable to Argonne operations could be detected.

The plutonium-239,240 concentrations in air averaged $18 \times 10^{-18} \mu\text{Ci/ml}$,* about three times the 1980 value, and were nearly identical at all sampling locations. The monthly variations showed a "spring maximum" in stratospheric fallout of plutonium similar to that observed for beryllium-7 (a naturally-occurring nuclide produced in the stratosphere by cosmic-ray interactions) and several fission products. The results indicated that the airborne plutonium was from atmospheric nuclear test detonations and there was no evidence that any of the plutonium originated at Argonne. The average plutonium concentration was equivalent to 0.0018% of the Concentration Guide (CG).**

* The radioactivity units are described in Section III.

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

The major airborne radionuclides released from the Laboratory were argon-41 and krypton-85. The maximum dose from these two nuclides at the site boundary was in the north-northeast direction and was less than 0.001 mrem/yr, 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) northeast of the site boundary, was less than 0.0005 mrem/yr. These releases constitute an insignificant addition to the dose received from the natural background radiation, which is about 90 mrem/yr.

Argonne waste water is discharged into Sawmill Creek, and this stream was sampled above and below the site to evaluate the effect of Argonne operations on its radioactive content. The nuclides (for which analyses were made) added to the Creek in the waste water, and the percent of CG of their net average Creek concentrations, were hydrogen-3, 0.004%; strontium-90, 0.04%; neptunium-237, 0.00025%; plutonium-239,240, 0.00017%; americium-241, 0.00009%; and curium-244 and/or californium-249, 0.0000 . Although Sawmill Creek is not a source of potable water, the dose to an individual using water at these concentrations as his sole source of drinking water would be less than 1.2 mrem/yr.

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

Plutonium concentrations in soil showed the same general range and average at the site perimeter and off the site as in the past years. The average plutonium-239,240 content of the top 5 cm (2 in) of soil was $1.1 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ at the site perimeter and $0.9 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ off the site. The corresponding plutonium-238 averages were $0.11 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ and $0.10 \times 10^{-3} \mu\text{Ci}/\text{m}^2$, respectively. The plutonium content of grass was similar to that found in previous years and was about a factor of 10^4 less than soil from the same locations. The results were within the range reported by other laboratories for fallout from test detonations, and the plutonium found in soil and grass is attributed to this source. The plutonium content of samples from beds of streams and ponds ranged from $2 \times 10^{-9} \mu\text{Ci}/\text{g}$ to $7 \times 10^{-9} \mu\text{Ci}/\text{g}$ of plutonium-239,240, a range found in previous years to be normal for fallout plutonium in such materials.

Milk from a dairy farm near the Laboratory was analyzed for several fission products, including hydrogen-3. Hydrogen-3 concentrations averaged $< 115 \times 10^{-9} \mu\text{Ci}/\text{ml}$. The strontium-90 concentration ($4.7 \times 10^{-9} \mu\text{Ci}/\text{ml}$), and the cesium-137 concentration ($1.7 \times 10^{-9} \mu\text{Ci}/\text{ml}$) increased by 50% compared to 1980.

Measurements of penetrating radiation were made at several locations at the site boundary and off the site. The off-site results averaged 92 mrem/yr with a standard deviation of 6 mrem/yr, which is in the normal range for the area. At three locations at the site boundary, above-normal readings were recorded that were attributable to Argonne operations. At two locations at the south fence the dose rates averaged, respectively, about 490 mrem/yr above normal (grid 7I in Figure 1) and 5-10 mrem/yr above normal (grid 8H) as a result of radiation from an on-site temporary storage facility for radioactive waste. About 300 m (0.2 mi) south of the fence, the measured dose rate dropped to 105 ± 10 mrem/yr, which is slightly above the normal range. Along the north side of the site, the dose at the fence at location 14I was about 24 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 0.06 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 0.004 mrem/yr. Thus, doses to individuals near the site from these sources will not exceed 0.01% of the 500 mrem/yr limit.

Concentrations of total suspended particulates (TSP) were determined upwind and downwind of the coal burning steam plant. Results were essentially the same as those obtained in previous years and were at or below the primary federal standard. Both continuous and 24-hour TSP samplers yielded average results which were statistically identical. Results for sulfur dioxide were also below the standard. On the basis of these results, the initiation of coal burning in February, 1981, did not have any effect on these air quality parameters. Concentrations of airborne iron, copper, lead, and bromine differed slightly from previous years, but were independent of location and unrelated to Argonne operations.

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

Results from the nine* NPDES permit locations indicated general compliance, with the exception of mercury at location 001 (7M) and pH at location 009 (14K). The elevated mercury levels resulted from occasional releases which occurred even though reasonable care is taken to prevent them. New limits recently adopted by Illinois for effluents of the type released by Argonne are substantially higher than levels found at location 001. The pH values that exceeded the limit resulted from the alkaline water conditioning process used for Argonne domestic water. They have no impact on the receiving stream since the relative volume and buffering capacity of the effluent are low.

The average values in Sawmill Creek for dissolved oxygen and chemical constituents, except ammonia nitrogen and dissolved solids, were within the State of Illinois Water Quality Standards. The average ammonia nitrogen level above the Argonne waste-water outfall was 1.8 times the State standard, and exceeded this value in 54% of the samples, while levels below the outfall were 1.5 times the State standard and exceeded this value in 50% of the samples. The average concentration of dissolved solids above the waste-water outfall exceeded the State standard by 12%, while below the outfall it was 2% above the standard. The elevated levels of ammonia nitrogen and total dissolved solids were due to effluent from a county sewage treatment plant upstream of the Laboratory. Individual values for copper, iron, manganese, and mercury exceeded State standards from 2% to 25% of the time. Hexavalent chromium levels were not in excess of the State standard at any time. Concentrations of mercury, iron, and zinc in the Des Plaines River were not affected by the amounts released in Argonne effluent water.

*There are ten permit locations, but no water was present at one location during 1981.

III. MONITORING RESULTS

A. Radiological

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

The results of radioactivity measurements are expressed in this report in terms of microcuries per milliliter ($\mu\text{Ci}/\text{ml}$) for water, air, and milk and microcuries per gram (g) and square meter (m^2) for soil 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 (\pm) limit value. Unless otherwise stated, this value is the standard error at the 95% confidence level calculated from the standard deviation of the average, and is a measure of the range in the concentrations encountered at that location. It does not represent the conventional uncertainty in the average of repeated measurements on the same or identical samples. Since many of the variations observed in environmental radioactivity are not random but occur for specific reasons (e.g., nuclear testing), samples collected from the same location at different times are not replicates. The more random the variation in activity at a particular location, the closer the confidence limits will represent the actual distribution of values at that location. The averages and confidence limits should be interpreted with this in mind. When a plus-or-minus figure accompanies an individual result in this report, it represents the statistical counting error at the 95% confidence level.

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

The new dose assessment and evaluation procedures and systems recommended by the International Commission on Radiological Protection (ICRP) in their reports ICRP-26,⁶ *et seq.*, are not used in this report for the following reasons. These recommendations have not yet been officially adopted by DOE, although it is anticipated that they will be used by DOE and all regulatory agents in the future. The annual whole body dose limit recommended in ICRP-26 for both occupational and public exposure has not changed from their earlier recommendations, and these are already in effect as DOE standards; and the dose to

off-site individuals and the population from Argonne operations, as will be shown, is very small and entirely whole body. In addition, whole body equivalent doses calculated from the annual limit of intake and the corresponding derived air and water concentrations are not different in any significant way from the organ doses calculated from the Concentration Guides and dose conversion factors used in this report.

1. Air

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

The total alpha and beta activities in the individual weekly samples are summarized in Table 4. These measurements were made in low-background gas-flow proportional counters, and the counting efficiencies used to convert counting rates to disintegration rates were those measured for radon decay products on filter paper. The average concentrations of a number of gamma-ray emitters, as determined by gamma-ray spectrometry performed on composite weekly samples are given in Table 5. The gamma-ray detector is a shielded 74 cm³ lithium-drifted germanium diode, calibrated for each gamma-ray emitting nuclide listed in Table 5.

The alpha activities, principally due to naturally-occurring nuclides, averaged the same as the past several years and were in their normal range.

*The site perimeter samplers are placed at the nearest location to the site boundary fence that provides electrical power and shelter.

TABLE 4
TOTAL ALPHA AND BETA ACTIVITIES IN AIR-FILTER SAMPLES, 1981
(CONCENTRATIONS IN 10^{-15} MICROCURIES/ML)

MONTH	LOCATION	NO. OF SAMPLES	AV.	ALPHA ACTIVITY			AV.	BETA ACTIVITY		
				MIN.	MAX.			MIN.	MAX.	
JANUARY	OFF-SITE PERIMETER	22 35	2.1 2.2	1.3 1.6	3.9 2.8		116 111	65 68	249 224	
FEBRUARY	OFF-SITE PERIMETER	20 35	1.8 1.7	1.0 0.7	5.2 3.0		104 103	59 62	206 170	
MARCH	OFF-SITE PERIMETER	21 34	2.0 1.8	0.9 1.0	3.9 5.0		187 205	88 100	422 402	
APRIL	OFF-SITE PERIMETER	21 31	2.2 2.0	1.0 0.9	7.4 4.4		293 303	190 182	428 498	
MAY	OFF-SITE PERIMETER	22 38	2.7 2.2	0.5 1.3	6.8 3.4		277 274	67 118	595 384	
JUNE	OFF-SITE PERIMETER	22 34	2.0 1.8	0.7 0.9	3.6 4.0		155 159	104 112	247 279	
JULY	OFF-SITE PERIMETER	20 35	1.9 1.7	0.3 0.8	3.9 2.5		107 107	29 53	175 165	
AUGUST	OFF-SITE PERIMETER	20 34	1.8 1.6	0.3 0.9	4.6 3.1		54 55	16 15	96 85	
SEPTEMBER	OFF-SITE PERIMETER	21 34	2.0 2.0	0.6 0.8	4.9 4.2		33 31	20 15	55 56	
OCTOBER	OFF-SITE PERIMETER	24 38	2.1 1.6	1.0 0.2	3.5 3.9		31 29	12 15	73 46	
NOVEMBER	OFF-SITE PERIMETER	17 32	2.2 2.6	0.9 0.9	4.2 7.4		33 30	21 21	53 48	
DECEMBER	OFF-SITE PERIMETER	22 27	1.7 1.7	0.7 0.7	3.3 3.2		31 31	20 22	44 46	
ANNUAL SUMMARY	OFF-SITE PERIMETER	252 407	2.0 ± 0.03 1.9 ± 0.03	0.3 0.2	7.4 7.4		118 ± 12 120 ± 9.6	12 15	595 498	
% CG	OFF-SITE PERIMETER	-	(0.002) (0.002)	(0.0003) (0.0002)	(0.007) (0.007)		0.118 0.120	0.011 0.015	0.595 0.498	

* 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.

-15
NOTE: ($10^{-15} = 10^{-18}$)

TABLE 5
GAMMA-RAY ACTIVITY IN AIR-FILTER SAMPLES, 1981
(CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

MONTH	LOCATION	BE7	MN54	Y88	ZR95	HB95	RU103	RU106-RH106	SB125	CS137	CE141	CE144
JANUARY	OFF-SITE PERIMETER	70 70	0.2 0.2	< 0.1 < 0.1	15.2 15.4	20.7 21.1	10.9 12.1	3.5 2.7	0.3 0.3	0.4 0.4	9.0 9.3	6.5 6.8
FEBRUARY	OFF-SITE PERIMETER	52 54	0.2 0.2	< 0.1 < 0.1	13.8 14.5	21.0 22.6	8.8 9.9	2.9 3.4	0.3 0.5	0.4 0.5	6.3 6.7	7.3 8.0
MARCH	OFF-SITE PERIMETER	58 55	0.6 0.5	0.2 0.2	27.7 26.4	47.7 46.1	13.9 13.9	8.3 8.2	1.1 0.8	1.2 0.9	8.7 8.4	20.0 18.7
APRIL	OFF-SITE PERIMETER	87 77	1.1 0.8	0.6 0.3	42.3 40.1	77.1 75.9	18.3 18.4	17.4 15.9	2.2 2.2	2.4 2.4	10.4 10.0	39.4 36.8
MAY	OFF-SITE PERIMETER	87 90	1.2 1.3	0.3 0.3	34.3 38.3	68.6 75.6	13.0 15.0	16.9 19.8	2.0 2.2	2.8 3.1	6.6 7.5	41.7 45.2
JUNE	OFF-SITE PERIMETER	68 60	0.7 0.7	0.2 0.2	14.8 15.4	31.0 31.3	5.1 5.0	10.4 11.2	1.0 1.2	1.8 1.7	2.3 2.3	23.7 23.8
JULY	OFF-SITE PERIMETER	72 67	0.4 0.5	0.1 0.1	7.9 7.8	17.7 19.3	2.0 2.6	6.8 7.5	1.0 0.9	1.3 1.5	0.9 0.9	15.9 17.1
AUGUST	OFF-SITE PERIMETER	74 77	0.3 0.3	< 0.1 < 0.1	2.7 3.2	6.0 6.8	0.5 0.6	3.5 3.9	0.6 0.4	0.7 0.8	0.2 0.2	7.5 8.6
SEPTEMBER	OFF-SITE PERIMETER	51 59	0.1 0.1	< 0.1 < 0.1	0.7 0.8	1.4 1.5	0.1 0.2	1.4 1.1	0.2 0.2	0.3 0.3	0.1 0.1	2.1 2.6
OCTOBER	OFF-SITE PERIMETER	60 55	< 0.1 0.1	< 0.1 < 0.1	0.3 0.4	0.7 0.6	0.1 0.1	0.5 0.4	0.2 0.1	0.2 0.2	0.1 0.1	1.3 1.4
NOVEMBER	OFF-SITE PERIMETER	50 56	< 0.1 < 0.1	< 0.1 < 0.1	0.3 0.2	0.3 0.3	< 0.1 < 0.1	0.6 0.5	0.2 0.1	0.1 0.2	0.1 0.1	1.3 1.0
DECEMBER	OFF-SITE PERIMETER	43 44	0.1 < 0.1	< 0.1 < 0.1	0.2 0.2	0.1 0.2	< 0.1 < 0.1	0.5 0.6	< 0.1 < 0.1	0.1 0.1	< 0.1 < 0.1	1.0 0.7
ANNUAL SUMMARY	OFF-SITE PERIMETER	64± 9 64± 8	0.4±0.2 0.4±0.2	< 0.19 < 0.15	13.4±13.4 13.6±13.6	24.4±16.3 25.1±16.7	6.1±6.0 6.5±4.2	6.1±3.7 6.3±3.9	0.8±0.4 0.7±0.5	1.0±0.6 1.0±0.6	3.7±2.5 3.8±2.5	14.0±8.8 14.2±8.9
% CG (X 1E-3)	OFF-SITE PERIMETER	(0.16) (0.16)	0.04 0.04	- *	1.3 1.4	8.0 8.3	2.0 2.1	3.0 3.1	0.08 0.07	0.20 0.20	0.1 0.1	7.0 7.1

* NO CG LISTED IN DOE ORDER 5480.1 CHAPTER XI

The average beta activity for the year, $1.2 \times 10^{-13} \mu\text{Ci}/\text{ml}$, was about four times higher than the 1980 average and this increase was due to stratospheric fallout from the October 16, 1980, atmospheric nuclear test by the People's Republic of China. The increased beta activity occurred primarily in the spring and as Table 5 indicates, was principally due to the intermediate half-life radionuclides manganese-54, yttrium-88, zirconium-95, niobium-95, ruthenium-103, ruthenium-106-rhodium-106, cerium-141, and cerium-144. These radionuclides and the stratospherically-produced beryllium-7 exhibit the same spring increase in concentrations, indicating their stratospheric origin. Concentrations of iodine-131 and barium-140 were less than their respective detection limits of $5 \times 10^{-15} \mu\text{Ci}/\text{ml}$ and $1 \times 10^{-16} \mu\text{Ci}/\text{ml}$, in all samples.

Samples for radiochemical analyses were collected at perimeter locations 12N and 7I (Fig. 1) and off the site in Downers Grove (Fig. 2). Collections were made on a polystyrene filter medium. 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. This treatment has been found in our laboratory to solubilize plutonium that has been ignited at 1000°C.

Plutonium and thorium were separated on an anion exchange column and the uranium was extracted from the column effluent. Following the extraction, the aqueous phase was analyzed for radiostrontium by a standard radiochemical procedure. The separated plutonium, thorium, and uranium fractions were electrodeposited and measured by alpha spectrometry. The chemical recoveries were monitored by adding known amounts of plutonium-242, thorium-234, and uranium-232 tracers prior to ignition. Since alpha spectrometry cannot distinguish between plutonium-239 and plutonium-240, it should be understood that when plutonium-239 is mentioned in this report, the alpha activity due to the plutonium-240 isotope is also included. The results are given in Table 6.

The average strontium-90 and plutonium-239 concentrations increased by a factor of three compared to 1980 and are consistent with the increase observed in the annual average of the long-lived nuclide cesium-137 in Table 5. The factor of ten increase in average strontium-89 concentrations is also consistent with similar half-life nuclides in Table 5. Figure 4 shows the

TABLE 6
STRONTIUM, THORIUM, URANIUM AND PLUTONIUM CONCENTRATIONS IN AIR-FILTER SAMPLES, 1981
(CONCENTRATIONS IN 1E-18 MICROCURIES/ML)

MONTH	LOCATION	SR-89	SR-90	TH-228	TH-230	TH-232	U-234	U-235	U-238	PU-238	PU-239
JANUARY	7I	7770 \pm 160	290 \pm 20	5 \pm 4	12 \pm 3	6 \pm 2	15 \pm 2	0.3 \pm 0.3	14 \pm 2	< 0.3	7.7 \pm 1.0
	12N	9970 \pm 320	230 \pm 30	9 \pm 2	13 \pm 1	8 \pm 1	17 \pm 2	< 0.1	16 \pm 2	< 0.3	8.0 \pm 1.0
	OFF-SITE	6760 \pm 300	170 \pm 30	3 \pm 1	6 \pm 1	3 \pm 1	6 \pm 1	0.4 \pm 0.4	6 \pm 1	< 0.3	6.3 \pm 1.0
FEBRUARY	7I	7830 \pm 360	310 \pm 60	15 \pm 1	18 \pm 1	9 \pm 1	21 \pm 1	1.0 \pm 0.3	17 \pm 1	< 0.3	9.0 \pm 1.3
	12N	9300 \pm 340	310 \pm 50	15 \pm 1	22 \pm 1	11 \pm 1	28 \pm 2	0.9 \pm 0.3	26 \pm 2	< 0.3	11 \pm 1.4
	OFF-SITE	6030 \pm 160	260 \pm 20	16 \pm 1	19 \pm 1	10 \pm 1	22 \pm 1	0.2 \pm 0.1	22 \pm 1	< 0.3	8.5 \pm 1.4
MARCH	7I	15350 \pm 240	860 \pm 50	21 \pm 2	22 \pm 1	15 \pm 1	27 \pm 2	1.4 \pm 0.4	23 \pm 2	0.4 \pm 0.4	26 \pm 1.8
	12N	17570 \pm 290	930 \pm 70	23 \pm 2	24 \pm 2	19 \pm 1	39 \pm 2	0.7 \pm 0.3	23 \pm 2	< 0.3	28 \pm 2.0
	OFF-SITE	12430 \pm 70	670 \pm 20	9 \pm 1	7 \pm 1	4 \pm 1	13 \pm 1	< 0.1	9 \pm 1	0.5 \pm 0.4	15 \pm 1.4
APRIL	7I	18220 \pm 350	1460 \pm 110	39 \pm 4	52 \pm 3	38 \pm 3	52 \pm 3	0.4 \pm 0.7	42 \pm 2	1.3 \pm 0.5	44 \pm 2.6
	12N	22260 \pm 300	1560 \pm 90	35 \pm 2	40 \pm 2	29 \pm 2	43 \pm 2	0.3 \pm 0.6	37 \pm 2	0.8 \pm 0.4	44 \pm 2.5
	OFF-SITE	17620 \pm 140	1340 \pm 40	22 \pm 2	23 \pm 1	16 \pm 1	25 \pm 2	0.5 \pm 0.4	21 \pm 3	1.1 \pm 0.6	39 \pm 2.6
MAY	7I	14840 \pm 300	1700 \pm 120	18 \pm 2	27 \pm 2	17 \pm 1	35 \pm 2	1.1 \pm 0.3	30 \pm 2	1.2 \pm 0.4	45 \pm 2.4
	12N	16810 \pm 400	1860 \pm 160	21 \pm 2	25 \pm 2	16 \pm 1	28 \pm 2	0.8 \pm 0.3	26 \pm 2	0.9 \pm 0.5	56 \pm 3.0
	OFF-SITE	13310 \pm 220	1580 \pm 90	16 \pm 2	20 \pm 1	11 \pm 1	29 \pm 2	1.2 \pm 0.3	23 \pm 2	1.1 \pm 0.4	43 \pm 2.5
JUNE	7I	4960 \pm 520	920 \pm 40	13 \pm 2	18 \pm 2	12 \pm 1	22 \pm 2	0.7 \pm 0.3	20 \pm 1	0.9 \pm 0.4	25 \pm 1.8
	12N	7250 \pm 430	1360 \pm 30	12 \pm 2	16 \pm 2	11 \pm 1	26 \pm 2	1.1 \pm 0.3	22 \pm 2	0.6 \pm 0.4	34 \pm 2.1
	OFF-SITE	4200 \pm 380	750 \pm 30	8 \pm 2	10 \pm 2	6 \pm 1	14 \pm 2	0.3 \pm 0.3	13 \pm 2	0.8 \pm 0.4	20 \pm 1.6
JULY	7I	3020 \pm 240	840 \pm 30	6 \pm 6	14 \pm 3	5 \pm 2	15 \pm 1	0.8 \pm 0.3	14 \pm 1	< 0.3	25 \pm 1.7
	12N	4110 \pm 450	1040 \pm 50	10 \pm 2	10 \pm 1	6 \pm 1	20 \pm 5	0.9 \pm 1.3	15 \pm 4	< 0.3	29 \pm 1.8
	OFF-SITE	2790 \pm 760	980 \pm 90	5 \pm 2	9 \pm 1	5 \pm 1	13 \pm 1	0.6 \pm 0.3	10 \pm 1	< 0.3	25 \pm 1.8
AUGUST	7I	870 \pm 310	480 \pm 50	6 \pm 1	10 \pm 1	5 \pm 1	9 \pm 2	0.5 \pm 0.5	8 \pm 1	< 0.3	12 \pm 1.2
	12N	1180 \pm 320	480 \pm 50	2 \pm 1	4 \pm 1	2 \pm 1	8 \pm 2	0.3 \pm 0.3	9 \pm 1	0.5 \pm 0.6	13 \pm 1.7
	OFF-SITE	800 \pm 230	370 \pm 40	2 \pm 1	4 \pm 1	2 \pm 1	8 \pm 2	0.7 \pm 0.5	10 \pm 2	0.4 \pm 0.7	13 \pm 2.7
SEPTEMBER	7I	560 \pm 20	180 \pm 90	7 \pm 2	10 \pm 2	7 \pm 1	23 \pm 2	0.5 \pm 0.8	26 \pm 2	0.5 \pm 0.7	6.7 \pm 0.9
	12N	240 \pm 80	180 \pm 20	5 \pm 2	9 \pm 2	5 \pm 1	15 \pm 1	0.5 \pm 0.3	12 \pm 1	< 0.3	7.0 \pm 0.9
	OFF-SITE	220 \pm 90	170 \pm 20	2 \pm 2	4 \pm 1	2 \pm 1	6 \pm 1	0.3 \pm 0.2	7 \pm 1	< 0.3	8.2 \pm 1.3
OCTOBER	7I	< 100	160 \pm 30	9 \pm 3	13 \pm 2	8 \pm 3	20 \pm 2	0.7 \pm 0.3	13 \pm 1	0.9 \pm 0.5	6.9 \pm 1.0
	12N	< 100	140 \pm 30	9 \pm 4	10 \pm 4	9 \pm 4	23 \pm 2	0.4 \pm 0.2	12 \pm 1	0.5 \pm 0.5	6.0 \pm 0.9
	OFF-SITE	< 100	160 \pm 40	2 \pm 2	2 \pm 1	1 \pm 1	9 \pm 3	< 0.1	6 \pm 2	0.5 \pm 0.5	4.6 \pm 0.8
NOVEMBER	7I	< 100	130 \pm 20	9 \pm 2	13 \pm 2	8 \pm 2	21 \pm 2	0.7 \pm 0.3	21 \pm 2	0.4 \pm 0.5	3.2 \pm 1.1
	12N	< 100	120 \pm 10	4 \pm 2	10 \pm 1	4 \pm 1	28 \pm 2	1.0 \pm 0.4	27 \pm 2	0.6 \pm 0.5	7.1 \pm 1.3
	OFF-SITE	< 100	90 \pm 40	2 \pm 3	4 \pm 1	2 \pm 1	26 \pm 2	0.8 \pm 1.0	24 \pm 2	0.6 \pm 0.9	10 \pm 2.5
DECEMBER	7I	< 100	160 \pm 80	12 \pm 2	24 \pm 3	12 \pm 2	-	-	0.9 \pm 0.8	2.7 \pm 1.0	
	12N	< 100	100 \pm 30	9 \pm 3	17 \pm 2	9 \pm 1	29 \pm 2	0.8 \pm 0.3	28 \pm 2	0.8 \pm 0.6	2.7 \pm 2.4
	OFF-SITE	< 100	70 \pm 30	3 \pm 1	5 \pm 1	2 \pm 1	8 \pm 2	0.4 \pm 0.4	8 \pm 1	< 0.3	1.6 \pm 0.6
ANNUAL	7I	6130 \pm 4050	620 \pm 320	13 \pm 6	19 \pm 7	12 \pm 5	22 \pm 8	0.7 \pm 0.2	19 \pm 7	0.6 \pm 0.3	18 \pm 9
	12N	7400 \pm 4780	690 \pm 380	14 \pm 5	18 \pm 6	11 \pm 4	26 \pm 6	0.6 \pm 0.2	21 \pm 5	0.5 \pm 0.2	20 \pm 10
	OFF-SITE	5360 \pm 3670	550 \pm 310	7 \pm 4	9 \pm 4	5 \pm 3	15 \pm 5	0.5 \pm 0.2	13 \pm 4	0.5 \pm 0.2	16 \pm 8
ZCG (X 1E-3)	7I	0.61	0.31	(6.6)	(6.4)	(1.2)	(0.54)	(0.02)	(0.38)	0.06	1.8
	12N	0.74	0.35	(6.9)	(5.9)	(1.1)	(0.64)	(0.02)	(0.42)	0.05	2.0
	OFF-SITE	0.54	0.27	(3.7)	(3.1)	(0.5)	(0.38)	(0.01)	(0.26)	0.05	1.6

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 10^{12} FOR URANIUM-238 AND BY 10^9 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).

monthly plutonium-239 air concentrations for the past nine years. The arrows at the bottom of the figure 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.

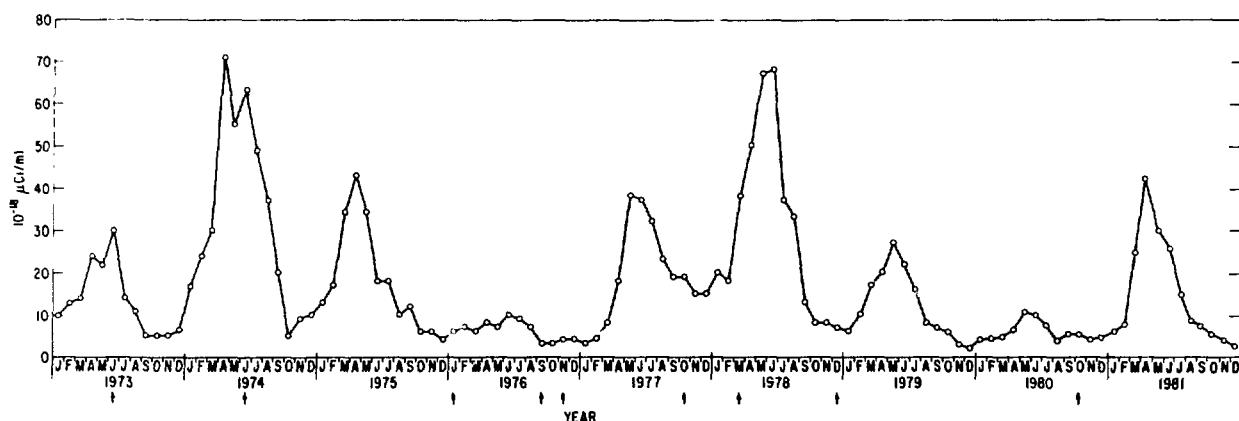


FIG. 4. Plutonium-239,240 in Air Concentrations, 1973-1981

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

The thorium and uranium concentrations are in the same range found during the past several years and are considered to be of natural origin. The percent of CG for the averages is included for completeness; the values are placed in parentheses since the concentrations are considered to be background levels. The amounts of thorium and uranium in a sample were proportional to the mass of material collected on the filter paper, and the bulk of these elements in the air was due to resuspension of soil. For example, the uranium-238 concentrations in March at 7I and off-site differed by a factor of about 2.5 in terms of air volume, but were nearly identical, 1.28×10^{-12} $\mu\text{Ci/g}$ at 7I and $1.34 \mu\text{Ci/g}$ off site, in terms of mass. In contrast, the amount of plutonium in the air samples contributed by soil, if the resuspended soil has the same plutonium concentrations as the first centimeter on the ground, ranged from about 0.5% in July to 13% in December of the total plutonium in the samples.

With the termination of the CP-5 reactor operation (Building 330, location 9H) on September 30, 1979, the principal source of tritiated water vapor and argon-41 in air was eliminated. The only measurable source of argon-41 was from the Janus reactor (Building 202, location 13I). Argon-41 was collected in the exhaust stack of this reactor by filling an evacuated "Marinelli-type" container with air and measuring the argon-41 concentration by gamma-ray spectrometry. The total amount released during 1981 was about 0.4 Ci.

The only other major effluent released was krypton-85 from Building 212 (location 12I), estimated to be about 6.6 Ci during 1981. Several other fission products were also released in millicurie or smaller amounts. Since the CP-5 reactor no longer operates, the concentration of radionuclides released to the air by the Laboratory has been reduced by several orders of magnitude.

2. Surface Water

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

Argonne waste water is discharged into Sawmill Creek, a small stream that runs through the Laboratory grounds, drains surface water from much of the site, and flows into the Des Plaines River about 500 m (0.3 mi) downstream from the waste-water outfall. Sawmill Creek was sampled upstream from the Argonne site and downstream from the waste-water outfall to determine if

radioactivity was added to the stream by Argonne waste water or surface drainage. The sampling locations are shown in Figure 1. Below the waste-water outfall, daily samples were collected by a continuous sampler, which operated about 73% of the year. When the continuous sampling device was not functioning, a grab sample was collected each working day. Equal portions of the daily samples collected each week were combined and analyzed to obtain an average weekly concentration. Above the site, samples were collected once a month and were analyzed for the same radionuclides as the below-outfall samples.

Annual summaries of the results obtained for Sawmill Creek are given in Table 7. Comparison of the results, and 95% confidence limits of the averages, for the two sampling locations shows that the nuclides whose presence in 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 67% for hydrogen-3; 94% for neptunium-237; 94% for plutonium-239, and 80% for americium-241. The concentrations of all these nuclides were low compared to the CGs. The principal radionuclide added to the Creek by Argonne waste water, in terms of concentration, was hydrogen-3. Its average net concentration (equivalent to 0.004% of the CG) was the lowest since such measurements were made. The decrease in the hydrogen-3 is related to the cessation of the operation of the CP-5 reactor. The total concentration, regardless of source, must be used in assessing the hazard of a radionuclide not naturally present, so the percent CG in the table was calculated on this basis.

The hydrogen-3 in the Creek above the site was similar in concentration to levels found away from the Laboratory site and is characteristic of the current ambient levels in surface water. During 1981, the hydrogen-3 content of 22 other lakes and streams ranged from $< 100 \times 10^{-9}$ $\mu\text{Ci}/\text{ml}$ to 280×10^{-9} $\mu\text{Ci}/\text{ml}$ and averaged 130×10^{-9} $\mu\text{Ci}/\text{ml}$.

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 0.9 Ci of hydrogen-3, 1.0 mCi of strontium-90, 0.08 mCi of plutonium-239, 0.07 mCi of neptunium-237, 0.04 mCi of americium-241, and < 0.05 mCi of curium and californium nuclides.

TABLE 7
RADIONUCLIDES IN SAMMILL CREEK, 1981

TYPE OF ACTIVITY	LOCATION #	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/ML)	Avg.	Min.	Max.	Avg.	Percent CG	Min.	Max.
ALPHA (NONVOLATILE)	16K 7H	12 248	1.7 ± 0.1 1.4 ± 0.2	1.0 0.5	3.1 3.3	(0.056) (0.045)	(0.033) (0.017)	(0.103) (0.110)		
BETA (NONVOLATILE)	16K 7H	12 248	22 ± 1 17 ± 1	13 10	28 25	(0.73) (0.57)	(0.45) (0.34)	(0.93) (0.83)		
HYDROGEN-3	16K 7H	12 248	< 126 221 ± 35	< 100 < 100	210 691	< 0.0042 0.0074	< 0.003 < 0.003	0.0070 0.0230		
STRONTIUM-90	16K 7H	12 248	0.24 ± 0.01 0.35 ± 0.05	< 0.25 < 0.25	0.44 0.86	0.080 0.118	< 0.08 < 0.08	0.15 0.29		
IODINE-131	16K 7H	12 248	< 1.1 < 1.7	< 1 < 1	2.0 10.7	< 0.36 0.56	< 0.33 < 0.33	0.67 3.56		
BARIUM-140	16K 7H	12 119	-	-	< 2 < 2	-	-	-	< 0.007 < 0.007	
RADIUM-226	7H	248	0.26 ± 0.05	0.08	0.94	(0.860)	(0.27)	(5.13)		
** URANIUM (NATURAL)	16K 7H	12 253	1.5 ± 0.1 1.5 ± 0.1	1.0 0.8	1.9 2.9	(0.0038) (0.0038)	(0.0025) (0.0020)	(0.0047) (0.0072)		
NEPTUNIUM-237	16K 7H	12 248	0.0076 ± 0.0015	< 0.0010	< 0.001 0.03	0.00025	< 0.00003	< 0.000033 0.0011		
PLUTONIUM-238	16K 7H	12 248	< 0.0020	< 0.001	< 0.001 0.0122	0.00004	< 0.00002	< 0.00002 0.00024		
PLUTONIUM-239	16K 7H	12 248	< 0.00053 0.0085 ± 0.0027	< 0.0005 < 0.0005	0.00079 0.04640	< 0.000011 0.000017	< 0.000010 0.000010	0.000016 0.0009		
AMERICIUM-241	16K 7H	11 243	0.0037 ± 0.0010	< 0.001	< 0.001 0.017	0.00009	< 0.000025	< 0.000025 0.0004		
CURIUM-242 AND/OR CALIFORNIUM-252	16K 7H	11 243	< 0.0011	< 0.001	< 0.001 0.0029	< 0.00006	< 0.000005	< 0.000005 0.000014		
CURIUM-244 AND/OR CALIFORNIUM-249	16K 7H	11 243	0.0016 ± 0.0005	< 0.001	< 0.001 0.0090	0.000023	< 0.000014	< 0.000014 0.00013		

* LOCATION 16K IS UPSTREAM FROM THE ARGONNE SITE AND LOCATION 7H 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 .
THE AVERAGE CONCENTRATION IN THE CREEK THEN BECOMES 2.2 MICROGRAMS/L.

Since Sawmill Creek empties into the Des Plaines River, which in turn flows into the Illinois River, the radioactivity in the latter two streams is important in assessing the contribution of Argonne waste water to the environmental radioactivity. The Des Plaines River was sampled twice a month below, and monthly above, the mouth of Sawmill Creek to determine if the radioactivity in the Creek had any effect on the activity in the River. Annual summaries of the results obtained for these two locations are given in Table 8. The average nonvolatile alpha, beta, and uranium concentrations in the River were very similar to past averages and remained in the normal range. Results were quite similar above and below the Creek for all radionuclides since the activity in Sawmill Creek was reduced by dilution so that it was not detectable as such in the Des Plaines River. The average nonvolatile alpha and beta activities, $1.2 \times 10^{-9} \mu\text{Ci}/\text{ml}$ and $9 \times 10^{-9} \mu\text{Ci}/\text{ml}$, respectively, of 22 off-site surface water samples collected this year (excluding the Des Plaines River) were similar to the levels found in previous years.

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

3. Soil, Grass, and Bottom Sediment

The radioactive content of soil, grass, and bottom sediment was measured at the site perimeter and off the site. The purpose of the off-site sampling was to measure deposition from weapons testing for comparison with perimeter samples, and with results obtained by other organizations for samples collected at large distances from nuclear installations. Such comparisons are useful in determining if the soil activity near Argonne is normal. For this purpose, the site selection criteria and sample preparation techniques of the DOE Environmental Measurements Laboratory⁹ were used. Sites were selected in several directions and at various distances from the Laboratory. Each site was selected on the basis that the soil appeared, or was known to have been, undisturbed for a number of years. Attempts were made to select open, level, grassy areas that were mowed at reasonable intervals. Public parks were selected when available.

Each soil sample consisted of ten cores totaling 864 cm^2 in area by

TABLE 8
RADIONUCLIDES IN DESPLAINES RIVER WATER, 1981

TYPE OF ACTIVITY	LOCATION *	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/ML)			AVG.	PERCENT CG	
			AVG.	MIN.	MAX.		MIN.	MAX.
ALPHA (NONVOLATILE)	A	12	1.7 ± 0.1	0.9	2.6	(0.055)	(0.030)	(0.080)
	B	24	1.6 ± 0.2	0.6	2.9	(0.052)	(0.020)	(0.097)
BETA (NONVOLATILE)	A	12	16 ± 1	10	35	(0.52)	(0.32)	(1.15)
	B	24	14 ± 2	8	24	(0.48)	(0.27)	(0.79)
HYDROGEN-3	A	12	115 ± 4	< 100	225	0.0038	< 0.003	0.0075
	B	24	115 ± 23	< 100	214	0.0038	< 0.003	0.0071
STRONTIUM-90	A	12	0.55 ± 0.02	0.34	1.59	0.18	0.11	0.53
	B	24	0.47 ± 0.09	< 0.25	1.13	0.16	< 0.08	0.38
IODINE-131	A	12	< 1.0	< 1	1.5	< 0.35	< 0.33	0.50
	B	24	< 1.3	< 1	4.0	< 0.43	< 0.33	1.33
BARIUM-140	A	12	-	-	< 2	-	-	< 0.007
	B	12	-	-	< 2	-	-	< 0.007
** URANIUM (NATURAL)	A	12	1.4 ± 0.1	0.8	2.4	(0.0036)	(0.0020)	(0.0060)
	B	24	1.4 ± 0.2	0.5	2.3	(0.0035)	(0.0012)	(0.0057)
NEPTUNIUM-237	A	12	-	-	< 0.001	-	-	< 0.000033
	B	12	-	-	< 0.001	-	-	< 0.000033
PLUTONIUM-238	A	12	-	-	< 0.001	-	-	< 0.00002
	B	12	-	-	< 0.001	-	-	< 0.00002
PLUTONIUM-239	A	12	< 0.00056	< 0.0005	0.0008	< 0.000011	< 0.000010	0.000016
	B	12	< 0.00064	< 0.0005	0.0012	< 0.000013	< 0.000010	0.000024
AMERICIUM-241	A	11	-	-	< 0.001	-	-	< 0.000025
	B	11	-	-	< 0.001	-	-	< 0.000025
CURIUM-242 AND/OR CALIFORNIUM-252	A	11	-	-	< 0.001	-	-	< 0.000005
	B	11	-	-	< 0.001	-	-	< 0.000005
CURIUM-244 AND/OR CALIFORNIUM-249	A	11	-	-	< 0.001	-	-	< 0.000014
	B	11	-	-	< 0.001	-	-	< 0.000014

* LOCATION A, NEAR ROUTE 45, 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 .
THE AVERAGE CONCENTRATION IS 2.1 MICROGRAMS/L.

TABLE 9

Radionuclides in Illinois River Water, 1981
 (Concentrations in 10^{-9} $\mu\text{Ci}/\text{ml}$)

Date Collected	Location	Alpha *	Beta *	Hydrogen-3	Uranium ** (natural)	Plutonium-239
June 19	McKinley Woods State Park	0.6	7.2	< 100	0.9	0.00047
June 19	Below Dresden Power Station	0.5	4.7	< 100	0.6	0.00029
June 19	Morris	1.6	5.8	186	0.9	-
June 19	Starved Rock State Park	0.5	6.0	< 100	0.9	-
October 13	McKinley Woods State Park	0.5	9.9	219	0.6	0.00023
October 13	Below Dresden Power Station	1.4	5.3	281	0.8	0.00024
October 13	Morris	1.3	5.9	171	0.9	-
October 13	Starved Rock State Park	1.0	6.3	110	0.9	-

* Nonvolatile activity.

** Uranium concentrations in units of $\mu\text{g}/\text{l}$ can be obtained by multiplying the concentration by 1.48×10^3 .

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 should be more sensitive to changes in current deposition. The grass samples were obtained by collecting the grass from a 1 m² area in the immediate vicinity of a soil sample. A grab sample technique was used to obtain bottom sediment. After drying, grinding, and mixing, 100 g portions of soil, bottom sediment, and grass were analyzed by the same method described in Section III.A. for air-filter residue. Results are given in terms of the oven-dried sample weight.

Soil samples were analyzed by gamma-ray spectrometry and radiochemically for plutonium. The results for the gamma-ray nuclides in soil is presented in Table 10. These radionuclides are present primarily as a result of the October 16, 1980, Chinese test and the decrease in concentration due to decay of the intermediate half-life fission products is obvious when comparing results of samples collected in the spring and fall. The annual average concentrations for the perimeter and off-site samples are similar. The plutonium concentrations are given in Table 11. The range and concentration of plutonium in soil is similar at both perimeter and off-site locations and consistent with results obtained in previous years.

Composite monthly precipitation samples were analyzed for plutonium-239. The results are given in Table 12, along with results since 1973 for comparison. The 1981 deposition by precipitation was equivalent to 0.4% of the total plutonium deposited through 1976, which is $2.2 \times 10^{-3} \mu\text{Ci}/\text{m}^2$.¹⁰ Deposition since 1976 has not changed this value significantly. The data in Table 12 is illustrated in Figure 5. The arrows at the bottom of the figure

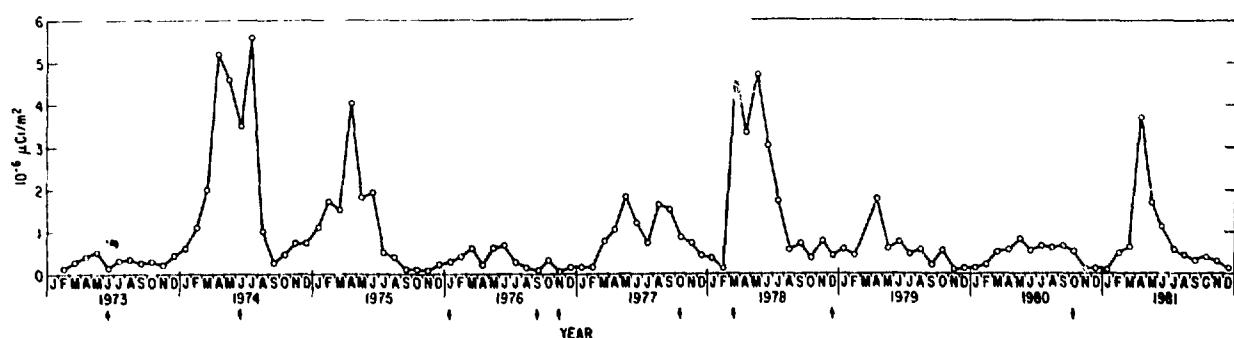


FIG. 5. Plutonium-239, 240 Surface Deposition, 1973-1981

TABLE 10

 Gamma-Ray Emitting Radionuclides in Soil, 1981
 (Concentrations in 10^{-6} $\mu\text{Ci/g}$)

Date Collected	Location	^{95}Zr	^{95}Nb	^{103}Ru	^{106}Ru	^{125}Sb	^{137}Cs	^{141}Ce	^{144}Ce
<u>Perimeter*</u>									
April 29	12-0	0.08 ± 0.05	0.31 ± 0.06	< 0.05	< 0.1	< 0.05	1.31 ± 0.04	0.09 ± 0.05	0.09 ± 0.07
June 24	8N	0.05 ± 0.02	0.16 ± 0.02	< 0.05	0.17 ± 0.09	0.05 ± 0.03	0.64 ± 0.03	< 0.05	0.16 ± 0.07
June 24	14L	0.05 ± 0.02	0.27 ± 0.03	0.05 ± 0.02	0.14 ± 0.11	< 0.05	1.04 ± 0.03	0.06 ± 0.02	0.16 ± 0.02
June 24	12C	0.05 ± 0.02	0.23 ± 0.03	< 0.05	0.17 ± 0.11	0.07 ± 0.03	0.98 ± 0.04	0.06 ± 0.02	0.09 ± 0.06
June 24	8G	0.08 ± 0.02	0.14 ± 0.02	< 0.05	< 0.1	< 0.05	1.23 ± 0.04	< 0.05	0.15 ± 0.06
October 20	14E	< 0.05	< 0.05	< 0.05	0.12 ± 0.10	< 0.05	1.14 ± 0.04	0.10 ± 0.05	0.14 ± 0.07
October 20	14N	< 0.05	< 0.05	< 0.05	0.24 ± 0.11	0.07 ± 0.04	1.11 ± 0.04	< 0.05	0.09 ± 0.06
October 20	10P	0.06 ± 0.02	0.10 ± 0.05	< 0.05	< 0.1	< 0.05	1.50 ± 0.04	< 0.05	0.25 ± 0.06
October 20	4EF	< 0.05	< 0.05	< 0.05	< 0.1	0.06 ± 0.04	1.43 ± 0.04	0.06 ± 0.04	0.13 ± 0.07
October 20	7EF	0.07 ± 0.05	0.12 ± 0.04	< 0.05	< 0.1	0.07 ± 0.04	1.00 ± 0.03	0.12 ± 0.05	0.16 ± 0.06
	Average	0.05 ± 0.01	0.14 ± 0.07	< 0.05	0.11 ± 0.04	< 0.06	1.14 ± 0.16	0.06 ± 0.02	0.14 ± 0.03
<u>Off-Site</u>									
June 11	Woodridge, IL	0.11 ± 0.03	0.26 ± 0.03	< 0.05	< 0.1	< 0.05	1.01 ± 0.04	0.06 ± 0.02	0.19 ± 0.07
June 11	Naperville, IL	0.10 ± 0.03	0.26 ± 0.03	< 0.05	< 0.1	< 0.05	1.02 ± 0.03	< 0.05	0.12 ± 0.07
June 19	Channahon, IL	0.09 ± 0.02	0.26 ± 0.03	0.06 ± 0.02	0.22 ± 0.10	< 0.05	0.56 ± 0.03	0.06 ± 0.02	0.23 ± 0.07
June 19	Starved Rock State Park, IL	0.07 ± 0.02	0.17 ± 0.03	< 0.05	0.20 ± 0.10	< 0.05	0.97 ± 0.03	< 0.05	0.16 ± 0.06
June 19	Morris, IL	0.26 ± 0.03	1.16 ± 0.05	0.12 ± 0.02	0.34 ± 0.11	< 0.05	0.42 ± 0.02	0.11 ± 0.02	0.49 ± 0.07
October 8	Lemont, IL	0.05 ± 0.03	0.09 ± 0.05	< 0.05	0.22 ± 0.11	< 0.05	0.69 ± 0.03	< 0.05	< 0.05
October 8	Romeoville, IL	0.06 ± 0.02	0.10 ± 0.03	< 0.05	0.26 ± 0.13	0.10 ± 0.04	1.44 ± 0.04	< 0.05	0.38 ± 0.08
October 8	Downers Grove, IL	< 0.05	< 0.05	< 0.05	0.14 ± 0.11	< 0.05	0.88 ± 0.03	< 0.05	0.15 ± 0.07
October 13	McKinley Woods State Park, IL	< 0.05	0.15 ± 0.04	< 0.05	< 0.1	< 0.05	2.18 ± 0.05	< 0.05	0.20 ± 0.07
October 13	Dresden Lock and Dam, IL	< 0.05	0.10 ± 0.04	< 0.05	< 0.1	< 0.05	1.38 ± 0.04	< 0.05	0.13 ± 0.07
	Average	0.08 ± 0.04	0.26 ± 0.21	< 0.06	0.16 ± 0.07	< 0.06	1.06 ± 0.32	< 0.06	0.21 ± 0.09

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

TABLE 11

Plutonium in Soil, 1981

Date Collected	Location	Plutonium-238		Plutonium-239		
		10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	$^{238}\text{Pu}/^{239}\text{Pu}$
<u>Perimeter*</u>						
April 29	12-0	1.4 \pm 0.3	0.063 \pm 0.012	22.4 \pm 1.3	0.97 \pm 0.06	0.065
June 24	8N	1.7 \pm 0.3	0.099 \pm 0.017	11.2 \pm 0.8	0.66 \pm 0.05	0.150
June 24	14L	1.7 \pm 0.3	0.103 \pm 0.017	18.7 \pm 1.2	1.11 \pm 0.07	0.093
June 24	12C	1.8 \pm 0.3	0.090 \pm 0.016	16.3 \pm 1.1	0.84 \pm 0.06	0.108
June 24	8G	1.9 \pm 0.3	0.096 \pm 0.016	21.8 \pm 1.3	1.12 \pm 0.07	0.086
October 20	14E	2.5 \pm 0.4	0.131 \pm 0.020	24.9 \pm 1.2	1.29 \pm 0.06	0.101
October 20	14N	1.9 \pm 0.3	0.112 \pm 0.017	20.4 \pm 0.9	1.22 \pm 0.06	0.092
October 20	10P	2.6 \pm 0.4	0.131 \pm 0.018	30.1 \pm 1.2	1.51 \pm 0.06	0.087
October 20	4EF	3.3 \pm 0.4	0.148 \pm 0.019	23.9 \pm 1.1	1.08 \pm 0.05	0.138
October 20	7EF	-	-	-	-	-
	Average	2.1 \pm 0.4	0.108 \pm 0.017	21.1 \pm 3.6	1.09 \pm 0.17	0.102
<u>Off-Site</u>						
June 11	Woodridge, IL	2.1 \pm 0.3	0.095 \pm 0.015	18.0 \pm 1.1	0.82 \pm 0.05	0.117
June 11	Naperville, IL	2.3 \pm 0.4	0.081 \pm 0.013	18.1 \pm 1.1	0.65 \pm 0.04	0.125
June 19	Channahon, IL	0.3 \pm 0.4	0.016 \pm 0.026	10.7 \pm 0.9	0.68 \pm 0.06	0.024
June 19	Starved Rock State Park, IL	1.9 \pm 0.3	0.127 \pm 0.020	17.8 \pm 1.1	1.18 \pm 0.07	0.108
June 19	Morris, IL	1.6 \pm 0.3	0.078 \pm 0.013	9.7 \pm 0.7	0.46 \pm 0.04	0.169
October 8	Lemont, IL	1.3 \pm 0.3	0.064 \pm 0.014	12.6 \pm 0.8	0.64 \pm 0.04	0.101
October 8	Romeoville, IL	3.0 \pm 0.4	0.112 \pm 0.016	32.7 \pm 1.9	1.20 \pm 0.07	0.093
October 8	Downers Grove, IL	2.2 \pm 0.3	0.109 \pm 0.016	15.9 \pm 1.0	0.79 \pm 0.05	0.138
October 13	McKinley Woods State Park, IL	1.9 \pm 0.4	0.085 \pm 0.017	41.1 \pm 1.7	1.87 \pm 0.08	0.045
October 13	Dresden Lock and Dam, IL	4.2 \pm 0.6	0.195 \pm 0.026	24.3 \pm 1.3	1.13 \pm 0.06	0.172
	Average	2.1 \pm 0.7	0.096 \pm 0.029	20.1 \pm 6.3	0.94 \pm 0.26	0.109

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

TABLE 12

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

Month	1973	1974	1975	1976	1977	1978	1979	1980	1981
January	-	0.60 \pm 0.10	1.1 \pm 0.18	0.25 \pm 0.06	0.17 \pm 0.08	0.37 \pm 0.11	0.57 \pm 0.20	0.15 \pm 0.09	0.06 \pm 0.09
February	0.14 \pm 0.04	1.1 \pm 0.16	1.7 \pm 0.19	0.38 \pm 0.09	0.15 \pm 0.07	0.12 \pm 0.09	0.43 \pm 0.14	0.21 \pm 0.07	0.43 \pm 0.17
March	0.27 \pm 0.06	2.0 \pm 0.22	1.5 \pm 0.17	0.58 \pm 0.09	0.74 \pm 0.15	4.7 \pm 0.37	-	0.50 \pm 0.12	0.48 \pm 0.20
April	0.40 \pm 0.07	5.2 \pm 0.37	4.0 \pm 0.43	0.21 \pm 0.07	1.0 \pm 0.17	3.3 \pm 0.30	1.78 \pm 0.25	0.56 \pm 0.13	3.70 \pm 0.34
May	0.50 \pm 0.06	4.6 \pm 0.31	1.8 \pm 0.18	0.58 \pm 0.11	1.8 \pm 0.21	4.7 \pm 0.42	0.58 \pm 0.14	0.81 \pm 0.14	1.43 \pm 0.21
June	0.16 \pm 0.04	3.5 \pm 0.26	1.9 \pm 0.18	0.65 \pm 0.12	1.2 \pm 0.18	3.0 \pm 0.32	0.73 \pm 0.17	0.54 \pm 0.15	1.11 \pm 0.22
July	0.32 \pm 0.10	5.6 \pm 0.37	0.48 \pm 0.10	0.26 \pm 0.07	0.71 \pm 0.15	1.7 \pm 0.24	0.45 \pm 0.15	0.67 \pm 0.18	0.51 \pm 0.20
August	0.34 \pm 0.14	1.0 \pm 0.19	0.38 \pm 0.09	0.15 \pm 0.06	1.6 \pm 0.21	0.54 \pm 0.17	0.54 \pm 0.14	0.62 \pm 0.15	0.40 \pm 0.21
September	0.27 \pm 0.17	0.25 \pm 0.11	0.10 \pm 0.06	0.06 \pm 0.05	1.5 \pm 0.20	0.69 \pm 0.18	0.21 \pm 0.12	0.65 \pm 0.14	0.29 \pm 0.15
October	0.30 \pm 0.12	0.45 \pm 0.10	0.12 \pm 0.08	0.30 \pm 0.10	0.85 \pm 0.15	0.34 \pm 0.14	0.53 \pm 0.22	0.53 \pm 0.09	0.36 \pm 0.11
November	0.22 \pm 0.10	0.73 \pm 0.12	0.08 \pm 0.07	0.05 \pm 0.04	0.72 \pm 0.14	0.74 \pm 0.16	0.09 \pm 0.11	0.13 \pm 0.07	0.24 \pm 0.15
December	0.46 \pm 0.12	0.71 \pm 0.12	0.21 \pm 0.08	0.12 \pm 0.07	0.43 \pm 0.10	0.40 \pm 0.14	0.13 \pm 0.10	0.12 \pm 0.12	0.07 \pm 0.10
Average Monthly Deposition	0.31 \pm 0.07	2.1 \pm 1.2	1.1 \pm 0.7	0.30 \pm 0.13	0.91 \pm 0.31	1.7 \pm 1.0	0.55 \pm 0.27	0.46 \pm 0.14	0.76 \pm 0.26
Annual Deposition	3.38	25.74	13.37	3.59	10.87	20.60	6.04	5.49	9.08
Percent Added to Existing	0.2	1.2	0.6	0.2	0.5	0.9	0.3	0.3	0.4

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 nine years in Figure 4 shows excellent correlation.

The results of gamma-ray emitting nuclides in grass are given in Table 13. The annual averages were similar between perimeter and off-site locations, while the same spring-fall relationship observed in soil samples also holds for the plant samples. Other radionuclides measured in grass samples are collected in Table 14. 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. Concentration ranges were the same at the perimeter and off-site, and similar to previous years, indicating no contribution from ANL operations.

Results of analysis of bottom sediment samples for gamma-ray emitters and plutonium are given in Table 15. The annual average and seasonal concentrations of the fission products are similar to those observed in soil samples. Plutonium results vary widely between locations and are strongly dependent on the retentiveness of the bottom material. However, the plutonium concentrations are in the same range as off-site samples collected in previous years.

4. Milk

Raw milk was collected monthly from a local dairy farm south of Lemont and analyzed for several radioactive nuclides. Iodine-131 was analyzed with a detection limit of $1 \times 10^{-10} \mu\text{Ci}/\text{ml}$ by an ion-exchange separation followed by beta counting. Cesium-137 was analyzed with a detection limit of $5 \times 10^{-10} \mu\text{Ci}/\text{ml}$ 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 16. The average strontium-90 and cesium-137 concentrations increased by about 50% compared to 1980. These nuclides are fission products from nuclear tests and their presence in milk is not related to Argonne operations. No short-lived fission products, i.e., strontium-89, iodine-131, or barium-140 were detected in milk.

The concentrations given in Table 16 may be compared to the CGs for drinking water given in the Appendix, Part B. The drinking water CGs are based on an intake of 2.2 liters per day. The consumption of one liter of milk per day would result in an average intake of 0.7% of the strontium-90

TABLE 13

 Gamma-Ray Emitting Radionuclides in Grass Samples, 1981
 (Concentrations in 10^{-6} $\mu\text{Ci/g}$)

Date Collected	Location	^{95}Zr	^{95}Nb	^{103}Ru	^{106}Ru	^{137}Cs	^{141}Ce	^{144}Ce
<u>Perimeter*</u>								
June 24	8N	0.33 ± 0.03	0.78 ± 0.03	< 0.05	0.22 ± 0.09	< 0.05	0.09 ± 0.02	0.62 ± 0.05
June 24	14L	0.14 ± 0.04	0.35 ± 0.04	< 0.05	< 0.1	0.05 ± 0.02	< 0.05	0.27 ± 0.09
June 24	8G	0.20 ± 0.04	0.47 ± 0.04	< 0.05	< 0.1	< 0.05	< 0.05	0.31 ± 0.08
June 24	12C	0.20 ± 0.05	0.47 ± 0.04	< 0.05	< 0.1	< 0.05	0.08 ± 0.03	0.51 ± 0.09
October 20	14E	< 0.05	< 0.05	< 0.05	< 0.1	< 0.05	< 0.05	0.15 ± 0.11
October 20	14N	< 0.05	< 0.05	< 0.05	< 0.1	< 0.05	< 0.05	0.45 ± 0.13
October 20	10P	< 0.05	< 0.05	< 0.05	< 0.1	< 0.05	< 0.05	< 0.05
October 20	4EF	< 0.05	< 0.05	< 0.05	< 0.1	< 0.05	< 0.05	0.30 ± 0.12
October 20	7EF	< 0.05	< 0.05	< 0.05	< 0.1	< 0.05	< 0.05	0.24 ± 0.12
	Average	< 0.12	< 0.26	< 0.05	< 0.1	< 0.05	< 0.06	0.26 ± 0.11
<u>Off-Site</u>								
June 11	Woodridge, IL	0.28 ± 0.05	0.44 ± 0.04	0.09 ± 0.02	0.31 ± 0.17	< 0.05	0.06 ± 0.02	0.45 ± 0.09
June 11	Naperville, IL	0.20 ± 0.04	0.55 ± 0.04	0.05 ± 0.02	0.18 ± 0.14	0.06 ± 0.02	0.05 ± 0.02	0.45 ± 0.08
June 19	Channahon, IL	0.10 ± 0.05	0.34 ± 0.05	< 0.05	< 0.1	< 0.05	< 0.05	0.28 ± 0.09
June 19	Starved Rock State Park, IL	0.29 ± 0.05	0.62 ± 0.04	< 0.05	< 0.1	0.07 ± 0.03	0.06 ± 0.02	0.76 ± 0.09
June 19	Morris, IL	0.11 ± 0.03	0.20 ± 0.03	< 0.05	< 0.1	0.05 ± 0.02	< 0.05	0.07 ± 0.07
October 8	Lemont, IL	< 0.05	< 0.05	< 0.05	< 0.1	< 0.05	< 0.05	0.24 ± 0.12
October 8	Romeoville, IL	< 0.05	0.09 ± 0.04	< 0.05	< 0.1	< 0.05	< 0.05	0.19 ± 0.12
October 8	Downers Grove, IL	< 0.05	< 0.05	< 0.05	< 0.1	< 0.05	< 0.05	0.31 ± 0.13
October 13	McKinley Woods State Park, IL	< 0.05	0.06 ± 0.04	< 0.05	< 0.1	< 0.05	< 0.05	< 0.05
October 13	Dresden Lock and Dam, IL	< 0.05	0.33 ± 0.06	< 0.05	< 0.1	0.10 ± 0.04	< 0.05	0.91 ± 0.16
	Average	< 0.11	0.27 ± 0.14	< 0.05	< 0.1	< 0.06	< 0.05	0.36 ± 0.18

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

TABLE 14

Radionuclides in Grass Samples, 1981

Date Collected	Location	Concentrations in 10^{-9} $\mu\text{Ci/g}$					10^{-6} $\mu\text{Ci/m}^2$
		Thorium-228	Thorium-230	Thorium-232	Uranium (natural)	Plutonium-238	
<u>Perimeter*</u>							
June 24	8N	0.3 \pm 0.9	2.3 \pm 0.4	1.7 \pm 0.4	35 \pm 3	< 0.1	0.94 \pm 0.16
June 24	14L	0.3 \pm 0.4	0.9 \pm 0.2	0.5 \pm 0.2	30 \pm 3	< 0.1	1.14 \pm 0.19
June 24	8G	0.7 \pm 0.4	1.8 \pm 0.3	1.2 \pm 0.2	27 \pm 3	< 0.1	0.38 \pm 0.11
June 24	12C	1.1 \pm 0.4	2.3 \pm 0.3	1.4 \pm 0.2	40 \pm 3	< 0.1	0.59 \pm 0.13
October 20	14E	6.5 \pm 1.4	9.0 \pm 2.6	5.4 \pm 2.2	89 \pm 7	< 0.1	0.58 \pm 0.12
October 20	14N	2.0 \pm 0.2	2.9 \pm 0.4	1.9 \pm 0.2	80 \pm 5	< 0.1	0.76 \pm 0.14
October 20	10P	1.8 \pm 0.2	2.7 \pm 0.2	1.5 \pm 0.1	64 \pm 5	< 0.1	0.63 \pm 0.18
October 20	4EF	1.0 \pm 0.1	1.2 \pm 0.1	0.7 \pm 0.1	47 \pm 4	< 0.1	0.31 \pm 0.10
October 20	7EF	4.5 \pm 0.6	6.7 \pm 0.7	3.6 \pm 0.5	28 \pm 2	< 0.1	0.34 \pm 0.10
	Average	2.0 \pm 1.4	3.3 \pm 1.8	2.0 \pm 1.0	49 \pm 16	< 0.1	0.63 \pm 0.19
							0.143 \pm 0.077
<u>Off-Site</u>							
June 11	Woodridge, IL	0.7 \pm 0.7	1.9 \pm 0.4	1.0 \pm 0.3	34 \pm 2	< 0.1	0.71 \pm 0.14
June 11	Naperville, IL	0.8 \pm 0.9	1.9 \pm 0.4	0.8 \pm 0.3	36 \pm 4	< 0.1	0.50 \pm 0.14
June 19	Channahon, IL	4.4 \pm 0.3	5.5 \pm 0.3	4.3 \pm 0.3	62 \pm 17	< 0.1	0.78 \pm 0.19
June 19	Starved Rock State Park, IL	0.7 \pm 0.7	3.0 \pm 0.5	2.2 \pm 0.4	35 \pm 3	< 0.1	0.97 \pm 0.23
June 19	Morris, IL	2.6 \pm 0.4	6.0 \pm 0.4	3.7 \pm 0.3	58 \pm 3	< 0.1	0.89 \pm 0.17
October 8	Lemont, IL	5.0 \pm 0.6	7.6 \pm 0.7	5.2 \pm 0.5	81 \pm 6	< 0.1	1.84 \pm 0.17
October 8	Romeoville, IL	5.0 \pm 0.5	5.8 \pm 0.5	3.9 \pm 0.4	27 \pm 2	< 0.1	0.62 \pm 0.32
October 8	Downers Grove, IL	3.0 \pm 0.7	5.8 \pm 0.8	3.5 \pm 0.6	60 \pm 5	< 0.1	0.73 \pm 0.13
October 13	McKinley Woods State Park, IL	0.8 \pm 0.1	1.1 \pm 0.1	0.7 \pm 0.1	41 \pm 6	< 0.1	0.47 \pm 0.12
October 13	Dresden Lock and Dam, IL	3.8 \pm 0.2	5.1 \pm 0.2	4.1 \pm 0.2	40 \pm 3	< 0.1	0.33 \pm 0.03
	Average	2.7 \pm 1.6	4.4 \pm 1.4	2.9 \pm 1.0	47 \pm 11	< 0.1	0.78 \pm 0.27
							0.104 \pm 0.030

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

TABLE 15

Radionuclides in Bottom Sediment, 1981

Date Collected	Off-Site Location	Concentrations in $10^{-6} \mu\text{Ci/g}$							Concentrations in $10^{-9} \mu\text{Ci/g}$		
		^{95}Zr	^{95}Nb	^{103}Ru	^{106}Ru	^{125}Sb	^{137}Cs	^{141}Ce	^{144}Ce	^{238}Pu	
June 11	DuPage River, Naperville, IL	0.48 ± 0.04	1.15 ± 0.04	0.15 ± 0.02	0.16 ± 0.10	< 0.05	0.23 ± 0.02	0.07 ± 0.02	0.52 ± 0.07	0.1 ± 0.2	4.9 ± 0.7
June 11	DuPage River, West Chicago, IL	0.36 ± 0.04	0.94 ± 0.04	0.08 ± 0.02	0.15 ± 0.10	0.05 ± 0.03	0.19 ± 0.02	0.07 ± 0.02	0.55 ± 0.07	0.2 ± 0.2	4.0 ± 0.5
June 19	Illinois River, Starved Rock State Park, IL	0.30 ± 0.04	0.72 ± 0.04	0.10 ± 0.02	0.37 ± 0.11	< 0.05	0.29 ± 0.02	< 0.05	0.33 ± 0.07	1.8 ± 0.3	6.5 ± 0.6
June 19	Illinois River, Morris, IL	0.11 ± 0.03	0.15 ± 0.02	0.06 ± 0.02	0.16 ± 0.08	0.06 ± 0.03	0.09 ± 0.02	0.05 ± 0.02	0.21 ± 0.06	0.4 ± 0.1	4.0 ± 0.4
October 8	Long Run Creek, Lemont, IL	< 0.05	< 0.05	< 0.05	0.16 ± 0.09	< 0.05	0.22 ± 0.02	< 0.05	< 0.05	0.1 ± 0.3	5.2 ± 0.6
October 8	Des Plaines River, Romeoville, IL	0.07 ± 0.02	0.08 ± 0.02	< 0.05	< 0.1	< 0.05	0.12 ± 0.02	< 0.05	0.20 ± 0.05	0.2 ± 0.6	3.3 ± 0.8
October 13	DuPage River, Channahon, IL	0.16 ± 0.03	0.23 ± 0.03	< 0.05	< 0.1	< 0.05	0.22 ± 0.02	< 0.05	0.23 ± 0.07	0.4 ± 0.4	5.4 ± 0.5
October 13	Illinois River, McKinley Woods State Park, IL	< 0.05	< 0.05	< 0.05	< 0.1	< 0.05	0.07 ± 0.01	< 0.05	< 0.05	0.4 ± 0.1	3.4 ± 0.4
October 13	Illinois River, Dresden Lock & Dam, IL	< 0.05	< 0.05	< 0.05	< 0.1	< 0.05	0.08 ± 0.01	< 0.05	< 0.05	0.6 ± 0.2	2.5 ± 0.3
Average		0.17 ± 0.11	0.37 ± 0.29	< 0.07	0.13 ± 0.07	< 0.05	0.17 ± 0.05	< 0.05	0.24 ± 0.13	0.5 ± 0.4	4.4 ± 0.8

and 0.004% of the cesium-137 Concentration Guides.

TABLE 16

Radionuclides in Milk, 1981
(Concentrations in 10^{-9} $\mu\text{Ci}/\text{ml}$)

Date Collected	Hydrogen-3	Strontium-90	Cesium-137
January 7	< 100	2.8	1.2
February 4	< 100	3.1	1.0
March 4	< 100	2.2	0.8
April 1	< 100	2.5	0.9
May 6	< 100	1.8	1.2
June 3	105	9.6	5.5
July 1	107	8.5	2.2
August 5	< 100	8.9	1.9
September 2	< 100	5.3	1.4
October 7	223	5.3	1.6
November 4	147	3.7	1.5
December 2	< 100	2.4	1.4
Average	< 115	4.7 \pm 1.6	1.7 \pm 0.7

5. External Penetrating Radiation

Measurements were made with calcium fluoride (dysprosium activated) and lithium fluoride thermoluminescent dosimeter (TLD) chips. Each measurement was the average of four chips exposed in the same packet. The response of the chips was calibrated with a National Bureau of Standards standard radium-226 source, and the results calculated in terms of air dose. Dosimeters were exposed at a number of locations at the site boundary to determine any dose that might be due to Argonne operations at the closest uncontaminated approaches to the Laboratory, and at several locations on the site. The latter were chosen for two purposes: to determine where abnormal doses might be encountered, and where the results might be useful in determining the origin of any abnormal dose readings obtained at the boundary. Readings were also taken at five off-site locations for comparison purposes. These

locations are shown in Figure 2.

The results are summarized in Tables 17 and 18, and the site boundary and on-site readings are also shown in Figure 6. Measurements were made in four successive exposure periods that varied in length from 82 to 98 days, and in total covered the period from January 12, 1981, to January 12, 1982. The results for each period were calculated in terms of annual dose for ease in comparing measurements made for different elapsed times, and were weighted according to their exposure times in calculating the annual average at each location. The uncertainty given in the tables for an average is the 95% confidence limit calculated from the standard error.

TABLE 17

Environmental Penetrating Radiation at Off-Site Locations, 1981

Location	Dose Rate (mrem/year)				Average
	1/12-4/16	4/16-7/7	7/7-10/6	10/6-1/12	
Downers Grove	89	88	96	88	90 \pm 6
Lemont	91	95	98	96	95 \pm 4
Lombard	101	96	97	95	97 \pm 4
Oakbrook	92	88	99	101	95 \pm 8
Oak Lawn	80	77	88	81	82 \pm 7
Average	91 \pm 9	89 \pm 9	96 \pm 5	92 \pm 9	92 \pm 7

The off-site results averaged 92 ± 7 mrem/yr. From 1973 to 1980, the corresponding averages and standard errors varied from 89 ± 7 mrem/yr (1979) to 100 ± 2 mrem/yr (1973). The eight-year average was 93 mrem/yr, with a standard deviation of 4 mrem/yr. Thus, the background radiation has been quite consistent and the 1981 average is within the range found previously. The variations from year-to-year have also been consistent at each location. The annual averages at Downers Grove, Lemont, and Lombard have been within 5 mrem/yr of the average of all five off-site locations (usually within 2 mrem/yr). At Oak Lawn, the results have been from 6 to 11 mrem/yr below and at Oakbrook (except in 1981), 6 to 11 mrem/yr above, the overall average.

The reasons for the unusually low readings in Oakbrook during the first half of 1981 are being investigated. The differences between sampling periods at the same location have also been relatively small, except in 1979, when heavy snow cover resulted in a lower dose during the winter.

If the off-site locations are an accurate sample of the radiation background in the area, then annual averages at the site boundary in the range of 92 ± 7 mrem/yr may be considered normal with a 95% probability, or 95% of the boundary results should have averages in this range. To compare boundary results for individual sampling periods, the standard deviation of the mean of the 20 individual off-site results is useful. This value is 6.9 mrem/yr, so that individual results in the range of 92 ± 14 mrem/yr may be considered normal with a 95% probability, unless there are obvious, known reasons to the contrary.

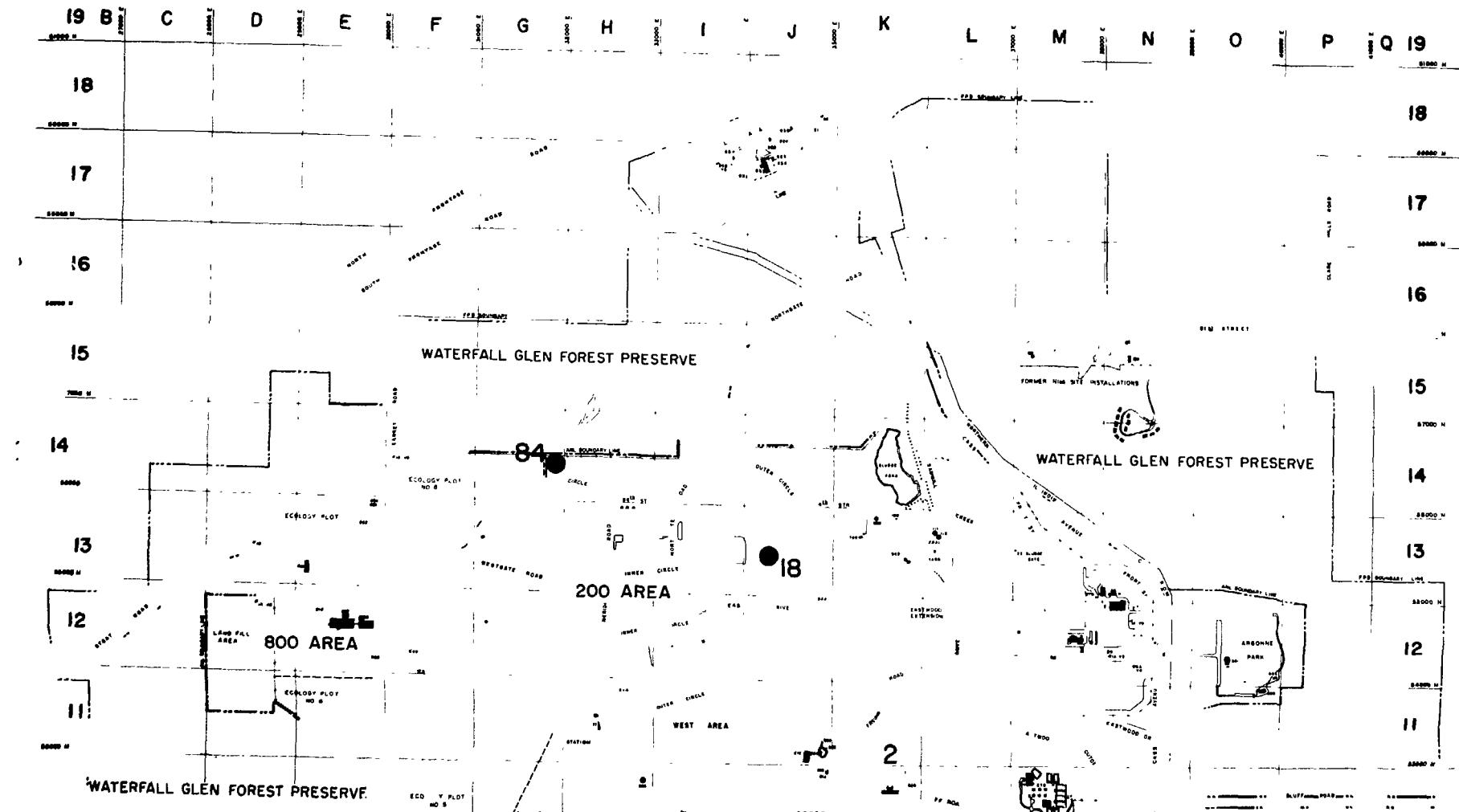
At two site boundary locations, 7I (south) and 14I (north), the dose rates were consistently above the normal range. At 7I this was due to radiation from a Radioactive Waste Storage Facility in the northern half of grid 7I. Waste is packaged and temporarily kept in this area prior to removal for permanent storage elsewhere. The net above-normal dose at this location was about 490 mrem/yr; in previous years, this value has ranged from 560 mrem/yr in 1980 to 114 mrem/yr in 1977. About 300 m (0.2 mi) south of the fence, the measured dose dropped to 105 ± 10 mrem/yr, which is close to normal levels. The dose at the 7I boundary from the Waste Storage Facility is consistent with the dose at the center of the Facility; the two values usually vary in a parallel manner, and the changes with time reflect the amount of radioactive waste in storage and its gamma-ray intensity. At 14I, the average dose rate was about 24 mrem/yr above normal, about two-thirds of the 1980 rate. This dose is attributed to the use of cobalt-60 irradiation sources in Building 202.

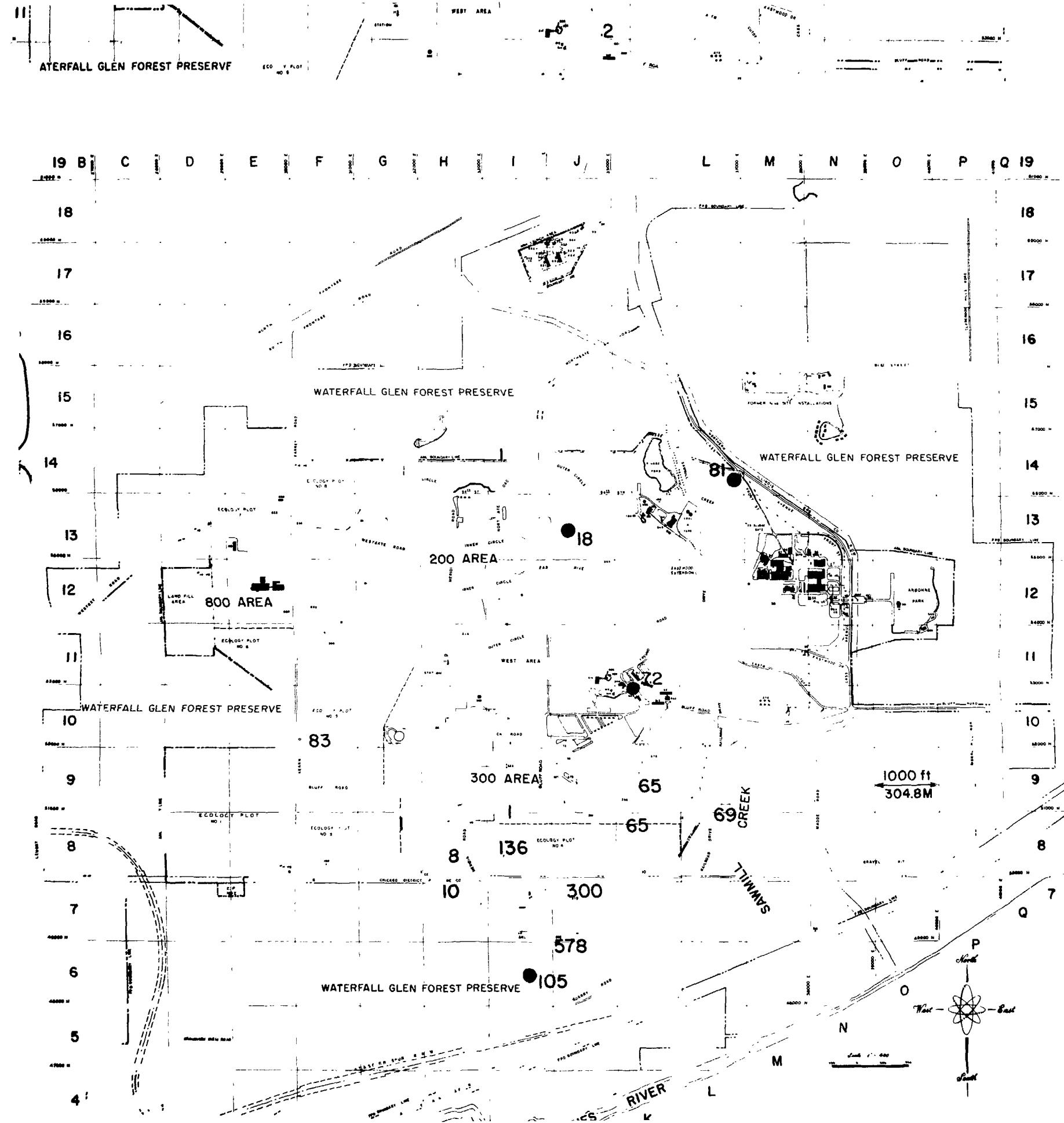
Dose variations from period to period at the same location, which are at least in part statistical, and natural dose differences between locations make it difficult to determine with high certainty when site boundary doses are only a few mrem/yr above normal and due to Argonne operations. Three criteria are used to identify such locations: 1) the results for each sampling period are frequently above the off-site average for the same period; 2) the annual average at a location exceeds the off-site average plus the

TABLE 18

Environmental Penetrating Radiation at ANL, 1981

Location	Dose Rate (mrem/year)				
	1/12-4/16	4/16-7/7	7/7-10/6	10/6-1/12	Average
14L - Boundary	83	75	83	84	81 ± 6
14I - Boundary	100	110	125	118	116 ± 11
14G - Boundary	-	80	86	86	84 ± 4
13D - Boundary	64	62	66	63	64 ± 3
9/10EF - Boundary	83	77	84	86	83 ± 6
8H - Boundary	87	-	-	-	87
8H - Boundary, 65 m E of Above	87	112	-	-	99 ± 25
8H - Boundary, Center, St. Patrick's Cemetery	93	109	109	108	105 ± 11
7I - Boundary	515	599	548	648	578 ± 78
6I - 200 m N of Quarry Road	98	99	111	111	105 ± 10
9I - Boundary	64	69	72	70	69 ± 4
9H - 50 m SE of CP-5	1160	1070	800	800	953 ± 258
8H - 65 m S of Building 316	103	107	107	115	108 ± 7
8H - 200 m NW of Waste Storage Area (Heliport)	131	142	133	139	136 ± 7
7I - Center, Waste Storage Area	-	14,900	12,500	18,300	15,300 ± 4030
10/11K - Lodging Facilities	70	72	75	71	72 ± 3
9J - Between ZGS Condenser and Building 370	64	62	65	68	65 ± 4
9K - 12 m SW of Building 367	-	72	63	62	65 ± 8
13J - 135 m E of Building 202	182	168	186	196	182 ± 7
12M - 30 m W of Building 55	57	64	62	70	63 ± 7
9I - 65 m NE of Building 350 230 m NE of Building 316	104	109	108	132	114 ± 8





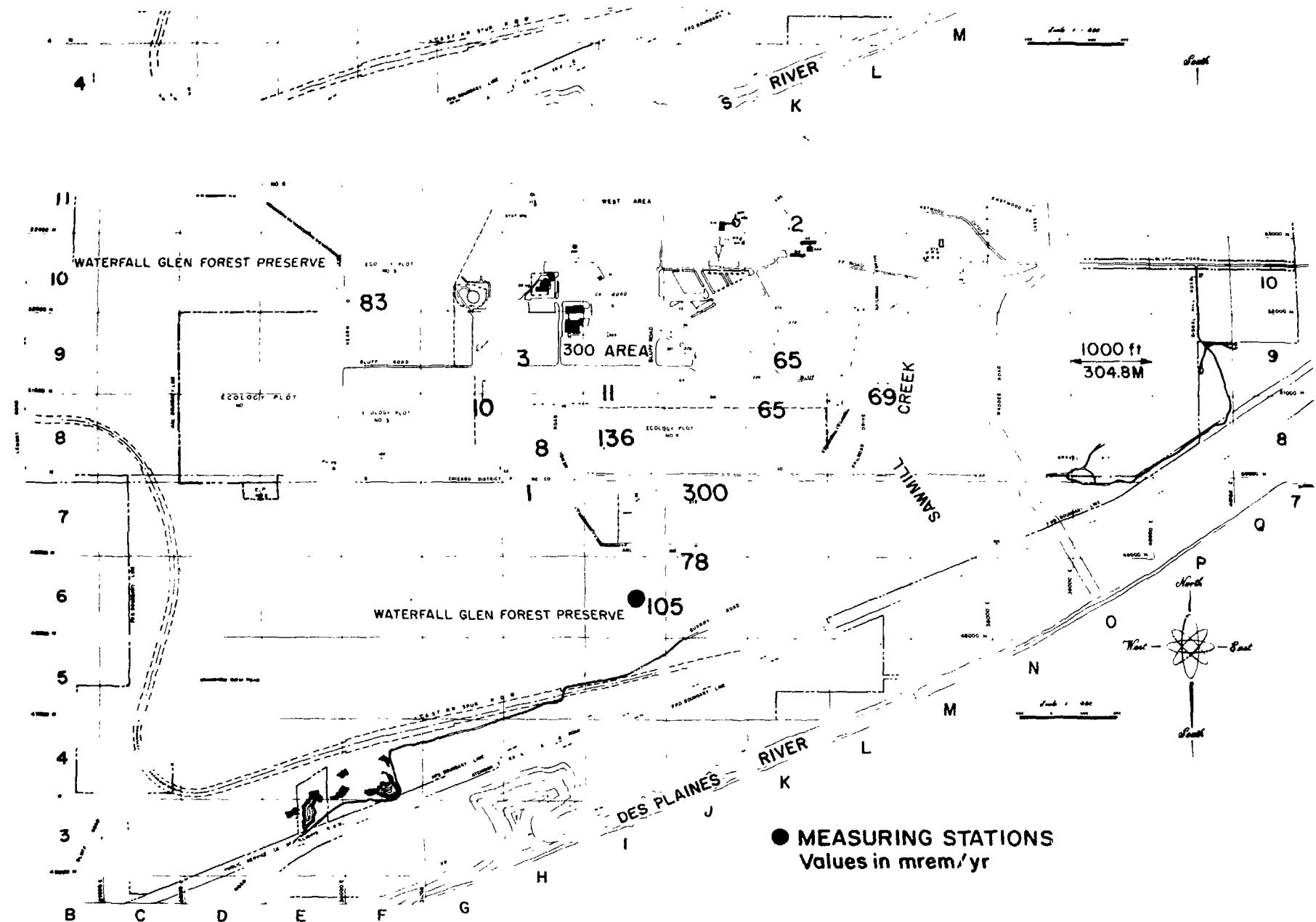


FIG. 6. Penetrating Radiation Measurements at the ANL Site, 1981

standard error of the average at the 95% confidence limit (99 mrem/yr); or 3) occasional results at a particular location are significantly above the normal value for that location, but do not exceed the off-site normal range. The last criterion could apply to a location such as 13D, where dose rates have been consistently below the average. At this location, the ground contains considerable gravel, which has less radium than the usual clay soil, and this probably accounts for the low dose rates, which averaged 64 mrem/yr in 1981. Application of the first two criteria and the difficulties in interpreting small differences from natural background by TLD measurements are illustrated below.

The dose at 8H is of interest since this area includes St. Patrick's Cemetery, which was in use before Argonne was constructed and is open to visitors. The dose at the center of the Cemetery was 13 mrem/yr above the off-site average and 6 mrem/yr above the 95% upper confidence limit of the normal range (99 mrem/yr). Two sampling locations on the boundary line between ANL and the Cemetery (8H) were essentially in the normal range. The dose rates at the center of the Cemetery are higher than at the fence line at 8H because of the topography. The higher values were obtained at a location that has a direct line of sight to a radiation source. Based on this comparison and the average individual results, the dose at 8H is estimated to be 5 to 10 mrem/yr above normal for that location. Similar results were obtained in previous years.

At the south end of the Laboratory site, three possible sources of external radiation exist: direct radiation from the Waste Storage Facility and direct radiation from a Dynamitron accelerator and low-power reactors in Building 316 (location 9GH). The contribution from Building 316 to the dose at 8H is considered negligible since dose rates measured in other directions from these sources at the same distance were less than at 8H. For this reason, the excess dose is attributed to the material stored at 7I, rather than to sources north of the Cemetery.

6. Potential Radiation Dose Estimates

a. Air and Waterborne Radionuclides

The radiation doses at the site boundary and off the site that could have been received by the public from radioactive materials leaving the site

were calculated by two methods. Where measured radionuclide concentrations in water were available, conversion of concentration to dose was based on the ratio of environmental concentrations to the Concentrations Guides given in Table 19. This table gives the annual radiation doses that would result from continuous exposure at the specified concentrations for those nuclides whose presence in the environment are attributable to Argonne. For argon-41 and krypton-85 released from stacks, doses were calculated from an atmospheric dispersion model which made use of a source term and meteorological data.

The principal exposure pathway for radioactive substances released from Argonne is directly from air to man by submersion. Although Sawmill Creek water is not used for drinking purposes, the dose that would be received by an individual ingesting water at the concentrations found in the Creek is calculated. A minor exposure route is from water to man by ingestion 710 km (370 mi) downstream from Argonne (Section I.E.), but the dilution of Sawmill Creek water at this point is so great that the dose calculation is meaningless. No other exposure pathways are significant.

Argon-41 and krypton-85 represent the major gaseous radioactive effluents released from the Laboratory. The concentrations and dose rates were calculated for those two nuclides by a computer program based on an atmospheric dispersion model.^{11,12} The following parameters were used in the calculations:

- a) annual release rates: argon-41, 0.4 Ci and krypton-85, 6.6 Ci.
- b) meteorological data: the wind velocity data shown in Figure 3.
- c) the usual parameters for stack height, building, wake, plume, momentum, temperature, etc.

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

The highest perimeter dose rates are in the north to east sectors. The closest full-time resident, who would receive the largest dose, is located approximately 0.5 km (0.3 mi) northeast of the site boundary. The results are summarized in Table 20.

TABLE 19

Concentration-to-Dose Conversion Factors				
Nuclide	Medium	Concentration*	Dose*	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
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
Iodine-131	Air	1×10^{-10}	1.5	Thyroid
Krypton-85	Air	3×10^{-7}	0.5	Whole Body
Neptunium-237	Water	3×10^{-6}	3	Bone
Plutonium-238	Water	5×10^{-6}	3	Bone
Plutonium-239	Water	5×10^{-6}	3	Bone
Strontium-90	Water	3×10^{-7}	3	Bone

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

TABLE 20

Whole-Body Dose From Airborne Emissions, 1981
(Millirems)

Nuclide	Pathway	Maximum Perimeter		Maximum Individual	
		Annual	50-Year	Annual	50-Year
Argon-41	Submersion	0.0001	0.0001	0.00005	0.00005
Krypton-85	Submersion	0.0005	0.0005	0.00025	0.00025

The population data in Table 2 was used to calculate the cumulative population dose from argon-41 and krypton-85. The results are given in Table 21, 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.

TABLE 21

80 km Whole-Body Population Dose

Source	Man-Rems	
	Annual	50-Year
Argon-41	0.002	0.002
Krypton-85	0.025	0.025
Natural	7.15×10^5	-

The total argon-41 or krypton-85 released by the Laboratory during 1981 is too small to permit their estimation by indirect methods such as has been done in the past for argon-41 with TLD measurements.

The only location where radionuclides attributable to Argonne operations could be found in off-site water was Sawmill Creek below the waste-water out-fall. The nuclides added to Sawmill Creek by Argonne waste water, their net concentrations in the Creek, and the corresponding dose rates if water at these concentrations were used as the sole water supply by an individual are

given in Table 22. 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 22

Radionuclide Concentrations and Dose Estimates
for Sawmill Creek Water, 1981

Nuclide	Conc. (net avg.) 10^{-9} $\mu\text{Ci}/\text{ml}$	Dose mrem/year	Percent of Standard*
Hydrogen-3	95	0.016	0.003
Strontium-90	0.11	1.1	0.037
Neptunium-237	0.0076	0.0076	0.00025
Plutonium-239	0.0085	0.0051	0.00017
Americium-241	0.0037	0.0014 (kidney) 0.0022 (bone)	0.00009
Curium-244	0.0016	0.0007 (bone)	0.00002
Californium-249	0.0016	0.0012 (bone)	0.00004

* DOE Order 5480.1 Standard.

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

The Environmental Protection Agency (EPA) has established drinking water standards based on a dose of 4 mrem/yr for man-made beta particle and photon emitting radionuclides.¹⁴ For hydrogen-3, the EPA standard is 2×10^{-5} $\mu\text{Ci}/\text{ml}$ and for strontium-90, it is 8×10^{-9} $\mu\text{Ci}/\text{ml}$. The concentrations in Table 22 correspond to 0.5% for hydrogen-3 and 1.4% for strontium-90 of the EPA

standards. No specific EPA standards exist for the transuranic nuclides.

b. External Penetrating Radiation Dose

Above normal fence-line doses attributable to Argonne operations were found at the south boundary near the Waste Storage Facility (location 7I), at the north boundary near Building 202 (14I), and at the south boundary adjacent to St. Patrick's Cemetery (8H). The results are discussed in Section III.A.5.

At location 7I, the fence-line dose from Argonne was about 490 mrem/yr. Approximately 300 m (0.3 mi) south of the fence line (grid 6I), the measured dose averaged 105 ± 10 mrem/yr, slightly above the upper end of the normal range, 99 mrem/yr. There are no individuals living in this area. The closest residents are about 1.6 km (1 mi) south of the fence line. At this distance, the calculated dose rate from the Waste Storage Area is 0.02 mrem/yr, if the energy of the radiation were 0.66 MeV, and 0.05 mrem/yr, if the energy were 1.3 MeV. The energy spectrum of the radiation varies with the composition of the stored waste and is not known, so it is necessary to assume an energy to make the calculations. Since cesium-137 and cobalt-60 are common radionuclides, the energies of the gamma-rays from these nuclides were used in the calculations.

At St. Patrick's Cemetery (8H), the estimated dose attributable to Argonne was estimated at 5-10 mrem/yr. An individual spending an average of 1 hr/week at this location would receive an annual dose in the range of 0.003 mrem/yr to 0.006 mrem/yr. In the area north of the site, where the fence-line radiation dose from the cobalt-60 sources in Building 202 was measured at about 24 mrem/yr, the nearest residents are 750 m (0.47 mi) to the north-northwest. The calculated dose at that location was about 0.04 mrem/yr.

The applicable Radiation Protection Standards for whole body external radiation dose to the general population is a maximum of 500 mrem/yr to critical individuals, or if individual doses are not known, 170 mrem/yr to a suitable sample of the exposed population.⁵ The latter criterion assumes that the maximum dose to individuals in the sample will not exceed the average by more than a factor of three. Thus, the external penetrating radiation doses to individuals living near the site will not exceed 0.01% of

the 500 mrem/yr limit or 0.03% of the "suitable sample" limit. At the fence line, where higher doses were measured, the land is wooded and unoccupied.

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

c. Summary

The total dose received by off-site residents was combined from the separate pathways that contribute to this total: argon-41 and krypton-85 submersion dose and cobalt-60 external radiation dose. The highest dose was about 0.06 mrem/yr to individuals living south of the site if they were outdoors at that location during the entire year. The total annual population dose to the entire area within an 80 km (50 mi) radius is < 0.1 man-rem.

B. Chemical and Biological Pollutants

1. Air

The environmental air data reported for the past several years has been obtained to determine concentrations of total suspended particulates (TSP) and trace metals prior to the burning of coal at Argonne as a replacement fuel for oil and gas. Coal use began on February 10, 1981, at Boiler No. 5, and monitoring was directed toward demonstrating compliance with the USEPA Order No. EPA-5-80-A¹⁵ as well as continuing previous studies. The Order provides that samples for TSP be collected for 24-hour periods at six-day intervals and that these results be reported monthly. For this purpose, samplers were maintained at location 8F, generally sampling air upwind of the boiler, and at location 12M, generally sampling air downwind of the boiler. Continuous samplers were also maintained at these locations from January through October. A third continuous sampler was maintained at 14N, also frequently downwind of the boiler.

Twenty-four-hour samples were obtained using samplers equipped with constant flow regulation and temperature programming. Air flow rates were maintained at 1.2 m³/minute and this flow was verified by comparison with a Roots meter which served as a primary air flow standard. All other experimental parameters and locations for TSP and trace metals are as previously described.⁴ Trace metal analyses were performed on the continuous samples by x-ray fluorescence.

A sample was maintained at 12M to measure sulfur dioxide on the 24-hour, six-day interval schedule. Analysis for sulfur dioxide was performed using the West-Gaeke method and reliability was established by participation in the USEPA intercomparison studies.

State and federal standards for TSP, lead, and sulfur dioxide appear in Table 23. While only TSP results are required for reports to the USEPA, other parameters are useful in determining boiler effluent effects. Results for 1981 are shown in Table 24. Trace metal results are for the period January-July 1981. Both continuous and 24-hour samples at 8F and 12M had similar geometric means for TSP. The results at 14N averaged 60 $\mu\text{g}/\text{m}^3$, which is equal to the secondary standard. The TSP results at 12M and 14N were about 10% higher than 1980, which is not considered to be a significant difference. The secondary 24-hour standard of 150 $\mu\text{g}/\text{m}^3$ was not exceeded at any time. During the period from April 4 to 9, 1981, extremely high TSP levels were measured. Continuous samples ranged from 181-198 $\mu\text{g}/\text{m}^3$. A 24-hour sample at 8F obtained on April 7, 1981 (not on the six-day schedule due to equipment malfunction), contained 411 $\mu\text{g}/\text{m}^3$, by far the highest level ever measured. This was a period of unusually severe dust storms.

TABLE 23

Summary of National and Illinois
Ambient Air Quality Standards¹⁶

Constituent	Time of Average	Primary Standard	Secondary Standard
Particulate Matter (TSP)	Annual Geometric Mean	75 $\mu\text{g}/\text{m}^3$	60 $\mu\text{g}/\text{m}^3$
	24 Hour	260 $\mu\text{g}/\text{m}^3$	150 $\mu\text{g}/\text{m}^3$
Lead (Pb)	Quarterly Arithmetic Mean	1.5 $\mu\text{g}/\text{m}^3$	1.5 $\mu\text{g}/\text{m}^3$
Sulfur Dioxide	Annual Arithmetic Mean	0.03 ppm	None
	24 Hour	0.14 ppm	None

Note: Illinois Air Quality Standards are identical to National Air Quality Standards with the exception of lead, for which no State standard exists. All standards with averaging time of 24 hours or less are not to be exceeded more than once per year.

TABLE 24

Environmental Air Data, 1981
(Concentrations in $\mu\text{g}/\text{m}^3$)

Location	Constituent	No. of Samples	Geometric Mean ± S.D.	Arithmetic Mean
8F	TSP	38	46 ± 1.4	49
8F	TSP (24-hour)	54	49 ± 1.6	54
12M	TSP	38	52 ± 1.4	55
12M	TSP (24-hour)	54	53 ± 1.5	58
14N	TSP	36	60 ± 1.4	64
8F	Iron	25	1.0 ± 1.9	1.3
12M	Iron	26	1.3 ± 1.8	1.5
14N	Iron	25	1.5 ± 1.8	1.7
8F	Copper	25	0.12 ± 2.2	0.16
12M	Copper	26	0.13 ± 1.7	0.15
14N	Copper	25	0.15 ± 2.4	0.21
8F	Zinc	25	0.12 ± 3.2	0.24
12M	Zinc	26	0.13 ± 3.3	0.26
14N	Zinc	25	0.18 ± 3.3	0.36
8F	Lead	25	0.14 ± 1.6	0.16
12M	Lead	26	0.19 ± 1.6	0.21
14N	Lead	25	0.19 ± 2.0	0.24
8F	Bromine	25	0.02 ± 2.3	0.02
12M	Bromine	26	0.03 ± 2.6	0.04
14N	Bromine	25	0.02 ± 2.8	0.04
12M	Sulfur Dioxide	49	0.005* ± 2.2	0.008*

* Parts per million (ppm).

Results for iron were higher than in 1980 by 30%. Results during the April period of high particulate levels ranged from 3.7 $\mu\text{g}/\text{m}^3$ to 4.0 $\mu\text{g}/\text{m}^3$ and the 8F 24-hour sample described above contained 9.0 $\mu\text{g}/\text{m}^3$. The higher levels of iron for 1981 can be ascribed at least partially to increased dust loading. Results for lead and bromine are lower than in 1980. Both of these elements are present in leaded gasoline and this could be related to a lower usage of fuel of this type. Results for sulfur dioxide were below the primary 24-hour and annual standards and rank in the middle of values reported Statewide.¹⁶ The highest value observed in a single sample was 0.02 ppm. For the parameters studied, there is no indication of any effect on air quality due to burning coal at Argonne.

2. Water

The environmental water data contained in this section have been collected in an effort to ascertain Argonne's compliance with State of Illinois regulations on surface stream and effluent water quality, as well as to verify the adequacy of Argonne's effluent pollution controls. The appropriate standards are listed in the *Illinois Pollution Control Board Rules and Regulations*, Chapter 3.¹⁷ 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 appropriate State standards, which are listed in Table 25. Minimum detectable amounts are included for comparison. The detection limits for atomic absorption methods represent twice the background variation, which is commonly used for this purpose. Detection limits for ion selective methods are those listed by the manufacturer. The lower limit of detection for x-ray fluorescence is based on the appropriate counting statistics.

The testing procedures for water analysis have been described in previous reports in this series.⁴ The results of the measurement of 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 (\pm) limit value. This value is the standard error at the 95% confidence limit and it is calculated from the standard deviation of the yearly average. Only when sample concentrations are random does this value approach the actual distribution occurring at the sampling location.

As in the past, emphasis has been placed on three areas: Sawmill Creek,

TABLE 25

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

Constituent	State Standard Stream	Standard Effluent	Detection Limit
Ammonia Nitrogen (as N)	1.5	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	0.1
Arsenic	1.0	0.25	0.005
Barium	5.0	2.0	0.005
Cadmium	0.05	0.15	0.0004
Chromium(VI)	0.05	0.3	0.003
Chromium(III)	1.0	1.0	-
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

a tributary of the Des Plaines River, since this is the principal route for waste water leaving the Argonne site; the study of NPDES permit locations (including several cooling tower blowdowns); and the control of mercury releases in the effluent.

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 2.7 megaliters (0.72 million gallons) per day and in 1981 was comprised of 30% sanitary waste water and 70% water from laboratory operations. Laboratory waste is directed to a large holding tank with an overflow which limits the discharge flow at about 1300 liters/minute (350 gallons/minute).

Effluents from the waste treatment plant were monitored 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.

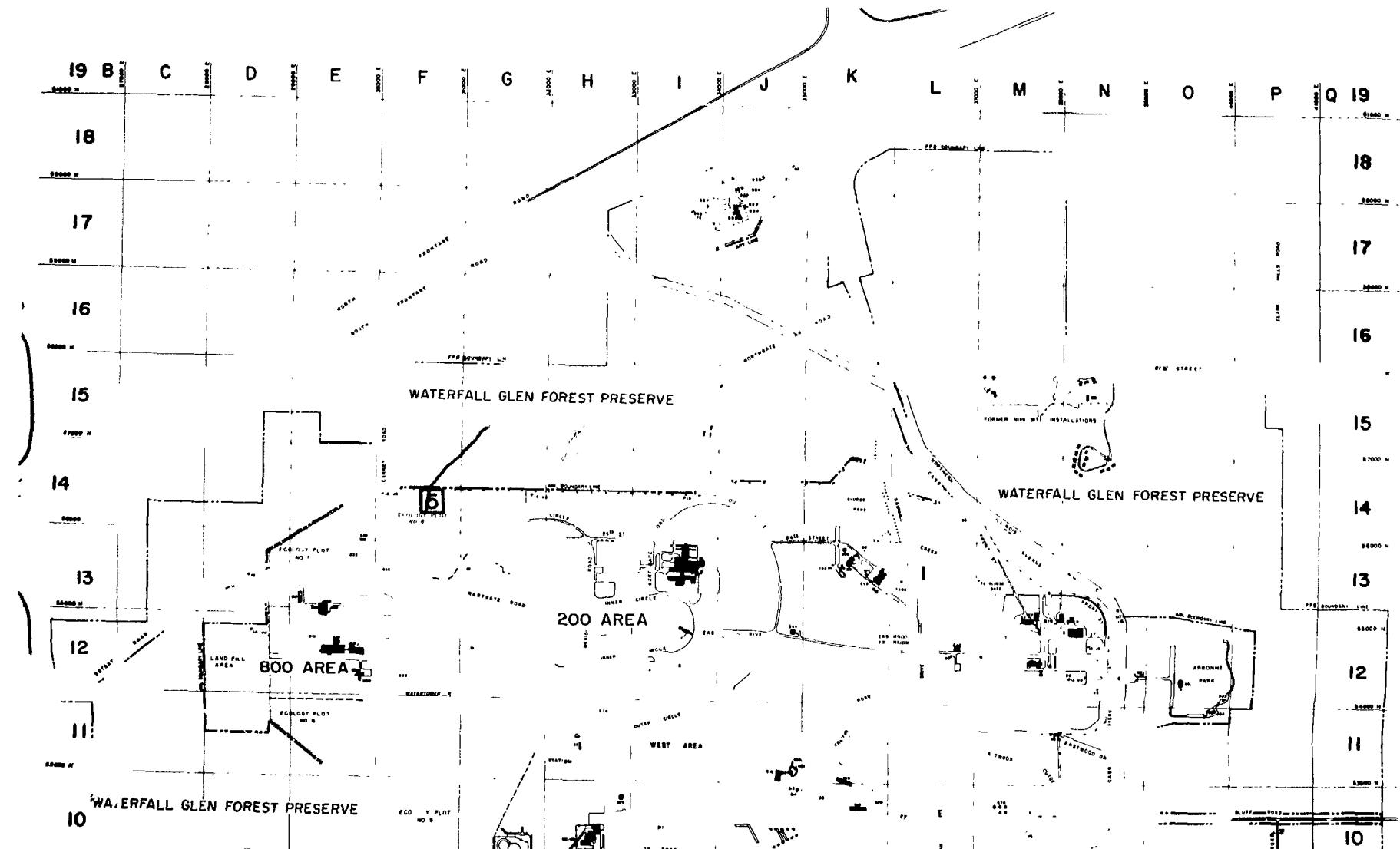
The results obtained for chemical constituents in the waste treatment plant effluent are shown in Table 26. All of the average concentrations were at or below the State standards. The average value for mercury was 75% of the State standard and 19% of the samples exceeded this value. Efforts are continuing to lower these levels. Concentrations of all the other constituents are similar to last year's values and are probably a measurement of background levels.

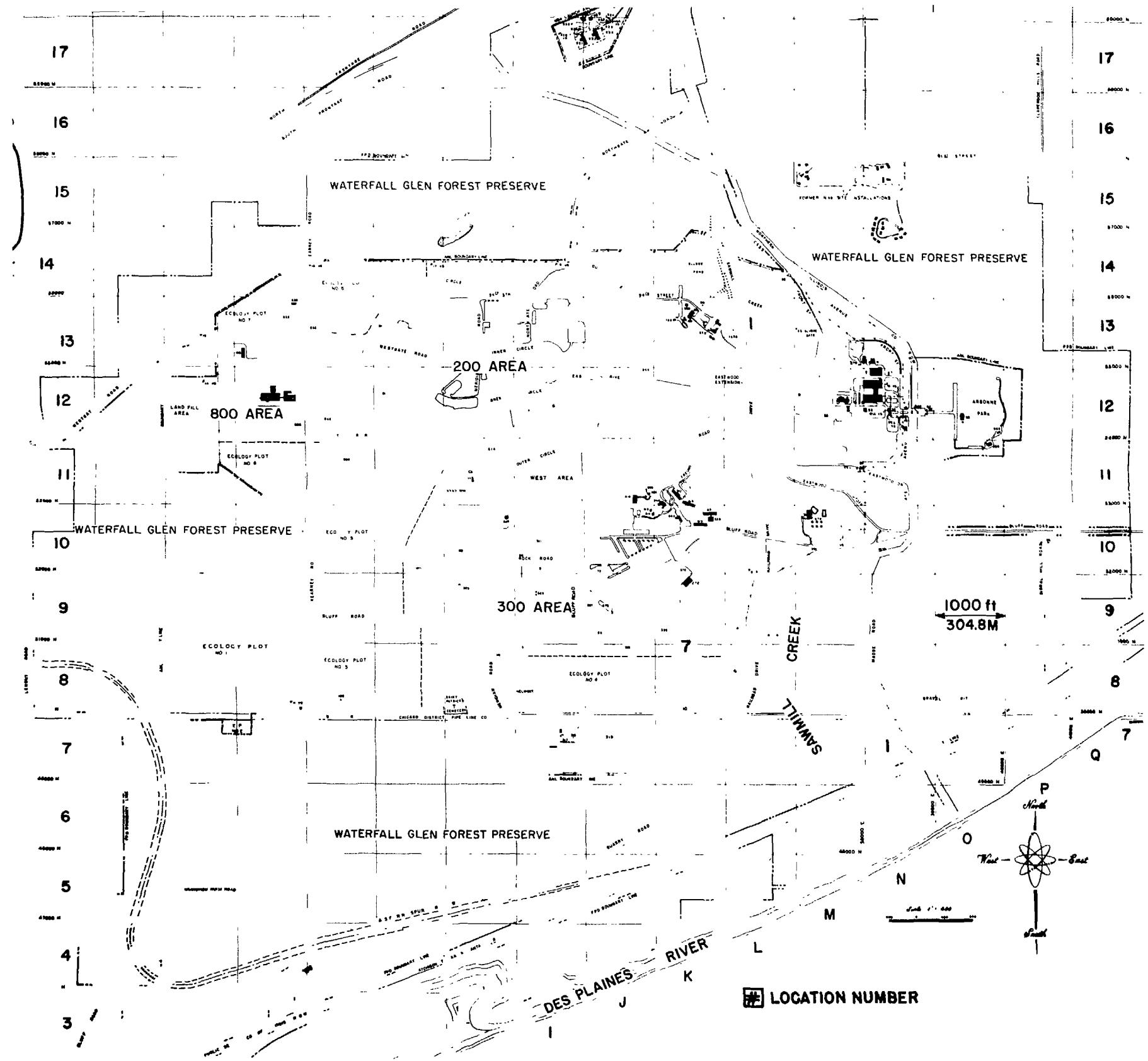
b. National Pollution Discharge Elimination System (NPDES)

A new and expanded NPDES permit program was begun in March, 1981. The permit provides for 10 sampling locations, which are shown in Figure 7. Locations 001 and 002 are the same as reported for 1980.⁴ Other locations cited in previous reports as cooling tower effluents are related to current permit locations as follows. Permit location 003 monitors water from the combined 200 Area blowdown channels and is former location 12L. Location 004 (formerly 14J) monitors water from Building 202 and contains cooling water. Location 005 monitors water from former locations 12F and 14G and now includes water from the entire 800 Area. Location 006 monitors the canal treatment plant outfall and is former location 8J. Locations 007, 008, and 009 are new sampling points and monitor water from ZGS cooling, East Area cooling, and

TABLE 26
CHEMICAL CONSTITUENTS IN EFFLUENTS FROM ANL TREATMENT PLANT, 1981
(CONCENTRATIONS IN MICROGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.	PERCENT OF STANDARD (AVG.)	PERCENT EXCEEDING STATE STANDARD
ARSENIC	52	< 5	-	< 5	< 2.0	0
BARIUM	43	14 ± 2	3	25	0.7	0
BERYLLIUM	12	0.020 ± 0.00	0.02	0.02	-	-
CADMIUM	51	0.6 ± 0.1	< 0.2	1.1	0.4	0
CHROMIUM(III)	51	4 ± 1	< 3	13.0	0.4	0
CHROMIUM(VI)	51	< 5	-	< 5	< 1.0	0
COPPER	51	26 ± 2	17	41	2.6	0
FLUORIDE	49	249 ± 18	192	512	1.7	0
IRON	51	296 ± 39	54	785	14.8	0
LEAD	51	3 ± 0.4	< 2	9.9	2.7	0
MANGANESE	51	39 ± 11	6	226	3.9	0
MERCURY	243	0.38 ± 0.05	0.05	3.0	75.2	19
NICKEL	51	9 ± 1	3	20	0.9	0
PH	244	-	6.8	8.4	-	0
SELENIUM	51	< 5	-	< 5	< 0.5	0
SILVER	51	3 ± 0.4	1	12	3.1	0
ZINC	51	131 ± 15	74	282	13.1	0





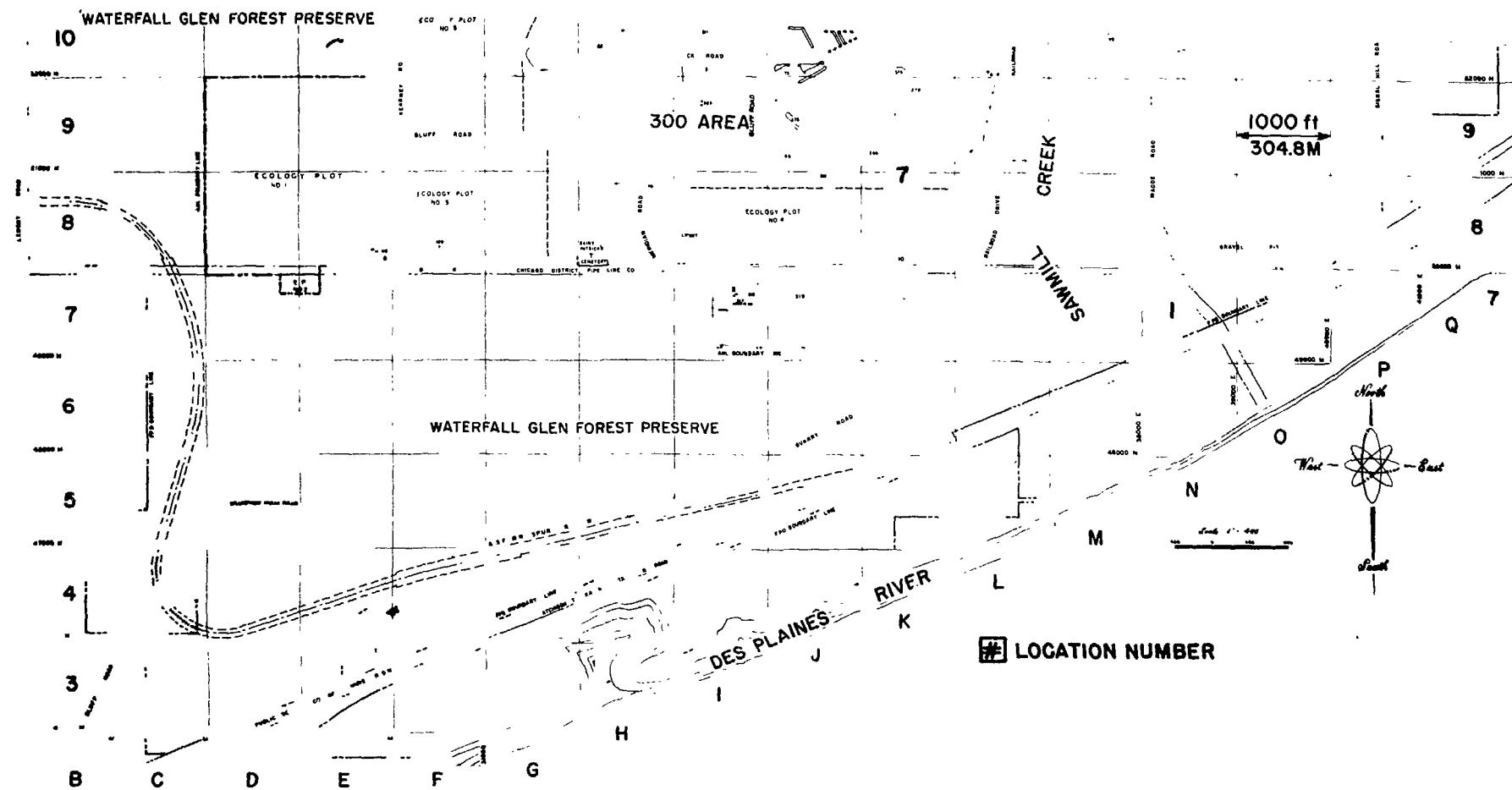


FIG. 7. NPDES Permit Locations

the water plant and pond overflow, respectively. Permit location 010 is designed to monitor coal pile runoff, but contained no water in 1981. Monitoring at locations 002-009 was carried out on a once per month basis, and were grab samples. Sampling at location 001 was performed once per week. The new permit at 001 does not include a limit for ammonia nitrogen. It does, however, add a limit for mercury in the laboratory waste stream before combination with the sanitary waste stream, as well as for three radioactive constituents, total beta activity, radium-226, and strontium-90.

The results obtained for the permit parameters for NPDES locations 001-009 are listed in Tables 27 to 35. At location 001, there was one coliform bacteria value that exceeded the limit and this was traced to low chlorine residual and was quickly alleviated. The level of 0.5 $\mu\text{g/l}$ for mercury was exceeded on twenty occasions by various amounts. The annual average of 0.8 $\mu\text{g/l}$ compares well with the annual average of 0.38 $\mu\text{g/l}$ previously listed as the waste treatment plant, Table 26, since there is about a one to two dilution. In December, 1981, new effluent regulations were adopted by the Illinois Pollution Control Board which provide a monthly limit, by exemption, of 3.0 $\mu\text{g/l}$ for mercury discharges from research facilities. If this limit were applied to the NPDES parameters, all the results would be in compliance. Occasional violations of a few parameters occurred at other locations for a variety of reasons, such as suspended solids due to runoff from heavy precipitation. At location 009, the pH was consistently above the upper limit of 9.0. This high pH is probably the result of the use of alkaline water softening compounds by the Laboratory. During the course of this report period, the pH control level was changed from 6.0-9.5 to 6.0-9.0. About half of the monthly samples were in compliance with the old standard, but exceeded the upper limit of the new standard.

c. Sawmill Creek

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

TABLE 27

ANL National Pollution Discharge Elimination
System Data Summary, Location 001 (7M), 1981

Parameter	Results Reported Permit Condition	Quantity or Conc.				Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.	Units			
Flow	Reported	0.72	-	-	MGD	-	Cont.	N/A
	Permit Condition	0.84	-	-		-	Cont.	N/A
B.O.D.	Reported	3.0	-	7.0	mg/l	0	1/7	24 Hr.
	Permit Condition	30	-	75		-	1/7	24 Hr.
Suspended Solids	Reported	4.0	-	20	mg/l	0	2/7	24 Hr.
	Permit Condition	30	-	75		-	1/7	24 Hr.
Fecal Coliform Bacteria	Reported	-	-	12,200	Number/ 100 ml	1	1/7	Grab
	Permit Condition	200	-	400		-	1/7	Grab
Residual Chlorine	Reported	0.40	-	-	mg/l	0	1/7	Grab
	Permit Condition	0.75	-	-		-	1/7	Grab
Mercury	Reported	0.00082	-	0.0059	mg/l	20	1/7	Grab
	Permit Condition	-	-	0.0005		-	1/7	Grab
Nitrate (N)	Reported	5.2	-	26.8	mg/l	-	1/7	24 Hr.
	Permit Condition	-	-	-		-	-	Grab
pH	Reported	-	6.9	8.5	Units	0	1/7	24 Hr.
	Permit Condition	-	6.0	9.0		-	1/7	Grab
Gross Beta	Reported	16.7	-	24.8	pCi/l	0	1/7	Grab
	Permit Condition	-	-	100		-	1/7	Grab
Radium-226	Reported	0.26	-	0.94	pCi/l	0	1/7	Grab
	Permit Condition	-	-	1.0		-	1/7	Grab
Strontium-90	Reported	0.37	-	0.69	pCi/l	0	1/7	Grab
	Permit Condition	-	-	2.0		-	1/7	Grab

TABLE 28

ANL National Pollution Discharge Elimination
System Data Summary, Location 002 (13L), 1981

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
Flow	Reported Permit Condition	0.33	-	-	MGD	0	1/30	Single approxima- tion
		-	-	-		-	1/30	
Total Suspended Solids	Reported Permit Condition	-	-	86 15	mg/l	1	1/30	Grab
		-	-			-	1/30	Grab
Temperature	Reported Permit Condition	All Within Range Natural Temp. +5°			°F	0	1/30	Grab
						-	1/30	Grab
pH	Reported Permit Condition	6.7 6.5	12.1 9.0		Units	1	1/30	Grab
						-	1/30	Grab

TABLE 29

ANL National Pollution Discharge Elimination
System Data Summary, Location 003 (12L), 1981

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported Permit Condition	1.14	0.34	2.8	MGD	0	Single Reading	N/A
		-	-	-		-	Single Reading	N/A
Total Suspended Soils	Reported Permit Condition	7.9	2.4	14.1	mg/l	0	1/30	Grab
		-	-	15		-	1/30	Grab
Total Chlorine Residual	Reported Permit Condition	-	-	0.10	mg/l	0	1/30	Grab
		-	-	0.75		-	1/30	Grab
Total Dissolved Solids	Reported Permit Condition	585	332	789	mg/l	0	1/30	Grab
		-	-	3500		-	1/30	Grab
Temperature	Reported Permit Condition	All Results Within Limits Natural Temp. +5°			°F	-	1/30	Grab
pH	Reported Permit Condition	-	7.6	8.9	Units	0	1/30	Grab
		-	6.5	9.0		-	1/30	Grab

TABLE 30

ANL National Pollution Discharge Elimination
System Data Summary, Location 004 (14J), 1981

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported Permit Condition	0.093	0.015	0.14	MGD	0	Single Approximation	
		-	-	-		-	Single Approximation	
Suspended Solids	Reported Permit Condition	3.9	0.5	12.8	mg/l	0	1/30	Grab
		-	-	15		-	1/30	Grab
Total Dissolved Solids	Reported Permit Condition	522	425	680	mg/l	0	1/30	Grab
		-	-	3500		-	1/30	Grab
Temperature	Reported Permit Condition	All Results Within Limits Natural Temp. +5°			°F	0	1/30	Grab
						-	1/30	Grab
pH	Reported Permit Condition	-	7.6	8.1	Units	0	1/30	Grab
		-	6.0	9.0		-	1/30	Grab

TABLE 31

ANL National Pollution Discharge Elimination
System Data Summary, Location 005 (14F), 1981

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported Permit Condition	0.12	0.05	0.18	MGD	0	Single Approximation	
		-	-	-		-	Single Approximation	
Suspended Solids	Reported Permit Condition	9.0	1.6	32	mg/l	1	1/30	Grab
		-	-	15		-	1/30	Grab
Fat, Oil & Grease	Reported Permit Condition	<5	-	<5	mg/l	0	1/30	Grab
		-	-	30		-	1/30	Grab
Temperature	Reported Permit Condition	All Results Within Limits Natural Temp. +5°			°F	-	1/30	Grab
						-	1/30	Grab
pH	Reported Permit Condition	-	7.5	7.9	Units	0	1/30	Grab
		-	6.0	9.0		-	1/30	Grab

TABLE 32

ANL National Pollution Discharge Elimination
System Data Summary, Location 006 (8J), 1981

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported Permit Condition	0.13	0.03	0.52	MGD	0	Single Approximation	Single Approximation
	-	-	-	-		-		
Suspended Solids	Reported Permit Condition	12.0	1.8	33.5	mg/l	2	1/30	Grab
	-	-	15	-		-		
pH	Reported Permit Condition	-	6.1	11.0	Units	1	1/30	Grab
	-	6.0	9.0	-		-		

TABLE 33

ANL National Pollution Discharge Elimination
System Data Summary, Location 007 (9K), 1981

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported Permit Condition	0.03	0.01	0.04	MGD	0	Single	Approximation
	-	-	-	-		-	Single	Approximation
pH	Reported Permit Condition	-	8.5	9.4	Units	1	1/30	Grab
	-	6.0	9.0	-		-	1/30	Grab

TABLE 34

ANL National Pollution Discharge Elimination
System Data Summary, Location 008 (13M), 1981

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported Permit Condition	0.02	0.002	0.061	MGD	0	Single Approximation	Single Approximation
pH	Reported Permit Condition	-	7.6	8.9	Units	0	1/30	Grab
		-	6.0	9.0		-	1/30	Grab

TABLE 35

ANL National Pollution Discharge Elimination
System Data Summary, Location 009 (14K), 1981

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported Permit Condition	0.02	0.001	0.034	MGD	0	Single Approximation	
		-	-	-		-	Single Approximation	
Suspended Solids	Reported Permit Condition	1.5	0.5	3.8	mg/l	0	1/30	Grab
		-	-	15		-	1/30	Grab
pH	Reported Permit Condition	-	9.4	9.9	Units	9	1/30	Grab
		-	6.0	9.0		-	1/30	Grab

The data from these studies are in Table 36. The average level for ammonia nitrogen upstream is 1.8 times the State of Illinois standard and individual samples exceeded the standard 54% of the time. The downstream sample averaged 1.5 times the State standard and exceeded this standard 50% of the time. All of the samples obtained downstream that violated the standard were due to contamination from upstream sources. The dissolved oxygen levels obtained during 1981 were all above the State minimum of 5 mg/l. The total dissolved solids above the outfall exceeded the State standard frequently, and increased the solids content of the downstream sample. Similar results for these constituents were obtained in 1980.

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 37.

Individual samples for arsenic, barium, cadmium, chromium, fluoride, lead, nickel, selenium, silver, and zinc did not exceed the State standards. The levels of mercury averaged 21% of the State limit and exceeded this limit 2% of the time. As in the past, high iron levels reflect high turbidity conditions and are not related to Argonne releases. The levels of copper exceeded State limits 20% of the time, down from 62% of the time in 1980.

Fuel oil spills occurred at the Argonne Magnetohydrodynamics Process Engineering Laboratory (AMPEL) in Building 145 at location 13L on August 8 and 9, 1981, due to a failure within the oil-slurry recirculation system. A total of 910 gallons of No. 2 fuel oil was released. Although prompt and concerted efforts were applied to contain the oil, some portion was released to Sawmill Creek. To evaluate oil levels in Sawmill Creek, two grab samples collected at 10M on August 9, 1981, and the continuous Creek sample collected during the period from August 7, 1981, through August 10, 1981, at 7M were analyzed.

The oil content of the grab samples was 40 and 290 mg/l, respectively. Oil was not detected in the continuous sample; the detection limit is 10 mg/l. From these data, it appears that elevated levels were localized and elevated levels did not appear downstream of the Laboratory. For comparison purposes, the NPDES discharge limit at location 005 for fats, oil, and grease is 30 mg/l.

TABLE 36

Sawmill Creek - Effect of Sanitary Waste, 1981

Constituent	Location *	No. of Samples	Concentration (mg/l)			Avg. Percent of Standard	Percent Exceeding State Standard
			Avg.	Min.	Max.		
Ammonia Nitrogen	7M (up)	48	2.8 ± 0.8	0.3	12.0	184	54
	7M (down)	48	2.3 ± 0.6	0.3	9.2	150	50
Dissolved Oxygen	7M (up)	48	9.7 ± 0.6	4.6	13.6	-	-
	7M (down)	48	9.8 ± 0.6	5.4	13.2	-	-
pH	7M (up)	48	-	7.6	8.5	-	-
	7M (down)	48	-	7.5	8.5	-	-
Sulfate	7M (up)	12	137 ± 28	78	192	28	0
	7M (down)	12	138 ± 28	82	196	28	0
Temperature	7M (up)	48	15.4 ± 1.9	1.2	25.7	-	-
	7M (down)	48	15.6 ± 1.9	2.4	25.2	-	-
Total Dissolved Solids	7M (up)	48	1120 ± 100	380	1780	112	63
	7M (down)	48	1020 ± 80	390	1440	102	60

* Location 7M (up) is 15 m (50 ft) upstream from the waste-water outfall. All other samples were collected 60 m (200 ft) downstream from the outfall.

TABLE 37
 CHEMICAL CONSTITUENTS IN SAMMILL CREEK LOCATION 7H, 1981
 (CONCENTRATIONS IN MICROGRAM/LITER OR MILLIGRAM/LITER)

CONSTITUENT	NO. OF SAMPLES	AVG.	CONCENTRATION MIN.	MAX.	PERCENT OF STANDARD (AVG.)	PERCENT EXCEEDING STATE STANDARD
ARSENIC	51	< 5	-	< 5	< 0.5	0
BARIUM	43	43 ± 5	5	85	0.9	0
BERYLLIUM	12	0.056 ± 0.02	0.02	0.10	-	-
CADMIUM	51	1 ± 0.1	0.3	3.1	1.4	0
** CHLORIDE	14	418 ± 69	220	600	-	-
CHROMIUM(III)	51	6 ± 2	3	24	0.6	0
CHROMIUM(VI)	51	< 3	-	< 3	< 6.0	0
COPPER	51	13 ± 2	4	38	66.4	20
FLUORIDE	51	361 ± 22	200	580	25.8	0
IRON	51	869 ± 232	116	3580	86.9	25
LEAD	51	7 ± 2	2	33	6.8	0
MANGANESE	51	144 ± 43	32	1030	14.4	2
MERCURY	248	0.11 ± 0.02	0.05	1.3	21.8	2
NICKEL	51	10 ± 2	3	27	1.0	0
PH	248	-	7.0	8.5	-	0
SELENIUM	51	< 5	-	< 5	< 0.5	0
SILVER	51	1 ± 0.2	0.2	4.3	27.4	0
ZINC	51	51 ± 7	10	119	5.1	0

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

** CONCENTRATION IN MILLIGRAM/LITER.

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 38. Two of the samples had very high levels of suspended material, as evidenced by iron levels of 3.2 and 6.7 mg/l. In no case was there any indication that hexavalent chromium, mercury, or zinc levels were affected by the Argonne effluent.

TABLE 38

Chemical Constituents in the Des Plaines River, 1981

Constituent	Location *	No. of Samples	Concentration (mg/l)		
			Avg.	Min.	Max.
Chromium(VI)	A	12	< 0.01	-	< 0.01
	B	24	< 0.01	-	< 0.01
Iron	A	12	1.7 ± 0.5	0.98	3.2
	B	24	1.6 ± 0.6	0.45	6.7
Mercury	A	12	< 0.0001	-	< 0.0001
	B	24	< 0.0001	-	< 0.0001
Zinc	A	12	0.05 ± 0.02	0.03	0.10
	B	24	0.05 ± 0.02	0.01	0.24

* Location A, near Route 45, is upstream and Location B, near Lemont, is downstream from the mouth of Sawmill Creek. See Figure 2.

IV. APPENDIX

A. References

1. U. S. Department of Energy, *Effluent and Environmental Monitoring and Reporting*, DOE Order 5484.1, Chapter III (1981).
2. *Draft Environmental Assessment Related to Operation of Argonne National Laboratory*, Argonne National Laboratory, Argonne, Illinois, December, 1981, Review Copy.
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 1980," U.S.D.O.E. Report ANL-81-23 (March 1981).
5. U. S. Department of Energy, *Standards 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 and J. Sedlet, "Radiochemical Determination of Plutonium in Environmental Water Samples," *Radiochem. Radioanaly. Letters*, 12, 215 (1972).
8. 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.
9. J. H. Harley (ed.), "HASL Procedure Manual," U.S.A.E.C. Report HASL-300 (1972).
10. 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).

11. 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).
12. 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).
13. 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.
14. "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.
15. "Delayed Compliance Order for the Department of Energy, Argonne National Laboratory," 40 CFR Part 55, *Federal Register*, Vol. 46, No. 17, Tuesday, January 27, 1981.
16. *Annual Air Quality Report 1980*, Illinois Environmental Protection Agency.
17. *Illinois Pollution Control Board, Rules and Regulations*, Chapter 3, 1972 (Chicago).
18. G. A. Welford, I. M. Fisenne, and C. Sanderson, "Summary Report of the Department of Energy, Division of Operational and Environmental Safety - Quality Assurance Programs 9 through 12," U.S.D.O.E. Report EML-369 (1980).

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

1. Standards

All of the standards and detection limits for chemical constituents, and some of those for radionuclides and external radiation, are given in the main body of the report. In addition, in Table 39 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

TABLE 39

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

Nuclide or Activity	Concentration Guide		Detection Limit	
	Water	Air	Water	Air
Americium-241	4×10^{-6}	4×10^{-12}	1×10^{-12}	5×10^{-19}
Antimony-125	-	9×10^{-10}	-	5×10^{-16}
Argon-41	-	4×10^{-8}	-	1.5×10^{-8}
Barium-140	3×10^{-5}	1×10^{-9}	2×10^{-9}	5×10^{-16}
Beryllium-7	-	4×10^{-8}	-	5×10^{-15}
Californium-249	4×10^{-6}	-	1×10^{-12}	-
Californium-252	7×10^{-6}	-	1×10^{-12}	-
Cerium-141	-	5×10^{-9}	-	5×10^{-16}
Cerium-144	-	2×10^{-10}	-	1×10^{-15}
Cesium-137	2×10^{-5}	5×10^{-10}	-	5×10^{-16}
Cobalt-60	-	3×10^{-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}	2×10^{-7}	1×10^{-7}	1×10^{-13}
Iodine-131	3×10^{-7}	1×10^{-10}	3×10^{-9}	5×10^{-15}
Krypton-85	-	3×10^{-7}	-	-
Neptunium-237	3×10^{-6}	-	1×10^{-12}	-
Plutonium-238	5×10^{-6}	1×10^{-12}	1×10^{-12}	1×10^{-19}
Plutonium-239	5×10^{-6}	1×10^{-12}	5×10^{-13}	1×10^{-19}
Radium-226	3×10^{-8}	-	1×10^{-13}	-
Ruthenium-103	-	3×10^{-9}	-	5×10^{-16}
Ruthenium-106	-	2×10^{-10}	-	1×10^{-15}
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}
Zirconium-95	-	1×10^{-9}	-	5×10^{-16}
Alpha **	3×10^{-6} to Beta **	1×10^{-10} to 1×10^{-13}	2×10^{-10} 1×10^{-9}	2×10^{-16} 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.

concentration of a radioactive nuclide. The Order distinguishes two CGs, one for occupational exposure in controlled areas and one for uncontrolled areas, beyond the site boundary where individuals can be exposed nonoccupationally, for 168 hours per week. The CGs in the table are for uncontrolled areas. For water, the standard selected was for the soluble form of the radionuclide; for air, the standard for the insoluble form was selected (except for iodine-131, for which the soluble form was chosen as a more conservative standard).

2. Detection Limits

The detection limits were chosen so that the error at the 95% confidence level is equal to the detection limit. 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 10%.

3. Quality Assurance Program

a. Radiochemical Analysis and Radioactivity Measurements

All nuclear instrumentation is calibrated with standard sources obtained from the U. S. National Bureau of Standards (NBS), if possible. If NBS standards were not available for particular nuclides, standards from the Amersham Corporation were used. The equipment is usually checked on a daily basis with secondary counting standards to insure proper operation. Samples are 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 DOE Environmental Measurements Laboratory (EML) are analyzed regularly. The DOE Environmental Measurements Quality Assurance Program (DOE-EML-QAP) is a semi-annual distribution of four or five different sample matrices containing various combinations of radionuclides.¹⁸ Results of our participation in this program during 1981 are given in Table 40. In the table, the comparison is made between the EML value, which is the result of replicate determinations by that Laboratory, and the value obtained in our laboratory. More than 95% of all the intercomparison samples received were analyzed for the radionuclides for which results were requested. To assist in judging the quality of the results, typical errors for our analyses are 2-50% and the error in the EML results is 1-30% (depending on the nuclide and the amount present). For most analyses for which the differences are large

TABLE 40

Summary of DOE-EML-QAP Samples, 1981

Nuclide	Average Difference From EML Value				
	Air Filters	Water	Soil	Tissue	Vegetation
Hydrogen-3	-	5% (2)	-	-	-
Beryllium-7	5% (2)	-	-	-	-
Potassium-40	-	-	4% (2)	1% (1)	7% (2)
Chromium-51	-	2% (1)	-	-	-
Manganese-54	2% (1)	3% (1)	-	-	-
Iron-59	-	3% (1)	-	-	-
Cobalt-57	6% (1)	21% (1)	-	-	-
Cobalt-58	14% (1)	-	-	-	-
Cobalt-60	16% (1)	1% (1)	-	-	-
Strontium-89	9% (1)	-	-	-	-
Strontium-90	15% (2)	5% (2)	8% (1)	67% (1)	24% (2)
Zirconium-95	7% (1)	-	-	-	-
Antimony-125	4% (2)	-	-	-	-
Cesium-134	3% (1)	-	-	-	-
Cesium-137	1% (1)	3% (1)	5% (2)	-	4% (2)
Cerium-144	-	11% (1)	-	-	-
Radium-226	-	-	3% (2)	9% (1)	75% (1)
Thorium-228	-	-	-	17% (1)	-
Uranium-234	-	7% (2)	21% (2)	-	-
Uranium-238	-	11% (2)	23% (2)	-	-
Plutonium-238	5% (1)	-	45% (2)	-	-
Plutonium-239	5% (2)	18% (2)	9% (2)	-	-
Americium-241	41% (1)	12% (2)	-	-	-

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

(> 20%), the concentrations were quite low and the differences were within the measurement uncertainties.

b. Penetrating Radiation

Our laboratory participated in the Fifth International Environmental Dosimeter Intercomparison Project conducted in 1980 by the DOE Environmental Measurements Laboratory and the School of Public Health, University of Texas (Houston). Participants supplied four sets of each type of dosimeter they wished to evaluate. One set was given approximately a four-month field exposure to a mixture of natural and artificial radiation at the Idaho National Engineering Laboratory; one set each was exposed in the laboratory to a known (but unrevealed) quantity of radiation at the beginning and one set at the end of the field exposure period, and one set served as a control. The laboratory exposures were to a cesium-137 source, and were done for a fading experiment. After exposure, the dosimeters were returned to the participant for measurement. The natural radiation field exposure was measured by an EML high-pressure ion chamber for comparison. The results, in mR, were:

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

The uncertainties listed are the standard deviations as estimated by the School of Public Health for their values and by us for our results. All results agree with the standard or accepted School of Public Health values within the measurement error at the 95% confidence level. The most important comparison is for the field exposures. Here the differences between the ion chamber result and our results were 3.1 mR \pm 3.5 mR (10.3% \pm 11.7%) for calcium fluoride and 1.0 mR \pm 3.4 mR (3.3% \pm 11.3%) for lithium fluoride.

c. Chemical Analysis

With each set of atomic absorption analyses, standard amounts of trace metals were analyzed in blank solutions at concentrations corresponding to 50% and 100% of the current State standards. Recoveries were determined by

comparing these results to results obtained by analyzing stream and effluent samples to which identical concentrations were added. Average recoveries ranged from about 85-100%, with a standard deviation of about 10%. Recovery studies were also performed for ammonia nitrogen, fluoride, and hexavalent chromium analyses, and similar results were obtained. All trace metals in air were referenced to Urban Air Particulate SRM 1648 from the Bureau of Standards.

Results of our participation in intercomparison studies during 1981 are summarized in Table 41. Samples distributed by the Department of Energy's Environmental Measurements Laboratory (EML) were analyzed for cations and anions in water. Agreement with EML results are acceptable for all species except ammonia nitrogen and chloride. Results for both of these species reported by other laboratories who performed these analyses were quite variable. Their ammonia nitrogen results averaged 86% of the EML value with a standard deviation of 28%; the chloride results averaged 110% with a standard deviation of 11%. The reasons for both the poor accuracy and precision for these two species are not known but a more extensive study should be performed.

Intercomparison air-filter samples furnished by the National Institute of Occupational Safety and Health (NIOSH) were analyzed for cadmium, lead, and zinc. The results were in excellent agreement with the reference values. The Environmental Protection Agency's sulfur dioxide (in potassium tetrachloromercurate) intercomparison samples were also analyzed. Results for samples containing from 0.03 ppm (the primary air standard) to 0.07 ppm were in excellent agreement. Results at 0.018 ppm and 0.021 ppm gave agreement within \pm 16%.

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

TABLE 41

Summary of Quality Assurance Studies

Constituent	Average Difference From Reference Value		
	NIOSH	EPA	EML
Cadmium	4% (4)	-	-
Copper	-	-	8% (2)
Lead	1% (4)	-	3% (1)
Manganese	-	-	5% (1)
Zinc	0% (4)	-	1% (2)
Ammonia Nitrogen	-	-	26% (1)
Chloride	-	-	16% (1)
Nitrate Nitrogen	-	-	6% (1)
Sulfate	-	-	4% (1)
Sulfur Dioxide	-	1.5% (9)	-

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

meter primary standard.

Samples are pre-treated in a manner designed to maintain the integrity of the constituent sought. For example, samples for trace radionuclide analysis are acidified immediately after collection to prevent hydrolytic loss of metal ions, but aliquots for radioiodine analyses are withdrawn first, since trace iodine is unstable in acid solution.