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ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY

ANNUAL REPORT FOR 1977

by

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

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY ANNUAL REPORT FOR 1977

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ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1977 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, fallout, Argonne, and other) and are compared with applicable environmental quality standards. The potential radiation dose to off-site population groups is also estimated.

I. INTRODUCTION

A. General

This report is prepared to provide the U. S. Department of Energy (DOE) and the public with information on the levels of radioactive and non-radioactive pollutants in the environment of Argonne National Laboratory (ANL) and on the amounts of pollutants, 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 substances in the environment. Of special interest is the detection of any such substances released to the environment by Argonne. One important

function of the program is to verify the adequacy of Argonne's effluent 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, and solar energy utilization. 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 surrounding the site (Waterfall Glen Forest Preserve) was formerly Argonne property, but was deeded to the DuPage County Forest Preserve District in 1973 for their use as a public recreational area, nature preserve, and demonstration forest.

The terrain is gently rolling, partially-wooded, former prairie and farmland. The grounds contain a number of small ponds and streams, the principal one being Sawmill Creek, which runs through the site in a southerly direction and enters the Des Plaines River about 2.1 km (1.3 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°, reaching 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.

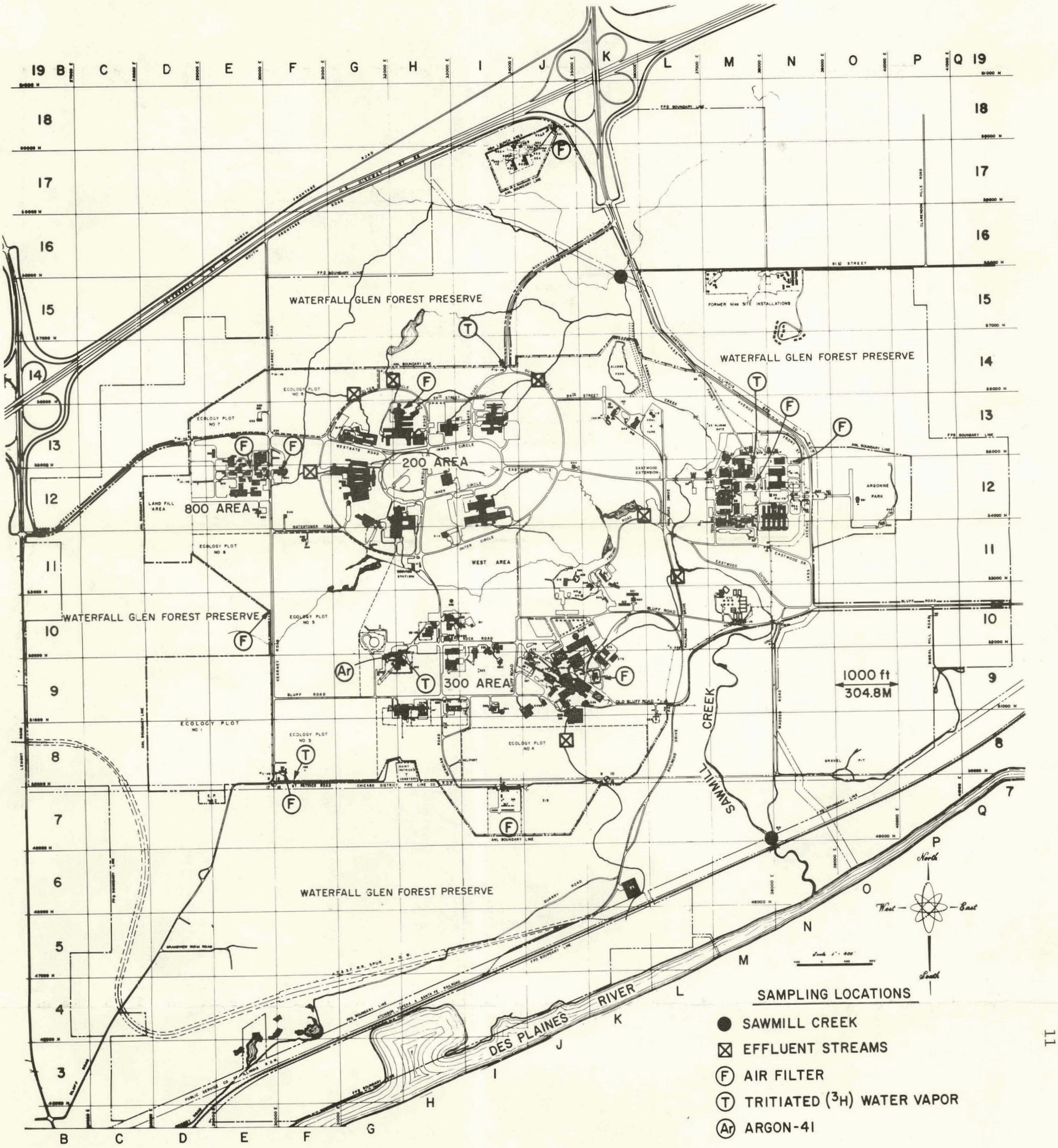

Fig. 1. Sampling Locations at Argonne National Laboratory

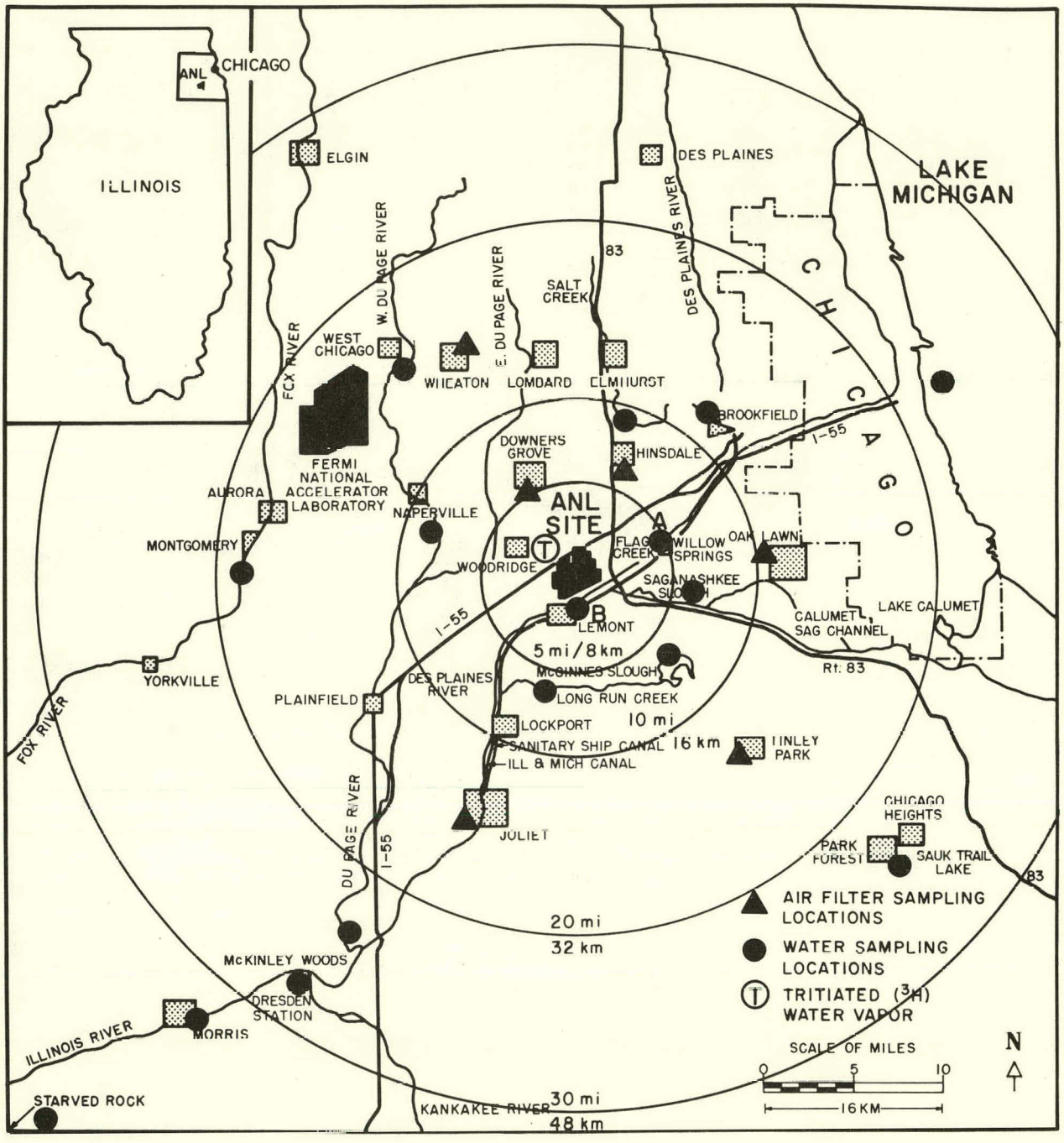

Fig. 2. Sampling Locations Near Argonne National Laboratory

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.

The most important meteorological parameters for the purposes of the report are wind direction, wind speed, temperature, and precipitation. These are given here for 1977 and were provided by the Atmospheric Physics Section of the Radiological and Environmental Research Division at Argonne. The average monthly and annual wind roses are shown in Figure 3. The wind roses are polar coordinate plots in which the lengths of the radii represent the percentage frequency of wind speeds in classes of 2.01-6 m/s (4.5-13.4 mph), 6.01-10 m/s (13.4-22.4 mph), and greater than 10.01 m/s (22.4 mph). The direction of the radii represents the direction from which the wind blows. Sixteen radii are shown on each plot at 22.5° intervals; each radius represents the average wind speed for the direction covering 11.25° on either side of the radius. For example, in the plot labeled "77 total", the predominant wind blows from the west. The length and direction of this vector shows that, of the total wind observations, about 5% were in the 2.01-6 m/s range, about 5% were in the 6.01-10 m/s range, about 2% were greater than 10.01 m/s; and about 12% of the observations were from the direction between 268.75° and 281.25° (west). The number in the center represents the percent of observations of wind speed less than 2 m/s in all directions.

The roses show that the predominant winds were from the west and southwest on the average, but seasonal variations are apparent. The winds were primarily westerly in January and February. In the spring, the northeast lake breezes are evident. The warm southwesterly winds, brought up by the high pressure areas in the southeastern U.S., began in July and continued for the remainder of the year. The winds during 1977 were characterized by a much stronger westerly component than the usual southwest dominance in past years. The winds are sufficiently variable so that monitoring for any airborne releases must be carried out in all directions from the site.

The monthly average wind speed at 44.5 m (146 ft) above ground level varied from 5.0 m/s (11 mph) in September to 7.0 m/s (15.4 mph) in March. Average monthly wind speeds at 1.5 m (4.9 ft) and 6.0 m (19.7 ft) above ground

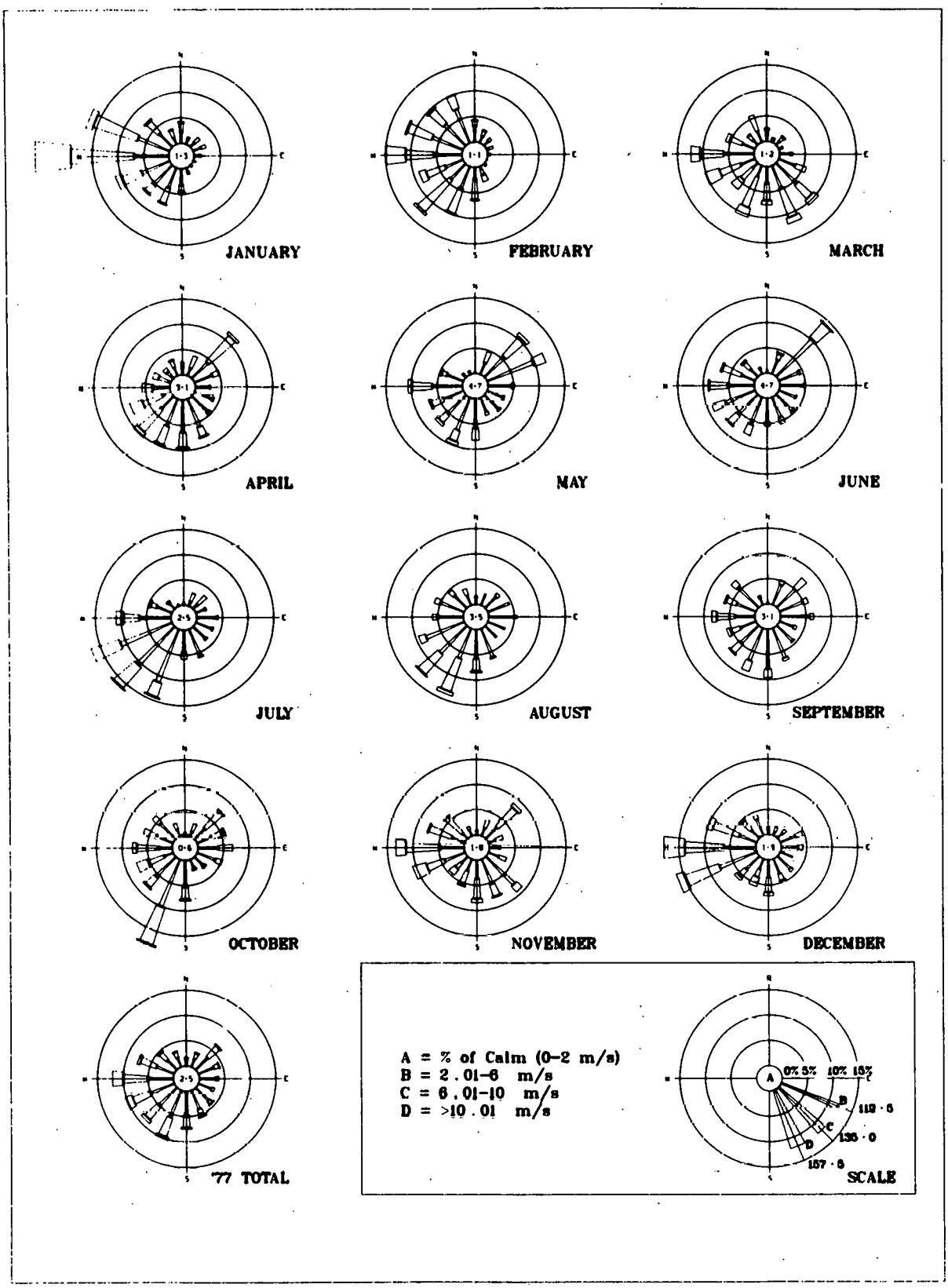

Fig. 3. Monthly and Annual Wind Roses at Argonne National Laboratory, 1977

level had maxima and minima in the same months. At 1.5 m the speeds ranged from 2.6 m/s (5.7 mph) to 4.1 m/s (9.0 mph) and at 6.0 m the range was from 3.1 m/s (6.8 mph) to 4.8 m/s (10.6 mph).

The precipitation and temperature data for 1977 are shown in Table 1. Precipitation in August and September was well above average while July was one of the driest months in history. Overall, the annual precipitation was 10% above average. The monthly temperature mean was well below average in January and well above average from March to June. The annual mean, however, was essentially normal.

TABLE 1

Weather Summary, 1977

Month	Precipitation (cm)		Temperature (°C)	
	Amount	Normal	Monthly Average	Normal
January	1.16	4.70	-12.4	-4.3
February	1.49	4.04	-3.1	-2.6
March	9.44	6.93	6.7	2.7
April	9.72	9.52	13.0	9.9
May	5.86	8.66	19.8	15.6
June	12.36	10.03	19.8	15.6
July	3.86	10.39	24.7	23.7
August	17.34	7.98	21.1	23.2
September	19.15	7.62	18.0	18.8
October	4.67	6.65	10.4	13.0
November	6.62	5.59	4.8	4.7
December	4.67	5.36	-4.5	-1.9

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 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. The distribution, centered on the CP-5 reactor, was obtained by

TABLE 2

Incremental Population Data in the Vicinity of ANL, 1974

Distance, miles Distance, km	0 - 1 0-1.6	1 - 2 1.6-3.2	2 - 3 3.2-4.8	3 - 4 4.8-6.4	4 - 5 6.4-8.0	5-10 8-16	10-20 16-32	20-30 32-48	30-40 48-64	40-50 64-80
Direction										
N	0	250	2150	3100	3560	36570	188820	294520	107730	194880
NNE	0	25	1530	4530	3210	39600	336130	545820	113100	0
NE	0	160	1650	1170	1240	29400	803170	1071375	0	0
ENE	0	0	4390	755	1410	29850	737740	333865	0	0
E	0	0	130	0	10	31220	514440	252870	13720	24720
ESE	0	0	160	275	100	10760	225710	287970	295700	43080
SE	0	0	170	125	75	15230	48900	113780	21270	9140
SSE	0	20	485	440	80	1120	11820	7270	13770	18580
S	0	5	565	550	700	2910	13670	1270	26770	39320
SSW	0	15	4080	1030	730	16300	107590	8680	15130	7370
SW	0	600	150	50	100	12220	28170	4990	14460	7420
WSW	0	100	10	155	315	9140	5080	1980	5820	9990
W	0	1350	115	6480	9510	1570	47760	17720	17380	7510
WNW	0	740	35	1965	1960	21470	91000	8390	4190	50590
NW	0	180	200	2350	4995	13510	42110	74900	14910	12100
NNW	0	150	1575	1340	3650	37980	111420	146640	90780	67920
Total	0	3595	17405	24325	31645	308850	3313530	3172040	754730	492620
Cumulative Total	0	3595	21000	45325	76970	385820	3699350	6871390	7626120	8118740

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. The figures were adjusted in the first few kilometers by actual inspection of the area, and over the entire region with the use of information from the Northeastern Illinois Planning Commission (NIPC), the Northwestern Indiana Regional Planning Commission, and the individual County planning offices outside of these areas. The NIPC provided population forecasts by quarter section for 1974 in the six county area around Chicago. The Northwestern Indiana Regional Planning Commission provided the same type of data for Lake and Porter Counties. The other Illinois Counties provided their best estimates of the 1974 population in their respective areas. This count was usually a locally adjusted estimate using the State of Illinois Bureau of the Budget forecast.

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 7.6 megaliters (2 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 is discharged into Sawmill Creek at location 7M in Figure 1. In 1977, the water flow in the Creek averaged about 29 megaliters (7.8 million gallons) per day upstream of the Argonne waste-water outfall; Argonne waste-water effluent averaged 3 megaliters (0.8 million gallons) per day. The Argonne waste water is composed of sanitary waste and laboratory waste. During 1977, the laboratory waste comprised 56% and sanitary waste 44% of the combined Argonne effluent.

Seven streams (or channels) that carry blowdown discharge water from cooling towers are considered effluents since they leave the site boundary. These are shown in Figure 1. Two streams from the 200 Area leave the site at location 14J and 14G, respectively, and enter the Creek at 15K; one from the 300 Area enters Sawmill Creek at location 11L (the Creek crosses the site

boundary shortly thereafter); another stream from the 300 Area crosses the site boundary at location 7K and soaks into the ground south of the site. The channel at 14G carries water principally from Building 208 and the 14H channel serves Building 203. The 14G channel enters Sawmill Creek at 15K, while the 14H channel enters a pond at 15H. The outflow of this pond is an intermittent stream which flows into Sawmill Creek at 15K when sufficient water is present. The effluent at 12L is on-site and drains the Freund ponds located at 11K and 12K. Cooling tower effluents from Buildings 200, 205, and 212 enter these ponds upstream from the 12L sampling point. Sawmill Creek flows into the Des Plaines River as described earlier.

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 forms 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. The plant uses water from the Kankakee for cooling, and discharges the water into the Illinois River. The first use of water for drinking is an indirect one at 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 for hiking and skiing, and its development and increased utilization by the public is expected. Sawmill Creek flows through the south portion of the Preserve on its way to the Des Plaines River. This region of the Preserve (formerly named Rocky Glen) was used principally for picnicking,

hiking, and overnight camping by youth groups, but has been closed for rehabilitation since June 6, 1976. East and southeast of Argonne and the Des Plaines River are located several large forest preserves of the Cook County Forest Preserve District. The preserves include the two sloughs shown in Figure 2, McGinnis and Saganashkee, as well as other smaller lakes. These areas are used for picnicking, boating, fishing, and hiking. A small park located in the eastern part of the Argonne site (12-O in Figure 1) is for the use of Argonne and Department of Energy employees only.

The most recently available information on milk and principal agricultural production in a 10,600 sq km (4,100 sq mi) area around Argonne is shown by County in Table 3. These estimates were obtained from the Illinois Crop Reporting Service of the State Department of Agriculture and are for 1976.

TABLE 3

Agricultural Production Near ANL, 1976

	Milk		Corn	Soybeans	Wheat	Oats
County	No. of Cows	Million Pounds	Million Bushels			
DuPage	300	3.2	2.0	0.51	0.14	0.13
Cook	400	4.0	1.8	0.64	0.13	0.15
Will	3,400	33.7	15.2	3.1	0.73	0.58
Kane	6,400	63.4	14.1	2.0	0.30	0.64
Kendall	600	5.9	9.1	1.5	0.25	0.25
Grundy	1,000	9.9	12.6	2.2	0.075	0.16
Lake	1,400	13.9	1.8	0.47	0.32	0.30

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-77-13. Included in this report are the results of measurements obtained in 1977 for a number of radionuclides in air, surface water, soil, grass, bottom sediment, and foodstuffs; 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 the last four months of the year, and are attributed to fallout from the Chinese atmospheric nuclear tests of November 17, 1976, and September 17, 1977. No activity attributable to Argonne operations could be detected.

The plutonium-239,240 concentrations in air averaged 19×10^{-18} μCi/ml,* about three times the 1976 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 0.002% of the CG.**

Argon-41 and hydrogen-3 (as water vapor) represent the major airborne radioactivity released from the Laboratory. The argon-41 concentration and

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*The radioactivity units are described in Section III.

**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.(3) The pertinent CGs are listed in the Appendix, Section IV.B.

corresponding radiation dose at the site boundary was less than the detection limit (50% of the CG) of the present sampling system, but was calculated from an atmospheric dispersion model to be 4.5×10^{-10} μCi/ml and 5.7 mrem/yr in the predominant east to northeast wind directions. 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, is 3.0 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 11×10^{-12} μCi/ml, which is 0.006% of the CG and about seven times greater than the off-site concentration. The corresponding dose is 0.02 mrem/yr.

Argonne waste water is discharged into Sawmill Creek, and this stream was sampled above and below the site to evaluate the effect of Argonne operations on its radioactive content. The nuclides (for which analyses were made) added to the Creek in the waste water, and the percent CG of their average Creek concentrations, were hydrogen-3, 0.039%; strontium-90, 0.01%; neptunium-237, 0.0006%; plutonium-239,240, 0.0002%; americium-241, 0.0001%; and curium-244 and/or californium-249, < 0.00003%. Although Sawmill Creek is not a source of potable water, the dose to an individual using water at these concentrations as his sole source of drinking water would be less than 0.28 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 0.7×10^{-3} μCi/m^2 at the site perimeter and 0.8×10^{-3} μCi/m^2 off the site. The corresponding plutonium-238 averages were both 0.04×10^{-3} μCi/m^2. 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 x 10^{-9} μCi/g to 45 x 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 were all < 200 x 10^{-9} μCi/ml (the detection limit). The strontium-90 and cesium-137 concentrations were similar to 1976. Strontium-89, iodine-131, and barium-140 were detected late in the year as a result of fallout from a Chinese atmospheric nuclear test.

Measurements of penetrating radiation were made at several locations at the site boundary and off the site. The off-site readings averaged 92 mrem/yr, with a standard deviation of 7 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 114 mrem/yr above normal at location 7I in Figure 1 and about 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 within the normal range, 101 mrem/yr. Along the north side of the site, the dose at the fence at location 14I was 119 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 to the residents closest to the south boundary, about 1.6 km (1 mile) from the fence line, was about 0.002 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 0.2 mrem/yr. Thus, doses to individuals living near the site will not exceed 0.04% 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, manganese, and zinc in the Des Plaines River were measured to determine whether any contribution from Argonne waste water could be seen.

The Biochemical Oxygen Demand (B.O.D.) in the Argonne combined sanitary and laboratory waste-water effluent exceeded the State of Illinois standard in two of the weekly samples. The ammonia nitrogen exceeded the standard in January, February, and April. Suspended solids did not exceed the State limit at any time. Average concentrations of each of the other chemical constituents measured in this effluent were at or below these standards. The concentration of mercury averaged 100% of the State standard and exceeded this value 16% of the time (i.e., in 16% 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 average concentration at one location (14H) exceeded the standard but extremely low water flow conditions prevented normal dilution and runoff, thus causing persistent high levels. The levels in the other six channels represent a marked reduction from 1976.

The average values in Sawmill Creek for dissolved oxygen and chemical constituents, except ammonia nitrogen and dissolved solids, were within the State of Illinois standards. The average ammonia nitrogen level above the Argonne waste-water outfall was 2.8 times the State standard, and exceeded this standard in 50% of the samples. The average level below the outfall was 1.5 times the State standard and exceeded this value in 40% of the samples. The average concentration of dissolved solids above the Argonne outfall exceeded the State limit by 20%, but below the outfall it was less than the standard. On two occasions, levels 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. Individual values for copper, cyanide, iron, mercury, and silver exceeded State standards from 6% to 28% of the time. Hexavalent chromium levels were in excess of the State standard 1% of the time. 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.

Studies of lead, beryllium, and particulate matter in air were carried out. The values obtained for particulate matter at four locations met both primary and secondary U. S. Environmental Protection Agency (USEPA) standards. Values obtained for beryllium and lead agreed with ambient air studies performed elsewhere.

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 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 the Concentration Guides (CGs) and annual dose limits (Radiation Protection Standards) given in DOE Manual Chapter 0524.[3] 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.

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 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

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

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, 1.5×10^{-13} µCi/ml, was about three times the 1976 average. The results in Table 5 indicate that the increase was principally due to intermediate half-life fission products (zirconium-95-niobium-95, ruthenium-103, ruthenium-106-rhodium-106, cerium-141, and cerium-144). Their presence in the air during the spring of the year was the result of stratospheric fallout from an atmospheric nuclear test by the People's Republic of China on November 17, 1976. Fission products (e.g., ruthenium-103, iodine-131, barium-140-lanthanum-140, and cerium-141) from a Chinese atmospheric nuclear test on September 17, 1977, were detected in the last four months of the year. In addition to the gamma-ray emitting nuclides listed in Table 5, yttrium-88 was detected during the spring at concentrations up to 0.3×10^{-15} µCi/ml and technetium-99m, tellurium-132, and neodymium-147 were observed shortly after the test of September 17, 1977. The maximum concentrations for each of these nuclides were: 1.1×10^{-15} µCi/ml for technetium-99m, 2.8×10^{-15} µCi/ml for tellurium-132, and 6.2×10^{-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 45% of the gamma-ray activity was due to beryllium-7, principally produced in the stratosphere by cosmic-ray interactions. The remaining activity was primarily fission and activation products from nuclear test detonations.

TABLE 4

Total Alpha and Beta Activities in Air-Filter Samples, 1977*

(Concentrations in 10^{-15} μCi/ml)

Month	Location	No. of Samples	Alpha Activity Av.	Alpha Activity Min.	Alpha Activity Max.	Beta Activity Av.	Beta Activity Min.	Beta Activity Max.
January	perimeter	42	2.6	1.2	5.1	50	25	78
	off-site	15	3.1	1.6	5.2	55	25	83
February	perimeter	36	2.0	0.6	3.4	41	30	53
	off-site	14	1.9	1.5	3.1	39	30	54
March	perimeter	38	2.1	0.5	3.9	73	34	134
	off-site	15	2.0	1.0	4.3	74	29	134
April	perimeter	37	3.3	0.9	8.9	180	49	317
	off-site	19	3.1	0.7	9.8	183	71	338
May	perimeter	39	2.6	1.4	6.2	328	114	516
	off-site	18	2.7	0.6	5.1	285	111	467
June	perimeter	35	2.5	1.1	7.3	290	217	435
	off-site	15	2.7	0.9	7.6	293	115	581
July	perimeter	42	2.3	0.8	4.4	219	159	304
	off-site	16	2.0	0.6	5.8	202	138	307
August	perimeter	39	1.3	0.5	2.8	163	68	280
	off-site	14	1.1	0.2	1.6	151	57	233
September	perimeter	36	1.5	0.6	3.7	136	46	487
	off-site	18	1.4	0.2	3.2	148	33	480
October	perimeter	42	1.9	0.6	3.4	201	86	523
	off-site	19	1.9	1.0	3.0	194	84	486
November	perimeter	36	2.0	0.6	4.5	99	65	145
	off-site	21	2.1	0.9	3.8	108	50	159
December	perimeter	41	2.3	0.9	5.2	80	48	117
	off-site	19	2.4	1.5	4.2	82	55	128
Annual Summary	perimeter	463	2.2 ± 0.3	0.5	8.9	155 ± 54	25	523
	off-site	203	2.2 ± 0.4	0.2	9.8	151 ± 49	25	581
Percent CG	perimeter	-	(0.022)	(0.005)	(0.089)	1.55	0.25	5.2
	off-site	-	(0.022)	(0.002)	(0.098)	1.51	0.25	5.8

*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, 1977

(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	89	< 0.1	4.3	2.4	0.8	< 0.5	< 5	0.4	0.9	1.8	1.0
	off-site	78	< 0.1	4.3	2.1	1.0	< 0.5	< 5	0.4	1.0	1.7	1.0
February	perimeter	110	< 0.1	6.5	2.2	0.8	< 0.5	< 5	0.4	0.7	1.6	1.4
	off-site	109	< 0.1	6.8	2.0	0.6	< 0.5	< 5	0.4	0.5	1.5	1.4
March	perimeter	124	< 0.1	25	6.6	2.7	< 0.5	< 5	0.7	0.7	4.3	5.0
	off-site	110	< 0.1	26	6.4	3.2	< 0.5	< 5	1.2	0.1	4.0	5.0
April	perimeter	155	0.2	98	18	9.0	1.0	< 5	1.6	0.3	11	21
	off-site	138	< 0.1	86	16	7.9	0.6	< 5	1.7	< 0.1	9.5	19
May	perimeter	151	0.3	187	26	22	1.9	< 5	3.3	0.1	14	49
	off-site	148	0.4	188	25	20	1.8	< 5	3.3	< 0.1	14	48
June	perimeter	128	0.4	159	17	25	2.2	< 5	3.6	< 0.1	9.2	56
	off-site	131	0.4	166	17	25	2.3	< 5	3.4	< 0.1	7.9	52
July	perimeter	109	0.2	112	8.1	17	1.7	< 5	2.5	< 0.1	3.8	39
	off-site	109	0.4	109	8.4	17	1.7	< 5	2.7	< 0.1	3.6	43
August	perimeter	95	0.2	76	5.0	15	1.8	< 5	2.5	< 0.1	2.0	36
	off-site	82	< 0.1	64	3.6	14	0.9	< 5	2.0	< 0.1	1.9	34
September	perimeter	62	0.2	28	3.8	9.0	< 0.5	17	1.2	8.3	2.9	18
	off-site	64	0.1	30	4.1	8.9	0.9	17	1.5	7.7	3.2	18
October	perimeter	83	0.2	30	11	10	0.9	29	1.7	22	11	21
	off-site	74	0.1	29	10	9.0	1.1	23	1.9	16	10	20
November	perimeter	81	0.2	19	3.4	8.4	0.8	< 5	1.6	1.8	2.9	17
	off-site	79	0.2	21	3.4	8.8	1.0	< 5	1.7	1.7	3.0	18
December	perimeter	64	0.1	11	0.9	5.6	0.5	< 5	1.2	0.3	0.8	12
	off-site	70	0.2	12	1.0	7.0	0.7	< 5	1.5	0.2	0.7	14
Annual Summary	perimeter	104 ± 18	0.2 ± 0.1	63 ± 36	8.7 ± 4.5	13 ± 4.6	1.0 ± 0.4	< 8	1.7 ± 0.6	2.9 ± 3.7	5.4 ± 2.6	23 ± 11
	off-site	99 ± 16	0.2 ± 0.1	62 ± 36	8.2 ± 4.3	13 ± 4.3	1.0 ± 0.4	< 8	1.8 ± 0.6	2.3 ± 2.8	5.1 ± 2.4	23 ± 10
Percent CG ($\times 10^{-3}$)	perimeter	(0.26)	0.02	6.3	0.29	5.0	0.11	< 8	0.34	0.29	0.11	12
	off-site	(0.25)	0.02	6.2	0.27	5.0	0.11	< 8	0.36	0.23	0.10	12

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 analysis 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 and radiostrontium concentrations increased at all locations by a factor of about three from 1976. The general increase in the spring or early summer of the year is also seen in Figure 4, which shows the monthly plutonium-239 air concentrations for the past five years. The arrows at the bottom of the figure indicate the approximate dates of atmospheric nulcear tests.

The monthly variations in the concentrations of strontium-89, strontium-90, and plutonium-239 correlate with the results for the fission products in Table 5. The presence of these radionuclides are considered to be due to fallout from the atmospheric nuclear tests mentioned previously. There is

TABLE 6

Strontium and Plutonium Concentrations in Air-Filter Samples, 1977

(Concentrations in 10^{-18} μCi/ml)

Month	Location[1]	Strontium-89	Strontium-90	Plutonium-238	Plutonium-239[2]
January	12N	1430 ± 80	140 ± 20	< 0.5	3.0 ± 0.5
	7I	1450 ± 80	170 ± 20	< 0.5	2.3 ± 0.5
	off-site	1580 ± 140	200 ± 40	< 0.5	4.2 ± 0.6
February	12N	1640 ± 70	140 ± 30	< 0.5	3.6 ± 0.6
	7I	1760 ± 150	180 ± 60	< 0.5	3.8 ± 0.7
	off-site	1580 ± 180	140 ± 70	< 0.5	2.6 ± 0.5
March	12N	5180 ± 210	390 ± 60	< 0.5	8.2 ± 0.9
	7I	4710 ± 240	370 ± 70	< 0.5	8.3 ± 0.9
	off-site	5430 ± 190	300 ± 50	< 0.5	8.3 ± 0.9
April	12N	13770 ± 860	980 ± 230	< 0.5	18 ± 2
	7I	13020 ± 750	1030 ± 190	< 0.5	21 ± 2
	off-site	13120 ± 1250	1160 ± 330	< 0.5	20 ± 2
May	12N	24830 ± 480	1840 ± 160	0.6 ± 0.3	38 ± 2
	7I	22200 ± 260	1880 ± 80	0.9 ± 0.5	36 ± 2
	off-site	-	-	0.9 ± 0.3	37 ± 2
June	12N	16100 ± 230	1820 ± 90	0.6 ± 0.3	37 ± 2
	7I	15960 ± 440	1820 ± 180	0.8 ± 0.4	33 ± 2
	off-site	17090 ± 440	2020 ± 160	0.8 ± 0.4	38 ± 2
July	12N	10750 ± 930	1560 ± 270	0.5 ± 0.3	32 ± 2
	7I	8720 ± 600	1380 ± 150	0.7 ± 0.3	31 ± 2
	off-site	10890 ± 1070	1510 ± 310	0.6 ± 0.3	34 ± 2
August	12N	4190 ± 230	1080 ± 70	< 0.5	23 ± 2
	7I	2820 ± 300	1170 ± 80	< 0.5	15 ± 2
	off-site	4900 ± 250	1230 ± 70	0.6 ± 0.3	23 ± 2
September	12N	5740 ± 350	800 ± 70	1.3 ± 0.5	19 ± 2
	7I	2940 ± 1540	1080 ± 300	< 0.5	21 ± 4
	off-site	6950 ± 860	870 ± 180	0.6 ± 0.4	16 ± 2
October	12N	11500 ± 780	940 ± 230	1.0 ± 0.3	19 ± 1
	7I	8530 ± 300	820 ± 90	0.8 ± 0.3	16 ± 1
	off-site	8440 ± 250	870 ± 80	0.9 ± 0.3	16 ± 1
November	12N	3510 ± 330	900 ± 140	1.3 ± 0.4	15 ± 1
	7I	3130 ± 360	920 ± 150	1.0 ± 0.4	15 ± 1
	off-site	2980 ± 1560	680 ± 550	0.6 ± 0.2	14 ± 1
December	12N	1150 ± 60	770 ± 20	0.4 ± 0.2	15 ± 1
	7I	900 ± 70	680 ± 30	0.7 ± 0.3	13 ± 1
	off-site	1060 ± 90	710 ± 40	0.5 ± 0.4	12 ± 1
Annual Summary	12N	8320 ± 4190	950 ± 330	0.6 ± 0.2	19 ± 7
	7I	7180 ± 3900	960 ± 330	0.5 ± 0.2	18 ± 6
	off-site	6730 ± 3150	880 ± 350	0.5 ± 0.1	19 ± 5
Percent CG ($\times 10^{-3}$)	12N	0.83	0.48	0.06	1.9
	7I	0.72	0.48	0.05	1.8
	off-site	0.67	0.44	0.05	1.9

[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, 1977

(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	36 ± 2	48 ± 2	39 ± 2	57 ± 4	6.5 ± 1.2	58 ± 4
	7I	19 ± 2	28 ± 2	20 ± 1	39 ± 3	3.6 ± 0.8	41 ± 3
	off-site	21 ± 2	38 ± 2	23 ± 2	54 ± 3	4.6 ± 0.8	57 ± 3
February	12N	12 ± 1	18 ± 1	11 ± 1	33 ± 2	0.7 ± 0.3	29 ± 2
	7I	9 ± 1	14 ± 1	8 ± 1	22 ± 3	2.5 ± 0.9	35 ± 3
	off-site	4 ± 1	6 ± 1	4 ± 1	12 ± 1	2.0 ± 0.5	17 ± 1
March	12N	17 ± 2	28 ± 2	16 ± 1	26 ± 2	0.8 ± 0.3	26 ± 2
	7I	9 ± 1	14 ± 1	9 ± 1	14 ± 1	0.4 ± 0.2	13 ± 1
	off-site	5 ± 1	15 ± 1	6 ± 1	12 ± 1	0.3 ± 0.2	12 ± 1
April	12N	24 ± 2	34 ± 2	17 ± 1	36 ± 2	1.9 ± 0.4	39 ± 2
	7I	9 ± 1	17 ± 1	9 ± 1	21 ± 2	0.8 ± 0.3	25 ± 2
	off-site	6 ± 1	14 ± 1	6 ± 1	15 ± 2	0.6 ± 0.3	17 ± 1
May	12N	20 ± 2	38 ± 2	24 ± 1	38 ± 2	1.2 ± 0.4	38 ± 2
	7I	6 ± 2	20 ± 2	12 ± 1	23 ± 2	0.8 ± 0.6	26 ± 2
	off-site	15 ± 1	26 ± 1	16 ± 1	23 ± 2	1.0 ± 0.3	25 ± 2
June	12N	10 ± 1	18 ± 1	11 ± 1	23 ± 2	1.1 ± 0.4	22 ± 2
	7I	7 ± 1	11 ± 1	6 ± 1	12 ± 1	0.2 ± 0.3	15 ± 1
	off-site	7 ± 1	20 ± 1	9 ± 1	20 ± 2	1.2 ± 0.4	20 ± 1
July	12N	9 ± 1	18 ± 1	10 ± 1	34 ± 2	0.6 ± 0.3	26 ± 1
	7I	7 ± 1	13 ± 1	7 ± 1	20 ± 1	0.5 ± 0.2	15 ± 1
	off-site	4 ± 1	12 ± 1	5 ± 1	11 ± 1	0.6 ± 0.2	13 ± 1
August	12N	12 ± 1	24 ± 1	14 ± 1	24 ± 2	0.4 ± 0.3	26 ± 2
	7I	2 ± 1	6 ± 1	2 ± 1	7 ± 1	0.3 ± 0.3	8 ± 1
	off-site	4 ± 1	8 ± 1	4 ± 1	8 ± 1	0.3 ± 0.2	8 ± 1
September	12N	8 ± 1	13 ± 1	8 ± 1	16 ± 1	0.7 ± 0.3	13 ± 1
	7I	2 ± 2	5 ± 2	3 ± 1	-	< 0.1	12 ± 2
	off-site	2 ± 1	4 ± 1	3 ± 1	7 ± 1	< 0.1	7 ± 1
October	12N	10 ± 1	14 ± 1	9 ± 1	17 ± 1	0.3 ± 0.2	16 ± 1
	7I	6 ± 1	12 ± 1	6 ± 1	10 ± 1	0.2 ± 0.2	10 ± 1
	off-site	10 ± 1	14 ± 1	9 ± 1	8 ± 1	0.2 ± 0.2	8 ± 1
November	12N	4 ± 1	8 ± 1	4 ± 1	13 ± 1	0.2 ± 0.4	13 ± 1
	7I	10 ± 1	12 ± 1	9 ± 1	8 ± 1	0.2 ± 0.3	10 ± 1
	off-site	7 ± 1	13 ± 1	12 ± 1	6 ± 1	0.2 ± 0.2	6 ± 1
December	12N	6 ± 1	9 ± 1	11 ± 1	22 ± 2	0.9 ± 0.3	23 ± 2
	7I	4 ± 1	5 ± 1	6 ± 1	11 ± 1	0.3 ± 0.2	12 ± 1
	off-site	8 ± 1	12 ± 1	11 ± 1	26 ± 2	1.8 ± 0.4	28 ± 2
Annual Summary	12N	14 ± 5	22 ± 7	14 ± 5	28 ± 7	1.3 ± 1.0	27 ± 7
	7I	8 ± 3	13 ± 4	8 ± 3	17 ± 6	0.8 ± 0.6	18 ± 6
	off-site	8 ± 3	15 ± 5	9 ± 3	17 ± 8	1.1 ± 0.7	18 ± 8
Percent CG ($\times 10^{-3}$)	12N	(7.0)	(7.3)	(1.4)	(0.70)	(0.03)	(0.54)
	7I	(4.0)	(4.3)	(0.8)	(0.42)	(0.02)	(0.36)
	off-site	(4.0)	(5.0)	(0.9)	(0.42)	(0.03)	(0.36)

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

[2]The concentrations in units of μg/m^3 can be obtained by multiplying the value in μCi/ml by 2.96×10^{12} for uranium-238 and by 9×10^{12} for thorium-232. The mass of the other thorium isotopes in comparison to thorium-232 and the other uranium isotopes in comparison to uranium-238 are negligible.

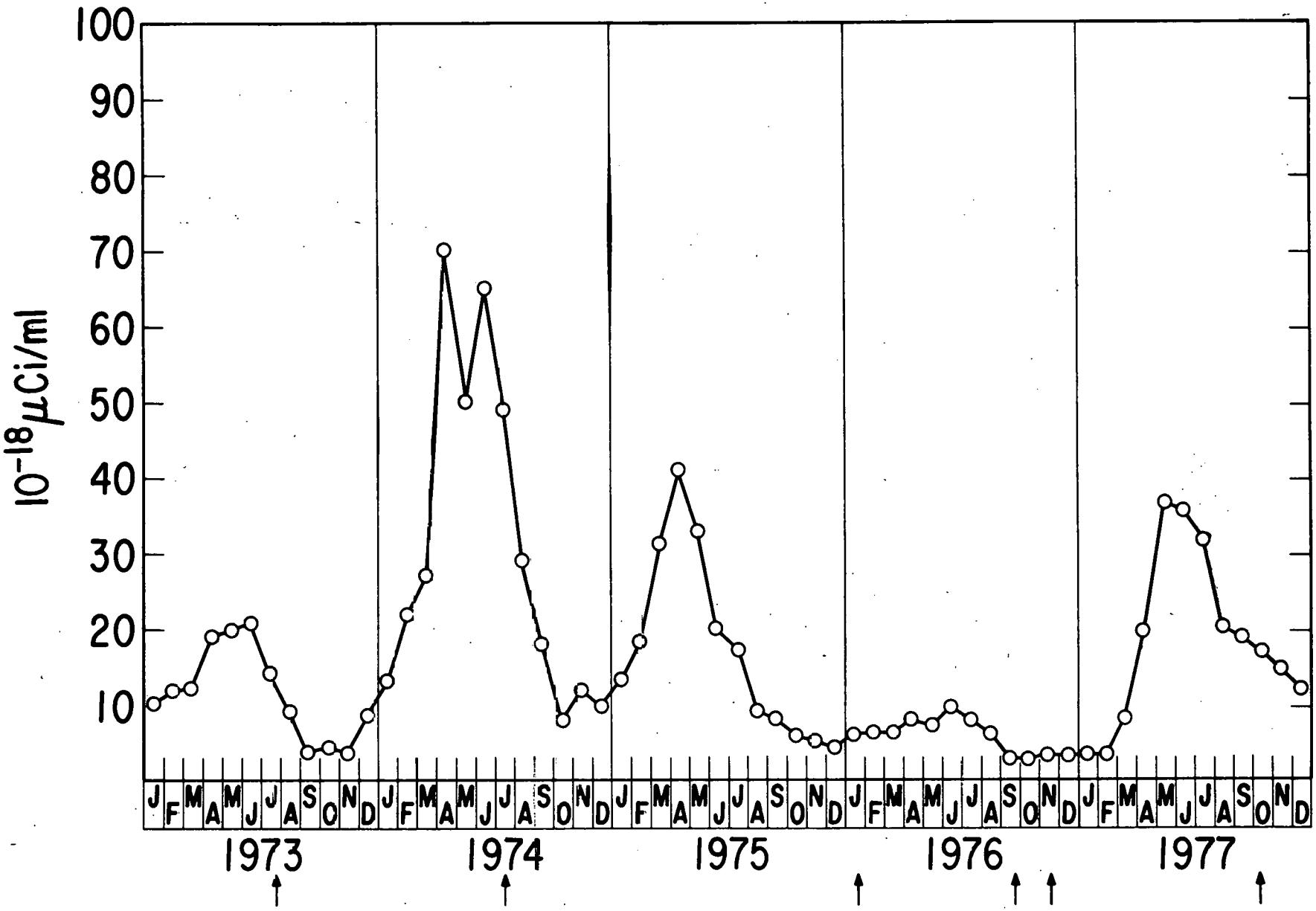

Fig. 4. Plutonium-239 Concentrations in Air

no indication that they originated from Argonne operations, and their concentrations were similar at the site perimeter and off the site. A pronounced spring peak is evident in the plutonium and strontium activities, indicating their stratospheric-fallout origin, and not in the thorium and uranium concentrations.

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 5 cm on the ground, ranged from about 1% in August to 33% in January of the total plutonium in the samples.

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. 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 8. 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 similar to previous years' results. 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 0.006% and 0.12% of the CG, respectively.

TABLE 8

Hydrogen-3 Perimeter and Off-Site Concentrations, 1977

Month	Location*	No. of Samples	Concentrations in 10^{-12} µCi/ml Av.	Min.	Max.
January	8F	8	7.6	0.1	40
	14I	8	4.2	1.9	8.3
	12M	2	5.4	3.1	7.6
	off-site	2	1.3	0.6	1.9
February	8F	8	3.0	< 0.1	12
	14I	8	8.0	0.5	31
	12M	2	3.3	3.2	3.5
	off-site	2	0.5	< 0.1	0.9
March	8F	9	3.4	0.1	15
	14I	9	6.8	0.3	44
	12M	2	13	13	13
	off-site	2	0.9	0.4	1.4
April	8F	9	26	1.0	119
	14I	9	9.7	1.0	30
	12M	2	12	5.4	19
	off-site	2	0.8	0.7	0.9
May	8F	8	47	2.4	162
	14I	8	13	1.6	44
	12M	2	4.7	1.3	8.1
	off-site	2	2.1	1.5	2.8
June	8F	9	50	1.8	240
	14I	9	12	1.0	42
	12M	2	1.6	1.0	2.1
	off-site	2	2.9	2.5	3.2
July	8F	9	14	1.0	60
	14I	9	23	0.9	56
	12M	2	14	1.0	28
	off-site	2	1.2	0.1	2.3
August	8F	9	12	< 0.1	64
	14I	8	7.4	3.6	26
	12M	2	4.7	< 0.1	9.3
	off-site	2	1.5	0.5	2.4
September	8F	8	11	1.2	31
	14I	8	8.4	2.4	26
	12M	2	7.0	1.6	12
	off-site	2	1.6	0.1	3.2
October	8F	9	10	0.4	28
	14I	9	13	1.9	39
	12M	2	3.5	2.0	5.0
	off-site	2	0.8	0.7	0.9
November	8F	9	8.0	0.5	49
	14I	9	4.6	0.3	13
	12M	2	38	26	49
	off-site	2	1.1	0.7	1.4
December	8F	8	2.8	0.4	11
	14I	9	5.3	0.4	16
	12M	2	30	6.6	53
	off-site	2	0.3	< 0.1	0.5
Annual Summary	8F	103	16 ± 9.4	< 0.1	240
	14I	103	9.6 ± 3.0	0.3	56
	12M	24	11 ± 6.6	< 0.1	53
	off-site	24	1.2 ± 0.4	< 0.1	3.2
Percent CC	8F	-	0.0080	< 0.00005	0.12
	14I	-	0.0048	0.00015	0.028
	12M	-	0.0055	< 0.00005	0.026
	off-site	-	0.0006	< 0.00005	0.0016

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

The off-site concentrations, measured about 10 km (6.2 miles) northwest of the Laboratory, were also similar to levels observed at this location in 1976. This background level of hydrogen-3 should be subtracted from the other concentrations in Table 7 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 1977, the total amount of argon-41 discharged from the reactor was estimated to be 4.0×10^4 Ci, based on a measured release rate of 1.54 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 360 Ci, based on a measured release rate of 0.041 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 1.5 Ci in 1977, was released from the Janus reactor (Building 202, location 13I). In addition, other effluents were krypton-85, estimated to be 15 Ci and elemental tritium gas, estimated to be 0.4 Ci. The other nuclides, in millicuries or smaller amounts, were various fission products. 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 concentration 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 radiochemical separations followed by appropriate counting. One liter aliquots

were used for all analyses except hydrogen-3 and the transuranium nuclides. Hydrogen-3 analyses were performed by counting 10 ml in a gel system. Plutonium and neptunium analyses were performed on 10 or 50-liter samples by a plutonium chemical separation method,[4] modified to include neptunium, followed by alpha spectrometry. Plutonium-236 was used to determine the plutonium and neptunium yields. A further modification of this procedure allowed the group separation of a fraction containing the transplutonium elements.[5] Americium-243 was added to determine chemical recovery and individual nuclides were measured by alpha spectrometry.

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) downstream from the waste-water outfall. Sawmill Creek was sampled upstream from the Argonne site and downstream from the waste-water outfall to determine if radioactivity was added to the stream by Argonne waste water or 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 85% of the year. When the continuous sampling device was not operating, a grab sample was collected each working day. Equal portions of the samples collected each week were combined and analyzed. The results obtained in this way represent the average concentrations in the weekly samples. Above the site, samples were usually collected once a month and were analyzed for the same radionuclides as the below-outfall samples.

Annual summaries of the results obtained for Sawmill Creek are given in Table 9. 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 90% for neptunium-237 and plutonium-239; 80% for hydrogen-3; and 70% for americium-241. The concentrations of all these nuclides were low compared to the CGs. The principal radionuclide added to the Creek by Argonne waste water, in terms of concentration was hydrogen-3. Its average concentration (equivalent to 0.039% of the CG) was the lowest since 1972.

TABLE 9

Radionuclides in Sawmill Creek, 1977

Type of Activity	Location*	No. of Samples	Concentration (10^{-9} µCi/ml) Av.	Min.	Max.	Percent CG Av.	Min.	Max.
Alpha	16K	12	2.5 ± 0.6	1.6	4.8	(0.083)	(0.053)	(0.16)
(nonvolatile)	7M	252	1.8 ± 0.2	0.70	4.1	(0.060)	(0.023)	(0.14)
Beta	16K	12	23 ± 4	14	32	(0.77)	(0.47)	(1.1)
(nonvolatile)	7M	252	19 ± 1	11	33	(0.63)	(0.37)	(1.1)
Hydrogen-3	16K	11	< 324	< 200	720	< 0.011	< 0.007	0.024
	7M	252	1160 ± 560	< 200	8550	0.039	< 0.007	0.28
Strontium-89	16K	12	-	-	< 2	-	-	< 0.07
	7M	252	< 2.1	< 2	6.4	< 0.07	< 0.07	0.21
Strontium-90	16K	12	< 0.52	< 0.5	0.70	< 0.17	< 0.17	0.23
	7M	252	0.54 ± 0.09	< 0.5	1.5	0.18	< 0.17	0.50
Iodine-131	16K	12	-	-	< 3	-	-	< 1
	7M	252	-	-	< 3	-	-	< 1
Barium-140	16K	12	-	-	< 2	-	-	< 0.007
	7M	137	-	-	< 2	-	-	< 0.007
Uranium**	16K	12	2.4 ± 0.8	1.2	6.4	(0.0060)	(0.0030)	(0.016)
(natural)	7M	252	2.4 ± 0.2	1.2	4.6	(0.0060)	(0.0030)	(0.012)
Neptunium-237	16K	12	< 0.001	< 0.001	0.0012	< 0.00003	< 0.00003	0.00004
	7M	247	0.017 ± 0.012	< 0.001	0.29	0.00057	< 0.00003	0.0097
Plutonium-238	16K	12	-	-	< 0.002	-	-	< 0.00004
	7M	247	< 0.003	< 0.002	0.017	< 0.00006	< 0.00004	0.00034
Plutonium-239	16K	12	< 0.00053	< 0.0005	0.00089	< 0.000011	< 0.00001	0.000018
	7M	247	0.0088 ± 0.0039	< 0.0005	0.081	0.00018	< 0.00001	0.0016
Americium-241	16K	12	-	-	< 0.001	-	-	< 0.000025
	7M	252	0.0056 ± 0.0020	< 0.001	0.035	0.00014	< 0.000025	0.00088
Curium-242 and/or Californium-252	16K	12	-	-	< 0.001	-	-	< 0.000005
	7M	252	< 0.0016	< 0.001	0.0046	< 0.000008	< 0.000005	0.000023
Curium-244 and/or Californium-249	16K	12	-	-	< 0.001	-	-	< 0.000014
	7M	252	< 0.0017	< 0.001	0.011	< 0.000024	< 0.000014	0.00016

*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 3.6 µg/l.

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 1977, the hydrogen-3 content of other lakes and streams ranged from $< 200 \times 10^{-9}$ μCi/ml to 450×10^{-9} μCi/ml and averaged $< 223 \times 10^{-9}$ 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 added 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 6×10^{-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 as in 1976, while the fallout contribution increased by a factor of three. The total concentration, regardless of source, must be used in assessing the health hazard of a radionuclide not naturally present, and the percent of the CGs for all nuclides listed in Table 8 was low.

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 values are 10 Ci of hydrogen-3, 0.1 mCi of neptunium-237, 0.08 mCi of plutonium-239, 0.05 mCi of americium-241, and $<$ 0.05 mCi of curium and californium nuclides.

A pathway study was conducted to examine the distribution of plutonium and neptunium discharged into Sawmill Creek. Samples were collected of water, suspended sediments, algae, and fish and analyzed for plutonium and neptunium. The suspended sediment was separated into two fractions by filtration, coarse (particle sizes greater than 2.5 μm) and fine (particle sizes between 2.5 and 0.45 μm.) Water was collected at the point the Laboratory discharges its

waste water into Sawmill Creek and at various points downstream.

The water and sediment data (Table 10) indicate that the loss by sedimentation is greater for plutonium than for neptunium. Because of the relatively high velocity of the stream to a point about 200 m below the outfall, loss of both elements from the stream is by association with the coarse sediment fraction. Beyond the 200 m point, the stream velocity slows sufficiently so that the loss of plutonium and neptunium with fine sediment dominates.

Analysis of algae and fish samples for plutonium-239 and neptunium-237 and a comparison of their concentrations (wet weight) to the average annual concentrations of these nuclides in Sawmill Creek water yielded the following concentration factors (CF). In algae, CFs were 465 for plutonium-239 and 195 for neptunium-237. In fish samples, the CFs were 850 for plutonium-239 and 280 for neptunium-237. The fish sample was a composite of several species.

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 11. The average nonvolatile alpha and uranium concentrations in the River were very similar to the 1976 averages, and although higher than earlier years, still remain in the normal range. The beta activities were about 30% higher than 1976 due to increased fallout from nuclear testing. This is evident from the results obtained for fission product concentrations. 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, 6×10^{-9} μCi/ml, was due to fallout. The average nonvolatile alpha and beta activities, 1.0×10^{-9} μCi/ml and 11×10^{-9} μCi/ml, respectively, of 23 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 12, were similar to those found in other bodies of water in the area and to the

TABLE 10

Distribution of ^{239}Pu and ^{237}Np
Between Water and Suspended Sediment

(Concentrations in 10^{-12} μCi/ml)

Data Collected	Location	Fraction*	^{239}Pu	^{237}Np
October 10	Outfall (7M)	Water	4.5 (19%)	44.7 (90%)
		Fine Sediment	2.9 (12%)	< 1 -
		Coarse Sediment	16.6 (69%)	4.2 (10%)
			24.0	48.9
October 10	200 m Below Outfall	Water	4.0 (24%)	22.8 (72%)
		Fine Sediment	1.8 (11%)	< 1 -
		Coarse Sediment	10.9 (65%)	8.2 (28%)
			16.7	31.0
November 23	Outfall (7M)	Water	57.8 (32%)	82.6 (68%)
		Fine Sediment	15.5 (9%)	4.7 (4%)
		Coarse Sediment	107.2 (59%)	33.6 (28%)
			180.5	120.9
November 23	30 m Below Outfall	Water & Fines	35.2 (35%)	62.4 (68%)
		Coarse Sediment	66.4 (65%)	28.7 (32%)
			101.6	91.1
November 23	200 m Below Outfall	Water & Fines	7.1 (31%)	9.3 (48%)
		Coarse Sediment	15.7 (69%)	10.2 (52%)
			22.8	19.5
November 23	500 m Below Outfall	Water	3.3 (20%)	17.7 (68%)
		Fine Sediment	9.8 (60%)	6.2 (24%)
		Coarse Sediment	3.3 (20%)	2.0 (8%)
			16.4	25.9

*Suspended sediment results are given in terms of the volume of water filtered.

TABLE 11

Radionuclides in Des Plaines River Water, 1977

Type of Activity	Location*	No. of Samples	Concentration (10^{-9} µCi/ml) Av.	Min.	Max.	Percent CG Av.	Min.	Max.
Alpha	A	12	1.9 ± 0.4	1.0	3.3	(0.063)	(0.033)	(0.11)
(nonvolatile)	B	23	1.9 ± 0.3	0.87	4.1	(0.063)	(0.029)	(0.14)
Beta	A	12	20 ± 4	7.7	35	(0.67)	(0.26)	(1.17)
(nonvolatile)	B	23	20 ± 2	12	33	(0.67)	(0.40)	(1.10)
Hydrogen-3	A	12	< 267	< 200	490	< 0.0089	< 0.007	0.016
	B	23	< 223	< 200	435	< 0.0074	< 0.007	0.014
Strontium-89	A	12	< 2.4	< 2	5.9	< 0.08	< 0.07	0.20
	B	23	< 2	< 2	6.1	< 0.07	< 0.07	0.20
Strontium-90	A	12	< 0.56	< 0.5	0.73	< 0.19	< 0.17	0.24
	B	23	< 0.57	< 0.5	1.1	< 0.19	< 0.17	0.37
Iodine-131	A	12	< 3.3	< 3	6.3	< 1.1	< 1	2.1
	B	23	< 3.2	< 3	6.7	< 1.1	< 1	2.2
Barium-140	A	12	< 2.3	< 2	5.1	< 0.0077	< 0.007	0.017
	B	14	< 2.2	< 2	4.8	< 0.0073	< 0.007	0.016
Uranium**	A	12	1.9 ± 0.3	1.3	2.9	(0.0048)	(0.0032)	(0.0072)
(natural)	B	23	2.3 ± 0.3	1.0	4.1	(0.0058)	(0.0025)	(0.010)
Neptunium-237	A	11	< 0.001	< 0.001	0.0015	< 0.00003	< 0.00003	0.00005
	B	12	< 0.0011	< 0.001	0.0020	< 0.00004	< 0.00003	0.00007
Plutonium-238	A	11	-	-	< 0.002	-	-	< 0.00004
	B	12	-	-	< 0.002	-	-	< 0.00004
Plutonium-239	A	11	< 0.00056	< 0.0005	0.00084	< 0.000011	< 0.00001	0.000017
	B	11	< 0.00061	< 0.0005	0.00095	< 0.000012	< 0.00001	0.000019
Americium-241	A	12	-	-	< 0.001	-	-	< 0.000025
	B	12	< 0.001	< 0.001	0.0012	< 0.000025	< 0.000025	0.00003
Curium-242 and/or	A	12	-	-	< 0.001	-	-	< 0.000005
Californium-252	B	12	< 0.0011	< 0.001	0.0019	< 0.0000055	< 0.000005	0.000048
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 3.1 µg/l.

TABLE 12

Radionuclides in Illinois River Water, 1977

(Concentrations in 10^{-9} µCi/ml)

Location	Date Collected	Alpha*	Beta*	Hydrogen-3	Uranium**	Neptunium-237	Plutonium-239
McKinley Woods State Park	June 14	0.92	13	< 200	0.87	< 0.0005	0.00028
Below Dresden Power Station	June 14	0.89	7.6	< 200	1.2	< 0.0005	0.00066
Morris	June 14	0.13	6.8	< 200	1.0	-	-
Starved Rock State Park	June 14	0.56	9.9	< 200	1.2	-	-
McKinley Woods State Park	October 6	1.0	15	< 200	1.5	< 0.0005	0.00048
Below Dresden Power Station	October 6	1.3	6.9	< 200	1.3	< 0.0005	0.00026
Morris	October 6	1.6	8.0	< 200	1.3	-	-
Starved Rock State Park	October 6	1.0	7.4	< 200	1.3	-	-

*Nonvolatile activity.

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

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 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[6] 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. This is a departure from past practice. 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 13 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.

TABLE 13

Plutonium Content of Soil, 1977

Date Collected	Location	Plutonium-238 10^{-9} µCi/g	Plutonium-238 10^{-3} µCi/m^2	Plutonium-239 10^{-9} µCi/g	Plutonium-239 10^{-3} µCi/m^2	$^{238}Pu/^{239}Pu$
	Perimeter*					
June 16	10E	1.1 ± 0.1	0.05 ± 0.01	15.2 ± 0.5	0.68 ± 0.02	0.070
June 16	14I	0.9 ± 0.1	0.04 ± 0.01	18.5 ± 0.5	0.89 ± 0.03	0.046
June 16	14N	1.0 ± 0.2	0.06 ± 0.01	19.3 ± 0.5	1.03 ± 0.04	0.055
June 16	9N	0.8 ± 0.1	0.04 ± 0.01	9.0 ± 0.4	0.45 ± 0.02	0.084
June 16	6I	1.1 ± 0.1	0.06 ± 0.01	17.4 ± 0.5	1.03 ± 0.03	0.062
November 15	8N	0.5 ± 0.1	0.04 ± 0.01	8.0 ± 0.4	0.53 ± 0.03	0.066
November 15	7EF	0.7 ± 0.4	0.04 ± 0.02	11.7 ± 1.4	0.58 ± 0.07	0.063
November 15	13N	0.7 ± 0.3	0.03 ± 0.02	11.8 ± 1.4	0.56 ± 0.06	0.061
November 15	14E	0.6 ± 0.4	0.03 ± 0.02	8.8 ± 1.2	0.42 ± 0.06	0.064
	Average	0.8 ± 0.1	0.04 ± 0.01	13.3 ± 2.9	0.69 ± 0.16	0.063 ± 0.007
	Off-Site					
June 14	Channahon, IL	0.7 ± 0.1	0.04 ± 0.01	13.9 ± 0.8	0.86 ± 0.05	0.047
June 14	Morris, IL	0.8 ± 0.1	0.06 ± 0.01	18.3 ± 0.9	1.42 ± 0.07	0.044
June 14	Starved Rock State Park, IL	0.5 ± 0.1	0.03 ± 0.01	11.0 ± 0.6	0.70 ± 0.04	0.048
June 15	Lemont, IL	0.6 ± 0.1	0.03 ± 0.01	14.1 ± 0.7	0.72 ± 0.04	0.040
June 15	Romeoville, IL	1.0 ± 0.2	0.04 ± 0.01	11.2 ± 0.5	0.48 ± 0.02	0.087
October 6	McKinley Woods State Park, IL	0.8 ± 0.2	0.05 ± 0.01	15.7 ± 0.9	1.03 ± 0.06	0.048
October 6	Dresden Lock and Dam, IL	1.1 ± 0.5	0.06 ± 0.03	15.3 ± 1.9	0.85 ± 0.10	0.073
October 27	Saganashkee Slough, IL	0.7 ± 0.1	0.04 ± 0.01	12.9 ± 0.5	0.81 ± 0.03	0.053
October 27	McGinnis Slough, IL	0.5 ± 0.1	0.03 ± 0.01	14.7 ± 0.6	0.78 ± 0.03	0.037
	Average	0.7 ± 0.1	0.04 ± 0.01	14.1 ± 1.5	0.85 ± 0.17	0.053 ± 0.011

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

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 14 along with results since 1973 for comparison. The total 1977 deposition by precipitation was about three times that of 1976, a reflection of the increased fallout from atmospheric nuclear tests, and was equivalent to 0.5% of the total plutonium previously deposited, which is reported to be 2.2×10^{-3} $\mu Ci/m^2$.[7] The data in Table 14 is illustrated in Figure 5. The arrows at the bottom of the figure indicate the approximate dates of the Chinese atmospheric nuclear tests. Comparison of the results in Figure 5 with the plutonium air concentrations for the past five years in Figure 4 shows excellent agreement. A log-normal distribution plot of the monthly plutonium deposition results in a straight line best fit, indicating a single plutonium source, evidently stratospheric fallout.

The thorium, uranium, and gamma-ray emitters in soil samples were also measured, and the concentrations in the perimeter and off-site samples (Table 15) were similar. These are expected levels of the naturally-occurring thorium and uranium activities and nuclides attributed to fallout. In terms of mass, the average thorium concentrations were 1.9 µg/g and 2.2 µg/g at the perimeter and off the site, respectively, while the uranium concentrations were 2.0 µg/g at the perimeter and 2.6 µg/g off-site.

The results of radioactivity measurements in grass are given in Tables 16 and 17. 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. The presence and concentration of some of the short-lived fission products strongly depends on the time of sample collection relative to an atmospheric test.

TABLE 14

Ground Deposition of Plutonium-239,240

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

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

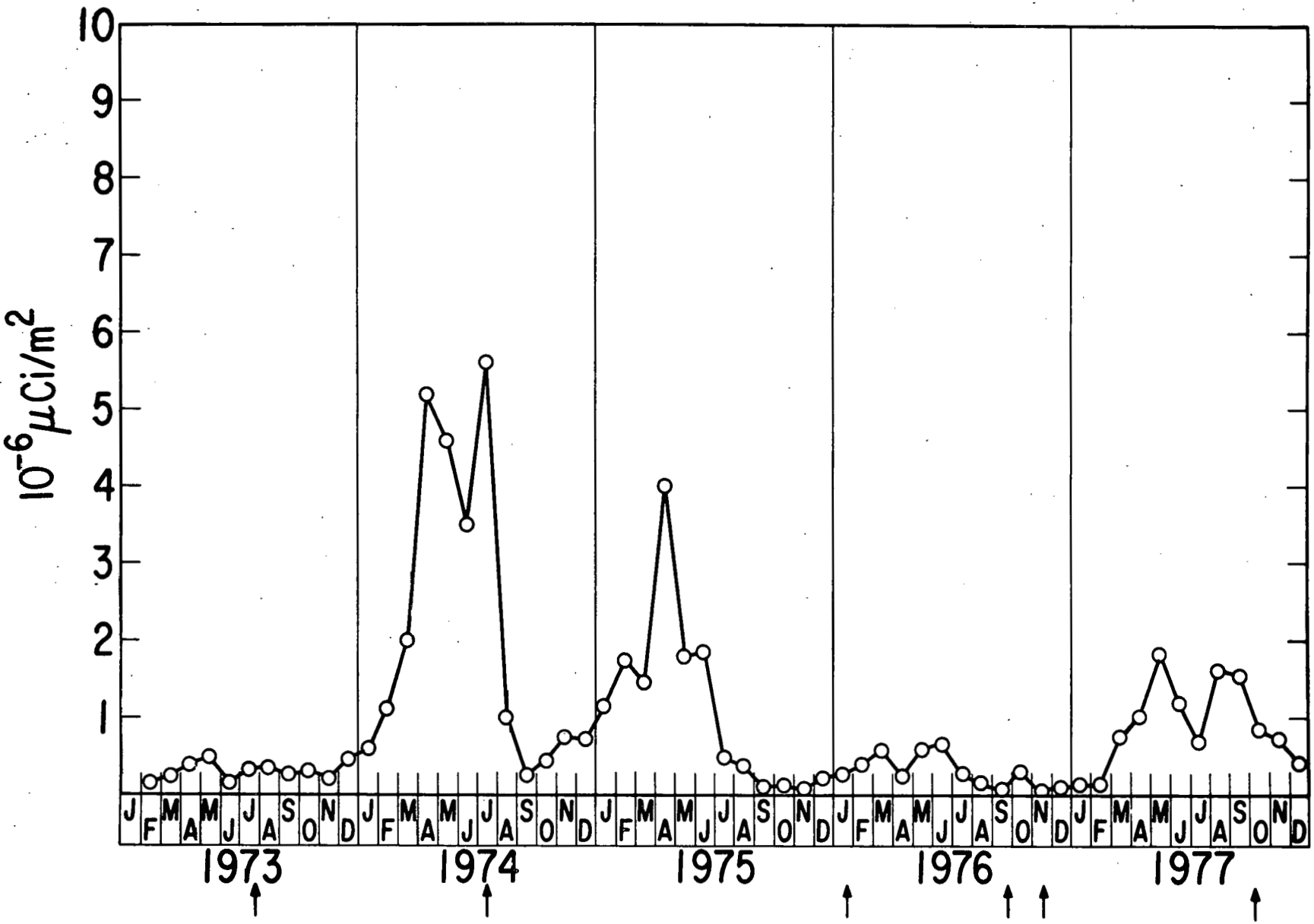

Fig. 5. Plutonium-239 Surface Deposition

TABLE 15

Radionuclides in Soil, 1977

(Concentrations in 10^{-6} μCi/g)

Date Collected	Location	Zirconium-95-Niobium-95	Cesium-137	Cerium-141	Cerium-144	Thorium-228	Thorium-230	Thorium-232	Uranium (natural)
	Perimeter*								
June 16	10E	0.2 ± 0.1	1.0 ± 0.3	< 0.1	0.1 ± 0.1	0.36 ± 0.01	0.46 ± 0.01	0.34 ± 0.01	1.6 ± 0.1
June 16	14I	0.3 ± 0.1	0.9 ± 0.3	< 0.1	< 0.1	0.24 ± 0.01	0.29 ± 0.01	0.23 ± 0.01	1.8 ± 0.1
June 16	14N	0.5 ± 0.2	1.1 ± 0.3	< 0.1	< 0.1	0.19 ± 0.01	0.19 ± 0.01	0.15 ± 0.01	1.2 ± 0.1
June 16	9N	0.2 ± 0.1	0.5 ± 0.2	< 0.1	< 0.1	0.36 ± 0.03	0.48 ± 0.03	0.36 ± 0.03	1.6 ± 0.1
June 16	6I	< 0.1	1.0 ± 0.3	< 0.1	0.1 ± 0.1	0.39 ± 0.04	0.29 ± 0.04	0.33 ± 0.04	1.1 ± 0.1
November 15	8N	0.2 ± 0.1	0.4 ± 0.1	< 0.1	< 0.1	0.22 ± 0.01	0.29 ± 0.01	0.25 ± 0.01	1.1 ± 0.1
November 15	7EF	0.4 ± 0.1	1.1 ± 0.3	0.1 ± 0.1	0.3 ± 0.1	0.05 ± 0.01	0.06 ± 0.01	0.07 ± 0.01	0.9 ± 0.1
November 15	13N	0.3 ± 0.1	1.1 ± 0.3	0.1 ± 0.1	0.2 ± 0.1	0.05 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	1.4 ± 0.1
November 15	14E	0.3 ± 0.1	0.9 ± 0.3	0.1 ± 0.1	0.2 ± 0.1	0.07 ± 0.01	0.09 ± 0.01	0.07 ± 0.01	1.2 ± 0.1
	Average	0.3 ± 0.1	0.9 ± 0.2	< 0.1	0.1 ± 0.1	0.21 ± 0.09	0.24 ± 0.11	0.21 ± 0.08	1.3 ± 0.2
	Off-Site								
June 14	Channahon, IL	0.1 ± 0.1	0.9 ± 0.3	< 0.1	0.3 ± 0.1	-	-	-	1.3 ± 0.1
June 14	Morris, IL	0.2 ± 0.1	1.2 ± 0.4	0.1 ± 0.1	0.1 ± 0.1	0.18 ± 0.02	1.02 ± 0.06	0.32 ± 0.03	3.0 ± 0.2
June 14	Starved Rock State Park, IL	0.2 ± 0.1	0.6 ± 0.2	0.1 ± 0.1	0.3 ± 0.1	0.25 ± 0.01	0.32 ± 0.01	0.26 ± 0.01	0.5 ± 0.1
June 16	Lemont, IL	0.4 ± 0.1	0.7 ± 0.2	0.1 ± 0.1	0.1 ± 0.1	0.34 ± 0.01	0.51 ± 0.01	0.38 ± 0.01	1.2 ± 0.1
June 16	Romeoville, IL	0.3 ± 0.1	0.5 ± 0.2	0.1 ± 0.1	0.1 ± 0.1	0.12 ± 0.02	0.13 ± 0.02	0.17 ± 0.02	3.4 ± 0.2
October 6	McKinley Woods State Park, IL	0.2 ± 0.1	1.0 ± 0.3	0.2 ± 0.1	0.3 ± 0.1	0.13 ± 0.01	0.17 ± 0.01	0.13 ± 0.01	1.0 ± 0.1
October 6	Dresden Lock and Dam, IL	0.3 ± 0.1	1.2 ± 0.4	0.1 ± 0.1	0.2 ± 0.1	-	-	-	2.0 ± 0.1
October 27	Saganashkee Slough, IL	0.3 ± 0.1	0.8 ± 0.3	0.1 ± 0.1	0.1 ± 0.1	0.20 ± 0.01	0.39 ± 0.01	0.23 ± 0.01	1.6 ± 0.1
October 27	McGinnis Slough, IL	0.2 ± 0.1	0.9 ± 0.3	< 0.1	< 0.1	0.14 ± 0.01	0.28 ± 0.01	0.19 ± 0.01	1.5 ± 0.1
	Average	0.2 ± 0.1	0.9 ± 0.2	0.1 ± 0.03	0.2 ± 0.1	0.19 ± 0.06	0.40 ± 0.23	0.24 ± 0.07	1.7 ± 0.6

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

TABLE 16

Plutonium Content of Grass Samples, 1977

Date Collected	Location	Plutonium-238 10^{-9} µCi/g	Plutonium-238 10^{-6} µCi/m^2	Plutonium-239 10^{-9} µCi/g	Plutonium-239 10^{-6} µCi/m^2
	Perimeter*				
June 16	10E	0.12 ± 0.08	0.027 ± 0.018	0.84 ± 0.15	0.189 ± 0.034
June 16	14I	0.11 ± 0.09	0.016 ± 0.013	1.02 ± 0.17	0.153 ± 0.026
June 16	14N	< 0.01	< 0.001	0.34 ± 0.10	0.065 ± 0.019
June 16	9N	0.23 ± 0.19	0.046 ± 0.038	0.78 ± 0.20	0.155 ± 0.040
June 16	6I	0.57 ± 0.22	0.062 ± 0.024	0.98 ± 0.25	0.107 ± 0.027
November 15	8N	0.73 ± 0.16	0.106 ± 0.023	2.52 ± 0.28	0.365 ± 0.041
November 15	7EF	0.39 ± 0.10	0.093 ± 0.024	2.72 ± 0.25	0.650 ± 0.060
November 15	13N	0.47 ± 0.13	0.130 ± 0.036	2.24 ± 0.26	0.618 ± 0.072
November 15	14E	1.55 ± 0.36	0.339 ± 0.079	1.20 ± 0.32	0.263 ± 0.070
	Average	0.46 ± 0.31	0.091 ± 0.068	1.40 ± 0.57	0.285 ± 0.144
	Off-Site				
June 14	Channahon, IL	0.13 ± 0.08	0.020 ± 0.012	0.91 ± 0.15	0.137 ± 0.023
June 14	Morris, IL	0.08 ± 0.08	0.006 ± 0.006	0.72 ± 0.14	0.054 ± 0.011
June 14	Starved Rock State Park, IL	0.02 ± 0.07	0.002 ± 0.008	0.93 ± 0.15	0.109 ± 0.018
June 16	Lemont, IL	< 0.01	< 0.001	0.64 ± 0.12	0.100 ± 0.019
June 16	Romeoville, IL	0.03 ± 0.06	0.005 ± 0.011	0.87 ± 0.13	0.172 ± 0.026
October 6	McKinley Woods State Park, IL	0.86 ± 0.37	0.109 ± 0.047	0.99 ± 0.38	0.126 ± 0.048
October 6	Dresden Lock and Dam, IL	2.68 ± 0.54	0.541 ± 0.109	3.03 ± 0.57	0.612 ± 0.115
October 27	Saganashkee Slough, IL	1.28 ± 0.29	0.114 ± 0.026	3.20 ± 0.45	0.285 ± 0.040
October 27	McGinnis Slough, IL	0.98 ± 0.28	0.130 ± 0.037	2.66 ± 0.43	0.354 ± 0.057
	Average	0.67 ± 0.60	0.103 ± 0.115	1.55 ± 0.72	0.217 ± 0.117

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

TABLE 17

Radionuclides in Grass Samples, 1977

Date Collected	Location	Concentrations in 10^{-6} µCi/g						Concentrations in 10^{-9} µCi/g			
		^{95}Zr-^{95}Nb	^{103}Ru	^{137}Cs	^{140}Ba-^{140}La	^{141}Ce	^{144}Ce	^{228}Th	^{230}Th	^{232}Th	Uranium (natural)
	Perimeter*										
June 16	10E	4.0 ± 0.6	0.4 ± 0.1	0.1 ± 0.1	< 0.1	0.4 ± 0.1	1.6 ± 0.4	2 ± 1	3 ± 1	2 ± 1	29 ± 5
June 16	14I	4.2 ± 0.6	0.4 ± 0.1	0.1 ± 0.1	< 0.1	0.3 ± 0.1	2.2 ± 0.4	1 ± 1	3 ± 1	2 ± 1	15 ± 3
June 16	14N	0.8 ± 0.2	< 0.1	< 0.1	< 0.1	< 0.1	0.5 ± 0.2	1 ± 1	1 ± 1	1 ± 1	20 ± 3
June 16	9N	4.2 ± 0.6	< 0.1	< 0.1	< 0.1	< 0.1	1.5 ± 0.4	1 ± 1	1 ± 1	1 ± 1	23 ± 3
June 16	6I	3.7 ± 0.5	< 0.1	0.1 ± 0.1	< 0.1	0.5 ± 0.1	1.4 ± 0.3	3 ± 1	2 ± 1	2 ± 1	18 ± 4
November 15	8N	5.4 ± 0.7	0.7 ± 0.2	0.2 ± 0.1	1.4 ± 0.3	2.1 ± 0.4	3.6 ± 0.5	2 ± 1	1 ± 1	1 ± 1	45 ± 4
November 15	7EF	3.3 ± 0.5	0.4 ± 0.1	0.1 ± 0.1	< 0.1	1.3 ± 0.3	2.1 ± 0.4	4 ± 1	1 ± 1	1 ± 1	26 ± 2
November 15	13N	4.9 ± 0.6	0.8 ± 0.2	0.2 ± 0.1	1.5 ± 0.3	2.2 ± 0.4	3.5 ± 0.5	2 ± 1	1 ± 1	1 ± 1	36 ± 4
November 15	14E	3.0 ± 0.5	2.3 ± 0.4	< 0.1	0.9 ± 0.3	1.3 ± 0.3	1.5 ± 0.4	1 ± 1	1 ± 1	1 ± 1	36 ± 4
	Average	3.7 ± 0.9	0.6 ± 0.5	0.1 ± 0.04	< 0.5	0.9 ± 0.6	2.0 ± 0.7	2 ± 1	2 ± 1	1 ± 0.3	28 ± 7
	Off-Site										
June 14	Channahon, IL	3.3 ± 0.5	0.4 ± 0.1	0.4 ± 0.1	< 0.1	< 0.1	1.4 ± 0.3	5 ± 1	8 ± 1	5 ± 1	29 ± 4
June 14	Morris, IL	1.8 ± 0.4	< 0.1	< 0.1	< 0.1	< 0.1	0.6 ± 0.2	4 ± 1	10 ± 1	5 ± 1	13 ± 3
June 14	Starved Rock State Park, IL	1.5 ± 0.3	0.2 ± 0.1	0.1 ± 0.1	< 0.1	< 0.1	0.9 ± 0.3	5 ± 1	9 ± 1	6 ± 1	62 ± 8
June 16	Lemont, IL	1.8 ± 3	< 0.1	0.1 ± 0.1	< 0.1	< 0.1	0.6 ± 0.2	1 ± 1	2 ± 1	1 ± 1	37 ± 4
June 16	Romeoville, IL	2.0 ± 0.3	< 0.1	< 0.1	< 0.1	< 0.1	1.1 ± 0.3	3 ± 1	4 ± 1	3 ± 1	25 ± 6
October 6	McKinley Woods State Park, IL	4.0 ± 0.6	0.7 ± 0.2	0.1 ± 0.1	< 0.1	2.6 ± 0.4	1.2 ± 0.3	3 ± 1	2 ± 1	1 ± 1	64 ± 4
October 6	Dresden Lock & Dam, IL	5.8 ± 0.7	1.3 ± 0.3	0.1 ± 0.1	13.5 ± 1.4	4.1 ± 0.6	2.9 ± 0.4	3 ± 1	1 ± 1	1 ± 1	46 ± 4
October 27	Saganashkee Slough, IL	14.3 ± 1.5	1.8 ± 0.4	0.4 ± 0.1	10.4 ± 1.0	7.0 ± 0.8	4.7 ± 0.6	4 ± 1	8 ± 1	4 ± 1	83 ± 6
October 27	McGinnis Slough, IL	9.7 ± 1.0	1.4 ± 0.3	0.2 ± 0.1	4.3 ± 0.6	5.7 ± 0.7	4.0 ± 0.6	6 ± 1	9 ± 1	4 ± 1	54 ± 4
	Average	4.9 ± 2.9	0.7 ± 0.4	0.2 ± 0.1	< 3.2	< 2.2	1.9 ± 1.0	4 ± 1	6 ± 2	3 ± 1	46 ± 15

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

Bottom sediment samples were analyzed for thorium, uranium, plutonium, and gamma-ray emitters for comparison. The results are given in Tables 18 and 19. Plutonium results vary widely between locations and are strongly dependent on the retentiveness of the bottom material. In 1975, a bottom sediment sample from the pond at 15H indicated slightly elevated concentrations of plutonium. Comparison of the results of the four 15H pond samples collected on August 24, 1977, with the two off-site samples collected the same day from similar types of ponds demonstrates that the concentrations in the sediment of 15H are in the normal range. In general, the average concentrations of all the nuclides that were measured are similar. The uranium and thorium concentrations were normal and similar to those found in soil. In terms of mass, the uranium concentrations were 4.0 μg/g at the perimeter and 2.2 μg/g off the site, while the thorium concentrations were 2.3 μg/g and 1.8 μg/g at the perimeter and off-site, respectively.

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 1×10^{-10} μCi/ml by a batch ion-exchange separation followed by beta counting. Cesium-137 was analyzed with a detection limit of 5×10^{-10} μCi/ml by an ion-exchange separation followed by gamma-ray spectrometry. The other nuclides were analyzed by the same methods used for water and with the same detection limits. The results are given in Table 20. The concentration of hydrogen-3 in milk for all samples was less than the detection limit. The average strontium-90 concentration was similar to 1976, while the average cesium-137 concentration increased by about 35%. These nuclides are fission products from nuclear tests and their presence in milk is not related to Argonne operations. Strontium-89, iodine-131, and barium-140 concentrations above the detection limit and the increase in the cesium-137 and strontium-90 concentration late in the fall are attributed to the previously discussed atmospheric nuclear tests.

The concentrations given in Table 20 may be compared to the CGs for drinking water given in the Appendix, Part B. The drinking water CGs are based on an intake of 2.2 liters per day. The consumption of one liter of milk per day would result in an average intake of 0.5% of the strontium-90, and 0.007% of the cesium-137 Concentration Guides.

TABLE 18

Alpha-Particle Emitting Nuclides in Bottom Sediment, 1977

Date Collected	Location	Concentrations in 10^{-6} µCi/g				Concentrations in 10^{-9} µCi/g		
		Thorium-228	Thorium-230	Thorium-232	Uranium (natural)	Plutonium-238	Plutonium-239	$^{238}Pu/^{239}Pu$
	Perimeter*							
August 24	Inlet 15H Pond	0.19 ± 0.01	0.30 ± 0.01	0.19 ± 0.01	2.3 ± 0.1	0.4 ± 0.1	6.3 ± 0.3	0.071
August 24	South Center 15H Pond	0.32 ± 0.01	0.45 ± 0.01	0.32 ± 0.01	2.6 ± 0.1	2.1 ± 0.3	28.0 ± 1.0	0.075
August 24	North Center 15H Pond	0.30 ± 0.01	0.49 ± 0.02	0.30 ± 0.01	3.9 ± 0.2	1.5 ± 0.2	31.8 ± 1.3	0.048
August 24	Outlet 15H Pond	0.25 ± 0.01	0.38 ± 0.01	0.25 ± 0.01	2.1 ± 0.1	1.1 ± 0.2	21.2 ± 1.1	0.053
	Average	0.26 ± 0.06	0.41 ± 0.08	0.26 ± 0.06	2.7 ± 0.8	1.3 ± 0.7	22 ± 11	0.062 ± 0.013
	Off-Site							
June 14	DuPage River, Channahon, IL	0.26 ± 0.01	0.28 ± 0.01	0.27 ± 0.01	1.2 ± 0.1	0.3 ± 0.1	1.0 ± 0.2	-
June 16	Long Run Creek, Lemont, IL	0.15 ± 0.01	0.22 ± 0.01	0.16 ± 0.01	0.8 ± 0.1	0.3 ± 0.1	2.4 ± 0.2	-
August 24	Pond, Lemont Rd. at 76th St., IL	0.21 ± 0.01	0.35 ± 0.01	0.20 ± 0.01	2.4 ± 0.2	2.2 ± 0.2	45.3 ± 1.8	0.049
August 24	Pond, 83rd St. & Clynderven Rd., IL	0.28 ± 0.01	0.55 ± 0.01	0.27 ± 0.01	2.1 ± 0.1	1.3 ± 0.2	15.0 ± 0.9	0.087
October 6	DuPage River, Channahon, IL	0.17 ± 0.01	0.22 ± 0.02	0.19 ± 0.01	1.2 ± 0.1	0.6 ± 0.1	0.4 ± 0.1	-
October 27	Des Plaines River, Brookfield, IL	0.19 ± 0.01	0.27 ± 0.01	0.15 ± 0.01	1.6 ± 0.1	2.0 ± 0.2	10.6 ± 0.7	-
October 27	Salt Creek, Western Springs, IL	0.11 ± 0.01	0.23 ± 0.01	0.19 ± 0.01	1.1 ± 0.1	0.4 ± 0.1	0.7 ± 0.1	-
	Average	0.20 ± 0.04	0.30 ± 0.09	0.20 ± 0.04	1.5 ± 0.4	1.0 ± 0.6	10.8 ± 12.3	0.068 ± 0.038

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

TABLE 19

Gamma-ray Emitting Nuclides in Bottom Sediment, 1977

(Concentrations in 10^{-6} μCi/g)

Date Collected	Location	Zirconium-95-Niobium-95	Ruthenium-103	Ruthenium-106	Antimony-125	Cesium-137	Cerium-141	Cerium-144
	Perimeter*							
August 24	Inlet 15H Pond	0.3 ± 0.1	< 0.1	< 0.1	< 0.1	0.2 ± 0.1	< 0.1	< 0.1
August 24	South Center 15H Pond	0.5 ± 0.2	< 0.1	< 0.1	< 0.1	1.9 ± 0.4	< 0.1	0.5 ± 0.2
August 24	North Center 15H Pond	0.6 ± 0.2	< 0.1	< 0.1	0.2 ± 0.1	2.8 ± 0.4	0.1 ± 0.1	0.2 ± 0.1
August 24	Outlet 15H Pond	2.7 ± 0.4	0.2 ± 0.1	0.6 ± 0.2	0.1 ± 0.1	2.2 ± 0.4	0.1 ± 0.1	1.2 ± 0.3
	Average	1.0 ± 1.1	< 0.1	< 0.2	< 0.1	1.8 ± 1.1	< 0.1	0.5 ± 0.5
	Off-Site							
June 14	DuPage River, Channahon, IL	1.8 ± 0.4	< 0.1	< 0.1	< 0.1	0.1 ± 0.1	< 0.1	< 0.1
June 16	Long Run Creek, Lemont, IL	0.2 ± 0.1	< 0.1	< 0.1	< 0.1	0.1 ± 0.1	< 0.1	< 0.1
August 24	Pond, Lemont Rd. at 76th St., IL	1.7 ± 0.4	0.2 ± 0.1	0.7 ± 0.2	< 0.1	3.3 ± 0.5	< 0.1	0.8 ± 0.2
August 24	Pond, 83rd St. & Clynderven Rd., IL	0.4 ± 0.1	< 0.1	< 0.1	< 0.1	0.9 ± 0.3	< 0.1	0.5 ± 0.2
October 6	DuPage River, Channahon, IL	0.1 ± 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.1 ± 0.1	< 0.1
October 27	Des Plaines River, Brookfield, IL	1.2 ± 0.3	0.3 ± 0.1	< 0.1	< 0.1	0.6 ± 0.2	0.3 ± 0.1	0.5 ± 0.2
October 27	Salt Creek, Western Springs, IL	0.2 ± 0.1	< 0.1	< 0.1	< 0.1	0.1 ± 0.1	< 0.1	< 0.1
	Average	0.8 ± 0.6	< 0.1	< 0.2	< 0.1	0.7 ± 0.9	< 0.1	< 0.3

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

TABLE 20

Radionuclides in Milk, 1977

(Concentrations in 10^{-9} μCi/ml)

Date Collected	Hydrogen-3	Strontium-89	Strontium-90	Iodine-131	Cesium-137	Barium-140
January 5	< 200	< 2	2.6	< 0.1	1.7	< 2
February 2	< 200	< 2	1.4	< 0.1	1.6	< 2
March 2	< 200	< 2	1.9	< 0.1	1.2	< 2
April 7	< 200	< 2	2.3	< 0.1	2.0	< 2
May 4	< 200	< 2	1.8	< 0.1	2.3	< 2
June 1	< 200	4.3	4.5	< 0.1	2.7	< 2
July 6	< 200	5.5	5.5	< 0.1	3.0	< 2
August 5	< 200	< 2	2.9	< 0.1	5.3	< 2
September 7	< 200	< 2	3.3	< 0.1	3.8	< 2
October 5	< 200	22.6	5.6	37.4	4.2	15.8
November 2	< 200	13.1	4.5	2.9	6.1	5.7
December 7	< 200	< 2	2.7	< 0.1	5.9	< 2
Average	< 200	< 5.2	3.2 ± 0.8	< 3.5	3.3 ± 1.0	< 3.5

Several samples of garden vegetables grown 8-16 km (5-10 miles) from Argonne were obtained and analyzed for plutonium, uranium, and thorium. The results are given in Table 21, 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 1975 and 1976. The plutonium content, although extremely variable, covered about the same range of concentrations as in the last two 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.

5. External Penetrating Radiation

Measurements were made with calcium fluoride (dysprosium activated) thermoluminescent dosimeter (TLD) chips. Each measurement was the average of three or four chips exposed in the same packet. The response 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 obtained at the boundary. Readings were also taken at five off-site locations for comparison purposes.

The results are summarized in Tables 22 and 23, 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 55 to 78 days, and in total covered the period from January 4, 1977, to January 5, 1978. 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 calculated from the standard error.

TABLE 21

Radionuclides in Garden Vegetables, 1977

Food	Concentration (10^{-9} µCi/g)				Plutonium-239	
	Thorium-228	Thorium-230	Thorium-232	Uranium (natural)	Concentration (10^{-12} µCi/g)	CF
Beets	0.6 ± 0.2	1.2 ± 0.2	0.6 ± 0.1	4.4 ± 0.4	16 ± 9	6×10^{-4}
Cabbage	0.1 ± 0.1	0.1 ± 0.1	0.1 ± 0.1	2.7 ± 0.2	9 ± 3	4×10^{-4}
Corn (kernels)	1.4 ± 0.1	2.9 ± 0.8	2.3 ± 0.8	1.5 ± 0.6	< 35	$< 1.4 \times 10^{-3}$
Tomatoes	0.1 ± 0.1	0.2 ± 0.1	0.1 ± 0.1	2.1 ± 0.2	3 ± 2	1×10^{-4}

TABLE 22

Environmental Penetrating Radiation at Off-Site Locations, 1977

Location	Dose Rate (mrem/year)						
	Period of Measurement						
	1/4-3/7	3/7-5/24	5/24-7/19	7/19-9/16	9/16-11/10	11/10-1/5	Average
Downers Grove	87	90	84	90	97	95	90 ± 5
Lockport	82	99	90	99	101	96	95 ± 7
Lombard	95	97	89	96	95	89	94 ± 4
Oak Lawn	83	82	74	84	84	83	82 ± 4
Oakbrook	97	95	103	100	102	100	99 ± 3
Average	89 ± 6	93 ± 6	88 ± 9	94 ± 6	96 ± 6	93 ± 6	92 ± 6

TABLE 23

Environmental Penetrating Radiation at ANL, 1977

Location	Dose Rate (mrem/year)						
	Period of Measurement						
	1/4-3/7	3/7-5/24	5/24-7/19	7/19-9/16	9/16-11/10	11/10-1/5	Average
14L - Boundary	85	85	83	84	80	85	84 ± 2
14I - Boundary	297	259	238	126	128	133	201 ± 76
14G - Boundary	92	95	87	93	94	88	92 ± 3
13D - Boundary	68	70	64	62	69	67	67 ± 3
9/10 EF - Boundary	92	93	95	94	91	92	93 ± 1
8H - Boundary	97	94	105	94	94	91	96 ± 5
8H - Center, St. Patrick's Cemetery	100	104	102	107	107	102	104 ± 3
7I - Boundary	194	179	186	255	235	188	206 ± 31
6I - 200 m N of Quarry Road	103	100	105	102	100	93	101 ± 4
9L - Boundary	84	82	82	73	83	83	81 ± 4
9H - 50 m SE of CP-5	2040	1910	1810	1810	1700	1620	1820 ± 150
8H - 65 m S of 316	177	158	198	140	174	124	162 ± 27
9H - 23 m E of 316	430	347	613	312	463	230	374 ± 134
9I - 45 m NE of 350 210 m NE of 316	109	100	106	93	113	106	104 ± 7
8H - 200 m NW of Waste Storage Area (Heliport)	203	134	141	139	140	115	146 ± 30
7I - Center, Waste Storage Area	2060	2090	2470	4260	3200	1950	2640 ± 900
10/11K - Lodging Facilities	82	81	82	73	80	88	81 ± 5
9J - Between ZGS Condenser and 386	98	88	93	67	88	91	88 ± 5
13IJ - 75 m E of 202	272	265	-	-	-	-	268 ± 15
13J - 140 m NE of 202	1570	1410	1280	766	785	868	1110 ± 350
12M - 30 m W of 55	141	115	102	82	84	121	108 ± 23

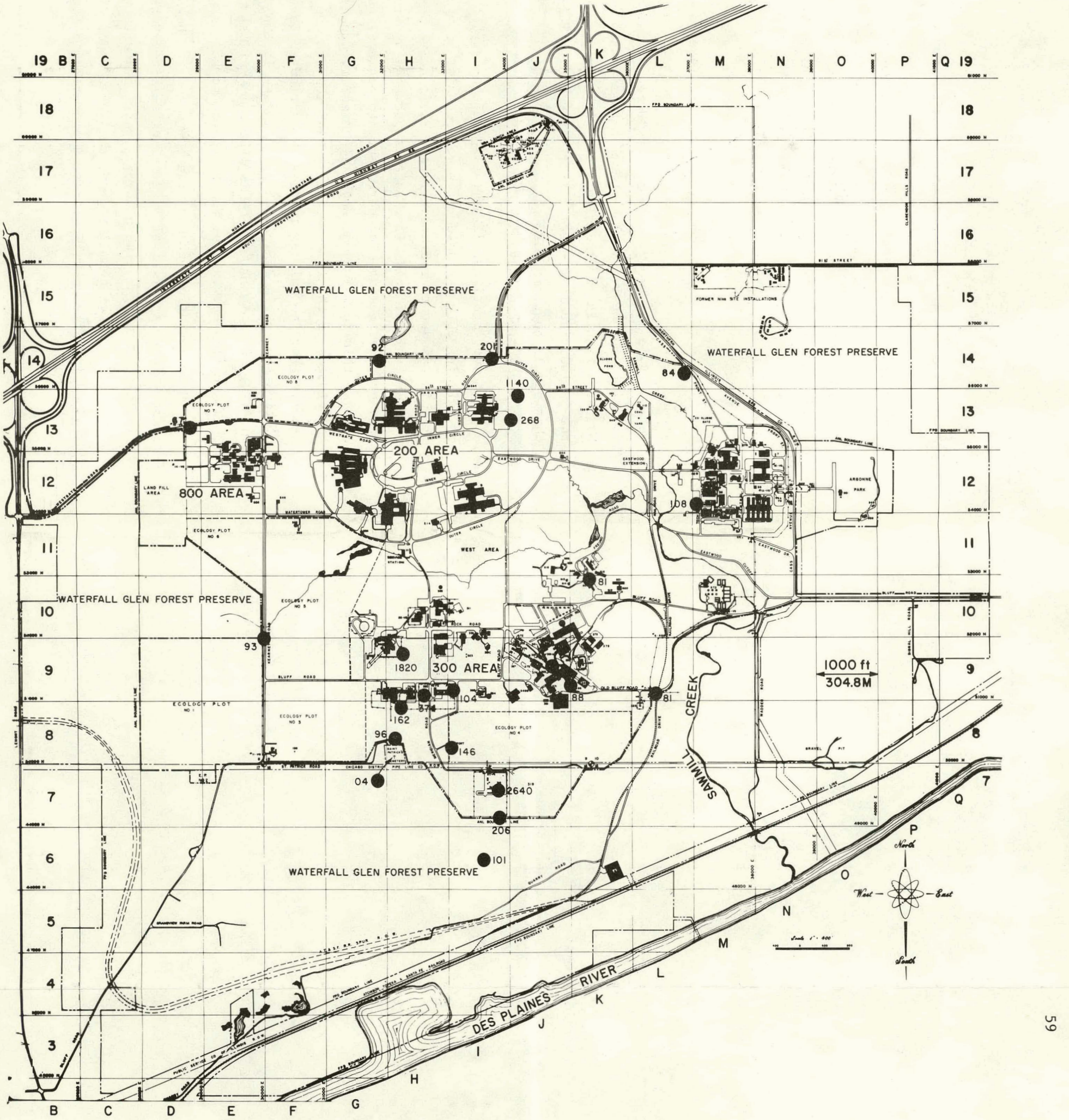

Fig. 6. Penetrating Radiation Measurements at the ANL Site, 1977

The off-site results averaged 92 mrem/yr. The standard error of the mean of all 30 individual results was 1.4 mrem/yr. (The standard error of the average of the five separate locations was 6 mrem/yr.) From 1972 to 1976 the corresponding averages and standard errors varied from 90 ± 2 mrem/yr (1976) to 105 ± 2 mrem/yr (1972). The five-year average was 98 ± 3 mrem/yr. The 1977 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 have been within 5 mrem/yr of the average of all five off-site locations (usually within 2 mrem/yr). At Oak Lawn the results have been from 8 to 11 mrem/yr below, and at Oakbrook the results have been 7 to 11 mrem/yr above, the overall average.

If the off-site locations are an accurate sample of the radiation background in the area, then annual averages at the site boundary ranging from 80 to 104 (92 ± 12) 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 7.4 mrem/yr, so that single boundary results in the range of 92 ± 15 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 114 mrem/yr, considerably less than in 1976 and similar to the 1975 value. At 14I the average dose rate was about 119 mrem/yr above normal. 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 difference in the measured dose rates at this location between the first and second halves of the year.

Dose variations from period to period at the same location, which are at least in part statistical, and the 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 (92 mrem/yr) plus twice the standard error in the average, 7 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 sub-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 about 12 mrem/yr above the off-site average but within the upper limit (106 mrem/yr) of the normal range. Results for some individual periods were at the 95% upper limit for such measurements, 107 mrem/yr. An upper limit for the excess dose at 8H is about 10 mrem/yr, based on this type of comparison, although statistically the dose at this location may be considered normal. 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 to the dose at 8H is considered negligible since dose rates measured in other directions from CP-5 at the same distance were less than at 8H. The contribution from Building 316 is also considered negligible since the dose at the Cemetery did not decrease when the dose close to the building decreased after the reactor in the building was provided with additional shielding. Since the dose at the center of the Cemetery averaged 8 mrem/yr above that at the boundary, 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, the conversion of concentration to dose was based on the ratio of environmental concentrations to the Concentration Guides given in Table 24. 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 making 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 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 released 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.[8] The following parameters were used in the calculations:

a) release rates (measured in the CP-5 exhaust stack): argon-41, 1.54 Ci/MW-hr; hydrogen-3, 0.041 Ci/hr.
b) metorological data: the wind velocity data 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 25. Doses were

TABLE 24

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.(9)

TABLE 25

Argon-41 Radiation Dose From CP-5 Reactor, 1977

(millirem/year)

Sector	Distance 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	4.2	10.4	1.9	0.8
NNE	5.6	13.9	2.6	1.1
NE	4.7	11.6	2.2	0.9
ENE	5.5	13.8	2.6	1.1
E	6.8	17.0	3.2	1.3
ESE	4.2	10.5	2.0	0.8
SE	2.8	6.9	1.3	1.2
SSE	1.7	4.2	0.8	0.3
S	1.4	3.5	0.6	0.3
SSW	2.0	4.9	0.9	0.4
SW	4.1	10.2	1.9	0.8
WSW	2.7	6.9	1.3	0.5
W	2.3	5.7	1.1	0.4
SNW	1.7	4.3	0.8	0.3
NW	2.7	6.6	1.1	0.5
NNW	3.2	8.0	1.5	0.6
Average	3.5	8.7	1.6	0.7

calculated for the mid-point of the annular interval. Thus, the dose for 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 receiving the largest dose, 3 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 5.6 mrem/yr if they were outdoors throughout the year at 1.6 km and 1.2 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 directions, including the predominant wind directions to the east and northeast, 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 92 ± 15 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 15 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 26, 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 at 1.5 km (0.93 mile), in the NNE direction, is 0.011 mrem/yr. The maximum individual exposure 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 0.007 mrem/yr. A summary of the results is given in Table 27. A comparison of the doses

calculated from the meteorological model with the measured data from Table 8 is given in Table 28. In both cases concentrations are converted to dose as described earlier. The agreement is good in view of the large number of variables and parameters involved in obtaining both the calculated and measured values.

TABLE 26

Argon-41 Average Individual and Population Dose From CP-5 Reactor, 1977

Distance (km)	Population	Dose Avg. mrem/year Argon-41	Natural	Man-rem/year Argon-41	Natural
0-1.6	0	-	-	-	-
1.6-3.2	3,595	1.6	92	5.8	331
3.2-4.8	17,405	0.7	92	12.2	1601
0-80	8,118,740	0.022	92	177.	7.17×10^5

TABLE 27

Hydrogen-3 Average Individual and Population Dose From CP-5 Reactor, 1977

Distance (km)	Population	Avg. mrem/year	Man-rem/year
0-1.6	0	-	-
1.6-3.2	3,595	0.0027	0.0096
3.2-4.8	17,405	0.0018	0.031
0-80	8,118,740	0.00014	1.13

TABLE 28

Comparison of Calculated and Measured Hydrogen-3 Dose Rates, 1977

Direction	Distance (km)	Calculated (mrem/yr)	Measured (mrem/yr)
NNE	1.5	0.010	0.021
ENE	1.9	0.006	0.024
SW	0.45	0.030	0.037

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 radioactivity attributable to Argonne operations could be found in off-site water was Sawmill Creek below the waste-water outfall. The nuclides added to Sawmill Creek by Argonne waste water, their 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 29. In the case of strontium-90, a small fraction (5%) was contributed by Argonne and the remainder by fallout, but the total concentration is included in the dose calculation since the total exposure is the figure of interest, regardless of source. 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 29

Radionuclide Concentrations and Dose Estimates
for Sawmill Creek Water, 1977

Nuclide	Conc. (avg.) 10^{-9} µCi/ml	Dose Rate mrem/year	Percent of Standard
Hydrogen-3	1.16×10^{3}	0.19	0.039
Strontium-90	0.54	5.4	0.18
Neptunium-237	0.017	0.017	0.0006
Plutonium-239	0.0088	0.0053	0.002
Americium-241	0.0056	0.0021 (kidney) 0.0034 (bone)	0.00014 0.00011
Curium-244 and/or Californium-249	< 0.0017	< 0.0007 or < 0.0013	< 0.00002 or < 0.00004

As indicated in Table 9, occasional Creek samples (less than 10) contained traces of plutonium-238 and curium-242 and/or californium-252, but the

averages were only slightly greater than the detection limit. The annual dose due to 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 are: 2×10^{-3} mrem/yr for plutonium-238 and from 3.6×10^{-4} to 2.4×10^{-5} mrem/yr for the other transplutonium nuclides.

b. External Penetrating Radiation

Above normal fence-line doses attributable to Argonne operations were found at the south boundary adjacent to the Waste Storage Facility (location 7I), at the north boundary near Building 202 (14I), and possibly 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 114 mrem/yr. Approximately 300 m (0.2 mile) south of the fence line the measured dose had decreased to within the normal range, 101 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 (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 0.003 mrem/yr, if the energy of the radiation was 0.66 MeV, and 0.014 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 about 12 mrem/yr. An individual spending an average of 1 hr/week at this location would receive an annual dose of about 0.007 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 119 mrem/yr, the nearest residents are 750 m (0.47 mile) to the north-northwest. The dose at that location (calculated as described above) was 0.20 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.[3] 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 0.04% of the 500 mrem/yr limit or 0.12% 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 individuals living NE of the site.

B. Chemical Pollutants

1. Water

The nonradioactive environmental water data contained in this report have been collected 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 Regulations, Chapter 3.[10] Stream Quality standards appear in Part II and Effluent Quality standards appear in Part IV of this document.

The determination of manganese was performed using conventional atomic absorption spectrophotometry, and results for this element are included for the first time. The other analytical techniques were previously described.[11]

The results of the measurement of chemical constituents are expressed as milligrams (mg) or micrograms (μg) per liter (l). Averages were calculated as described in Section III.A. Yearly averages are reported with a (±) 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 the 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 30. 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, since they depend on the solubility of the electrode material used.

As in the past, the major emphasis has been placed on Sawmill Creek, a tributary of the Des Plaines River, since this is the principal route for waste water leaving the Argonne site. A major effort was devoted to studying cooling tower blowdown effluents and a continuing emphasis was placed on 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 is approximately 3 megaliters (800,000 gallons) per day and in 1977 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.[12]

Release of chemical pollutants from the waste treatment plant was monitored on a continuous basis during the work week. A flow proportional 24-hour

TABLE 30

Water Quality Standards and Detection Limits

(Concentrations in mg/l)

Constituent	State Standard Stream	State Standard Effluent	Analytical Detection Limit
Ammonia Nitrogen (as N)	1.5	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	0.1
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
Cynanide	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
Mercury	0.0005	0.0005	0.0001
Nickel	1.0	1.0	0.05
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

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 31. The averages for January, February, and April exceeded the State of Illinois standard for ammonia nitrogen. The five-day biochemical oxygen demand and dissolved solid values were well below the standards.

TABLE 31

Performance of Sanitary Waste Treatment Plant, 1977

	Concentrations (mg/l)		
Month	$B.O.D._5$	Ammonia Nitrogen	Suspended Solids
January	4.9	14.7	2.00
February	3.5	12.2	2.75
March	6.4	2.6	3.11
April	7.5	3.5	4.75
May	1.6	0.4	4.29
June	1.6	0.1	1.50
July	3.4	0.3	2.44
August	3.6	-	-
September	1.4	0.6	6.44
October	1.6	2.0	4.00
November	4.2	0.7	1.00
December	2.6	1.7	2.40
State Limit	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 32. All of the average concentrations were below the State standards except mercury. The average concentration of mercury was 0.5 µg/l (100% of the State standard) which is higher than 1976 by 14%. However, one sample was extremely high (28.4 µg/l) and raised the average from 0.4 to 0.5 µg/l. The source of this release was not discovered. Studies to prevent future releases of this type are continuing. The level of hexavalent chromium was reduced by about a factor of three from 1976. The reduction was due in part to changes in treatment procedures which use non-chromate materials, although increased precipitation was also a factor. Levels for beryllium, cadmium, fluoride, iron, lead, nickel, silver, and zinc remained essentially the same as in the past. The levels of copper were increased approximately 30% from 1976.

Results obtained for the cooling tower effluents are shown in Table 33. Average chromium concentrations at all sampling locations were substantially lower than results obtained in 1976 with the exception of 14H. The primary reason for the decrease in these levels is as previously described. The stream at 11L contained trivalent chromium as well as silver. The chromium (III) results from reduction of hexavalent chromium by film processing chemicals, which are intermittently discharged together with silver, from film processing areas of the ZGS Complex. Samples at this location averaged 97% of the State standard for hexavalent chromium and exceeded this value 33% of the time. Although average concentrations of trivalent chromium and silver were well below State limits, levels of silver in the first quarter of the year were higher than normal. The efficiency of the silver removal system was improved, and this change reduced the silver levels during the last three quarters of the year.

Samples collected at 12L, 12F (a new location), and 14G never exceeded the State standard and averaged less than 3% of this value. The average level at location 14H exceeded the State standard and individual samples exceeded this value 14% of the time. Investigation of the flow patterns at this site revealed that the results obtained were due to a very low water flow. When high chromium(VI) levels were found, they persisted for long periods and were, in fact, more a measure of lack of dilution rather than continuous injection of chromium(VI) into the stream.

TABLE 32

Chemical Constituents in Effluent From ANL Treatment Plant, 1977

Constituent	No. of Samples	Concentration (mg/l or µg/l) Avg.	Min.	Max.	Percent of Standard (Avg.)	Percent Exceeding State Standard
Barium	56	< 0.1	-	-	< 5	0
Beryllium*	12	0.059 ± 0.016	0.022	0.104	-	-
Cadmium*	56	1.1 ± 0.2	0.3	2.4	0.7	0
Chromium(VI)	252	0.020 ± 0.002	< 0.01	0.082	6.7	0
Chromium(III)	56	0.013 ± 0.002	< 0.01	0.038	1.3	0
Copper*	56	25 ± 6	9	115	2.5	0
Fluoride	56	0.39 ± 0.06	0.27	1.90	2.6	0
Iron	56	0.20 ± 0.04	0.08	0.42	10	0
Lead*	56	< 2	< 2	6.2	< 2	0
Manganese*	43	14 ± 4	< 10	69	1.4	0
Mercury*	252	0.5 ± 0.2	< 0.1	28.36	100	16
Nickel	56	< 0.5	< 0.05	0.6	< 5	0
pH	252	-	6.60	8.15	-	0
Silver*	56	1.7 ± 0.6	< 0.2	4.8	1.7	0
Zinc	56	0.14 ± 0.03	0.05	0.51	14	0

*Concentrations in µg/l.

TABLE 33

Cooling Tower Effluents, 1977

Constituent	Location	No. of Samples	Concentration (mg/l or µg/l) Avg.	Min.	Max.	Percent of Standard (Avg.)	Percent Exceeding State Standard
Chromium(VI)	8J	52	0.15 ± 0.08	< 0.01	1.58	50	15
Chromium(VI)	11L	43	0.29 ± 0.09	< 0.01	1.22	97	33
Chromium(III)	11L	43	0.05 ± 0.03	< 0.01	0.49	5	0
Silver*	11L	43	12.7 ± 7.3	0.2	147	12.7	2.3
Chromium(VI)	12L	46	< 0.01	< 0.01	0.02	< 3	0
Chromium(VI)	12F	48	< 0.01	-	-	< 3	0
Chromium(VI)	14G	51	< 0.01	-	-	< 3	0
Chromium(VI)	14H	51	0.78 ± 0.80	< 0.01	16.74	260	14
Chromium(VI)	14J	52	0.07 ± 0.04	< 0.01	0.88	23	3.8

*Concentration in µg/l.

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)]. Additionally, samples to be examined for fecal coliform were collected at least five times per month at the 7M (up) location, but downstream the samples were collected at the same frequency 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 34. The average level for ammonia nitrogen upstream is 2.8 times the State of Illinois standard and individual samples exceeded this value 50% of the time. The downstream sample averaged 153% of the State standard and exceeded this value 40% of the time. All of the samples obtained downstream that exceeded the standard were due to upstream contamination. The dissolved oxygen levels obtained during 1977 averaged 107% and 102% 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 1976.

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 May, August, September, October, and December and there were nine instances when the 400 organisms/100 ml standard was exceeded. The sample obtained at 16K exceeded the 400 organisms/100 ml standard in 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 three occasions. These latter high levels were due to excessive chlorine demand (or requirements) at the treatment plant, and the cause was located and remedial action taken.

TABLE 34

Sawmill Creek - Effect of Sanitary Waste, 1977

Constituent	Location	No. of Samples	Concentration (mg/l) Avg.	Min.	Max.	Percent of Standard	Percent Exceeding State Standard
Ammonia Nitrogen	7M (up)	52	4.2 ± 1.1	< 0.1	18.5	280	50
	7M (down)	52	2.3 ± 0.7	< 0.1	10.3	153	40
Dissolved Oxygen	7M (up)	49	107 ± 9*	-	-	-	-
	7M (down)	49	102 ± 8*	-	-	-	-
Total Dissolved Solids	7M (up)	52	1196 ± 117	591	1851	120	77
	7M (down)	52	904 ± 38	586	1239	90	17

*Percent saturation.

Samples to evaluate the effect of combined sanitary and laboratory waste on the concentrations of chemical constituents in Sawmill Creek were collected five times per week. These were the same samples taken for radioactivity analyses (Section III.A.2.).

Results for the chemical constituents in Sawmill Creek are summarized in Table 35. Individual samples for hexavalent chromium at 7M (down) exceeded the State standard 1% of the time, while the standard was not exceeded in samples upstream of the outfall. Upstream samples contain contributions from effluents 11L, 12L, 14J, and 14G, as described earlier. Individual samples exceeded the State standard for mercury 6% of the time at the 7M (down) location, and the source is the Argonne outfall. However, this year's average is a factor of two less than in 1976 due to careful surveillance and treatment of effluent water containing mercury. The State standard for silver was exceeded in 22% of the samples. The source is the 11L effluent channel which contains photographic waste. The iron standard was exceeded in 26% of the samples. Much of this iron resulted from soil erosion during periods of heavy precipitation. As can be seen from the Argonne effluent levels in Table 35, the iron source must be in the stream area since effluent levels for iron were much lower. Individual samples exceeded the standard for copper 28% of the time, and it is believed that the copper is contributed largely by plumbing. Levels for cyanide exceeded the State standard in 7% of the samples. In at least one case, the upstream sample was as high as the downstream sample. The source is not known. The State has proposed raising the cyanide standard to 0.3 mg/l, and no sample was greater than 15% of this projected level. 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.

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, manganese, total iron, and total zinc. The results are in Table 36. Two of the samples had very high levels of suspended material, as evidenced by iron levels of 2.2 and 9.0 mg/l. In no case was there any indication that hexavalent chromium or mercury levels were affected by the Argonne effluent.

TABLE 35

Chemical Constituents in Sawmill Creek, 1977

Constituent	Location*	No. of Samples	Concentration (mg/l or µg/l) Avg.	Min.	Max.	Percent of Standard (Avg.)	Percent Exceeding State Standard
Barium	7M	54	< 0.1	< 0.1	0.2	< 2	0
Beryllium**	7M	12	0.076 ± 0.026	0.022	0.155	-	-
Cadmium**	7M	54	3.0 ± 0.4	0.8	8.2	6	0
Chromium(VI)	7M (Up)	22	< 0.01	-	-	< 20	0
	7M (Down)	199	< 0.01	< 0.01	0.09	< 20	1
Chromium(III)	7M	199	0.01 ± 0.001	< 0.01	0.05	1	0
Copper**	7M	54	18.3 ± 2.4	8	57	90	28
Cyanide	7M	52	< 0.02	< 0.02	0.04	< 80	7
Fluoride	7M	54	0.50 ± 0.03	0.33	0.85	36	0
Iron	7M	54	0.86 ± 0.17	0.17	2.56	86	26
Lead**	7M	54	4.7 ± 1.1	< 2.0	15.2	4.7	0
Manganese	7M	41	0.12 ± 0.03	0.01	0.30	12	0
Mercury**	7M (Up)	26	< 0.10	-	-	< 20	0
	7M (Down)	199	0.17 ± 0.03	< 0.01	1.61	34	6
Nickel	7M	54	< 0.05	-	-	< 5	0
pH	7M	199	-	7.38	8.78	-	0
Silver**	7M	54	4.6 ± 1.7	< 0.2	34.2	92	24
Zinc	7M	54	0.09 ± 0.01	0.04	0.2	9	0

*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.

**Concentrations in µg/l.

TABLE 36

Chemical Constituents in the Des Plaines River, 1977

Constituent	Location*	No. of Samples	Concentration (mg/l or µg/l) Avg.	Min.	Max.
Chromium (VI)	A	12	< 0.1	-	-
	B	22	< 0.1	-	0.01
Iron	A	12	1.63 ± 1.40	0.19	8.95
	B	22	0.93 ± 0.26	0.17	2.20
Manganese	A	9	0.13 ± 0.02	0.10	0.17
	B	17	0.12 ± 0.02	0.07	0.20
Mercury**	A	12	< 0.1	-	0.13
	B	22	< 0.1	-	-
Zinc	A	12	0.15 ± 0.09	0.02	0.58
	B	22	0.05 ± 0.01	0.03	0.16

*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/l.

2. Air

Studies were conducted in 1977 to assess background levels of particulate matter in air at Argonne prior to the beginning of several large scale coal-burning experiments. Studies of lead and beryllium levels in air have been conducted for several years to establish 1) the effect of ambient lead levels on the aquatic systems, and 2) to determine the effect of Argonne beryllium operations on the environment.

Samples for particulate matter determination were collected on glass fiber filters (20 cm x 25 cm) at four separate locations. High volume samplers were used (General Metals Corp.) and samples were collected for seven-day periods. The air flow rate varied from 1.27-1.70 m^3/min., depending on the sampler design, and was verified using a Root's 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. All weighings were conducted in this same dry box.

Weekly samples were collected at one location to determine ambient lead levels. The samples were collected using Millipore paper (0.8 μm pore size) at a flow rate of about 2 liters per minute. It has been shown that flow rates greater than this value, using this collection procedure, cause low results for lead. The filters were dissolved in nitric acid and analyzed for lead using conventional atomic absorption spectrophotometry. Appropriate blanks and standards were similarly treated.

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.

The results from these studies are shown in Table 37. The geometric mean (Gm) is reported since this best described the distribution of the data obtained. Average values for air particulates ranged from 43 μg/m^3 at 12M to 58 μg/m^3 at 12F. The data reported represent different time periods since sampling was established as equipment became available. Thus, samples at 18J and 12F included most of the spring and summer when particulate levels were high, while sampling at 12M began in late September when the particulate levels were decreasing.

TABLE 37

Total Particulate, Beryllium, and Lead in Air

Constituent	Location	Concentration ($\mu g/m^3$) Avg. (Gm)	Min.	Max.
Particulate	18J	52	32	102
	12F	58	35	129
	8F	47	31	75
	12M	43	31	68
Beryllium	12M	1.6×10^{-4}	0.6×10^{-4}	3.3×10^{-4}
Lead	12M	0.39	0.13	0.88

In general, ambient air particulate data are best described by a log-normal distribution, and the data obtained at 18J is presented in Figure 7 in this form. The straight line is a least squares fit. The data at the other three locations were similarly treated and the same results obtained.

The values obtained for beryllium are similar to results obtained previously and are in the range of values to be expected in ambient air, 1×10^{-4} $\mu g/m^3$ to 4×10^{-4} $\mu g/m^3$. The values obtained for lead in air are lower than previous years, which probably represents the effect of lower lead content of gasoline. A log-normal distribution plot of the lead results is shown in Figure 8. As can be seen, an excellent fit is obtained using this presentation. The levels of particulates, beryllium, and lead found in ambient air at Argonne are considered normal for this area and do not indicate an effect of Laboratory operations.

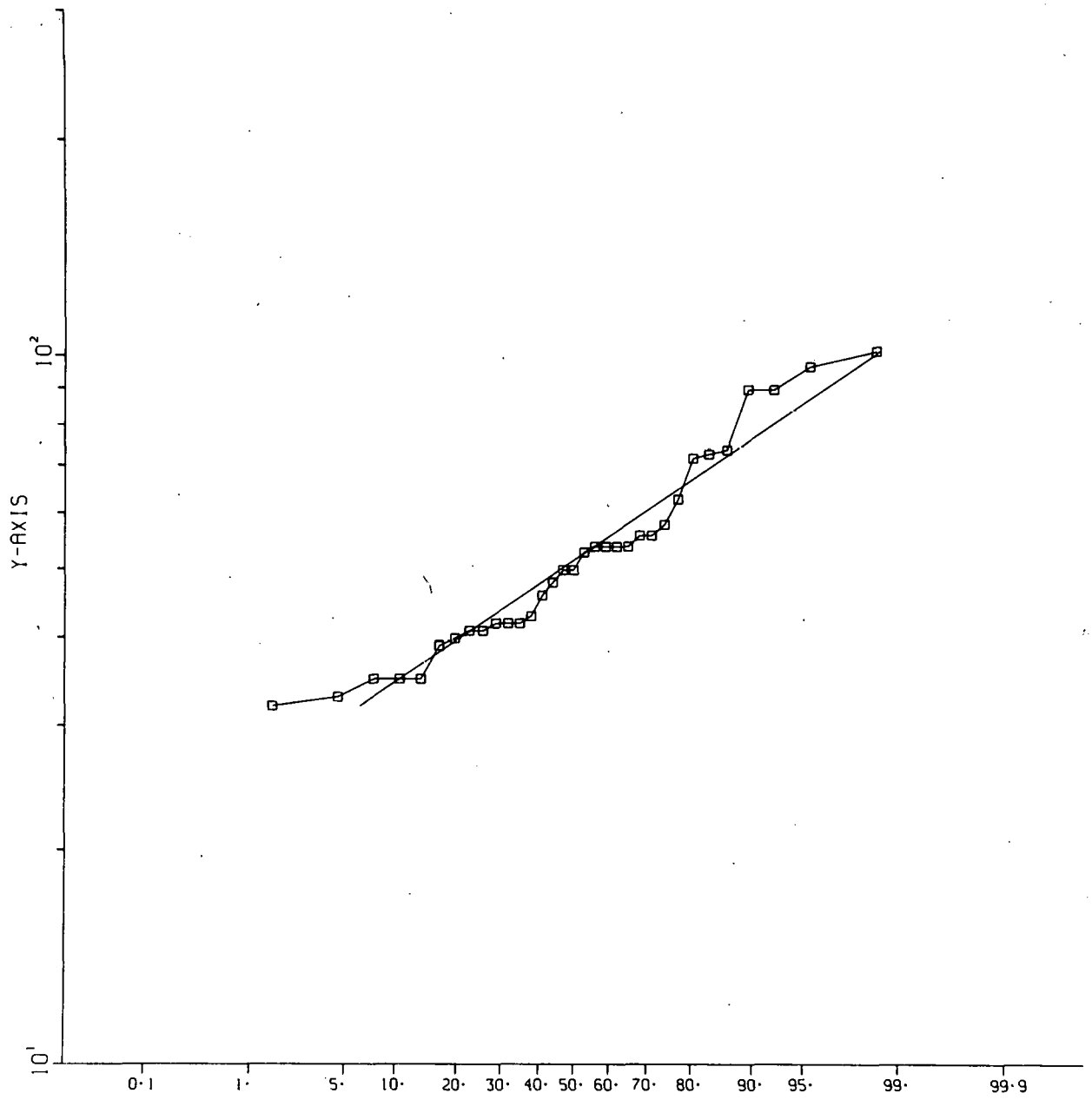

Fig. 7. Log-normal Distribution Plot of Particulate Matter in Air at ANL, 1977. Geometric mean - 51.6 $\mu g/m^3$; geometric standard deviation - 1.34 $\mu g/m^3$; arithmetic mean - 54.2 $\mu g/m^3$. The straight line is a least-squares fit to the data points.

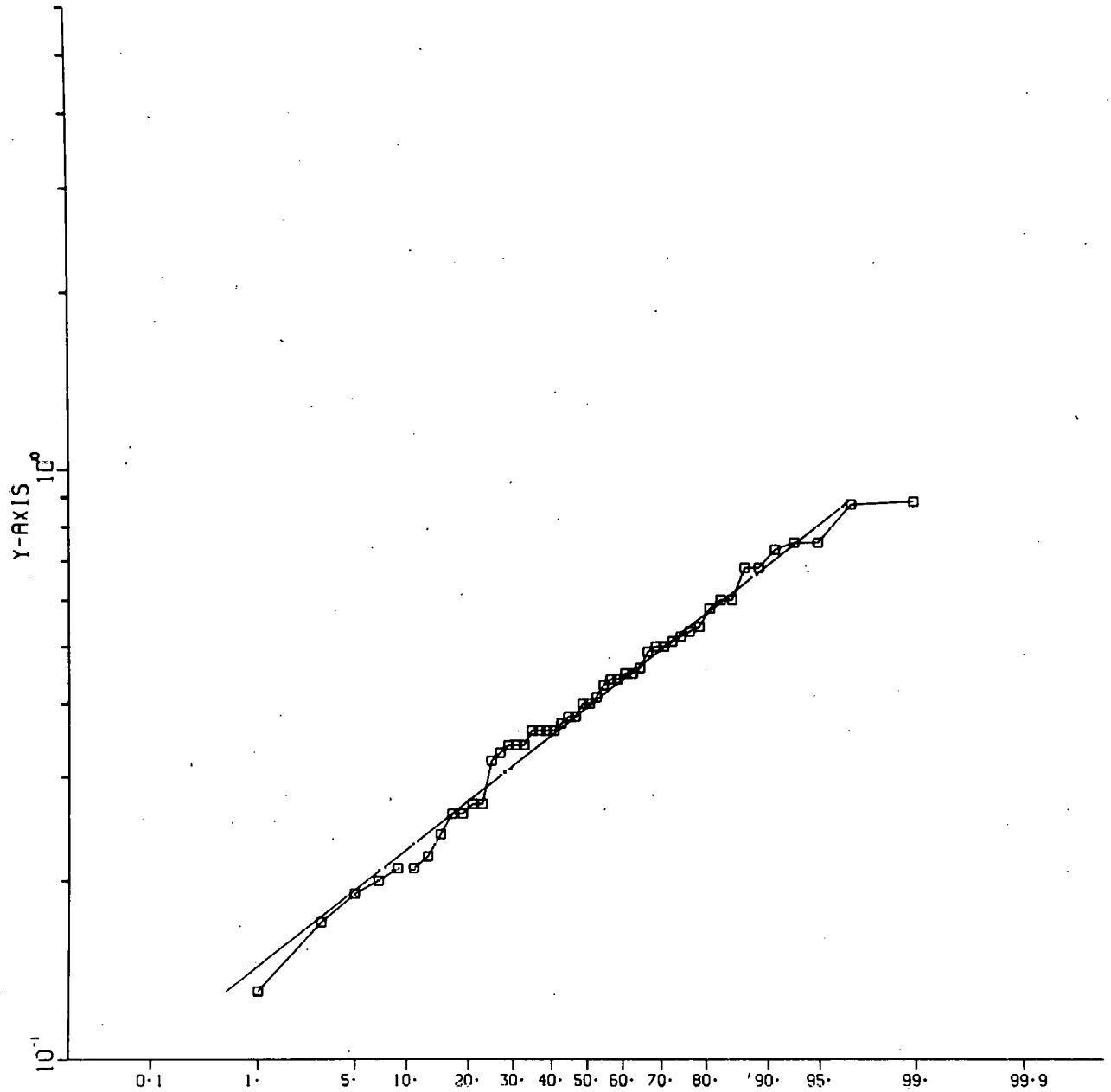

Fig. 8. Log-normal Distribution Plot of Lead Content of Air at ANL, 1977. Geometric mean - 0.39 $\mu g/m^3$; geometric standard deviation - 1.54 $\mu g/m^3$; arithmetric mean - 0.43 $\mu g/m^3$. The straight line is a least-squares fit to the data points.

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 38 are collected the 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, 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 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%.

TABLE 38

Concentration Guides and Detection Limits

(μCi/ml)

Nuclide or Activity	Concentration Guide Water	Concentration Guide Air	Detection Limit Water	Detection Limit Air
Americium-241	4×10^{-6}	4×10^{-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}	2×10^{-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}	5×10^{-10}	1×10^{-17}
Thorium-228	-	2×10^{-13}	-	1×10^{-18}
Thorium-230	-	3×10^{-13}	-	1×10^{-18}
Thorium-232	-	1×10^{-12}	-	1×10^{-18}
Uranium-234	-	4×10^{-12}	-	1×10^{-19}
Uranium-235	-	4×10^{-12}	-	1×10^{-19}
Uranium-238	-	5×10^{-12}	-	1×10^{-19}
Uranium - natural*	4×10^{-5}	4×10^{-12}	2×10^{-10}	2×10^{-17}
Zirconium-95	-	1×10^{-9}	-	5×10^{-16}
Alpha**	3×10^{-6} to 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.

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 run 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 1977 is shown in Table 39. The program and our participation in it is somewhat expanded compared to 1976. 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.[13] Results of our participation in this program during 1977 are given in Table 40. In the table the comparison is made between the EML value, which is the result of replicate determinations by that Laboratory, and the value obtained in our Laboratory. More than 95% of all the intercomparison samples received were analyzed for the radionuclides for which results were requested. To assist in judging the quality of the results, typical errors for our analyses are 2-25%, the error in the EML results is 1-30% (depending 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. 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

TABLE 39

Summary of EPA-QA Samples, 1977

Type of Sample	Analysis	Number Analyzed	Avg. Difference From Added
Air Filter	Total Alpha	5	23%
	Total Beta	5	6%
	Strontium-90	2	10%
	Cesium-137	5	13%
	Plutonium-239	1	10%
Water	Total Alpha	5	30%
	Total Beta	5	8%
	Hydrogen-3	2	5%
	Chromium-51	4	16%
	Cobalt-60	6	4%
	Zinc-65	6	6%
	Strontium-89	1	6%
	Strontium-90	1	6%
	Ruthenium-106	6	22%
	Cesium-134	6	10%
	Cesium-137	6	10%
	Plutonium-239	1	18%
Milk	Potassium-40	2	5%
	Strontium-89	1	23%
	Strontium-90	2	19%
	Iodine-131	1	16%
	Cesium-137	2	15%
	Barium-140	1	3%
Soil	Plutonium-239	1	16%

TABLE 40

Summary of DOE-EML-QAP Samples, 1977

Nuclide	Average Difference from EML Value				
	Air Filters	Water	Soil	Tissue	Vegetation
Hydrogen-3	-	8% (4)	-	-	-
Beryllium-7	6% (2)	18% (1)	-	-	-
Sodium-24	10% (2)	5% (1)	-	-	-
Potassium-40	-	-	8% (4)	14% (4)	7% (2)
Manganese-54	2% (2)	2% (2)	-	-	-
Cobalt-57	12% (2)	5% (1)	-	-	-
Cobalt-58	4% (1)	12% (1)	-	-	-
Cobalt-60	6% (3)	5% (3)	8% (3)	-	6% (1)
Iron-59	17% (1)	2% (2)	-	-	-
Zinc-65	18% (1)	8% (1)	-	-	-
Strontium-89	18% (1)	8% (3)	-	-	-
Strontium-90	17% (3)	7% (3)	9% (4)	22% (4)	10% (2)
Zirconium-95	2% (1)	-	-	-	-
Niobium-95	10% (1)	-	-	-	-
Ruthenium-103	16% (2)	-	-	-	-
Ruthenium-106	15% (1)	-	-	-	-
Antimony-125	17% (2)	-	-	-	-
Cesium-134	13% (3)	8% (1)	-	-	-
Cesium-137	14% (4)	5% (4)	6% (4)	24% (2)	11% (2)
Cerium-141	18% (1)	12% (1)	-	-	-
Cerium-144	1% (1)	4% (1)	-	-	-
Radium-226	-	-	14% (4)	-	10% (2)
Thorium-228	-	-	22% (4)	-	4% (1)
Uranium-234	6% (1)	17% (1)	2% (1)	-	-
Uranium-238	5% (1)	10% (1)	7% (1)	-	-
Uranium - total	-	-	2% (1)	-	-
Plutonium-238	10% (2)	29% (3)	33% (4)	-	-
Plutonium-239	4% (4)	39% (4)	22% (4)	-	-
Americium-241	37% (1)	24% (1)	27% (1)	-	-

The figure in parentheses is the number of samples.

samples to which identical concentrations were added. Average recoveries ranged from about 85-100%, with a standard deviation of about 10%.

Recovery studies were also performed for ammonia nitrogen, fluoride, and hexavalent chromium analyses, and similar results were obtained.

The laboratory also participated in the intercomparison program for the analysis of trace elements in water sponsored by the EPA (Cincinnati Laboratory).

The results for the three concentration levels distributed are shown in Table 41. In some cases the concentrations added by the EPA were below the normal limits of detection by direct analysis, e.g., iron and nickel. In other cases, e.g., beryllium and cadmium, levels are considerably higher than environmental levels normally encountered.

The results obtained for all of the elements, with the exception of mercury and cadmium, are in agreement within expected ranges. The cadmium results were obtained using a flameless atomic absorption technique, rather than a conventional flame. The reason for the somewhat higher than expected deviation is not known.

The initial results obtained for mercury (A) were considerably lower than expected. The samples were prepared by dilution of the EPA standard according to their directions. The fact that results approached better agreement as the amount increased indicated that stability problems were being encountered. Previous studies in this laboratory have shown that concentrations of mercury of less than 10 µg/l are difficult to maintain in solution. To test this, a small aliquot from each EPA-supplied concentrated solution was diluted by the procedure used in our laboratory to maintain a pH favorable for mercury stability. As can be seen, the results obtained from samples #2 and #3 are quite good (B). The results obtained for sample #1 are still low, which could indicate that stability problems still exist.

c. 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 installation of a continuous water sampler in Sawmill Creek during the year provides a representative sample for a critical

TABLE 41

Results of EPA Quality Assurance Samples
(Concentrations in µg/l)

Metal	Lower Limit of Detection	Added / Found #1	Added / Found #2	Added / Found #3	Avg. Found / Added
Beryllium	0.02	16/17	78/79.3	398/383	1.01
Cadmium	0.2	5.2/4.1	23/19	73/61	0.81
Chromium	10	16/16	154/144	209/186	0.94
Copper	5	16/12	72/66	102/95	0.86
Iron	80	26/36	417/428	678/698	1.12
Lead	2	22/24	265/298	352/343	1.05
Mercury (A)	0.10	0.8/0.4	4.5/2.2	9.4/7.2	0.56
Mercury (B)	0.10	0.8/0.5	4.5/4.7	9.4/9.1	0.83
Nickel	50	26/33	45/50	152/157	1.12
Zinc	10	11/20	30/42	174/176	1.33

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.

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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