ANL-79-24

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY

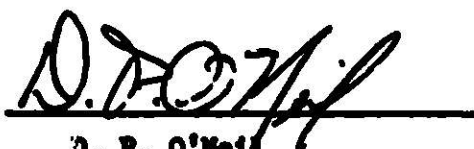ANNUAL REPORT FOR 1978

by

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

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March 1979

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ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY

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ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1978 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, Argonne effluent water, soil, grass, bottom sediment, and foodstuffs; 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, fall-out, 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 Manual Chapter 0513.[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 a multi-disciplinary research and development laboratory with several principal objectives. It carries out a broad program of research activities in the physical, biomedical, and environmental sciences and serves as an important center for energy research and development, both nuclear and non-nuclear. Some of the energy-related research projects are nuclear reactor safety studies, improvements in the utilization of coal for power production, coal liquefaction studies, the development of electric batteries for vehicles and off-peak energy storage, magnetohydrodynamic power generation, solar energy utilization, and ocean thermal energy conversion using ammonia as the working fluid. Environmental research studies include a Great Lakes radioecology program, which is primarily concerned with the effects of effluents from nuclear and fossil fuel power plants on Lake Michigan and other watersheds, studies on the dispersion and behavior of airborne pollutants under various meteorological conditions, and reclamation of strip-mined lands. Almost all of the work at the Laboratory is of an unclassified nature.

The principal nuclear facilities at the Laboratory are a 5 MW heavy-water cooled and moderated general-purpose research reactor (CP-5) fueled with fully-enriched uranium; 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 12.5 GeV proton accelerator, the Zero Gradient Synchrotron (ZGS); a 60-inch cyclotron; several other charged particle accelerators (principally of the Van de Graaff type), cobalt-60 irradiation sources; chemical and metallurgical plutonium laboratories; and several hot cells and laboratories designed for work with irradiated fuel elements and with multicurie quantities of the actinide elements.

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 miles) southwest of downtown Chicago, and 39 km (24 miles) 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 sq km (2,040-acre) area surround-
ing 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 miles) 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 miles)
southwest of the Laboratory to form the Illinois River.

The largest topographical feature is the Des Plaines River channel, about
1.6 km (1 mile) wide. This channel contains both the River and the Chicago
Sanitary and Ship Canal. Their presence extends the uninhabited area about
1.6 km (1 mile) south of the site. The elevation of the channel surface is
180 m (590 feet) above sea level. Bluffs, which comprise the south border of
the site, rise from the channel at varying slope angles of 15° to 60°, reach-
ing an average elevation of 200 m (650 feet) above sea level at the top. The
land then slopes gradually upward reaching the average site elevation of 220 m
(725 feet) above sea level at 940 m (3,000 feet) 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 feet). The remaining
portion of the site changes in elevation by no more than 7.6 m (25 feet) in a
distance of 150 horizontal m (500 feet). 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.
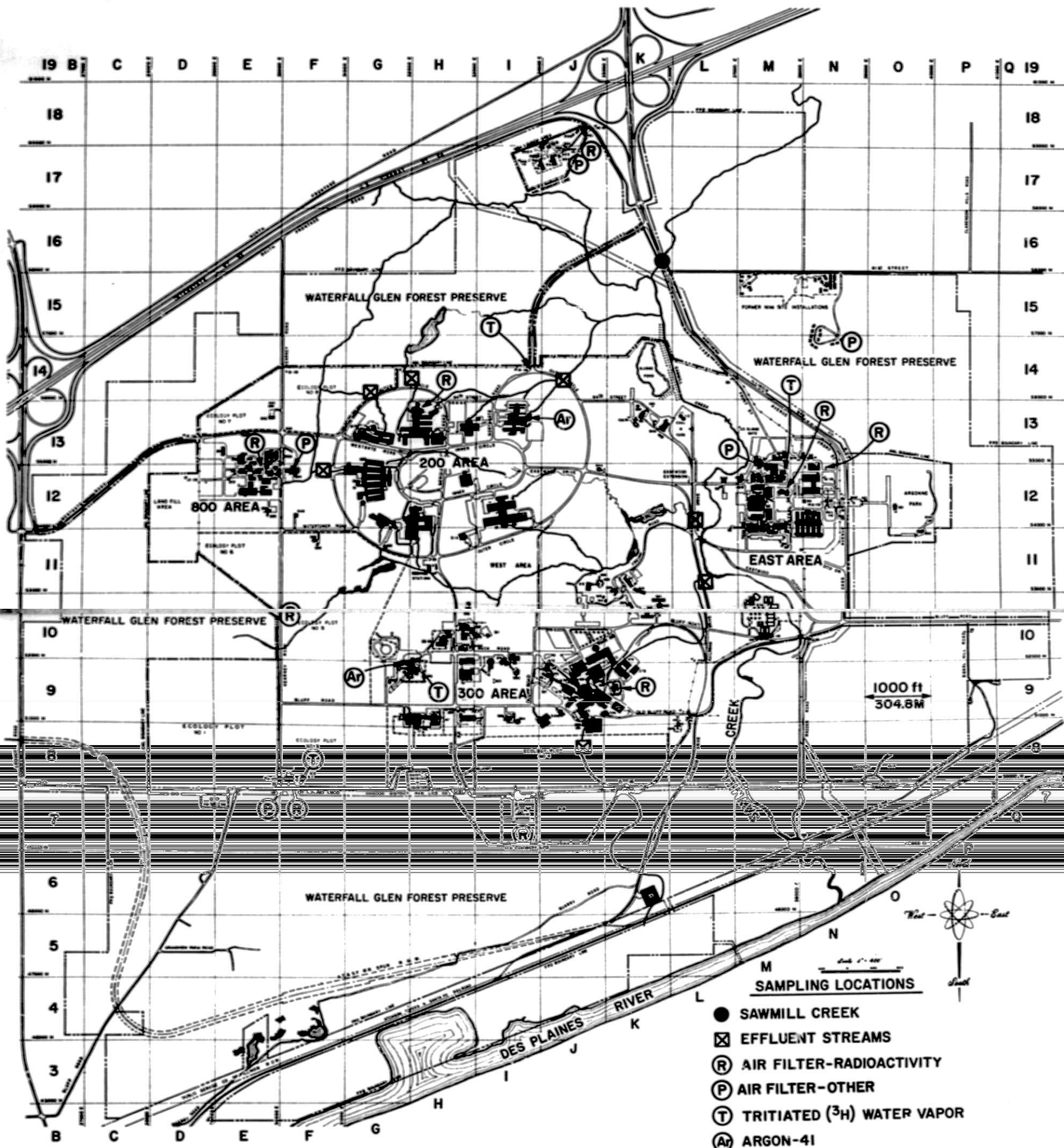
Fig. 1. Sampling Locations at Argonne National Laboratory

Fig. 2. Sampling Locations Near Argonne National Laboratory

C. Meteorology

The climate of the area is that of the upper Mississippi Valley, as moderated by Lake Michigan. A summary of the meteorological data collected on the site from 1950 to 1964 is available,[2] and provides an adequate sample of the climate. Similar data have been collected since 1964.

In the environmental monitoring reports for the past two years, the respective year's wind data were used in the dose calculations for airborne discharges. As a result of equipment failures, some due to lightning strikes on the meteorology tower, insufficient wind speed and direction data for 1978 are available for this purpose. Therefore, the 15-year summary,[2] shown as wind roses in Figure 3, was used this year for the dose calculations. The wind roses are essentially polar coordinate plots where the lengths of the radii represent percentage frequency of wind speeds in classes of 4 to 12 mph (6.4 to 19.3 km/hr), 4 to 24 mph (6.4 to 38.6 km/hr), and greater than 4 mph (6.4 km/hr). The direction of the radii represents the direction from which the wind blows. For example, on the 15-year summary rose for January in Figure 3 the three points plotted on the spoke extending horizontally to the left represent the wind speed distribution when the wind is blowing from the ten-degree sector, 266° to 275°. These points indicate that about 3.8% of the wind observations in January were from the west in the speed interval 4 to 12 mph, about 5.1% in the speed interval 4 to 24 mph, and about 5.2% in the speed interval greater than 4 mph. In the figure the curves for the latter two intervals are almost identical, and are not distinguishable on the drawing. The number 12.67 in the center of the rose represents the percent of observations of wind speed less than 4 mph in all directions.

The roses show that the predominant winds are from the southwest and west on the average, but seasonal variations are apparent. The winds are primarily westerly in January and February. In the spring, the lake breeze from the northeast becomes evident and continues through much of the summer. Southwesterly winds begin later in the spring, and become dominant in the fall. The winds are sufficiently variable so that monitoring for airborne releases must be carried out in all directions from the site. The daily average wind speed at 19 feet (5.8 m) above ground level varied from 5 mph (8 km/hr) in August to 10 mph (16.1 km/hr) in March. Gusts exceeded 50 mph

North

A = % of Calms (0-3 mph)
B = 4-12 mph
C = 4-24 mph
D = >4 mph
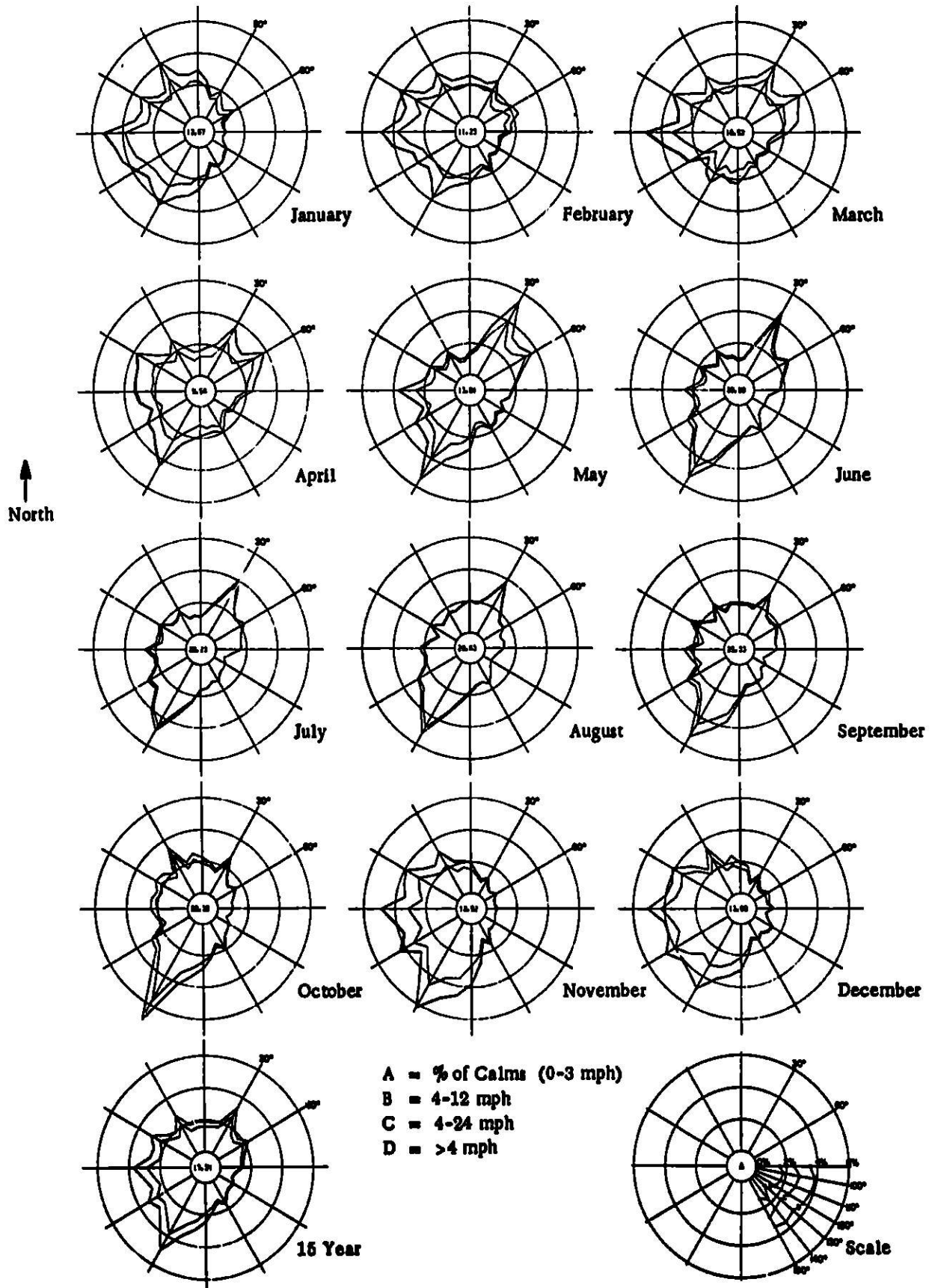
Fig. 3. Wind Roses Based on Hourly Observations,
January 1950-December 1964, 19-Foot Level

(80 km/hr) about once a year. The mean and median wind speeds for the 15-year period were 7.6 and 7.0 mph (12.2 and 11.3 km/hr), respectively.

The precipitation and temperature data for 1978 are shown in Table 1. Precipitation in July and December was well above average while the late summer and fall were below normal. Overall, the annual precipitation was 5% above average. The monthly temperature mean was well below average in the first three months of 1978 and December. The annual mean was about 15% below normal.

TABLE 1

Weather Summary, 1978

| Month | Precipitation (cm) | | Temperature (°C) | |
	Amount	Average[*]	Monthly Average	Average[**]
January	4.72	4.88	-9.0	-4.2
February	3.51	4.57	-8.5	-2.7
March	2.92	6.93	0.5	2.5
April	10.31	7.98	9.4	8.8
May	10.62	8.81	15.4	14.6
June	11.00	9.47	21.1	20.2
July	17.96	8.66	21.9	23.2
August	4.78	8.10	23.0	22.5
September	5.61	8.08	20.2	18.7
October	3.30	6.55	10.3	12.5
November	5.31	5.88	4.2	4.7
December	9.17	5.30	-4.1	-1.5

[*]Average precipitation, 1873-1977.

[**]Average temperature, 1871-1977.

D. Population

The area around Argonne has exhibited a large population growth in the past 15 years. Large areas of farmland have been converted into housing. A directional and annular 80-km (50-mile) population distribution for the area,

which is used for the population dose calculations later in this report, is shown in Table 2. [3] The distribution, centered on the CP-5 reactor, was obtained by modifying a similar distribution based on the 1970 U. S. Census and prepared by the Regional and Urban Studies Department at Oak Ridge National Laboratory. For distances within 8 km of the center, the updated values were largely based on quarter-section population data supplied by the Northeastern Illinois Planning Commission (NIPC). Some local adjustments were made on the basis of local observations. At distances beyond 8 km, but within the NIPC area, township forecasts were used, allocating population to each sector in proportion to the area of the township within the sector. The city of Chicago population was allocated in proportion to the area of each sector. Outside of the six county area around Chicago included in the NIPC program, the Northwestern Indiana Regional Planning Commission provided quarter-section estimates for Lake and Porter Counties and other Illinois Counties provided estimates of the population in their respective areas. Population counts were usually based on locally adjusted estimates made by the State of Illinois Bureau of the Budget and on special inter-term U. S. censuses for several municipalities. Included within the 80-km radius are portions of Lake and Porter Counties, Indiana, portions of Kankakee, Grundy, LaSalle, DeKalb, McHenry, and Lake Counties in Illinois, and all of the DuPage, Will, Cook, Kendall, and Kane Counties in Illinois.

E. 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 municipal sewage treatment plant (Marion Brook Treatment Plant) located a few kilometers north of the site and operating at about 11.4 megaliters (3 million gallons) per day. In addition, the residential development in the area has resulted in the collection and channeling of additional runoff water into the Creek. Treated sanitary and laboratory waste water from Argonne are combined and discharged into Sawmill Creek at location 7M in Figure 1. This effluent averaged 3 megaliters (0.8 million gallons) per day, while the water flow in the Creek upstream of the waste-water outfall averaged about 30 megaliters (8.1 million gallons) per day during 1978. The combined Argonne effluent consisted of 56% laboratory waste water and 44% sanitary waste water.

TABLE 2

Incremental Population Data in the Vicinity of ANL, 1978

Distance, miles	0 - 1	1 - 2	2 - 3	3 - 4	4 - 5	In Thousands				
Distance, km	0-1.6	1.6-3.2	3.2-4.8	4.8-6.4	6.4-8.0	5-10 8-16	10-20 16-32	20-30 32-48	30-40 48-64	40-50 64-80
Direction										
N	0	226	2171	3516	4859	44.0	163	309	168	193
NNE	0	124	1932	4314	2837	77.5	420	449	99	0
NE	0	347	1445	1233	1422	45.7	505	626	0	0
ENE	0	1729	3069	470	1617	45.0	684	289	0	0
E	0	9	212	0	12	19.2	653	508	13	26
ESE	0	0	88	275	118	18.6	213	320	308	46
SE	0	3	132	174	62	13.0	82	96	23	9
SSE	0	29	452	434	115	6.1	23	11	14	19
S	0	65	1305	688	772	3.9	31	4	27	40
SSW	0	39	3741	4557	665	11.8	97	8	18	7
SW	0	408	157	88	84	15.5	32	8	16	8
WSW	0	323	46	1199	2193	17.8	14	10	6	10
W	0	1242	740	7910	8852	13.4	42	18	15	8
WNW	0	662	136	2556	3960	24.5	73	46	6	52
NW	0	141	674	2267	6090	28.5	68	78	13	13
NNW	0	212	1390	1470	3414	37.0	101	135	91	66
Total	0	5559	17692	31151	37077	421.0	3202	2915	815	497
Cumulative Total	0		23251	54402	91479	512.0	3714	6629	7444	7941

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Seven streams (or channels) that carry cooling tower blowdown discharge water are considered effluents since the water eventually leaves the site boundary. Four streams from the 200 Area (sampling locations 14G, 12F, 14H, and 14J in Figure 1) leave the site at 14F, 14G, and 14J, respectively, and enter the Creek at 15K. The effluent sampled at 12L drains the Freund ponds at 11K and 12K and enters Sawmill Creek at 12L. The streams from the 300 Area (sampling locations 11L and 8J) cross the site boundary at 11L and 7K, respectively. The latter stream soaks into the ground south of the site, while the former enters the Creek at the release point.

Sawmill Creek and the Des Plaines River above Joliet, about 21 km (13 miles) 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 carries 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 miles) southwest of Argonne. The Dresden Nuclear Power Station complex is located at the confluence of the Kankakee, Des Plaines, and Illinois Rivers. This Station uses water from the Kankakee River for cooling, and discharges the water into the Illinois River. The first use of water for drinking is an indirect one at Peoria, on the Illinois River about 225 km (140 miles) downstream from Argonne, where River water is used to replenish groundwater supplies by infiltration. In the vicinity of the Laboratory, only subsurface water (from all aquifers, shallow and deep) 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. Most of this area has received little use thus far. It was available in 1978 for hiking, skiing, and equestrian sports, and its development and increased utilization by the public is expected. Very approximate estimates of usage are 600 individuals per day on weekends during the latter

part of the year, after a new automobile parking lot was opened, and 500 in-
dividuals for group educational purposes during 1978. 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 east-
ern part 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 agricul-
tural 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 and are for 1977.

TABLE 3

Agricultural Production Near ANL, 1977

County	Milk		Corn	Soybeans	Wheat	Oats
	No. of Cows	Million Pounds		Million Bushels		
DuPage	300	3.0	2.2	0.65	0.13	0.10
Cook	400	4.0	1.6	0.82	0.15	0.14
Will	3,300	33.0	15.8	4.6	0.70	0.55
Kane	6,600	66.0	15.1	2.6	0.32	0.57
Kendall	600	6.0	10.3	2.5	0.18	0.23
Grundy	900	9.0	12.3	3.0	0.13	0.15
Lake	1,400	14.0	2.3	0.72	0.28	0.26

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-78-26. Included in this report are the results of measurements obtained in 1978 for a number of radionuclides in air, surface water, soil, grass, bottom sediment, and food-stuffs; 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 the off-site and perimeter samples contained only radionuclides from natural sources and nuclear test detonations. Short-lived fission products were detected at all sampling locations in the spring and during December, and are attributed to fallout from the Chinese atmospheric nuclear tests of March 14, 1978, and December 14, 1978. No activity attributable to Argonne operations could be detected.

The plutonium-239,240 concentrations in air averaged 30×10^{-18} µCi/ml,* about 50% higher than the 1977 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) 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 0.003% of the CG.**

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*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 in Chapter 0524 of the DOE Manual.[4] The pertinent CGs are listed in the Appendix, Section IV.B. Comparison with other standards is given where appropriate.

Argon-41 and hydrogen-3 (as water vapor) represent the major airborne radioactivity released from the Laboratory. The argon-41 concentration and corresponding radiation dose at the site boundary was less than the detection limit (50% of the CG) of the sampling system, but was calculated from an atmospheric dispersion model to be 4.4×10^{-10} μCi/ml and 5.5 mrem/yr in the predominant east to northeast directions to which the wind is blowing. These values are 1.1% of the CG and the non-occupational dose limit to individuals (500 mrem/yr) for uncontrolled areas. The calculated dose at 2.2 km (1.4 miles) NE, where the closest full-time residents live, was 2.9 mrem/yr. These values are consistent with penetrating radiation dose measurements made at the site perimeter. The measured hydrogen-3 concentration at the site perimeter averaged about 16×10^{-12} μCi/ml, which is 0.008% of the CG and about eight times greater than the off-site concentration. The corresponding dose is 0.03 mrem/yr. These doses would only be received outdoors; no reduction for shielding by residences has been included.

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 CG of their average Creek concentrations, were hydrogen-3, 0.017%; strontium-90, 0.21%; neptunium-237, 0.0002%; plutonium-239,240, 0.0003%; americium-241, 0.0002%; and curium-244 and/or californium-249, 0.00002%. 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 1.8 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. The average plutonium-239,240 content of the top 5 cm of soil was 0.9×10^{-3} μCi/m^2 at the site perimeter and 1.2×10^{-3} μCi/m^2 off the site. The corresponding plutonium-238 averages were 0.04×10^{-3} μCi/m^2 and 0.05×10^{-3} μCi/m^2, respectively. The plutonium content of

grass was similar to that found in previous years and was about a factor of 10^4 less than soil from the same location. 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 1×10^{-9} µCi/g to 6×10^{-9} µCi/g of plutonium-239,240, a range found in previous years to be normal for fallout plutonium in such materials. The concentrations of uranium, thorium, and several gamma-ray emitters measured in soil, plant, and bottom sediment were normal.

Milk from a dairy farm near the Laboratory was analyzed for several fission products, including hydrogen-3. Hydrogen-3 concentrations averaged $< 220 \times 10^{-9}$ µCi/ml. The strontium-90 concentration (4.3×10^{-9} µCi/ml) increased by 35% compared to 1977, while the cesium-137 concentration (3.6×10^{-9} µCi/ml) was about the same as last year.

Measurements of penetrating radiation were made at several locations at the site boundary and off the site. The off-site readings averaged 93 mrem/yr, with a standard deviation of 5 mrem/yr, which is within 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 the south fence, the doses were about 190 mrem/yr above normal at location 7I in Figure 1 and 5-10 mrem/yr above normal at location 8H, as a result of radiation from an on-site temporary storage facility for radioactive waste. About 300 m south of the fence the measured dose dropped to 98 mrem/yr, which is within the normal range. Along the north side of the site, the dose at the fence at location 14I was 56 mrem/yr above normal as a result of radiation from cobalt-60 sources in Building 202. These locations are unoccupied, so there are no individuals receiving this dose. The calculated dose rate (outdoors) to the residents closest to the south boundary, about 1.6 km (1 mile) from the fence line, was about 0.02 mrem/yr; similarly, the dose rate to the residents closest to the north boundary, about 0.75 km (0.5 mile) from the fence was about 0.1 mrem/yr. Thus, doses to individuals living near the site will not exceed 0.05% of the 500 mrem/yr limit.

Levels 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. 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.

The weekly samples in the Argonne combined sanitary and laboratory waste-water effluent did not exceed the State of Illinois standard for Biochemical Oxygen Demand (B.O.D.) or suspended solids. Ammonia nitrogen exceeded the State standard in January, February, and March. Average concentrations of each of the other chemical constituents measured in this effluent were at or below these standards. The concentration of mercury averaged 98% of the State standard and exceeded this value 24% of the time (i.e., in 24% of the samples). No other constituent in this effluent exceeded the State standard at any time.

The average concentrations of hexavalent chromium from six of the seven cooling tower effluent channels used for blowdown were below the State standard. The levels in the effluent channel at 8J were consistently high, in part due to water diverted from the channel at 11L (Fig. 1). The channel at 11L is no longer used. All of the other channels averaged less than 3% of the State standard and in most cases contained non-detectable levels of hexavalent chromium.

The average values in Sawmill Creek for dissolved oxygen and chemical constituents, except ammonia nitrogen, dissolved solids, and copper, were within the State of Illinois standards. The average ammonia nitrogen level above the Argonne waste-water outfall was 1.7 times the State standard, and exceeded this standard in 49% of the samples. The average level below the outfall was 1.5 times the State standard and exceeded this value in 42% of the samples. The average concentration of dissolved solids above the Argonne waste-water outfall exceeded the State standard by 20%, but below the outfall it was less than the standard. The copper levels in the latter half of 1978 below the outfall exceeded the State standard in 56% of the samples and averaged 42% above the standard. The levels in the Argonne effluent were insufficient to account for these high levels and studies are underway to search for other sources. Individual values for cyanide, iron, mercury, and silver exceeded State standards from 1.2% to 13% of the time. Hexavalent chromium levels were not in excess of the State standard at any time. On one occasion, the level of fecal coliform in the Argonne waste water exceeded the individual sample standard of 400 organisms/100 ml, but at no time was the monthly

standard of 200 organisms/100 ml (geometric mean) exceeded. Samples collected in the Des Plaines River did not show any effect of Argonne effluent on levels of mercury, hexavalent chromium, iron, or zinc in the River.

Determination of total suspended particulate (TSP) and beryllium in air were continued. Studies were expanded to include iron, copper, lead, zinc, bromine, and strontium at the TSP locations. Results for TSP and beryllium were similar to last year. Results for the other trace elements ranged as follows: iron, 0.9-1.4 $\mu g/m^3$; copper, 0.07-0.5 $\mu g/m^3$; lead, 0.4-0.9 $\mu g/m^3$; zinc, 0.1-0.2 $\mu g/m^3$; bromine, 0.06-0.14 $\mu g/m^3$; and strontium, < 0.001-0.01 $\mu g/m^3$.

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 principally at the site perimeter and off the site for comparison purposes. Some on-site results are also reported when they are useful in interpreting perimeter and off-site results. Since radioactivity is usually spread by air and water, the sample collection program has concentrated on these media. In addition, soil, plants, foodstuffs, precipitation, and materials from the beds of lakes and streams were also collected and analyzed.

The results of radioactivity measurements are expressed in this report in terms of microcuries per milliliter (µCi/ml) for water, air, and milk and microcuries per gram (g) and 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 whose 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 true average concentration at locations where the concentrations not only varied greatly, but were at times not detectable.

Average values are usually accompanied by a plus-or-minus (±) limit value. Unless otherwise stated, this value is the 95% confidence limit calculated from the standard deviation of the average (standard error), and is a measure of the range in the concentrations encountered at that location. It does not represent the conventional error 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 Manual Chapter 0524.[4] 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 comparative purposes. Such values are enclosed in parentheses to indicate this. Where other standards are used, their source is identified in the text.

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 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

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

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 cm^3 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 average beta activity for the year, 0.97×10^{-13} µCi/ml, was about 60% of the 1977 average. The results in Table 5 indicate that the decrease was principally due to the decay of intermediate half-life fission products (zirconium-95-niobium-95, ruthenium-103, ruthenium-106-rhodium-106, cerium-141, and cerium-144) produced in nuclear test detonations. Fission products (e.g., ruthenium-103, iodine-131, barium-140-lanthanum-140 and cerium-141) from Chinese atmospheric nuclear tests on March 14, 1978, and December 14, 1978, were detected in the spring and again in December at all locations. In addition to the gamma-ray emitting nuclides listed in Table 5, technetium-99m, tellurium-132, and neodymium-147 were observed shortly after the test of March 14, 1978. The maximum concentrations for each of these nuclides were: 0.5×10^{-15} µCi/ml for technetium-99m, 2.7×10^{-15} µCi/ml for tellurium-132, and 1.9×10^{-15} µCi/ml for neodymium-147. These nuclides were produced in nuclear tests and their concentrations at the site perimeter and off the site were similar. About 65% of the gamma-ray activity was due to

TABLE 4

Total Alpha and Beta Activities in Air-Filter Samples, 1978[*]

(Concentrations in 10^{-15} µCi/ml)

Month	Location	No. of Samples	Alpha Activity			Beta Activity		
			Av.	Min.	Max.	Av.	Min.	Max.
January	perimeter	38	2.5	1.0	4.9	89	71	124
	off-site	14	1.6	0.6	2.6	85	57	122
February	perimeter	33	2.0	0.7	6.7	81	55	105
	off-site	15	2.1	0.9	3.4	81	49	104
March	perimeter	38	1.7	0.8	3.1	229	54	1017
	off-site	16	1.9	0.7	3.1	246	78	955
April	perimeter	39	2.2	0.6	5.0	203	125	306
	off-site	19	2.2	0.9	5.0	202	143	341
May	perimeter	39	2.4	0.1	5.4	123	72	230
	off-site	22	1.9	0.6	4.8	118	55	225
June	perimeter	37	2.5	0.8	4.9	132	62	193
	off-site	19	2.5	1.1	3.9	127	88	184
July	perimeter	37	1.6	0.5	3.6	79	40	122
	off-site	21	1.6	0.2	3.4	73	4	119
August	perimeter	39	2.1	0.8	4.4	61	41	87
	off-site	20	1.8	0.2	3.2	58	30	107
September	perimeter	37	2.3	0.9	4.0	45	21	94
	off-site	21	2.0	0.4	3.9	42	12	90
October	perimeter	42	2.9	0.9	10.2	33	20	49
	off-site	21	2.6	0.7	6.3	36	7	97
November	perimeter	35	2.3	0.7	9.5	46	30	73
	off-site	20	2.3	0.3	4.6	50	24	81
December	perimeter	41	1.6	0.4	3.4	41	20	67
	off-site	17	1.9	1.3	3.1	49	22	114
Annual Summary	perimeter	455	2.2 ± 0.2	0.1	10.2	97 ± 37	20	1017
	off-site	225	2.0 ± 0.1	0.2	6.3	97 ± 38	4	955
Percent CG	perimeter	–	(0.002)	(0.0001)	(0.010)	0.10	0.02	1.02
	off-site	–	(0.002)	(0.0002)	(0.006)	0.10	0.004	0.96

[*]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.

TABLE 5

Gamma-Ray Activity in Air-Filter Samples, 1978

(Concentrations in 10^{-15} μCi/ml)

Month	Location	^{7}Be	^{54}Mn	^{95}Zr-^{95}Nb	^{103}Ru	^{106}Ru-^{106}Rh	^{125}Sb	^{131}I	^{137}Cs	^{140}Ba-^{140}La	^{141}Ce	^{144}Ce
January	perimeter	83	0.2	9.9	0.4	8.1	0.9	< 5	1.7	0.1	0.2	16
	off-site	85	0.2	11	0.5	8.4	1.1	< 5	2.0	< 0.1	0.1	13
February	perimeter	68	0.2	7.4	0.2	8.1	1.1	< 5	1.7	< 0.1	0.1	16
	off-site	69	0.2	7.5	0.1	8.7	1.1	< 5	2.0	< 0.1	0.1	14
March	perimeter	94	0.2	10	7.8	14	1.8	84	2.9	19	5.3	27
	off-site	101	0.2	11	8.8	14	1.8	92	3.1	20	5.6	28
April	perimeter	103	0.4	11	11	18	2.2	36	4.2	19	6.4	26
	off-site	107	0.3	12	11	19	2.5	31	4.6	17	6.1	35
May	perimeter	95	0.3	6.0	0.9	14	2.5	< 5	4.0	0.7	0.3	33
	off-site	97	0.3	7.9	0.8	17	2.4	< 5	4.4	0.5	0.4	33
June	perimeter	116	0.3	5.6	0.2	18	2.6	< 5	4.7	0.1	0.1	33
	off-site	126	0.4	6.1	0.2	17	2.7	< 5	4.9	< 0.1	0.2	35
July	perimeter	89	0.2	2.6	0.1	10	1.6	< 5	2.9	0.2	< 0.1	20
	off-site	95	0.2	2.7	< 0.1	11	1.7	< 5	3.1	< 0.1	< 0.1	20
August	perimeter	101	0.2	1.2	< 0.1	6.0	1.0	< 5	1.9	0.1	< 0.1	11
	off-site	104	0.1	1.2	< 0.1	6.1	1.1	< 5	2.0	< 0.1	< 0.1	12
September	perimeter	93	0.1	0.3	< 0.1	3.2	0.5	< 5	1.0	0.2	< 0.1	5.2
	off-site	97	0.2	0.4	< 0.1	3.6	0.5	< 5	1.0	< 0.1	< 0.1	5.4
October	perimeter	71	< 0.1	0.1	< 0.1	1.7	0.3	< 5	0.6	< 0.1	< 0.1	3.0
	off-site	76	< 0.1	< 0.1	< 0.1	2.3	0.3	< 5	0.8	< 0.1	< 0.1	3.1
November	perimeter	82	< 0.1	< 0.1	< 0.1	1.6	0.4	< 5	0.6	< 0.1	< 0.1	2.8
	off-site	80	< 0.1	< 0.1	< 0.1	2.3	0.4	< 5	1.0	< 0.1	< 0.1	3.2
December	perimeter	69	< 0.1	0.1	0.2	1.4	0.3	6	0.6	1.6	0.3	2.1
	off-site	74	< 0.1	0.2	0.2	2.5	0.3	7	0.8	2.4	0.3	3.3
Annual Summary	perimeter	89 ± 8	0.2 ± 0.1	4.5 ± 2.5	1.8 ± 2.1	8.7 ± 3.6	1.3 ± 0.5	< 14	2.2 ± 0.9	3.4 ± 4.2	1.1 ± 1.3	16 ± 7
	off-site	93 ± 9	0.2 ± 0.1	5.0 ± 2.8	1.8 ± 2.2	9.3 ± 3.6	1.3 ± 0.5	< 15	2.5 ± 0.9	3.4 ± 4.1	1.1 ± 1.3	17 ± 7
Percent CG ($\times 10^{-3}$)	perimeter	(0.22)	0.02	0.45	0.06	4.4	0.14	< 14	0.44	0.34	0.02	8
	off-site	(0.23)	0.02	0.50	0.06	4.6	0.14	< 15	0.50	0.34	0.02	8

beryllium-7, produced in the stratosphere by cosmic-ray interactions. The remaining activity was primarily fission and activation products from nuclear test detonations.

The similarity of the annual averages of airborne alpha, beta, and gamma 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 in a localized source such as Argonne.

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 had been ignited at 1000°C.

Plutonium and thorium were separated on an anion exchange column. The acidity of the column effluent from this separation was adjusted to permit the extraction of uranium. 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 Tables 6 and 7.

The average plutonium-239 concentrations increased at all locations by about 50% over 1977; the increases occurred primarily during the first half of the year. Figure 4 shows the monthly plutonium-239 air concentrations for the past six years. The arrows at the bottom of the figure indicate the approximate dates of atmospheric nuclear tests. The average plutonium-238 concentration was similar to 1977, although monthly results were significantly higher during the first quarter of 1978.

TABLE 6

Strontium and Plutonium Concentrations in Air-Filter Samples, 1978

(Concentrations in 10^{-18} µCi/ml)

Month	Location[1]	Strontium-89	Strontium-90	Plutonium-238	Plutonium-239[2]
January	12N	610 ± 540	970 ± 120	0.9 ± 0.4	20 ± 2
	7I	720 ± 170	780 ± 40	0.6 ± 0.4	16 ± 2
	off-site	600 ± 250	810 ± 50	0.4 ± 0.4	16 ± 2
February	12N	460 ± 90	880 ± 30	0.2 ± 0.2	18 ± 1
	7I	540 ± 330	1050 ± 100	0.7 ± 0.4	20 ± 2
	off-site	320 ± 140	1020 ± 40	0.2 ± 0.2	19 ± 1
March	12N	6190 ± 450	1940 ± 170	0.6 ± 0.3	38 ± 2
	7I	7060 ± 100	1650 ± 40	1.0 ± 0.4	29 ± 2
	off-site	8610 ± 530	1740 ± 200	0.7 ± 0.3	32 ± 2
April	12N	9450 ± 150	2980 ± 60	1.3 ± 0.3	50 ± 3
	7I	7840 ± 420	2590 ± 150	1.2 ± 0.4	44 ± 2
	off-site	7300 ± 180	2680 ± 80	0.7 ± 0.3	51 ± 3
May	12N	650 ± 180	2370 ± 80	0.9 ± 0.5	67 ± 4
	7I	620 ± 90	2360 ± 40	1.9 ± 0.6	72 ± 4
	off-site	760 ± 130	2190 ± 540	1.2 ± 0.5	67 ± 4
June	12N	-	-	0.8 ± 0.6	68 ± 4
	7I	< 100	2550 ± 70	0.9 ± 0.4	68 ± 4
	off-site	< 100	2870 ± 50	1.5 ± 0.6	74 ± 4
July	12N	< 100	1490 ± 70	0.4 ± 0.4	37 ± 2
	7I	< 100	1590 ± 200	0.6 ± 0.4	38 ± 2
	off-site	< 100	1550 ± 40	0.7 ± 0.4	40 ± 2
August	12N	< 100	1110 ± 200	1.2 ± 0.6	33 ± 2
	7I	< 100	1090 ± 70	0.6 ± 0.4	24 ± 2
	off-site	< 100	1200 ± 50	0.7 ± 0.4	29 ± 2
September	12N	360 ± 260	420 ± 70	0.1 ± 0.3	13 ± 1
	7I	-	-	0.5 ± 0.3	12 ± 1
	off-site	-	-	0.1 ± 0.3	12 ± 1
October	12N	750 ± 540	270 ± 120	0.3 ± 0.4	8.3 ± 1.1
	7I	470 ± 110	310 ± 20	0.4 ± 0.3	8.1 ± 0.9
	off-site	520 ± 240	300 ± 50	0.4 ± 0.3	8.0 ± 0.9
November	12N	< 100	320 ± 50	0.2 ± 0.3	8.3 ± 1.1
	7I	< 100	370 ± 20	0.3 ± 0.3	7.7 ± 0.9
	off-site	< 100	210 ± 60	0.6 ± 0.5	8.0 ± 1.2
December	12N	470 ± 90	310 ± 40	0.1 ± 0.3	6.6 ± 1.0
	7I	430 ± 180	320 ± 70	0.7 ± 0.3	8.0 ± 0.9
	off-site	330 ± 260	200 ± 110	0.5 ± 0.3	5.4 ± 0.8
Annual Summary	12N	1750 ± 1870	1190 ± 550	0.6 ± 0.2	31 ± 13
	7I	1640 ± 1740	1330 ± 530	0.8 ± 0.2	29 ± 13
	off-site	1710 ± 1870	1340 ± 580	0.6 ± 0.2	30 ± 14
Percent CG ($\times 10^{-3}$)	12N	0.18	0.60	0.06	3.1
	7I	0.16	0.66	0.08	2.9
	off-site	0.17	0.67	0.06	3.0

[1]Perimeter locations are given in terms of the grid coordinates in Figure 1.

[2]Plutonium-240 is included (see text).

TABLE 7

Thorium and Uranium Concentrations in Air-Filter Samples, 1978

(Concentrations in 10^{-18} µCi/ml)

Month	Location[1]	Thorium-228	Thorium-230	Thorium-232[2]	Uranium-234	Uranium-235	Uranium-238[2]
January	12N	5 ± 1	8 ± 1	5 ± 1	15 ± 1	0.3 ± 0.2	12 ± 1
	7I	6 ± 1	8 ± 1	5 ± 1	12 ± 1	0.2 ± 0.2	13 ± 1
	off-site	14 ± 1	23 ± 1	13 ± 1	20 ± 1	1.3 ± 0.4	22 ± 1
February	12N	4 ± 1	5 ± 1	2 ± 1	10 ± 1	0.3 ± 0.2	12 ± 1
	7I	5 ± 1	6 ± 1	4 ± 1	11 ± 1	0.3 ± 0.3	14 ± 2
	off-site	8 ± 1	12 ± 1	7 ± 1	14 ± 1	0.5 ± 0.3	16 ± 1
March	12N	10 ± 1	10 ± 1	6 ± 1	12 ± 1	0.1 ± 0.2	11 ± 1
	7I	8 ± 1	8 ± 1	4 ± 1	11 ± 1	0.4 ± 0.2	9 ± 1
	off-site	9 ± 1	5 ± 1	4 ± 1	11 ± 1	0.4 ± 0.2	12 ± 1
April	12N	11 ± 1	14 ± 1	9 ± 1	16 ± 1	0.4 ± 0.2	18 ± 1
	7I	5 ± 1	5 ± 1	6 ± 1	12 ± 1	0.1 ± 0.2	12 ± 1
	off-site	8 ± 1	8 ± 1	6 ± 1	12 ± 1	0.6 ± 0.3	12 ± 1
May	12N	15 ± 2	15 ± 1	10 ± 1	23 ± 2	0.7 ± 0.4	15 ± 1
	7I	17 ± 2	20 ± 2	12 ± 1	23 ± 2	0.8 ± 0.3	20 ± 1
	off-site	13 ± 2	16 ± 2	9 ± 1	20 ± 2	0.5 ± 0.2	15 ± 1
June	12N	7 ± 1	20 ± 2	11 ± 1	27 ± 2	0.5 ± 0.4	20 ± 2
	7I	12 ± 1	20 ± 2	14 ± 1	26 ± 2	0.8 ± 0.3	24 ± 2
	off-site	12 ± 1	25 ± 2	20 ± 2	29 ± 2	0.6 ± 0.3	29 ± 2
July	12N	3 ± 1	8 ± 1	4 ± 1	12 ± 1	0.2 ± 0.2	10 ± 1
	7I	4 ± 1	9 ± 1	4 ± 1	13 ± 1	0.4 ± 0.3	10 ± 1
	off-site	11 ± 1	26 ± 2	15 ± 1	24 ± 2	1.1 ± 0.4	25 ± 2
August	12N	11 ± 2	15 ± 2	6 ± 1	13 ± 2	0.4 ± 0.4	12 ± 1
	7I	6 ± 1	9 ± 1	6 ± 1	11 ± 1	0.2 ± 0.2	10 ± 1
	off-site	21 ± 2	37 ± 2	20 ± 1	32 ± 2	2.3 ± 0.6	30 ± 2
September	12N	--	13 ± 1	6 ± 1	19 ± 2	0.6 ± 0.3	17 ± 2
	7I	--	14 ± 2	8 ± 1	17 ± 1	0.8 ± 0.3	15 ± 1
	off-site	--	31 ± 2	17 ± 1	28 ± 2	0.8 ± 0.3	29 ± 2
October	12N	--	13 ± 1	6 ± 1	16 ± 1	0.5 ± 0.3	14 ± 1
	7I	--	12 ± 1	6 ± 1	12 ± 2	0.5 ± 0.4	14 ± 2
	off-site	--	14 ± 1	7 ± 1	16 ± 1	1.0 ± 0.3 ·	19 ± 1
November	12N	6 ± 1	12 ± 2	7 ± 1	17 ± 2	0.7 ± 0.4	19 ± 2
	7I	4 ± 1	9 ± 1	5 ± 1	12 ± 1	0.1 ± 0.3	12 ± 1
	off-site	7 ± 1 ·	10 ± 1	4 ± 1	10 ± 1	0.3 ± 0.3	8 ± 1
December	12N	3 ± 1	9 ± 1	5 ± 1	14 ± 2	0.4 ± 0.4	13 ± 2
	7I	4 ± 1	9 ± 1	4 ± 1	14 ± 1	0.3 ± 0.3	12 ± 1
	off-site	5 ± 1	10 ± 1	5 ± 1	12 ± 1	0.4 ± 0.3	10 ± 1
Annual Summary	12N	8 ± 3	12 ± 2	6 ± 2	16 ± 3	0.4 ± 0.1	14 ± 2
	7I	7 ± 3	11 ± 3	6 ± 2	14 ± 3	0.4 ± 0.2	14 ± 2
	off-site	11 ± 3	18 ± 6	11 ± 4	19 ± 4	0.8 ± 0.3	19 ± 5
Percent CG (x 10^{-3})	12N	(4.0)	(4.0)	(0.6)	(0.40)	(0.01)	(0.28)
	7I	(3.5)	(3.7)	(0.6)	(0.35)	(0.01)	(0.28)
	off-site	(5.5)	(6.0)	(1.1)	(0.48)	(0.02)	(0.38)

[1]Perimeter locations are given in terms of the grid coordinates in Figure 1. --

[2]The concentrations in units of µg/m³ can be obtained by multiplying the value in µCi/ml by 2.96 x 10^{12} for uranium-238 and by 9 x 10^{12} for thorium-232. The mass of the other thorium isotopes in comparison to thorium-232 and the other uranium isotopes in comparison to uranium-238 is negligible.

Fig. 4. Plutonium-239,240 Air Concentrations, 1973-78

The annual average strontium-90 concentrations increased by about the same rate as the plutonium-239, while the strontium-89 concentrations decreased by a factor of about four as a result of radioactive decay. The strontium-89 was produced primarily in tests late in 1977 and the March test in 1978.

The long-lived radionuclides produced in nuclear detonations, plutonium-239, strontium-90, and cesium-137, showed similar monthly variations, and higher annual averages than shorter-lived fission products because of radioactive decay of the latter. There is no indication that these radionuclides originated from Argonne operations. Their concentrations were similar at the site perimeter and off the site and a pronounced spring peak is evident, indicating their stratospheric-fallout origin.

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. In contrast, the amount of plutonium in the air samples contributed by soil, if the resuspended soil has the same plutonium concentration as the first centimeter on the ground, ranged from about 0.4% in April to 5% in December of the total plutonium in the samples.

Using the thorium-228, -230, -232, uranium, and plutonium concentrations in air and soil for the past five years, resuspension factors (RF) were calculated and are presented in Table 8. The thorium and uranium air concentrations were measured directly, while the plutonium-239 concentration due to resuspended soil was calculated as above. All measurements are from the same air sampling location.

It is generally accepted that freshly deposited material will produce air concentrations of resuspended materials that decrease with time.[5] The resuspension factor should eventually reach a steady-state condition when the source has "aged" sufficiently. The thorium and uranium should represent the ultimate aged source and their resuspension factors, the equilibrium condition.

If it is assumed that most of the fallout plutonium was deposited in the early 1960's, the agreement between the thorium, uranium, and plutonium indicates that in 15 years the plutonium has reached an aged condition.

TABLE 8

Resuspension Factors

Nuclide	Air Concentration (aCi/m^3)	Ground Deposition (nCi/m^2)	RF (m^{-1})
Thorium-228	8.6	17	5.1×10^{-10}
Thorium-230	14.7	28	5.3×10^{-10}
Thorium-232	8.2	15	5.5×10^{-10}
Uranium	41.9	68	6.1×10^{-10}
Plutonium-239,240	0.7	1	7.0×10^{-10}

An Anderson 2000 high-volume cascade impactor was used to examine the distribution of thorium, uranium, and plutonium with particle size. The samples were collected at location 12M in Figure 1. The plutonium results are given in Table 9. The general trend for plutonium is that its concentration increases with decreasing particle size. This trend is consistent with the conclusion that most of the plutonium in air is of stratospheric-fallout origin. About 28% of the fallout plutonium would be expected to undergo pulmonary deposition, based upon the model of the ICRP Task Group on Lung Dynamics.

The thorium results were unusable because of the high thorium content of the sampling paper. The uranium results are in Table 10. The concentrations per volume of air decreased regularly with decreasing particle size, the opposite of the plutonium case. This is consistent with the conclusion that the source of the uranium in air is from resuspended soil.

Air sampling for argon-41 and hydrogen-3 (tritiated)water vapor was conducted in the exhaust stack of the CP-5 reactor (Building 330, 9H) because this reactor is the principal source of these nuclides at the Laboratory, and measurement of their concentrations at this location provides a source term. Tritiated water vapor measurements were also made at the site perimeter.

TABLE 9

Plutonium-239,240 Particle Size Distribution

Particle Range (µm)	Weight (g)	Particulate Concentration (µg/m³)	Plutonium-239,240 Concentration (aCi/m³)	Plutonium-239,240 Concentration (pCi/g) Fallout
> 7	0.440	19.3	0.97 (2%)	0.049
3.3-7	0.169	7.4	1.2 (3%)	0.16
2.0-3.3	0.097	4.3	3.0 (8%)	0.70
1.1-2.0	0.102	4.5	7.6 (20%)	1.66
< 1.1	0.445	19.5	25.9 (67%)	1.30
Total	1.253	55.0	38.7	

TABLE 10

Uranium Particle Size Distribution

Particle Range (μm)	Weight (g)	Particulate Concentration (μg/m^3)	Concentration (aCi/m^3)			Concentration (pCi/g)		
			^{234}U	^{235}U	^{238}U	^{234}U	^{235}U	^{238}U
> 7	0.440	19.3	30	1.6	29	1.55	0.083	1.50
3.3–7	0.169	7.4	18	0.6	18	2.43	0.081	2.43
2.0–3.3	0.097	4.3	16	< 0.5	16	3.76	< 0.2	3.76
1.1–2.0	0.102	4.5	12	< 0.5	11	2.68	< 0.2	2.46
< 1.1	0.445	19.5	4.5	< 0.5	3.5	0.23	< 0.1	0.18

Argon-41 was collected by filling an evacuated "Marinelli-type" container with air and was measured by gamma-ray spectrometry. Hydrogen-3 (tritiated) water vapor in air was collected by adsorption on silica gel and was measured by counting the desorbed water in a liquid scintillation counter.

Hydrogen-3 concentrations at three perimeter locations and off-site are given in Table 11. The 12M location (1900 m east-northeast of the CP-5 reactor) may be considered a perimeter sample for CP-5. The average hydrogen-3 concentrations at all perimeter locations are about 50% higher than in the past several years. The data show correlation with wind direction and indicate that, relative to the reactor, dilution to the background level occurs before reaching the site boundary in directions other than that from which the wind is blowing. The average and maximum perimeter concentrations were equivalent to 0.008% and 0.17% of the CG, respectively.

The off-site concentrations, measured about 10 km (6.2 miles) northwest of the Laboratory, were also slightly higher than levels observed at this location in the past. This background level of hydrogen-3 should be subtracted from the other concentrations in Table 11 to obtain the Argonne contribution.

Argon-41 and hydrogen-3 (in the form of tritiated water) from the CP-5 reactor constitute the major portion of the gaseous radioactive effluent released from the Laboratory. During 1978, the total amount of argon-41 discharged from the reactor was estimated to be 3.2×10^4 Ci, based on a measured release rate of 1.38 Ci/MW-hr. Since the half-life of this nuclide is only 110 minutes, about 5% will decay before reaching the site boundary if the argon-41 moves with an average wind speed of 3.4 m/s (7.6 mph). The total amount of hydrogen-3 (as tritiated water) discharged from the CP-5 reactor was 950 Ci, based on a measured release rate of 0.11 Ci/hr. These discharges and the corresponding doses will be discussed further in Section III.A.6.a.

Other airborne effluents were considerably lower. A small amount of argon-41, about 1.1 Ci in 1978, was released from the Janus reactor (Building 202, location 13I). The other major effluent was krypton-85, estimated to be 13 Ci. Several other fission products were also released in millicurie or smaller amounts. The release of iodine-131 (a nuclide of particular interest) in CP-5 exhaust air was estimated to be 0.015 Ci/yr, based on concentra-

TABLE 11

Hydrogen-3 Perimeter and Off-Site Concentrations, 1978

Month	Location*	No. of Samples	Concentrations in 10^{-12} µCi/ml		
			Avg.	Min.	Max.
January	8F	9	10	0.5	40
	14I	9	3.9	0.5	15
	12M	2	11	1.0	21
	off-site	2	0.5	0.3	0.7
February	8F	8	5.7	0.8	16
	14I	8	2.8	0.5	6.2
	12M	2	28	0.8	56
	off-site	2	1.0	0.6	1.5
March	8F	8	18	1.7	70
	14I	8	4.6	1.1	11
	12M	2	9.1	4.6	14
	off-site	2	1.2	0.8	1.5
April	8F	9	46	1.2	232
	14I	9	2.8	1.1	7.2
	12M	2	0.8	0.5	1.0
	off-site	2	0.7	0.6	0.8
May	8F	9	15	1.2	37
	14I	9	23	1.4	64
	12M	2	14	0.7	27
	off-site	2	1.0	0.7	1.3
June	8F	8	24	0.1	73
	14I	8	23	9.6	38
	12M	2	3.2	2.1	4.3
	off-site	2	2.4	2.0	2.9
July	8F	9	80	4.7	341
	14I	9	18	8.3	46
	12M	2	6.1	5.6	6.7
	off-site	2	7.1	3.7	10
August	8F	9	39	3.0	113
	14I	9	31	9.0	94
	12M	2	29	5.3	53
	off-site	2	3.0	2.7	3.4
September	8F	8	16	2.6	56
	14I	8	35	3.0	133
	12M	2	8.6	2.1	15
	off-site	2	1.4	0.1	2.8
October	8F	9	14	2.5	70
	14I	9	35	3.9	92
	12M	2	3.9	2.4	5.4
	off-site	2	2.8	1.6	4.1
November	8F	8	4.1	0.4	11
	14I	8	7.3	0.6	32
	12M	2	2.1	1.1	3.0
	off-site	2	1.0	0.2	1.8
December	8F	9	1.2	0.1	2.9
	14I	9	2.5	0.6	8.6
	12M	2	3.1	2.1	4.0
	off-site	2	0.3	0.2	0.3
Annual Summary	8F	103	23 ± 13	0.1	341
	14I	103	16 ± 7.6	0.5	133
	12M	24	9.9 ± 5.5	0.5	56
	off-site	24	1.9 ± 1.1	0.1	10
Percent CG	8F	-	0.012	0.00005	0.17
	14I	-	0.0080	0.00025	0.066
	12M	-	0.0050	0.00025	0.028
	off-site	-	0.0010	0.00005	0.0050

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

tion measurements in the stack. The maximum concentration at 1.5 km (0.93 mile), assuming no ground deposition, would be about 1×10^{-16} µCi/ml, or 10^{-4}% of the CG.

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 by liquid scintillation counting of 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 radio-chemical 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 counting 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.[6,7] 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 mile) down-stream from the waste-water outfall. Sawmill Creek was sampled upstream from the Argonne site and downstream from the waste-water outfall to deter-mine if radioactivity was added to the stream by Argonne waste water or from surface drainage. The sampling locations are shown in Figure 1. Below the waste-water outfall, daily samples were collected by a continuous sampler, which operated about 94% of the year. When the continuous sampling device was not functioning, a grab sample was collected each working day. Equal portions of the samples collected each week were combined and analyzed to obtain an average weekly concentration. Above the site, samples were usually collected once a month and were analyzed for the same radionculides as the below-outfall samples.

Annual summaries of the results obtained for Sawmill Creek are given in Table 12. 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 100% for plutonium-239; 90% for neptunium-237; 70% for americium-241; and 60% for hydrogen-3. 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 0.01% of the CG) was the lowest since 1972. However, 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 1978, the hydrogen-3 content of other lakes and streams ranged from $< 200 \times 10^{-9}$ µCi/ml to 290×10^{-9} µCi/ml and averaged 235×10^{-9} µCi/ml.

The average total alpha and beta activities were slightly higher above the site, which indicates that at times Argonne waste water contained less of these materials than Creek water. The higher activities above the site were probably due to the water added to the Creek by a large municipal sewage treatment plant. The large amount of dissolved solids present in the sewage water is naturally accompanied by a small amount of radioactive materials, and evidently increases the radioactivity in the Creek water.

In addition to the natural beta activity and that added by Argonne waste water at the outfall, beta activity from nuclear detonations was detected at both sampling locations. The normal nonvolatile beta activity is approximately 8×10^{-9} µCi/ml, while the contribution from the upstream municipal sewage treatment plant is another 8×10^{-9} µCi/ml. It is estimated that fallout activity added about 2×10^{-9} µCi/ml to the nonvolatile beta activity at both locations and that the Argonne contribution to the water below the outfall averaged about 1×10^{-9} µCi/ml. The Argonne contribution remained the same

TABLE 12

Radionuclides in Sawmill Creek, 1978

Type of Activity	Location[*]	No. of Samples	Concentration (10^{-9} µCi/ml)			Percent CG		
			Av.	Min.	Max.	Av.	Min.	Max.
Alpha (nonvolatile)	16K	12	1.6 ± 0.4	0.5	2.6	(0.053)	(0.017)	(0.087)
	7M	251	1.5 ± 0.2	0.2	5.5	(0.050)	(0.007)	(0.18)
Beta (nonvolatile)	16K	12	22 ± 3	15	32	(0.73)	(0.50)	(1.07)
	7M	251	18 ± 1	9	30	(0.60)	(0.30)	(1.00)
Hydrogen-3	16K	12	223 ± 73	< 200	406	0.0074	< 0.007	0.014
	7M	251	522 ± 111	< 200	1980	0.017	< 0.007	0.066
Strontium-89	16K	12	-	-	< 2	-	-	< 0.07
	7M	251	< 2.1	< 2	7.2	< 0.07	< 0.07	0.24
Strontium-90	16K	12	0.45 ± 0.18	< 0.3	1.2	0.15	< 0.10	0.40
	7M	251	0.63 ± 0.15	< 0.3	3.0	0.21	< 0.10	1.00
Iodine-131	16K	12	-	-	< 3	-	-	< 1.0
	7M	251	< 3.2	< 3	10	< 1.1	< 1.0	3.3
Barium-140	16K	11	-	-	< 2	-	-	< 0.007
	7M	135	-	-	< 2	-	-	< 0.007
Uranium[**] (natural)	16K	11	2.1 ± 0.3	1.5	3.5	(0.0052)	(0.0038)	(0.0088)
	7M	251	2.0 ± 0.1	1.0	3.3	(0.0050)	(0.0025)	(0.0082)
Neptunium-237	16K	12	-	-	< 0.001	-	-	< 0.00003
	7M	251	0.0059 ± 0.0018	< 0.001	0.0278	0.00020	< 0.00003	0.00093
Plutonium-238	16K	12	-	-	< 0.001	-	-	< 0.00002
	7M	251	0.0012 ± 0.0006	< 0.001	0.0101	0.000024	< 0.00002	0.00020
Plutonium-239	16K	12	-	-	< 0.0005	-	-	< 0.00001
	7M	251	0.016 ± 0.0104	< 0.0005	0.193	0.00032	< 0.00001	0.0039
Americium-241	16K	12	< 0.0011	< 0.001	0.0014	< 0.000028	< 0.000025	0.000036
	7M	251	0.0075 ± 0.0104	< 0.001	0.122	0.00019	< 0.000025	0.0030
Curium-242 and/or Californium-252	16K	12	< 0.0011	< 0.001	0.0018	< 0.0000055	< 0.000005	0.000009
	7M	251	< 0.0012	< 0.001	0.0056	< 0.0000060	< 0.000005	0.000028
Curium-244 and/or Californium-249	16K	12	< 0.0011	< 0.001	0.0011	< 0.000016	< 0.000014	0.000016
	7M	251	< 0.0012	< 0.001	0.0052	< 0.000017	< 0.000014	0.000074

[*]Location 16K is upstream from the Argonne site and location 7M is downstream from the Argonne waste-water outfall.

[**]Uranium concentrations in units of µg/l can be obtained by multiplying the concentration given by 1.48×10^9. The average concentration in the Creek then becomes 3.0 µg/l.

as in 1977, while the fallout contribution decreased by a factor of three.

The total radioactive effluent discharged to the Creek in Argonne waste water can be estimated from the average concentrations and the volume of water carried by the Creek. These totals are 3.7 Ci of hydrogen-3, 0.2 mCi of plutonium-239, 0.09 mCi of americium-241, 0.07 mCi of neptunium-237, and < 0.05 mCi of curium and californium nuclides.

Since Sawmill Creek empties into the Des Plaines River, which in turn flows into the Illinois River, the radioactivity in the latter two streams is important in assessing the contribution of Argonne waste water to the environmental radioactivity. The Des Plaines River was sampled twice a month below, and monthly above, the mouth of Sawmill Creek to determine if the radioactivity in the Creek had any effect on the activity in the River. Annual summaries of the results obtained for these two locations are given in Table 13. The average nonvolatile alpha, beta, and uranium concentrations in the River were very similar to past averages, except for the beta activity in 1976, and remain in the normal range. The beta activities were about 30% higher than 1976 due to increased fallout from nuclear testing. 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 natural nonvolatile beta activity was about 14×10^{-9} μCi/ml, and the excess, 2×10^{-9} μCi/ml, was due to fallout. The average nonvolatile alpha and beta activities, 1.7×10^{-9} μCi/ml and 13×10^{-9} μCi/ml, respectively, of 20 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 14, 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 plutonium content of soil, grass, and bottom sediment was measured at the site perimeter and off the site. The object of the off-site sampling was to determine the deposition of plutonium from weapons testing for comparison with perimeter samples, and with results obtained by other organizations

TABLE 13

Radionuclides in Des Plaines River Water, 1978

Type of Activity	Location*	No. of Samples	Concentration (10^{-9} µCi/ml) Av.	Min.	Max.	Percent CG Av.	Min.	Max.
Alpha	A	12	1.7 ± 0.3	0.9	2.8	(0.057)	(0.030)	(0.093)
(nonvolatile)	B	24	1.9 ± 0.3	0.9	4.7	(0.063)	(0.030)	(0.157)
Beta	A	12	15 ± 2	10	24	(0.50)	(0.33)	(0.80)
(nonvolatile)	B	24	16 ± 2	7	25	(0.53)	(0.23)	(0.83)
Hydrogen-3	A	12	< 207	< 200	286	< 0.007	< 0.007	0.0095
	B	24	< 206	< 200	265	< 0.007	< 0.007	0.0088
Strontium-89	A	12	-	-	< 2	-	-	< 0.07
	B	24	-	-	< 2	-	-	< 0.07
Strontium-90	A	12	0.7 ± 0.2	0.4	1.9	0.23	0.13	0.63
	B	24	0.7 ± 0.2	0.4	2.8	0.23	0.13	0.93
Iodine-131	A	12	< 4.0	< 3	15	< 1.3	< 1.0	5.0
	B	24	< 3.7	< 3	20	< 1.2	< 1.0	6.7
Barium-140	A	12	-	-	< 2	-	-	< 0.007
	B	12	-	-	< 2	-	-	< 0.007
Uranium**	A	12	2.1 ± 0.3	1.5	2.9	(0.0052)	(0.0038)	(0.0072)
(natural)	B	24	1.9 ± 0.2	1.0	2.6	(0.0048)	(0.0025)	(0.0065)
Neptunium-237	A	12	-	-	< 0.001	-	-	< 0.00003
	B	12	-	-	< 0.001	-	-	< 0.00003
Plutonium-238	A	12	-	-	< 0.001	-	-	< 0.00002
	B	12	-	-	< 0.001	-	-	< 0.00002
Plutonium-239	A	12	0.00062 ± 0.00023	< 0.0005	0.0014	0.000012	< 0.00001	0.000028
	B	12	0.00053 ± 0.00014	< 0.0005	0.0010	0.000011	< 0.00001	0.000020
Americium-241	A	12	< 0.0011	< 0.001	0.0022	< 0.000028	< 0.000025	0.000055
	B	12	< 0.0011	< 0.001	0.0017	< 0.000028	< 0.000025	0.000042
Curium-242 and/or	A	12	< 0.0011	< 0.001	0.0019	< 0.0000055	< 0.000005	0.000010
Californium-252	B	12	< 0.0011	< 0.001	0.0040	< 0.0000055	< 0.000005	0.000020
Curium-244 and/or	A	12	-	-	< 0.001	-	-	< 0.000014
Californium-249	B	12	-	-	< 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 µg/l can be obtained by multiplying the concentration given by 1.48×10^9. The average concentration is 3.0 µg/l.

TABLE 14

Radionuclides in Illinois River Water, 1978

(Concentrations in 10^{-9} μCi/ml)

Location	Date Collected	Alpha*	Beta*	Hydrogen-3	Uranium** (natural)	Neptunium-237	Plutonium-239
McKinley Woods State Park	June 20	0.87	10	< 200	1.0	< 0.001	0.00027
Below Dresden Power Station	June 20	0.99	6.8	< 200	1.3	< 0.001	0.00058
Morris	June 20	4.4	10	< 200	1.4	–	–
Starved Rock State Park	June 20	0.88	8.2	244	1.1	–	–
McKinley Woods State Park	October 19	0.80	12	254	0.74	< 0.001	0.00129
Below Dresden Power Station	October 19	0.60	9.2	252	1.1	< 0.001	0.00010
Morris	October 19	2.1	9.6	250	0.97	–	–
Starved Rock State Park	October 19	1.4	10	204	1.3	–	–

*Nonvolatile activity.

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

for samples collected at large distances from nuclear installations. This latter comparison is useful in determining if the soil activity near Argonne is normal. For this purpose, the site selection criteria and sample preparation techniques used by the DOE Environmental Measurements Laboratory[8] 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 five cores totaling 432 cm^2 in area by 5 cm deep. Through 1976, samples had been collected down to 30 cm to measure total plutonium deposition and as a result of five years of sample collection and analysis at this depth, the total plutonium deposited 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 m^2 area in the immediate vicinity of a soil sample. A grab sample technique was used to obtain bottom sediment. After drying, grinding, and mixing, 100 g portions of soil, bottom sediment, and grass were analyzed by the same method described in Section III.A. for air-filter residues. Results are given in terms of oven-dried soil, bottom sediment, or grass.

Comparison of the perimeter and off-site results in Table 15 shows that the same general range of concentrations exists in all areas for both plutonium isotopes, and it may be concluded that the plutonium in the perimeter samples resulted primarily from fallout of debris from nuclear detonations. Concentrations in the first 5 cm of soil are two to three times larger than in the first 30 cm; the deposition per unit area is two to three times smaller. That is, about one-third of the plutonium is in the top 5 cm.

Composite monthly precipitation samples were analyzed for plutonium-239. The results are given in Table 16 along with results since 1973 for comparison. The total 1978 deposition by precipitation was about twice that of 1977, a reflection of the increased fallout from atmospheric nuclear tests, and was equivalent to 0.9% of the total plutonium previously deposited, which is reported to be 2.2 x 10^{-3} $\mu Ci/m^2$.[9] The data in Table 16 is illustrated in Figure 5. The arrows at the bottom of the figure indicate the approximate

TABLE 15

Plutonium Content of Soil, 1978

Date Collected	Location	Plutonium-238		Plutonium-239		$^{238}Pu/^{239}Pu$
		10^{-9} µCi/g	10^{-3} µCi/m^2	10^{-9} µCi/g	10^{-3} µCi/m^2	
	Perimeter*					
June 23	7M	0.7 ± 0.2	0.04 ± 0.01	15.1 ± 0.9	0.83 ± 0.05	0.045
June 23	10N	0.5 ± 0.2	0.03 ± 0.01	5.3 ± 0.4	0.33 ± 0.03	0.101
June 23	14L	1.4 ± 0.3	0.06 ± 0.01	26.1 ± 1.2	1.09 ± 0.05	0.055
June 23	15H	0.9 ± 0.2	0.04 ± 0.01	22.7 ± 1.1	0.98 ± 0.05	0.040
June 23	12C	1.5 ± 0.3	0.07 ± 0.01	21.6 ± 1.1	1.01 ± 0.05	0.069
October 17	4EF	0.5 ± 0.1	0.02 ± 0.01	19.7 ± 0.8	0.93 ± 0.04	0.024
October 17	7EF	1.0 ± 0.2	0.04 ± 0.01	24.8 ± 1.2	1.02 ± 0.05	0.038
October 17	10P	0.4 ± 0.2	0.01 ± 0.01	19.8 ± 1.0	0.77 ± 0.04	0.018
October 17	14N	0.9 ± 0.2	0.04 ± 0.01	27.6 ± 1.3	1.23 ± 0.06	0.033
October 17	14E	0.9 ± 0.2	0.05 ± 0.01	22.2 ± 1.1	1.25 ± 0.06	0.042
	Average	0.9 ± 0.2	0.04 ± 0.01	20.5 ± 4.1	0.94 ± 0.17	0.046 ± 0.015
	Off-Site					
June 16	Naperville, IL	1.0 ± 0.2	0.05 ± 0.01	27.9 ± 1.4	1.56 ± 0.08	0.034
June 20	Channahon, IL	1.2 ± 0.2	0.08 ± 0.01	22.2 ± 1.1	1.34 ± 0.06	0.056
June 20	Morris, IL	1.2 ± 0.3	0.07 ± 0.02	24.8 ± 1.3	1.41 ± 0.07	0.047
June 20	Starved Rock State Park, IL	0.8 ± 0.2	0.05 ± 0.02	18.4 ± 1.0	1.17 ± 0.06	0.043
June 21	Willow Springs, IL	0.4 ± 0.2	0.02 ± 0.01	15.5 ± 0.8	0.73 ± 0.04	0.027
October 19	McKinley Woods State Park, IL	1.1 ± 0.2	0.06 ± 0.01	26.7 ± 1.3	1.48 ± 0.07	0.039
October 19	Dresden Lock and Dam, IL	1.2 ± 0.2	0.06 ± 0.01	23.8 ± 1.1	1.22 ± 0.06	0.051
October 20	Romeoville, IL	2.3 ± 0.4	0.07 ± 0.01	51.2 ± 2.4	1.55 ± 0.07	0.045
October 20	Lemont, IL	0.3 ± 0.3	0.02 ± 0.01	11.0 ± 0.8	0.58 ± 0.04	0.028
October 20	McGinnis Slough, IL	0.6 ± 0.2	0.03 ± 0.01	21.9 ± 1.1	1.15 ± 0.06	0.028
October 20	Saganashkee Slough, IL	0.2 ± 0.2	0.01 ± 0.01	15.6 ± 0.9	0.73 ± 0.04	0.016
	Average	0.9 ± 0.4	0.05 ± 0.01	23.5 ± 6.4	1.17 ± 0.21	0.038 ± 0.007

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

TABLE 16

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

Month	1973	1974	1975	1976	1977	1978
January	–	0.60 ± 0.10	1.1 ± 0.18	0.25 ± 0.06	0.17 ± 0.08	0.37 ± 0.11
February	0.14 ± 0.04	1.1 ± 0.16	1.7 ± 0.19	0.38 ± 0.09	0.15 ± 0.07	0.12 ± 0.09
March	0.27 ± 0.06	2.0 ± 0.22	1.5 ± 0.17	0.58 ± 0.09	0.74 ± 0.15	4.7 ± 0.37
April	0.40 ± 0.07	5.2 ± 0.37	4.0 ± 0.43	0.21 ± 0.07	1.0 ± 0.17	3.3 ± 0.30
May	0.50 ± 0.06	4.6 ± 0.31	1.8 ± 0.18	0.58 ± 0.11	1.8 ± 0.21	4.7 ± 0.42
June	0.16 ± 0.04	3.5 ± 0.26	1.9 ± 0.18	0.65 ± 0.12	1.2 ± 0.18	3.0 ± 0.32
July	0.32 ± 0.10	5.6 ± 0.37	0.48 ± 0.10	0.26 ± 0.07	0.71 ± 0.15	1.7 ± 0.24
August	0.34 ± 0.14	1.0 ± 0.19	0.38 ± 0.09	0.15 ± 0.06	1.6 ± 0.21	0.54 ± 0.17
September	0.27 ± 0.17	0.25 ± 0.11	0.10 ± 0.06	0.06 ± 0.05	1.5 ± 0.20	0.69 ± 0.18
October	0.30 ± 0.12	0.45 ± 0.10	0.12 ± 0.08	0.30 ± 0.10	0.85 ± 0.15	0.34 ± 0.14
November	0.22 ± 0.10	0.73 ± 0.12	0.08 ± 0.07	0.05 ± 0.04	0.72 ± 0.14	0.74 ± 0.16
December	0.46 ± 0.12	0.71 ± 0.12	0.21 ± 0.08	0.12 ± 0.07	0.43 ± 0.10	0.40 ± 0.14
Average Monthly Deposition	0.31 ± 0.07	2.1 ± 1.2	1.1 ± 0.7	0.30 ± 0.13	0.91 ± 0.31	1.7 ± 1.0
Annual Deposition	3.38	25.74	13.37	3.59	10.87	20.60
Percent Added to Existing	0.2	1.2	0.6	0.2	0.5	0.9

dates of the Chinese atmospheric nuclear tests. Comparison of the results in Figure 5 with the plutonium air concentrations for the past six years in Figure 4 shows excellent agreement. A log-normal distribution plot of the monthly plutonium deposition from 1973-1977 produced a straight line best fit, indicating a single plutonium source, evidently stratospheric fallout.

Thorium, uranium, and several gamma-ray emitting fission products in soil were also measured, and the concentrations in the perimeter and off-site samples (Table 17) were similar. These are expected levels of the naturally-occurring thorium and uranium activities and fission products which are attributed to fallout. In terms of mass, the average thorium concentrations were 3.4 µg/g and 3.2 µg/g at the perimeter and off the site, respectively, while the uranium concentrations were 4.2 µg/g at the perimeter and 4.2 µg/g off-site.

The results of radioactivity measurements in grass are given in Tables 18 and 19. The grass samples were washed before analysis to remove surface soil, which contains considerably more radioactivity per gram than grass. If the grass is analyzed without washing, results are more variable and difficult to compare between locations. The perimeter and off-site plutonium concentrations are similar to each other and to results of previous years. All the results, perimeter and off-site, were within the range expected and observed from fallout. In terms of deposition, the plutonium-239 concentration was a factor of about 10^4 less than in soil from the same location. Thorium, uranium, and the detectable gamma-ray emitting nuclides were similar at the perimeter and off-site, which indicates that their concentrations are due to naturally-occurring nuclides or fallout.

Results of analyses of bottom sediment samples for thorium, uranium, plutonium, and gamma-ray emitters are given in Table 20. Plutonium results vary widely between locations and are strongly dependent on the retentiveness of the bottom material. Concentrations were similar on and off the site. The uranium and thorium concentrations were normal and similar to those found in soil. In terms of mass, the uranium concentrations were 4.2 µg/g at the perimeter and 2.4 µg/g off the site, while the thorium concentrations were 2.6 µg/g and 2.5 µg/g at the perimeter and off-site, respectively.
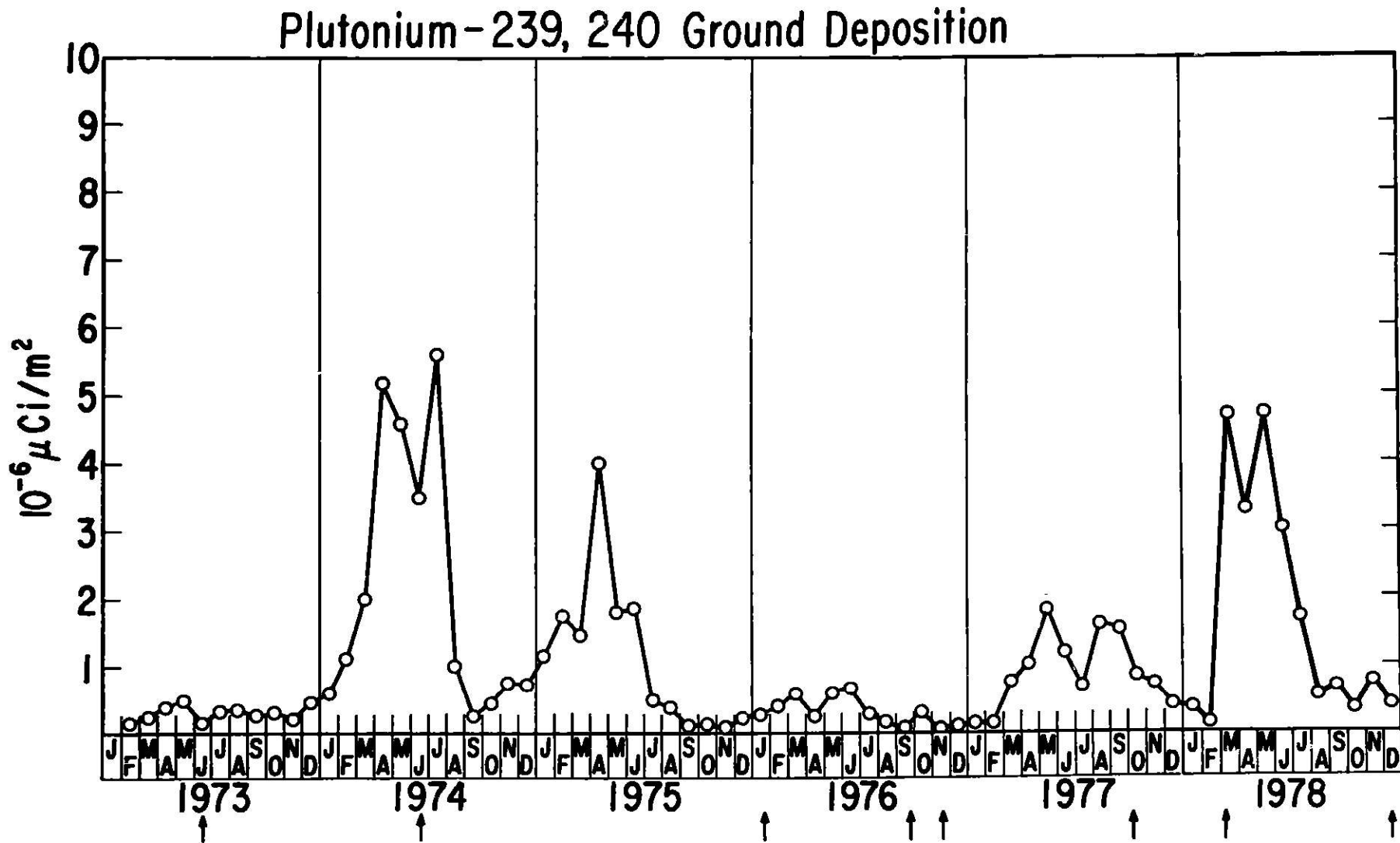
Fig. 5. Plutonium-239,240 Surface Deposition, 1973-78

TABLE 17

Radionuclides in Soil, 1978

(Concentrations in 10^{-6} µCi/g)

Date Collected	Location	Antimony-125	Cesium-137	Cerium-144	Thorium-228	Thorium-230	Thorium-232	Uranium (natural)
	Perimeter*							
June 23	7M	0.1 ± 0.1	0.8 ± 0.2	0.4 ± 0.1	0.28 ± 0.03	0.43 ± 0.03	0.26 ± 0.07	1.0 ± 0.1
June 23	10N	0.1 ± 0.1	0.3 ± 0.1	0.2 ± 0.1	0.55 ± 0.04	0.74 ± 0.04	0.60 ± 0.04	2.2 ± 0.2
June 23	14L	< 0.1	1.3 ± 0.3	0.3 ± 0.1	0.37 ± 0.03	0.46 ± 0.03	0.40 ± 0.03	1.3 ± 0.1
June 23	15H	< 0.1	1.2 ± 0.3	0.5 ± 0.2	0.39 ± 0.03	0.45 ± 0.03	0.38 ± 0.03	1.5 ± 0.1
June 23	12C	0.1 ± 0.1	1.2 ± 0.3	0.4 ± 0.1	0.39 ± 0.03	0.55 ± 0.03	0.38 ± 0.03	1.7 ± 0.1
October 17	4EF	0.1 ± 0.1	3.0 ± 0.7	0.1 ± 0.1	0.19 ± 0.02	0.22 ± 0.02	0.18 ± 0.02	1.2 ± 0.1
October 17	7EF	0.1 ± 0.1	1.3 ± 0.4	0.3 ± 0.1	0.30 ± 0.04	0.41 ± 0.04	0.36 ± 0.04	1.0 ± 0.1
October 17	10P	< 0.1	1.1 ± 0.3	0.3 ± 0.1	0.29 ± 0.03	0.46 ± 0.04	0.40 ± 0.04	1.2 ± 0.3
October 17	14N	< 0.1	1.5 ± 0.4	< 0.1	0.42 ± 0.04	0.58 ± 0.05	0.48 ± 0.04	1.3 ± 0.2
October 17	14E	0.1 ± 0.1	1.0 ± 0.3	0.4 ± 0.2	0.38 ± 0.02	0.54 ± 0.02	0.40 ± 0.02	1.5 ± 0.2
	Average	0.1 ± 0.02	1.3 ± 0.4	0.3 ± 0.1	0.36 ± 0.06	0.48 ± 0.08	0.38 ± 0.07	1.4 ± 0.2
	Off-Site							
June 16	Naperville, IL	< 0.1	1.2 ± 0.3	0.2 ± 0.1	0.44 ± 0.03	0.49 ± 0.03	0.53 ± 0.03	1.6 ± 0.2
June 20	Channahon, IL	< 0.1	1.1 ± 0.3	0.4 ± 0.1	0.35 ± 0.02	0.44 ± 0.02	0.36 ± 0.02	1.5 ± 0.1
June 20	Morris, IL	0.1 ± 0.1	1.2 ± 0.3	0.2 ± 0.1	0.27 ± 0.03	0.39 ± 0.04	0.27 ± 0.03	1.2 ± 0.2
June 20	Starved Rock State Park, IL	< 0.1	0.9 ± 0.3	0.2 ± 0.1	0.20 ± 0.02	0.22 ± 0.02	0.19 ± 0.02	0.6 ± 0.1
June 21	Willow Springs, IL	0.2 ± 0.1	0.9 ± 0.3	0.4 ± 0.1	0.31 ± 0.03	0.48 ± 0.03	0.31 ± 0.03	1.4 ± 0.1
October 19	McKinley Woods State Park, IL	< 0.1	1.3 ± 0.4	0.3 ± 0.1	0.36 ± 0.05	0.52 ± 0.06	0.35 ± 0.05	1.4 ± 0.3
October 19	Dresden Lock and Dam, IL	0.1 ± 0.1	1.6 ± 0.5	0.4 ± 0.2	–	–	–	1.3 ± 0.1
October 20	Romeoville, IL	0.1 ± 0.1	2.9 ± 0.7	0.5 ± 0.2	0.36 ± 0.04	0.61 ± 0.05	0.42 ± 0.04	2.2 ± 0.3
October 20	Lemont, IL	< 0.1	0.8 ± 0.3	0.2 ± 0.1	0.34 ± 0.04	0.48 ± 0.04	0.37 ± 0.04	1.1 ± 0.1
October 20	McGinnis Slough, IL	< 0.1	1.3 ± 0.4	0.3 ± 0.1	0.49 ± 0.05	0.57 ± 0.05	0.37 ± 0.04	1.6 ± 0.1
October 20	Saganashkee Slough, IL	< 0.1	1.1 ± 0.3	< 0.1	0.33 ± 0.04	0.59 ± 0.05	0.37 ± 0.04	1.8 ± 0.1
	Average	< 0.1	1.3 ± 0.3	0.3 ± 0.1	0.34 ± 0.05	0.48 ± 0.07	0.36 ± 0.06	1.4 ± 0.2

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

TABLE 18

Plutonium Content of Grass Samples, 1978

Date Collected	Location	Plutonium-238		Plutonium-239	
		10^{-9} µCi/g	10^{-6} µCi/m^2	10^{-9} µCi/g	10^{-6} µCi/m^2
	Perimeter*				
June 23	7M	0.02 ± 0.03	0.005 ± 0.008	1.59 ± 0.24	0.445 ± 0.067
June 23	10N	0.12 ± 0.27	0.031 ± 0.070	1.55 ± 0.25	0.401 ± 0.065
June 23	14L	0.19 ± 0.28	0.044 ± 0.064	0.93 ± 0.20	0.214 ± 0.046
June 23	15H	0.25 ± 0.47	0.080 ± 0.154	0.90 ± 0.31	0.296 ± 0.101
June 23	12C	0.10 ± 0.22	0.026 ± 0.055	1.18 ± 0.22	0.297 ± 0.055
October 17	4EF	0.63 ± 0.17	0.062 ± 0.017	2.96 ± 0.35	0.290 ± 0.034
October 17	7EF	0.75 ± 0.23	0.266 ± 0.082	2.52 ± 0.34	0.895 ± 0.121
October 17	10P	0.36 ± 0.16	0.094 ± 0.042	2.24 ± 0.32	0.587 ± 0.084
October 17	14N	1.22 ± 0.46	0.177 ± 0.067	2.42 ± 0.59	0.352 ± 0.086
October 17	14E	0.29 ± 0.49	0.065 ± 0.110	2.91 ± 0.44	0.649 ± 0.098
	Average	0.39 ± 0.24	0.085 ± 0.050	1.92 ± 0.50	0.443 ± 0.133
	Off-Site				
June 16	Naperville, IL	0.40 ± 0.24	0.038 ± 0.023	1.73 ± 0.23	0.165 ± 0.022
June 20	Channahon, IL	0.04 ± 0.20	0.004 ± 0.023	1.69 ± 0.21	0.192 ± 0.024
June 20	Morris, IL	0.13 ± 0.24	0.061 ± 0.113	0.85 ± 0.16	0.401 ± 0.075
June 20	Starved Rock State Park, IL	0.19 ± 0.25	0.046 ± 0.061	1.53 ± 0.21	0.373 ± 0.051
June 21	Willow Springs, IL	0.19 ± 0.26	0.029 ± 0.040	1.02 ± 0.21	0.158 ± 0.033
October 19	McKinley Woods State Park, IL	0.10 ± 0.08	0.007 ± 0.005	2.33 ± 0.33	0.163 ± 0.023
October 19	Dresden Lock and Dam, IL	1.15 ± 0.21	0.158 ± 0.029	3.57 ± 0.37	0.490 ± 0.051
October 20	Romeoville, IL	0.25 ± 0.47	0.062 ± 0.117	1.84 ± 0.34	0.456 ± 0.084
October 20	Lemont, IL	0.99 ± 0.14	0.131 ± 0.018	1.19 ± 0.16	0.157 ± 0.021
October 20	McGinnis Slough, IL	0.82 ± 0.12	0.172 ± 0.025	1.32 ± 0.15	0.277 ± 0.032
October 20	Saganashkee Slough, IL	0.78 ± 0.13	0.177 ± 0.030	1.76 ± 0.18	0.400 ± 0.041
	Average	0.46 ± 0.24	0.080 ± 0.040	1.71 ± 0.45	0.294 ± 0.080

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

TABLE 19

Radionuclides in Grass Samples, 1978

| Date Collected | Location | Concentrations in 10^{-6} µCi/g | | Concentrations in 10^{-9} µCi/g | | | Uranium (natural) |
		Cesium-137	Cerium-144	Thorium-228	Thorium-230	Thorium-232	
	Perimeter*						
June 23	7M	0.1 ± 0.1	1.0 ± 0.3	1.5 ± 0.4	1.7 ± 0.3	1.2 ± 0.2	16 ± 3
June 23	10N	0.1 ± 0.1	1.1 ± 0.3	3.2 ± 0.4	1.3 ± 0.2	0.9 ± 0.2	33 ± 6
June 23	14L	< 0.1	0.3 ± 0.1	3.0 ± 0.4	1.6 ± 0.3	0.9 ± 0.2	50 ± 4
June 23	15H	< 0.1	0.5 ± 0.2	2.1 ± 0.5	2.4 ± 0.5	1.2 ± 0.3	19 ± 3
June 23	12C	< 0.1	0.7 ± 0.2	1.3 ± 0.4	0.7 ± 0.2	0.9 ± 0.2	18 ± 3
October 17	4EF	< 0.1	0.7 ± 0.2	6.2 ± 0.5	8.8 ± 0.6	6.3 ± 0.5	40 ± 3
October 17	7EF	0.1 ± 0.1	1.2 ± 0.3	3.0 ± 0.4	3.7 ± 0.4	2.9 ± 0.3	35 ± 4
October 17	10P	0.1 ± 0.1	0.5 ± 0.2	-	-	-	50 ± 6
October 17	14N	0.1 ± 0.1	0.7 ± 0.2	5.2 ± 0.7	6.2 ± 0.5	4.9 ± 0.4	62 ± 4
October 17	14E	0.1 ± 0.1	0.8 ± 0.3	-	-	-	96 ± 9
	Average	0.1 ± 0.02	0.8 ± 0.2	3.2 ± 1.2	3.3 ± 2.0	2.4 ± 1.5	42 ± 15
	Off-Site						
June 16	Naperville, IL	0.1 ± 0.1	0.7 ± 0.2	4.9 ± 0.6	7.7 ± 0.7	5.2 ± 0.5	51 ± 4
June 20	Channahon, IL	0.1 ± 0.1	0.5 ± 0.2	2.1 ± 0.3	1.9 ± 0.3	1.6 ± 0.2	32 ± 3
June 20	Morris, IL	0.1 ± 0.1	0.4 ± 0.1	1.9 ± 0.4	1.7 ± 0.3	1.5 ± 0.2	28 ± 3
June 20	Starved Rock State Park, IL	0.1 ± 0.1	0.9 ± 0.3	3.9 ± 0.5	1.2 ± 0.2	2.0 ± 0.3	29 ± 4
June 21	Willow Springs, IL	< 0.1	0.7 ± 0.2	2.0 ± 0.4	3.1 ± 0.4	1.6 ± 0.2	26 ± 3
October 19	McKinley Woods State Park, IL	0.1 ± 0.1	0.4 ± 0.1	2.6 ± 0.2	4.2 ± 0.4	3.1 ± 0.3	123 ± 15
October 19	Dresden Lock & Dam, IL	0.1 ± 0.1	1.7 ± 0.4	1.6 ± 0.1	2.1 ± 0.2	1.9 ± 0.2	64 ± 8
October 20	Romeoville, IL	0.1 ± 0.1	0.5 ± 0.2	-	-	-	136 ± 12
October 20	Lemont, IL	< 0.1	0.2 ± 0.1	1.2 ± 0.1	1.2 ± 0.1	0.9 ± 0.1	90 ± 8
October 20	McGinnis Slough, IL	0.1 ± 0.1	0.3 ± 0.1	8.1 ± 0.7	7.0 ± 0.6	4.5 ± 0.4	54 ± 14
October 20	Saganashkee Slough, IL	0.1 ± 0.1	0.5 ± 0.2	10.1 ± 0.8	6.2 ± 0.6	5.1 ± 0.5	44 ± 6
	Average	0.1 ± 0.01	0.6 ± 0.2	3.8 ± 1.9	3.6 ± 1.6	2.7 ± 1.0	62 ± 23

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

TABLE 20

Radionuclides in Bottom Sediment, 1978

Date Collected	Location	Concentrations in 10^{-6} μCi/g						Concentrations in 10^{-9} μCi/g	
		Cesium-137	Cerium-144	Thorium-228	Thorium-230	Thorium-232	Uranium (natural)	Plutonium-238	Plutonium-239
	Perimeter*								
August 10	8J	0.1 ± 0.1	0.2 ± 0.1	0.31 ± 0.03	0.50 ± 0.04	0.29 ± 0.03	1.4 ± 0.1	1.2 ± 0.2	0.9 ± 0.2
	Off-Site								
June 16	DuPage River, Naperville, IL	0.2 ± 0.1	0.3 ± 0.1	0.68 ± 0.03	0.31 ± 0.02	0.64 ± 0.03	0.8 ± 0.1	1.6 ± 0.2	6.1 ± 0.5
June 16	DuPage River, West Chicago, IL	0.1 ± 0.1	0.2 ± 0.1	0.15 ± 0.01	0.15 ± 0.01	0.12 ± 0.01	0.8 ± 0.1	1.0 ± 0.2	1.9 ± 0.3
June 20	Illinois River, Morris, IL	< 0.1	< 0.1	0.11 ± 0.01	0.12 ± 0.01	0.08 ± 0.01	0.6 ± 0.1	0.6 ± 0.1	1.0 ± 0.2
October 19	Des Plaines River, McKinley Woods State Park, IL	0.1 ± 0.1	< 0.1	0.37 ± 0.04	0.35 ± 0.04	0.38 ± 0.04	0.7 ± 0.1	0.8 ± 0.2	0.6 ± 0.1
October 19	Illinois River, Starved Rock State Park, IL	0.1 ± 0.1	< 0.1	0.32 ± 0.03	0.67 ± 0.05	0.32 ± 0.03	0.4 ± 0.1	0.7 ± 0.2	0.5 ± 0.1
October 20	Prentiss Creek, Downers Grove, IL	< 0.1	< 0.1	0.29 ± 0.01	0.32 ± 0.01	0.28 ± 0.01	1.6 ± 0.1	1.0 ± 0.2	0.4 ± 0.2
October 20	Long Run Creek, Lemont, IL	0.2 ± 0.1	0.1 ± 0.1	0.09 ± 0.02	0.15 ± 0.02	0.13 ± 0.02	1.0 ± 0.1	0.1 ± 0.1	5.4 ± 0.5
	Average	0.1 ± 0.05	< 0.1	0.29 ± 0.16	0.30 ± 0.14	0.28 ± 0.15	0.8 ± 0.3	0.8 ± 0.3	2.3 ± 1.8

*The perimeter location is given in terms of the grid coordinates in Figure 1.

4. Foodstuffs

Raw milk was collected monthly from a local dairy farm south of Lemont and analyzed for several radioactive nuclides by methods similar to those used for water. Iodine-131 was analyzed with a detection limit of 1 x 10^{-10} µCi/ml by an ion-exchange separation followed by beta counting. Cesium-137 was analyzed with a detection limit of 5 x 10^{-10} µCi/ml by an ion-exchange separation followed by gamma-ray spectrometry. The other nuclides were analyzed by the same methods used for water and with the same detection limits. The results are given in Table 21. The average strontium-90 concentration increased by about 35% compared to 1977, while the average cesium-137 concentration was about the same as last year. 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 after the two atmospheric nuclear tests because the cows were being fed stored food during these times.

The concentrations given in Table 21 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 0.65% of the strontium-90 and 0.008% of the cesium-137 Concentration Guides.

Several samples of garden vegetables grown 8-16 km (5-10 miles) from Argonne were analyzed for plutonium, uranium, and thorium. The results are given in Table 22, and the concentrations are expressed in terms of air-dried weight. Compared to the results for grass, the plutonium-239 concentrations are about a hundred times lower, while the uranium concentrations are about ten times lower. The uranium results are in good agreement with analyses of the same types of vegetables measured during the past several years. The plutonium content, although extremely variable, covered about the same range of concentrations as in the last three years. Included with the plutonium results is a concentration factor (CF), which is defined as the ratio of plutonium concentration in the food divided by the average plutonium concentration in soil. As in the case of milk, the radioactivity is unrelated to Argonne operations, but the information is valuable as background data.

TABLE 21

Radionuclides in Milk, 1978

(Concentrations in 10^{-9} µCi/ml)

Date Collected	Hydrogen-3	Strontium-89	Strontium-90	Iodine-131	Cesium-137	Barium-140
January 4	< 200	< 2	2.9	< 0.1	3.9	< 2
February 1	< 200	< 2	5.5	< 0.1	3.9	< 2
March 1	< 200	< 2	2.6	< 0.1	3.0	< 2
April 5	213	< 2	4.1	< 0.1	3.0	< 2
May 3	< 200	< 2	3.6	< 0.1	3.3	< 2
June 7	205	< 2	3.8	< 0.1	4.6	< 2
July 5	< 200	< 2	6.1	< 0.1	4.7	< 2
August 2	282	< 2	7.8	< 0.1	3.9	< 2
September 6	326	< 2	5.2	< 0.1	3.2	< 2
October 4	< 200	< 2	4.7	< 0.1	2.1	< 2
November 1	< 200	< 2	3.3	< 0.1	3.2	< 2
December 6	< 200	< 2	2.0	< 0.1	3.9	< 2
Average	< 219	< 2	4.3 ± 1.0	< 0.1	3.6 ± 0.4	< 2

TABLE 22

Radionuclides in Garden Vegetables, 1978

| Food | Concentration (10^{-9} µCi/g) | | | | Plutonium-239 | |
	Thorium-228	Thorium-238	Thorium-232	Uranium (natural)	Concentration (10^{-12} µCi/g)	CF
Corn (kernels)	< 0.1	< 0.1	< 0.1	6 ± 1	16 ± 37	8×10^{-4}
Tomatoes	< 0.1	2.2 ± 0.5	1.2 ± 0.3	34 ± 6	3 ± 1	1×10^{-4}
Squash	1.4 ± 0.8	0.2 ± 0.4	0.6 ± 0.3	40 ± 6	6 ± 4	2×10^{-4}
Melon	< 0.1	< 0.1	0.4 ± 0.2	15 ± 4	10 ± 2	3×10^{-4}

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 three or four chips exposed in the same packet. The re-
sponse of the chips was calibrated with an NBS 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 the dose, if any, due
to Argonne operations at the closest uncontrolled approaches to the Laboratory,
and at several locations on the site. The latter were chosen for two 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 ob-
tained 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 23 and 24, and the site boundary
and on-site readings are also shown in Figure 6. Measurements were made in
six successive exposure periods that varied in length from 48 to 77 days,
and in total covered the period from January 5, 1978, to January 9, 1979.
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 error given for an average is the 95% confidence limit calcu-
lated from the standard error.

The off-site results averaged 93 mrem/yr. The standard error of the
mean of all 30 individual results was 1.5 mrem/yr. (The standard deviation
of the average of the five separate locations was 4.7 mrem/yr.) From 1972
to 1978, the corresponding averages and standard errors varied from 90 ± 2
mrem/yr (1976) to 105 ± 2 mrem/yr (1972). The six-year average was 97 ± 6
mrem/yr. Thus, the 1978 average is within the range found previously. The
variations from year-to-year have been consistent at each location. The
annual average at Downers Grove, Lockport, and Lombard has 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, 6 to 11 mrem/yr above, the overall average.

If the off-site locations are an accurate sample of the radiation back-
ground in the area, then annual averages at the site boundary ranging from

TABLE 23

Environmental Penetrating Radiation at Off-Site Locations, 1978

Location	Dose Rate (mrem/year)						
	1/5-3/8	3/8-5/24	Period of Measurement 5/24-7/11	7/11-9/12	9/12-11/9	11/9-1/9	Average
Downers Grove	81	99	92	97	99	92	94 ± 7
Lockport	80	90	96	93	109	97	94 ± 10
Lombard	82	91	91	94	100	92	92 ± 6
Oak Lawn	80	88	89	87	91	89	87 ± 4
Oakbrook	83	103	97	107	105	105	100 ± 9
Average	81 ± 2	94 ± 7	93 ± 4	96 ± 8	101 ± 8	95 ± 7	93 ± 5

TABLE 24

Environmental Penetrating Radiation at ANL, 1978

| Location	Dose Rate (mrem/year)						
	Period of Measurement						
	1/5-3/3	3/8-5/24	5/24-7/11	7/11-9/12	9/12-11/9	11/9-1/9	Average
14L - Boundary	69	86	82	83	89	82	82 ± 7
14I - Boundary	142	143	145	144	157	163	149 ± 9
14G - Boundary	76	90	92	88	93	88	88 ± 6
13D - Boundary	53	67	67	62	70	62	64 ± 6
9/10 EF - Boundary	77	101	99	96	98	91	93 ± 5
8H - Boundary	86	97	91	89	98	98	93 ± 5
8H - Center, St. Patrick's Cemetery	98	116	103	106	-	107	106 ± 8
7I - Boundary	197	228	243	295	362	374	281 ± 73
6I - 200 m N of Quarry Road	88	105	95	96	105	97	98 ± 7
9L - Boundary	68	88	80	78	85	78	80 ± 7
9H - 50 m SE of CP-5	2300	2810	2520	2510	2540	2610	2560 ± 165
8H - 65 m S of 316	151	120	109	110	114	135	124 ± 17
9H - 23 m E of 316	363	177	150	117	97	277	198 ± 103
9I - 45 m NE of 350 210 m NE of 316	112	124	101	107	105	96	108 ± 10
8H - 200 m NW of Waste Storage Area (Heliport)	113	133	135	141	155	141	136 ± 14
7I - Center, Waste Storage Area	2630	5650	3140	4150	5600	6840	4750 ± 1630
10/11K - Lodging Facilities	70	87	76	78	78	76	78 ± 6
9J - Between ZGS Condenser and 386	79	119	283	321	420	384	260 ± 140
13I/J - 75 m E of 202	-	-	302	315	323	358	325 ± 33
13J - 140 m NE of 202	838	777	718	717	769	800	770 ± 47
12M - 30 m W of 55	60	75	70	68	75	73	70 ± 6
13I - 60 m SE of 202	-	-	142	148	152	157	150 ± 9
12I - 160 m S of 202	-	-	77	82	82	83	81 ± 4
13I - 150 m W of 202	-	-	-	146	152	158	152 ± 11
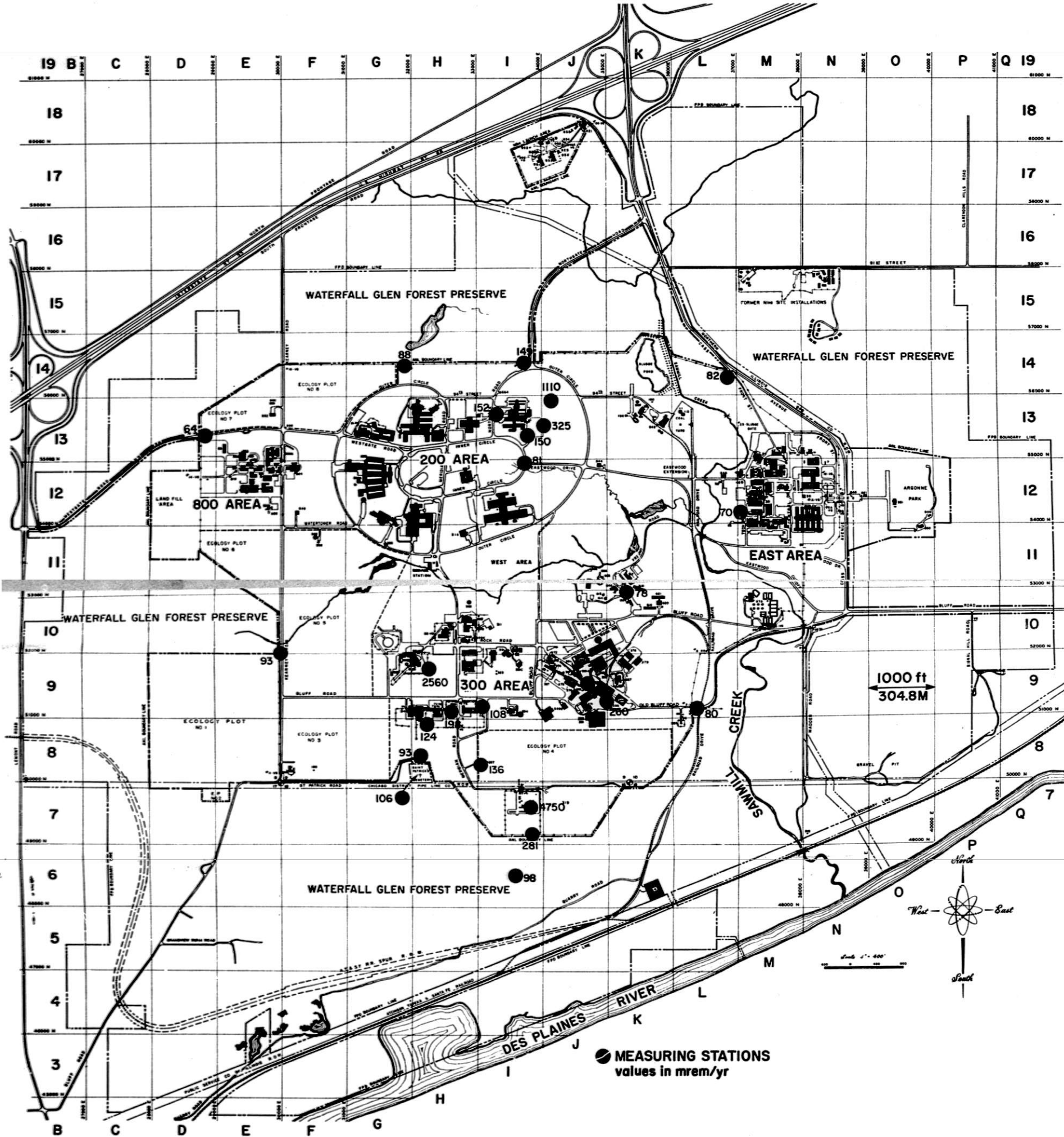
Fig. 6. Penetrating Radiation Measurements at the ANL Site, 1978

82 to 104 (93 ± 11) 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 30 individual off-site results is useful. This value is 8.0 mrem/yr, so that individual results in the range of 93 ± 16 may be considered normal with a 95% probability.

At two locations, 7I at the south boundary of the site, and at 14I north of Building 202, 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 190 mrem/yr; in previous years this value has ranged from 440 in 1976 to 114 in 1977. At 14I the average dose rate was about 55 mrem/yr above normal, about half of the 1977 rate. This dose is attributed to the use of cobalt-60 irradiation sources in Building 202. A change in the orientation of the sources in July, 1977, accounts for the decrease in the dose rates at the boundary observed since the change.

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 (93 mrem/yr) plus the standard deviation in the average at the 95% confidence limit (11 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 the results 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. Application of the first two criteria and the difficulties in interpreting small differences from natural background by TLD measurements are illustrated below.

The dose rate at 8H is of interest, since it lies in St. Patrick's Cemetery, which was in use before Argonne was constructed, and which is open

to visitors. The dose at the center of the Cemetery was 13 mrem/yr above the off-site average but only 2 mrem/yr above the upper limit (104 mrem/yr) of the normal range. The result for one individual period was above the 95% upper limit for such measurements, 109 mrem/yr. Based on this type of comparison, the excess dose at 8H was about 5 to 10 mrem/yr. Similar but somewhat higher 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, direct radiation from a tandem dynamitron and low-power reactors in Building 316 (location 9GH), and argon-41 from the CP-5 reactor at location 9H. The contribution from CP-5 and 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. In addition, the dose at the Cemetery did not decrease when the dose close to Building 316 decreased after the reactor in the building was provided with additional shielding. Since the dose at the center of the Cemetery averaged 13 mrem/yr above that at the 8H boundary station, 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 Water Borne 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 air and water are available, conversion of concentration to dose was based on the ratio of environmental concentrations to the Concentration Guides given in Table 25. 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, tritiated water vapor, and iodine-131 released from reactor 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 inhalation. Although Sawmill Creek water is not used for drinking purposes, the dose that would be received by

TABLE 25

Concentration-to-Dose Conversion Factors

Nuclide	Medium	Concentration* (μCi/ml)	Dose* (rem)	Critical Organ
Americium-241	Water	4×10^{-6}	1.5	Kidney
	Water	5×10^{-6}	3	Bone
Argon-41	Air	4×10^{-8}	0.5	Whole Body
Californium-249	Water	4×10^{-6}	3	Bone
Californium-252	Water	7×10^{-6}	1.5	GI (LLI)
	Water	2×10^{-5}	3	Bone
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
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 Radiation Protection Standards (RPS) specified in DOE Manual Chapter 0524 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 DOEM 0524, the concentrations used were one-tenth of the 168 hour occupational values specified by the ICRP.[11]

an individual ingesting water at the concentrations found in the Creek is calculated. A minor exposure route is from water to man by ingestion of Illinois River water 267 km (140 miles) 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 hydrogen-3 (in the form of tritiated water) from the CP-5 reactor represent the major portion of the gaseous radioactive effluent re- leased from the Laboratory. The concentrations and dose rates, as a function of distance from CP-5, were calculated for these two nuclides by a computer program based on an atmospheric dispersion model.[10] The following para- meters were used in the calculations:

a) release rates (measured in the CP-5 exhaust stack): argon-41, 1.38 Ci/MW-hr; hydrogen-3, 0.11 Ci/hr.

b) meteorological data: the wind velocity data is the 15-year average values shown in Figure 3.

c) the usual parameters for stack height, building, wake, plume, momentum, temperatures, etc.

The calculations were carried out to 80 km (50 miles). The argon-41 results for the first 4.8 km (3 miles) are given in Table 26. Doses were calculated for the mid-point of the annular interval. Thus, the dose for the 0-1.6 km (0-1 mile) average is the dose at 0.8 km (0.5 mile). The highest dose rates are in the N to E sectors. In this area, the full-time residents who receive the largest dose, 2.9 mrem/yr (outdoors), live 2.2 km (1.4 miles) from the reactor in the NE direction. This dose is less than 1% of the standard (500 mrem/yr) for individuals in uncontrolled areas. The dose varies greatly with distance in the first several kilometers. Thus, in the NE direction, individuals would receive 5.5 mrem/yr if they were outdoors throughout the year at 1.6 km and 1.1 mrem/yr if they were outdoors at 3.2 km.

The measurement technique for argon-41 is adequate in the vicinity of CP-5, but is not sufficiently sensitive to measure the concentration at the site boundary. However, an upper limit for the argon-41 dose at the site boundary can be estimated from the penetrating radiation dose measurements made with thermoluminescent dosimeters (TLD) and discussed in Section III.A.1. The measurements made south of the reactor and north of Building 202 are obscured by direct radiation from several gamma-ray sources, but in other

TABLE 26

Argon-41 Radiation Dose From CP-5 Reactor, 1978

(millirem/year)

	Distance			
Sector	1.5 km (0.93 mi)	0-1.6 km (0-1 mi)	1.6-3.2 km (1-2 mi)	3.2-4.8 km (2-3 mi)
N	5.1	12.4	2.3	1.0
NNE	5.7	14.4	2.6	1.2
NE	4.6	11.3	2.1	0.8
ENE	4.9	12.1	2.1	1.0
E	3.8	9.6	1.7	0.8
ESE	3.6	8.9	1.7	0.7
SE	3.5	8.6	1.6	0.7
SSE	3.0	7.2	1.4	0.7
S	3.1	7.9	1.4	0.7
SSW	3.6	8.9	1.7	0.7
SW	3.1	7.7	1.4	0.7
WSW	2.6	6.6	1.2	0.5
W	2.0	5.1	1.0	0.3
WNW	2.1	5.5	1.2	0.5
NW	2.6	6.4	1.2	0.5
NNW	3.1	7.7	1.4	0.7
Average	3.5	8.8	1.6	0.7

directions, including the predominant wind directions to the east and north-
east, the dose rates at the site boundary were in the normal range found
off-site. Increases in excess of two standard deviations of any single off-
site result (i.e., greater than 93 ± 16 mrem/yr) would have been recognized
as abnormal, and on this basis the dose from argon-41 at the site boundary
was less than about 16 mrem/yr. Thus, the calculated doses and those
measured by TLD agree within the ability of the TLD system to detect above-
normal doses.

The population data in Table 2 was used to calculate the population dose
from argon-41. The results are given in Table 27, together with the average
individual dose. For comparison, the table also gives the estimated natural
external radiation dose, which was calculated with the assumption that the
average off-site, outdoor radiation dose measured by TLD applies to the entire
area within an 80 km (50 miles) radius.

The dose rates due to hydrogen-3 from CP-5, calculated in the same manner
as for argon-41, are as follows. The maximum dose (outdoors) at 1.5 km (0.93
mile), in the NNE direction, is 0.029 mrem/yr. The maximum individual expo-
sure to full-time residents in the area occurs in the 1.6-3.2 km (1-2 miles)
annulus in the NNE direction, where the annual dose is calculated to be 0.018
mrem/yr. A summary of the results is given in Table 28. A comparison of the
doses calculated from the meteorological model with the measured data
from Table 11 is given in Table 29. In both cases, concentrations are con-
verted to dose as described earlier. The agreement is good in view of the
large number of variables and parameters involved in obtaining both the cal-
culated and measured values.

TABLE 27

Argon-41 Average Individual and Population Dose From CP-5 Reactor, 1978

Distance (km)	Population	Dose			
		Avg. mrem/year		Man-rem/year	
		Argon-41	Natural	Argon-41	Natural
0-1.6	0	-	-	-	-
1.6-3.2	5,559	1.6	93	8.9	517
3.2-4.8	17,692	0.7	93	12.4	1645
0-80	7,941,000	0.022	93	175.	7.39×10^5

TABLE 28

Hydrogen-3 Average Individual and Population
Dose From CP-5 Reactor, 1978

Distance (km)	Population	Avg. mrem/year	Man-rem/year
0-1.6	0	-	-
1.6-3.2	5,559	0.0071	0.039
3.2-4.8	17,692	0.0047	0.083
0-80	7,941,000	0.00037	2.94

TABLE 29

Comparison of Calculated and Measured Hydrogen-3 Dose Rates, 1978

Direction	Distance (km)	Calculated (mrem/yr)	Measured (mrem/yr)
NNE	1.5	0.026	0.035
ENE	1.9	0.016	0.020
SW	0.45	0.079	0.053

The iodine-131 released from CP-5 would result in an individual dose of 0.002 mrem/yr at 1.5 km (0.93 mile) in the NNE sector and a population dose of about 0.001 man-rem/yr in the 1.6-3.2 km (1-2 miles) annulus. These values are so small that the calculations for greater distances are not worthwhile.

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 30. In the case of strontium-90, a small fraction was contributed by Argonne and the remainder by fallout, but for the other nuclides, essentially all of the activity may be attributed to Argonne. 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 30

Radionuclide Concentrations and Dose Estimates
for Sawmill Creek Water, 1978

Nuclide	Conc. (net avg.) 10^{-9} µCi/ml	Dose mrem/year	Percent of Standard
Hydrogen-3	299	0.050	0.010
Strontium-90	0.18	1.8	0.060
Neptunium-237	0.0059	0.0059	0.0002
Plutonium-238	0.0012	0.0007	0.00002
Plutonium-239	0.016	0.0096	0.00032
Americium-241	0.0075	0.0028 (kidney) 0.0045 (bone)	0.0002 0.0002

As indicated in Table 12, occasional Creek samples (less than 10) contained traces of curium and californium 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 it should be noted that the method of averaging probably exaggerates the true concentration. These annual doses range from 2×10^{-4} to 5×10^{-5} mrem/yr for these nuclides.

The EPA[12] has recently established drinking water standards based on a dose of 4 mrem/yr. The standards in Table 25 are based on doses of 500 mrem/yr to 3000 mrem/yr, depending on the critical organ. Except for strontium-90, the percent of the EPA standard can be obtained by multiplying the value in Table 30 by the ratio 500/4 for hydrogen-3 and 3000 or 1500/4 (depending on the critical organ) for the actinide elements. Thus, for hydrogen-3, the value becomes 1.3%. For strontium-90, the EPA, using a different model than reference (11) developed a standard of 8×10^{-9} µCi/ml to produce a dose of 4 mrem/yr and therefore the concentration in Table 30 would give a 2.3% of that standard.

b. External Penetrating Radiation

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 probably 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 190 mrem/yr. Approximately 300 m (0.3 mile) south of the fence line (grid 6I), the measured dose had decreased to within the normal range, 98 mrem/yr. There are no individuals living in this area. The closest residents are about 1.6 km (1 mile) south of the fence line. At this distance the calculated dose rate due to the Waste Storage Area (based on exponential absorption of the radiation, a decrease in intensity with the square of the distance, and an increase in intensity with distance due to the buildup factor) is 0.005 mrem/yr, if the energy of the radiation was 0.66 MeV, and 0.023 mrem/yr, if the energy was 1.3 MeV. The energy spectrum of the radiation 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 upper limit of 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 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 56 mrem/yr, the nearest residents are 750 m (0.47 mile) to the north-northwest. The outdoor dose at that location (calculated as described above) was about 0.010 mrem/yr. In addition, the argon-41 dose here is about 2 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.[4] 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 doses to individuals living near the site will not exceed 0.06% of the 500 mrem/yr limit or 0.2% of the "suitable sample" limit. At the fence line where higher doses were measured, the land is wooded and unoccupied.

c. Summary

The total dose received by off-site residents was combined from the
separate pathways that contribute to this total: argon-41 immersion dose,
hydrogen-3 inhalation dose, and cobalt-60 external radiation dose. The
highest dose was about 3 mrem/yr (essentially all from argon-41) to indivi-
duals living NE of the site if they were outdoors at that location during the
entire year.

B. Chemical and Biological Pollutants

1. Water

The environmental water data contained in this section have been col-
lected in an effort to ascertain Argonne 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 Regu-
lations, Chapter 3.[13] Stream Quality standards appear in Part II and
Effluent Quality standards appear in Part IV of that document.

Several changes in methodology have been incorporated during this year.
Flameless atomic absorption techniques, using pyrolytic graphite, have been
adopted for copper, manganese, and nickel replacing flame techniques, and
have produced much lower detection limits. The tantalum ribbon flameless
technique for cadmium, lead, and silver has also been replaced by pyrolytic
graphite. An ion selective electrode method for ammonia nitrogen has been
substituted for the colorimetric technique and an oxygen sensitive electrode
is now used for dissolved oxygen. These changes were instituted in the last
half of the year and, where significant sensitivity changes resulted, are
indicated in the tables.

An x-ray fluorescence technique for arsenic has been devised and used for
both stream and effluent analyses and results are reported for the entire year.
Other analytical techniques used are as previously described.[14]

The results of the measurement of chemical constituents are expressed as
milligrams (mg) or micrograms (μg) per liter (1). Averages were calculated
as described in Section III.A. Yearly averages are reported with a (±) limit
value. This value is the standard error at the 95% confidence limit and 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. In some instances it appears that the measurements do represent a natural background concentration and the variation is representative of climatic conditions.

All of the results are compared to the appropriate State standards, which are listed in Table 31. Minimum detectable amounts are included for comparison. The detection limits for the 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.

As in the past, emphasis has been placed on three areas: Sawmill Creek, a tributary of the Des Plaines River, since this is the principal route for waste water leaving the Argonne site; the study of cooling tower blowdown effluents; and the control of mercury release in the effluent.

a. Effluent Water

The major discharge of waste water from Argonne operations is by way of the waste treatment plant. The water volume from this source was approximately 3 megaliters (800,000 gallons) per day and in 1978 was comprised of 44% sanitary waste water and 56% water from laboratory operations. The laboratory waste water is held in 0.26 megaliter (69,000 gallons) tanks and is checked for radioactivity before release. The release of these tanks occurs with some periodicity at a rate of about 4,200 liters (1,100 gallons) per minute. The sanitary waste water is released at a reasonably constant rate during the entire 24-hour period.

The performance of the sanitary waste treatment plant was monitored by the Reclamation Control Laboratory of the Plant Systems Division. This was done by analyzing twice weekly samples of the combined sanitary and laboratory waste systems for biochemical oxygen demand (B.O.D.), suspended solids, and ammonia nitrogen content. Each sample was a composite of eight separate grab samples taken approximately once per hour. All analyses were performed as outlined in Standard Methods.[15]

TABLE 31

Water Quality Standards and Detection Limits

(Concentrations in mg/l)

| Constituent | State Standard | | Analytical |
	Stream	Effluent	Detection Limit
Ammonia Nitrogen (as N)	1.5	2.5 (Apr.-Oct.)	0.1
		4.0 (Nov.-Mar.)	
Arsenic	1.0	0.25	0.01
Barium	5.0	2.0	0.1
Cadmium	0.05	0.15	0.0002
Chromium (hexavalent)	0.05	0.3	0.01
Chromium (trivalent)	1.00	1.00	-
Copper	0.02	1.0	0.005, 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	0.01, 0.001†
Mercury	0.0005	0.0005	0.0001
Nickel	1.0	1.0	0.05, 0.002†
pH	6.5-9.0	5.0-10.0	-
Silver	0.005	0.1	0.0002
Total Dissolved Solids	1000.	-	-
Zinc	1.0	1.0	0.01

†Flameless technique.

Release of chemical pollutants from the waste treatment plant was 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.

There are, in addition, seven effluent channels from cooling water operations which were monitored on a once-per-week schedule by grab sampling (see Figure 1). These channels carry blowdown water from various cooling towers and once-through cooling water systems. Additionally, the channel at 8J also carries some material from the treatment of Sanitary and Ship Canal water used for cooling at the ZGS Complex, and the channel at 11L contains some photographic wastes from the same source.

The results obtained for the sanitary waste parameters are shown in Table 32. The averages for January, February, and March exceeded the State of Illinois standard for ammonia nitrogen. The five-day biochemical oxygen demand and dissolved solid values were below the standards.

TABLE 32

Performance of Sanitary Waste Treatment Plant, 1978

Month	Concentrations (mg/l)		
	B.O.D.$_5$	Ammonia Nitrogen	Suspended Solids
January	1.32	4.68	3.75
February	6.6	6.85	3.00
March	6.2	7.53	4.40
April	1.3	1.6	2.22
May	4.80	2.48	1.50
June	1.58	0.26	1.50
July	0.20	0.14	3.71
August	1.53	0.57	3.50
September	1.45	0.75	3.33
October	1.08	0.93	2.50
November	1.35	1.91	7.50
December	2.15	2.59	1.60
State Standard	10	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	15

The results obtained for chemical constituents in the waste treatment plant effluent are shown in Table 33. All of the average concentrations were at or below the State standards. The average value for mercury was 98% of the State standard and 24% of the samples exceeded this value. Although the latter value is higher than last year (16%), the average was the same as 1977, and stringent controls have limited releases from known mercury sources. Occasional high releases from unknown sources have occurred. A single release increases subsequent samples by slow desorption of mercury from the system. For example, a concentration of 9.32 µg/l in July was followed by four subsequent samples which were above the limit and are believed to be related to the same release. The level of hexavalent chromium was essentially the same as in 1977, as were those for beryllium, cadmium, fluoride, iron, lead, nickel, and zinc. The levels of silver are somewhat higher, reflecting the diversion of photographic waste from 11L to this effluent. The levels of copper are higher in the latter six months of the year, but still below the State standard. Sources of copper are being investigated.

Results obtained for the cooling tower effluents are shown in Table 34. Except for 8J, average chromium concentrations at all stations were below the State standards. As a result of the replacement of chromium-containing corrosion inhibitors with non-chromate materials in some of the cooling towers, chromium levels at 14H and 14J were considerably lower than in 1977.

The channel at 11L was diverted in March, 1978. The photographic wastes were routed to the main effluent at 7M and the chromate wastes were routed to 8J. After March, chromium and silver levels at 11L were essentially not detectable and, during the latter part of the year, the channel was frequently dry. The average concentration at 8J was higher than in 1977, and reflected the diversion of additional chromate waste from 11L.

b. Sawmill Creek

Samples collected for evaluation of the effect of the sanitary wastes on stream quality were obtained once per week using specially constructed sampling bottles. The sampling bottles were designed to provide temperature measurement as well as to minimize changes in oxygen content during collection.

These samples were collected 15 m (50 feet) upstream of the Argonne outfall [7M (up)] and 60 m (200 feet) downstream of the outfall [7M (down)].

TABLE 33

Chemical Constituents in Effluent From ANL Treatment Plant, 1978

Constituent	No. of Samples	Concentration (mg/l or µg/l)			Percent of Standard (Avg.)	Percent Exceeding State Standard
		Avg.	Min.	Max.		
Arsenic	53	< 0.01	–	< 0.01	< 4	0
Barium	27	< 0.1	–	< 0.1	< 5	0
Beryllium*	12	0.046 ± 0.03	0.022	0.185	–	–
Cadmium*	53	0.90 ± 0.14	< 0.1	2.7	0.6	0
Chromium(VI)	69	0.02 ± 0.002	< 0.01	0.04	6.7	0
Chromium(III)	53	0.02 ± 0.002	< 0.01	0.05	2.0	0
Copper*	26	28 ± 7	14	86	2.8	0
	†27	63 ± 22	22	280	6.3	0
Fluoride	53	0.26 ± 0.02	0.16	0.52	1.7	0
Iron	53	0.23 ± 0.01	0.16	0.36	23	0
Lead*	53	3.3 ± 0.5	< 2	9.5	3.3	0
Manganese*	26	20.2 ± 3.5	< 10	46.0	2.0	0
	†27	11.9 ± 1.8	3.9	22.8	1.2	0
Mercury*	250	0.49 ± 0.3	< 0.1	9.3	98	24
Nickel*	26	< 50	–	< 50	< 5	0
	†27	14 ± 4	6.2	50	1.4	0
pH	250	–	6.49	7.57	–	–
Silver*	53	2.8 ± 0.7	0.5	18.1	2.8	0
Zinc	53	0.19 ± 0.03	0.05	0.51	19	0

*Concentrations in µg/l.

†Flameless technique.

TABLE 34

Cooling Tower Effluents, 1978

Constituent	Location[*]	No. of Samples	Concentration (mg/l or µg/l)			Percent of Standard (Avg.)	Percent Exceeding State Standard
			Avg.	Min.	Max.		
Chromium(VI)	8J	50	1.17 ± 29	< 0.01	3.39	390	70
Chromium(VI)	11L	39	0.15 ± 0.15	< 0.01	2.15	50	8
Chromium(III)	11L	24	0.17 ± 0.16	< 0.01	1.40	17	8
		†15	0.010 ± 0.008	0.001	0.06	10	0
Silver[**]	11L	39	22 ± 14	0.6	209	22	5
Chromium(VI)	12F	50	< 0.01	-	< 0.01	< 3	0
Chromium(VI)	12L	50	< 0.01	-	< 0.01	< 3	0
Chromium(VI)	14G	50	< 0.01	-	< 0.01	< 3	0
Chromium(VI)	14H	50	< 0.01	< 0.01	0.02	< 3	0
Chromium(VI)	14J	50	< 0.01	< 0.01	0.05	< 3	0

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

[**]Concentration in µg/l.

†Flameless technique.

Additionally, samples to be examined for fecal coliform were collected at least
five times per month at both 7M locations, but downstream the samples were col-
lected immediately in front of the outfall grating to minimize contamination
from Sawmill Creek. Once per month a sample was obtained as the water enters
the site (16K), which is downstream of the Marion Brook Treatment Plant.

Except for fecal coliform, the data from these studies are in Table 35.
The average level for ammonia nitrogen upstream is 1.7 times the State of
Illinois standard and individual samples exceeded this value 49% of the time.
The downstream sample averaged 1.5 times the State standard and exceeded this
value 42% of the time. All of the samples obtained downstream that exceeded
the standard were due to upstream contamination with one exception. One
sample, obtained on December 5, 1978, downstream, contained 23 mg/l ammonia
nitrogen. This represented a short-term high-level release. The dissolved
oxygen levels obtained during 1978 averaged 112% and 104% saturation for 7M
(up) and 7M (down) samples. The fact that values exceeded 100% saturation is
due primarily to photosynthetic activity. The total dissolved solids above
the outfall exceeded the State standard frequently, and increased the solids
content of the downstream samples. Similar results for these constituents
were obtained in 1977.

The fecal coliform standard requires that the monthly geometric mean not
exceed 200 organisms/100 ml and that no single sample exceed 400 organisms/
100 ml. Samples obtained above the 7M outfall exceeded the 200 organisms/100
ml standard in April, July, and November, and there were four instances when
the 400 organisms/100 ml standard was exceeded. The sample obtained at 16K
exceeded the 400 organisms/100 ml standard in March, April, and November.
Samples obtained at the Argonne outfall never exceeded the 200 organisms/100
ml monthly standard, but did exceed the 400 organisms/100 ml standard on one
occasion.

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 (Section III.A.2.). The results are
summarized in Table 36.

Individual samples for hexavalent chromium at 7M did not exceed the
State standard. This marks the first time that no results exceeded the State

TABLE 35

Sawmill Creek - Effect of Sanitary Waste, 1978

Constituent	Location*	No. of Samples	Concentration (mg/1)			Percent of Standard	Percent Exceeding State Standard
			Avg.	Min.	Max.		
Ammonia	7M (up)	50	2.6 ± 0.8	0.1	11.5	173	49
Nitrogen	7M (down)	50	2.3 ± 1	0.1	23	153	42
Dissolved	7M (up)	50	112 ± 8**	–	–	–	–
Oxygen	7M (down)	50	104 ± 6**	–	–	–	–
Total Dis-	7M (up)	50	1184 ± 102	516	1777	118	56
solved Solids	7M (down)	50	796 ± 76	508	1136	80	12

*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.

**Percent saturation.

TABLE 36

Chemical Constituents in Sawmill Creek, 1978

Constituent	Location[*]	No. of Samples	Concentration (mg/l or µg/l) Avg.	Min.	Max.	Percent of Standard (Avg.)	Percent Exceeding State Standard
Arsenic[**]	7M	55	< 10	-	< 10	< 10	0
Barium	7M	28	< 0.4	-	< 0.4	< 8	0
Beryllium[**]	7M	12	0.058 ± 0.02	< 0.02	0.096	-	-
Cadmium[**]	7M	55	1.4 ± 0.3	0.2	4.7	2.8	0
Chromium(VI)	7M	71	< 0.01	< 0.01	0.02	< 20	0
Chromium(III)	7M	71	0.01 ± 0.001	< 0.01	0.04	< 1	0
Copper[**]	7M	28	17.2 ± 3.5	8	50	86	18
	†27	28.4 ± 7.3	8	72	142	56
Cyanide	7M	52	< 0.02	< 0.02	0.03	< 80	2
Fluoride	7M	55	0.41 ± 0.02	0.26	0.56	29	0
Iron	7M	55	0.72 ± 0.20	0.09	3.90	72	13
Lead[**]	7M	55	7.0 ± 1.6	< 2	28	7	0
Manganese[**]	7M	28	113 ± 33	13	440	11.3	0
	†27	61 ± 20	7	219	6.1	0
Mercury[**]	7M	250	0.11 ± 0.08	< 0.1	0.73	22	1.2
Nickel[**]	7M	27	-	-	< 50	-	-
	†28	9.6 ± 7.2	3.3	31.5	9.6	0
pH	7M	250	-	7.3	8.9	-	-
Silver[**]	7M	55	2.5 ± 0.5	0.7	9.4	50	5.4
Zinc	7M	55	0.08 ± 0.01	0.02	0.22	8	0

[*]Location 7M is 60 m (200 ft) downstream from the waste-water outfall.

[**]Concentrations in µg/l.

†Flameless technique.

standard during the report period. This diminution is the direct result of substitution of non-chromate materials in cooling towers, and it is also reflected in Table 34. Individual samples exceeded the State standard for mercury 1.2% of the time, and the source is the Argonne outfall. This year's average is essentially the same as that obtained in 1977, but the number of samples in which the standard was exceeded was five times less this year. The State standard for silver was exceeded in 5.4% of the samples. All of the samples in excess were obtained in the period from January to March, 1978. At this time, the photographic wastes containing silver as well as the blow-down water containing chromium were diverted as previously described. The standard for silver has not been exceeded since March. The iron standard was exceeded in 13% of the samples. Much of this iron results from soil erosion during periods of heavy precipitation. As can be seen from the Argonne effluent levels in Table 36, the iron source must be in the stream area since effluent levels for iron were much lower. Individual samples for copper exceeded the State standards 18% of the time in the period January-June and 56% of the time in the period July-December. This is markedly higher than in previous years and a comparable increase can be seen in the effluent data (Table 33). However, at the average dilution ratio of 10 to 1, the concentrations of copper in the stream, due solely to the effluent levels, should have been much lower. The sampling program has been expanded to determine whether other sources of copper are adding to these increased stream levels. Levels for cyanide exceeded the State standard in 2% of the samples, and the source is not known. Levels for other constituents did not exceed the State standards in any individual sample. The annual average did not exceed the State standard for any of these constituents, except copper.

c. 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 37. Two of the samples had very high levels of suspended material, as evidenced by iron levels of 1.9 and 7.4 mg/l. In no case was there any indication that hexavalent chromium, mercury, or zinc levels were affected by the Argonne effluent.

TABLE 37

Chemical Constituents in the Des Plaines River, 1978

Constituent	Location*	No. of Samples	Concentration (mg/1 or µg/1)		
			Avg.	Min.	Max.
Chromium(VI)	A	12	< 0.1	–	< 0.1
	B	24	< 0.1	–	< 0.1
Iron	A	12	1.09 ± 0.34	0.29	1.90
	B	24	1.30 ± 0.66	0.21	7.40
Mercury**	A	12	< 0.1	–	< 0.1
	B	24	< 0.1	–	< 0.1
Zinc	A	12	0.06 ± 0.04	0.02	0.23
	B	24	0.09 ± 0.07	0.01	0.69

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

**Concentrations in µg/1.

2. Environmental Air Studies

Studies of background levels of total suspended particulates were continued during 1978 at four locations situated to study effluents from the Argonne power plant. This year these samples were also analyzed for iron, copper, zinc, lead, bromine, and strontium using energy dispersive x-ray fluorescence. The effect of Argonne operations on the levels of environmental beryllium was also studied.

Samples for particulate matter determination were collected on glass fiber filters (20 cm x 25 cm). High volume samplers (General Metals Corp.) were used and samples were collected for seven-day periods. The air flow rate varied from 1.1 to 1.9 m^3/min., depending on the sampler design, and was verified using a Roots meter as a primary standard. The papers were equilibrated before and after use in a dry box (relative humidity less than 35%) for a minimum of 24 hours.

A monthly sample was collected at a location near beryllium operations to evaluate their effect on ambient beryllium levels. Millipore filters were used at a flow rate of about 8.5 liters per minute and analyses were performed as for water.

Samples were collected for total suspended particulates at location 18J from January through July, 1978. In August the sampler was moved to location 14D to place it downwind of the boiler plant at 13L, and to remove it from close proximity to Interstate Highway 55. This sampler is located approximately 12 feet above ground level. The other three sampling locations are essentially the same as last year.

Sample location 12F is west of the power plant and is approximately 10 feet above ground. Sample location 8F is southwest of the power plant, at the Argonne meteorology station, and is 4 feet above the ground. This sampler also monitors effluent from power plants located southwest of the Laboratory. Sample location 12M is southeast of the power plant and located about 20 feet above ground. This is the nearest sample to the plant.

Trace element analyses were performed on the samples collected for total suspended particulates by removing a one centimeter circle and counting it for one hour using a cadmium-109 source to excite the x-rays. Studies have shown that distribution of these six elements across the filter is sufficiently

homogeneous so that counting this segment gives results representative of
the entire filter.

The results are shown in Table 38. All values are listed as geometric
means since this average best describes the results.[16] The average at 18J
for total suspended particulates is higher than in 1977, since samples were
not collected during the last half of the year when results tend to be lower.
Results at 14D were consistently lower than any of the other locations, prob-
ably because the location is a wooded area. The results obtained at 12F and
8F were the same as last year, while results at 12M were higher since sampling
was performed throughout the whole year as contrasted to only the fall in 1977.

Average results for iron ranged from 0.94 to 1.38 $\mu g/m^3$. The linear
correlation coefficient for iron and total suspended particulates at all
locations was 0.95, which shows a strong relationship between the two vari-
ables.

TABLE 38

Environmental Air Data

(Concentrations in $\mu g/m^3$)[*]

	Location				
Constituent	18J	14D	12F	8F	12M
Total Suspended Particulates	61	38	58	48	48
Iron	1.38	0.94	1.31	1.25	1.13
Copper	0.54	0.07	0.24	0.12	0.12
Lead	0.85	0.41	0.51	0.43	0.51
Zinc	0.15	0.10	0.17	0.13	0.11
Bromine	0.14	0.06	0.07	0.06	0.07
Strontium	0.010	< 0.001	0.006	0.006	0.005
Beryllium	–	–	–	–	1.9×10^{-4}

[*]Average geometric mean.

Average copper results ranged from 0.07 to 0.54 $\mu g/m^3$ and showed a linear
correlation coefficient of 0.84 when compared to total suspended particulate

averages. The copper results, however, are complicated to an unknown degree by contamination of air samples by copper abraded from the sampling motor. Presumably when the samplers are elevated and there is sufficient wind, this problem is minimized. Careful control of brush interval change has been instituted in an attempt to control this problem.

Average levels found for lead ranged from 0.85 $\mu g/m^3$ at 18J to 0.41 $\mu g/m^3$ at 14D. The average level at the only location sampled in 1977 was 0.39 $\mu g/m^3$. Lead and bromine levels correlate less well with total suspended particulate concentrations than do iron and copper. The linear correlation coefficients for lead and bromine are 0.77 and 0.71, respectively. Both lead and bromine are present in automobile exhaust as well as in soil. This is reflected in the high levels at 18J, which is close to heavy automobile traffic and the much lower results at 14D where automotive traffic is minimal. The linear correlation coefficient between average lead and average bromine levels is 0.99, which is highly significant. Levels of zinc and strontium appear to be related to total suspended particulates (linear correlation coefficients of 0.89 and 0.91, respectively).

The result obtained for beryllium at location 12M is essentially the same as in 1977. The use of beryllium at this location is being discontinued and sampling for this element will be continued at the other sampling sites since it is an element of interest in coal.

The levels of total suspended particulates and trace elements reported herein are normal constituents of ambient air and represent background levels useful for the evaluation of the effect of future coal-burning studies at Argonne on their concentrations in air.

IV. APPENDIX

A. __References__

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

1. Standards

All of the standards and detection limits for chemical constituents, and some of those for radionuclides and external radiation, are given in the main body of the report. In addition, in Table 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 Manual Chapter 0524,[4] and are used in this report to assess the hazard of a measured concentration of a radioactive nuclide. Chapter 0524 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

TABLE 39

Concentration Guides and Detection Limits

(µCi/ml)

Nuclide or Activity	Concentration Guide		Detection Limit	
	Water	Air	Water	Air
Americium-241	4×10^{-6}	4×10^{-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}	2×10^{-7}	1×10^{-13}
Iodine-131	3×10^{-7}	1×10^{-10}	3×10^{-9}	5×10^{-15}
Manganese-54	-	1×10^{-9}	-	5×10^{-16}
Neptunium-237	3×10^{-6}	-	1×10^{-12}	-
Plutonium-238	5×10^{-6}	1×10^{-12}	1×10^{-12}	5×10^{-19}
Plutonium-239	5×10^{-6}	1×10^{-12}	5×10^{-13}	5×10^{-19}
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}	3×10^{-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 1×10^{-7}	1×10^{-10} to 1×10^{-13}	2×10^{-10}	2×10^{-16}
Beta**			1×10^{-9}	5×10^{-16}

*Concentration Guides converted from the "special curie" used in DOEM 0524 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 CG is applicable.

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-Searle Co. 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), the Quality Assurance Branch of the U. S. Environmental Protection Agency (EPA-QA) at Las Vegas, and the International Atomic Energy Agency (IAEA) are analyzed regularly. The EPA-QA intercomparison program consists of analyzing a variety of samples, at intervals selected by the participant, to which known amounts of various radionuclides have been added by the EPA laboratory. A summary of all the EPA-QA samples analyzed in this laboratory in 1978 is shown in Table 40. The DOE Environmental Measurements Laboratory Quality Assurance Program (DOE-EML-QAP) is a quarterly distribution of four or five different sample matrices containing various combinations of radionuclides.[17] Results of our participation in this program during 1978 are given in Table 41. 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%, the error in the EML results is 1-30% (depending

TABLE 40

Summary of EPA-QA Samples, 1978

Type of Sample	Analysis	Number Analyzed	Avg. Difference From Added
Air Filter	Total Alpha	2	32%
	Total Beta	2	5%
	Strontium-90	2	14%
	Cesium-137	2	5%
Water	Total Alpha	7	39%
	Total Beta	7	9%
	Hydrogen-3	1	4%
	Chromium-51	3	8%
	Cobalt-60	5	5%
	Zinc-65	5	9%
	Strontium-89	4	6%
	Strontium-90	4	10%
	Ruthenium-106	5	7%
	Cesium-134	5	3%
	Cesium-137	3	4%
	Plutonium-239	2	18%
Milk	Potassium-40	1	0%
	Strontium-89	1	52%
	Strontium-90	1	33%
	Iodine-131	1	6%
	Cesium-137	1	20%
Soil	Plutonium-239	1	8%

TABLE 41

Summary of DOE-EML-QAP Samples, 1978

	Average Difference From EML Value				
Nuclide	Air Filters	Water	Soil	Tissue	Vegetation
Hydrogen-3	-	6% (4)	-	-	-
Sodium-22	9% (2)	6% (1)	-	-	-
Potassium-40	-	-	3% (4)	16% (4)	7% (3)
Manganese-54	3% (2)	4% (3)	-	-	-
Cobalt-57	7% (3)	14% (1)	-	-	-
Cobalt-58	-	11% (2)	-	-	-
Cobalt-60	6% (2)	8% (4)	-	7% (1)	2% (2)
Zinc-65	2% (2)	-	-	-	-
Strontium-89	21% (1)	-	-	-	-
Strontium-90	6% (4)	9% (4)	7% (4)	19% (4)	16% (3)
Ruthenium-106	2% (1)	-	-	-	-
Antimony-125	18% (1)	-	-	-	-
Cesium-134	7% (1)	-	-	-	-
Cesium-137	2% (3)	7% (4)	4% (4)	14% (3)	7% (3)
Cerium-144	8% (2)	3% (2)	-	-	-
Radium-226	-	-	7% (4)	6% (2)	4% (1)
Thorium-228	-	-	12% (2)	26% (2)	20% (1)
Uranium-234	25% (2)	23% (3)	91% (1)	-	-
Uranium-235	-	54% (1)	36% (1)	-	-
Uranium-238	30% (2)	24% (3)	102% (1)	-	-
Uranium-total	-	-	1% (3)	-	-
Plutonium-238	40% (3)	-	85% (3)	10% (1)	23% (3)
Plutonium-239	18% (4)	13% (3)	19% (3)	6% (1)	19% (3)
Americium-241	3% (2)	39% (3)	5% (2)	-	-

The figure in parentheses is the number of samples.

on the nuclide and the amount present), and the error in the added amount in the EPA-QA samples is 2-5% (our estimate).

b. Penetrating Radiation

Our laboratory participated in the Third International Comparison of Environmental Dosimeters conducted jointly by the DOE Environmental Measurements Laboratory, Oak Ridge National Laboratory, and the School of Public Health of the University of Texas. Participants supplied dosimeter sets (TLD chips in our case) which were exposed outdoors in an area that contained above-normal concentrations of cesium-137 in the soil, and to a known dose in the laboratory. They were then returned to the participant for measurement. The results, in mR, obtained were:

	Field Exposure	Laboratory Exposure
Oak Ridge Value	34.9	91.7
Our Value		
calcium fluoride	32.1	95.
lithium fluoride	33.4	95.

Agreement with the Oak Ridge values was within 3 mR or 8%.

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 identitical 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.

d. Sampling, Sample Storage, etc.

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 rate of discharge of waste water varies appreciably during each 24-hour period.

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

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

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D. Acknowledgements