ANL-77-13

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ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY ANNUAL REPORT FOR 1976

by

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

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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY ANNUAL REPORT FOR 1976

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

ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1976 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 surface 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 accepted 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. Energy Research and Development Administration (ERDA) 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 ERDA 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 a major effort in the fast breeder nuclear reactor (liquid-metal and gas-cooled), reactor safety studies, improvements in the utilization of coal for power production, coal liquefication 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 or 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.

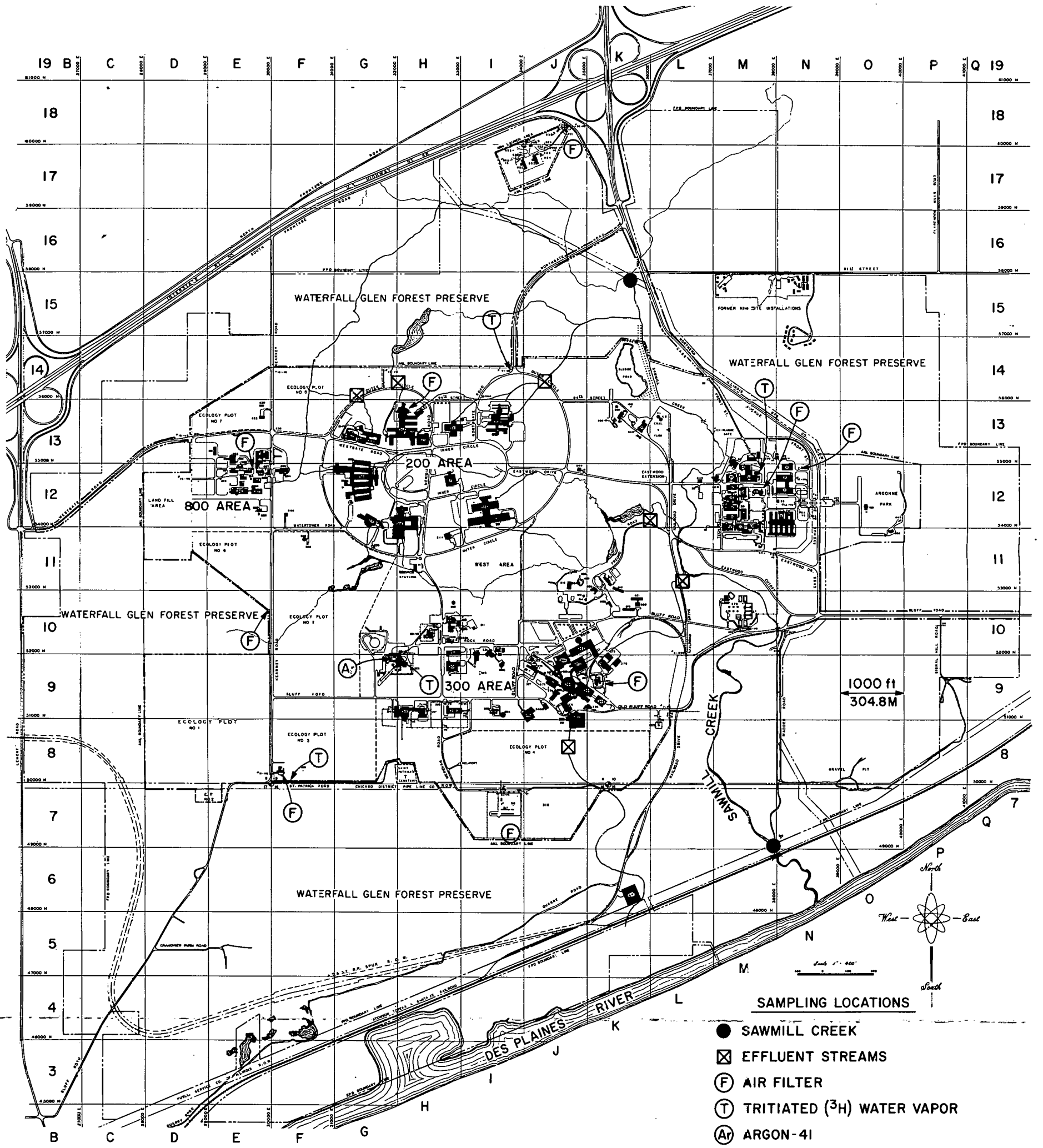

Fig. 1. Sampling Locations at Argonne National Laboratory

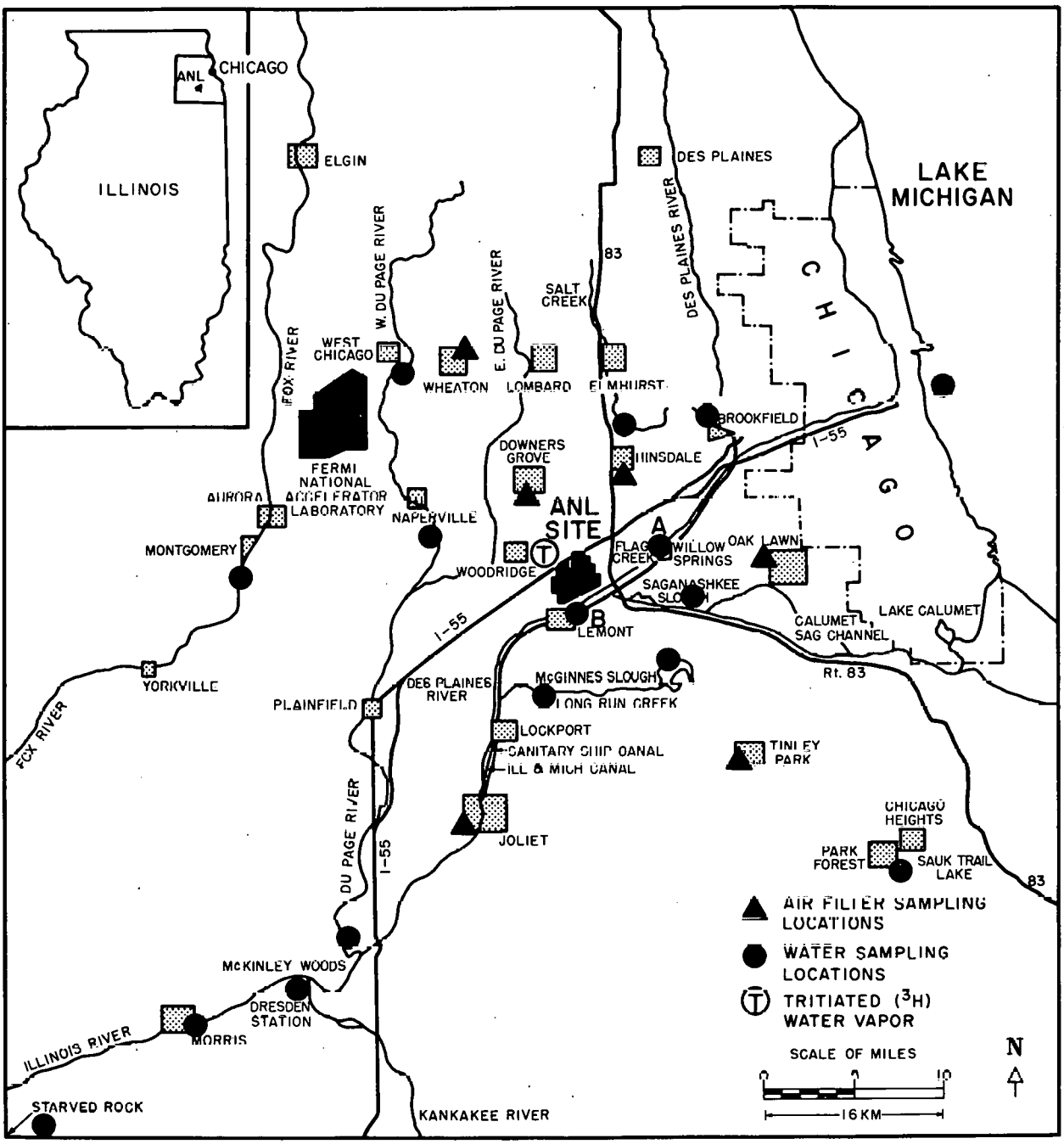

Fig. 2. Sampling Locations Near Argonne National Laboratory

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 variables for the purposes of the report, wind direction, wind speed, temperature, and precipitation, obtained from the Atmospheric Physics Section of the Radiological and Environmental Research Division at Argonne, are given here for 1976. 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 "76 total", the predominant wind blows from the southwest. This vector shows that, of the total wind observations, about 4% were in the 2.01-6 m/s range, about 5% were in the 6.01-10 m/s range, about 1% were greater than 10.01 m/s; and about 10% of the observations were from the direction between 213.75° and 236.25° (southwest). 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 southwest and west on the average, but seasonal variations are apparent. The winds were primarily westerly in January, while in February and March the southwest component was strong. In the spring, the northeast lake breeze became evident and continued through much of the summer. The warm southwesterly winds, brought up by the high pressure areas in the southeastern U.S., began later in the spring and continued for the remainder of the year. Strong north and west winds were noted during the last three months of the year in greater than average abundance. The winds are sufficiently variable so that monitoring for any airborne releases must be carried out in all directions from the site.

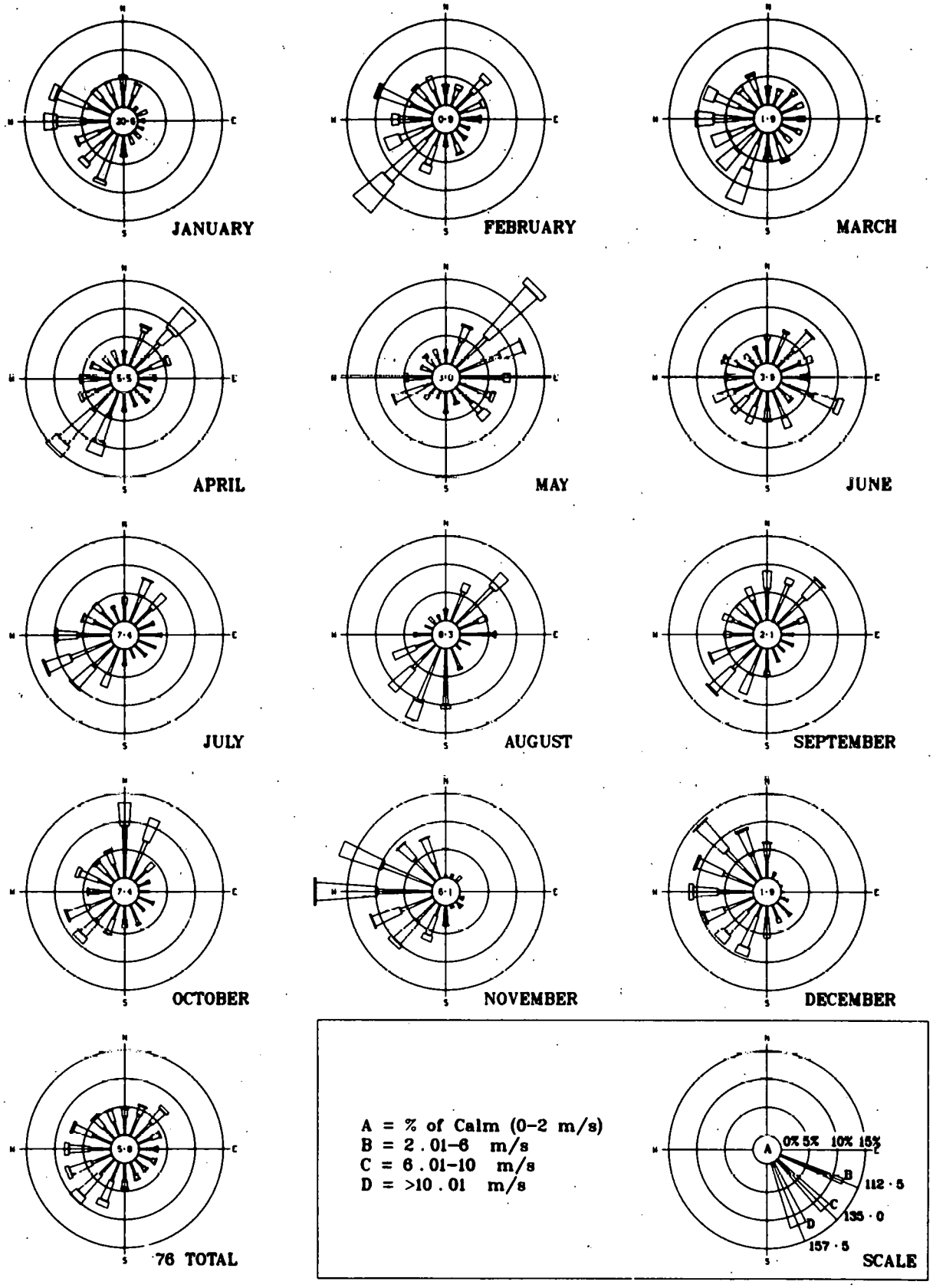

Fig. 3. Monthly Wind Roses at Argonne National Laboratory, 1976

The monthly average wind speed at 44.5 m (146 ft) above ground level varied from 4.7 m/s (10.5 mph) in August to 7.3 m/s (16.3 mph) in March. Average monthly wind speeds at 1.5 m (4.9 ft) and 6.0 m (19.7 ft) above ground level had maxima and minima in the same months. At 1.5 m the speeds ranged from 2.4 m/s (5.3 mph) to 4.6 m/s (10.3 mph) and at 6.0 m the range was from 3 m/s (6.7 mph) to 5.2 m/s (11.6 mph). A tornado passed through part of the Argonne site at about 6 p.m. on June 13, 1976. The maximum wind speed measured during this period was 27 m/s (60 mph) at the ANL Meteorology Tower, which was not in the direct path of the tornado.

Figure 4 shows the variation of the mean air temperature by month as compared to the 15-yr mean air temperature during the 1950-1964 period. During the first six months the temperatures generally exceeded the 15-yr mean and from August through December the temperature was less than the mean by approximately the same amount. The temperatures during the last quarter of 1976 were abnormally low, as would be expected from the wind data. A comparison of the 1976 wind roses to previous wind rose data shows a larger northerly component during this period in 1976.

The precipitation followed a similar pattern, showing above normal levels through July and very dry conditions during the remainder of the year. This will be discussed in more detail in Section I.E.

D. Population

The area around Argonne has exhibited a large population growth in the past 10 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 1. The distribution, centered on the CP-5 reactor, was obtained by modifying a similar distribution provided by the Regional and Urban Studies Department at Oak Ridge National Laboratory, which was based on the 1970 U.S. census figures. The figures were adjusted in the first few kilometers by actual inspection of the area, and over the entire region with the use of the most recent 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

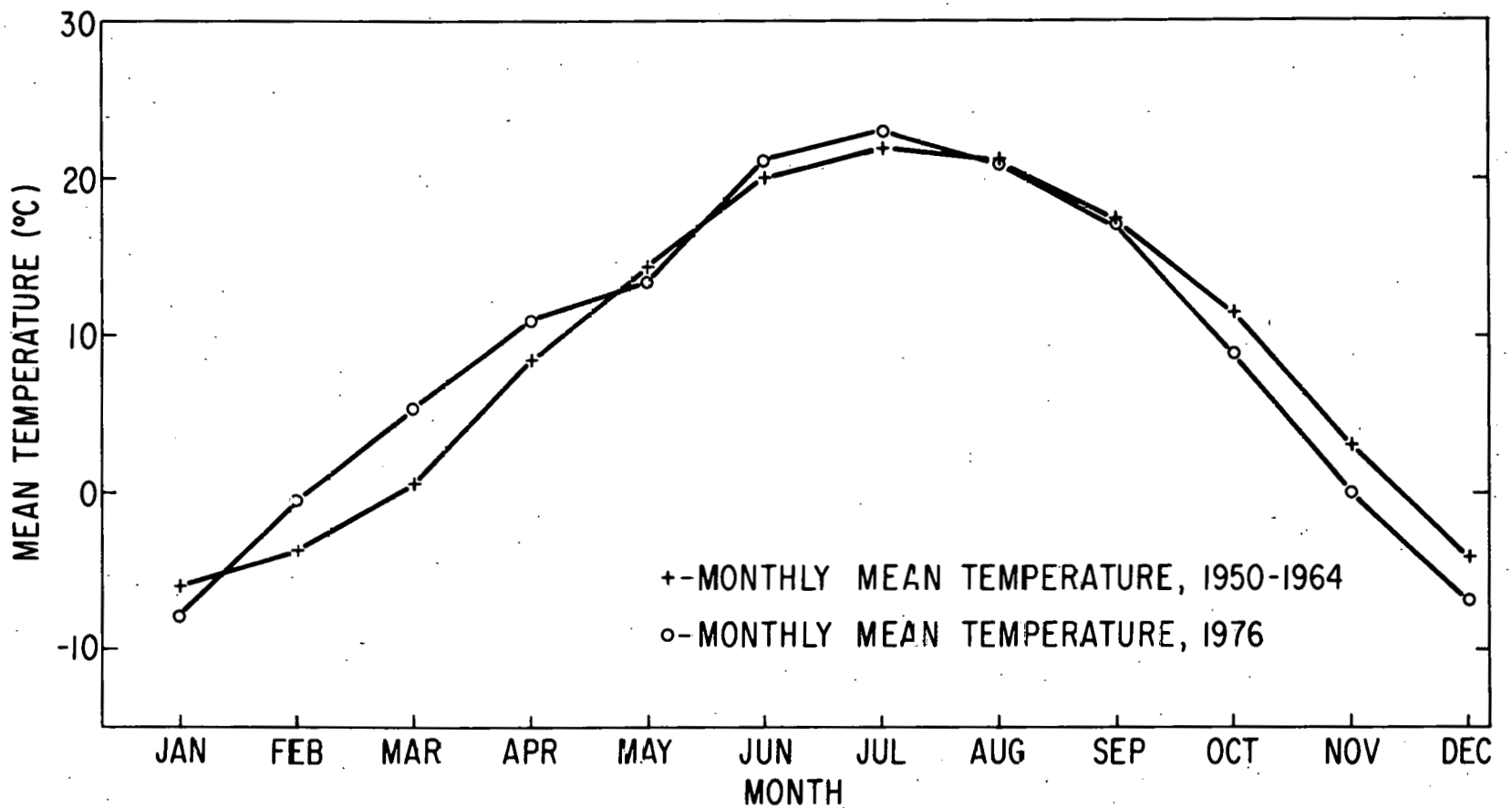

Fig. 4. Monthly Mean Air Temperatures at Argonne National Laboratory

TABLE 1

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	1660	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	560	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

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. The 1970 census of communities and townships in the area, and a map showing their relation to the Argonne site, are given in a preceding report in the series.[3]

E. Land and Water 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 addition 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 1976, the water flow in the Creek averaged about 24 megaliters (6.4 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 normally equally divided in volume between sanitary waste and laboratory waste. Figure 5 shows the relation between precipitation and the volume of sanitary waste water, and indicates that storm water enters the sanitary waste system. During the latter part of the year, when the amount of precipitation was abnormally low, the volume of sanitary waste water was about half the Laboratory volume. This changed the concentrations found previously for many elements by lowering normal dilution effects.

Six 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. One stream from the 200 Area leaves the site at location 14J and enters 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

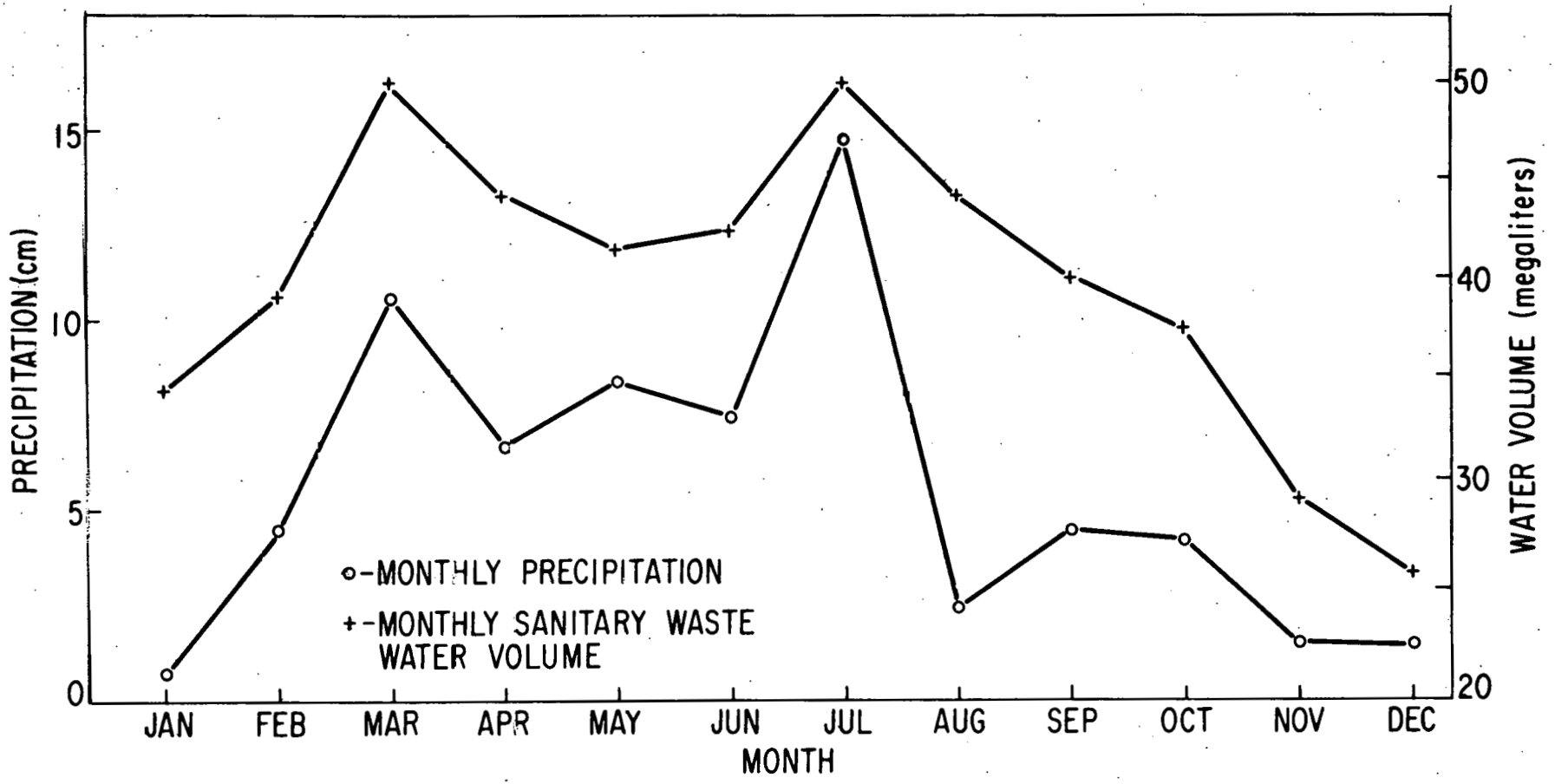

Fig. 5. Precipitation and Sanitary Waste Water Volume, Monthly Totals, 1976

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. 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 above the sampling point. The low water level indicated above would also severely concentrate materials released in these open channels.

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 ground water supplies by infiltration. In the vicinity of the Laboratory, only subsurface water (from all aquifers, shallow and deep) and Lake Michigan water is 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 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 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 2. The milk, corn, and soybean production is for 1976; the wheat and oat production is for 1975. These estimates were obtained from the Illinois Crop Reporting Service of the State Department of Agriculture.

TABLE 2

Agricultural Production Near ANL

	Milk		Corn	Soybeans	Wheat	Oats
County	No. of Cows	Million Pounds	Million Bushels			
DuPage	300	3.1	2.0	0.51	0.20	0.14
Cook	400	4.2	1.8	0.64	0.17	0.12
Will	4,500	46.7	15.2	3.1	0.86	0.59
Kane	7,700	79.9	14.2	2.0	0.37	0.59
Kendall	700	7.3	9.1	1.5	0.25	0.24
Grundy	1,300	13.5	12.6	2.2	0.84	0.15
Lake	1,400	14.5	1.8	0.47	0.45	0.32

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. Energy Research and Development Administration (ERDA) 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-76-29. Included in this report are the results of measurements obtained in 1976 for a number of radionuclides in air, surface water, soil, grass, bottom sediment, and foodstuffs; for a variety of chemical constituents in water; and for the external penetrating radiation dose.

Total alpha and beta activities, fission and activation products, plutonium, thorium, and uranium were measured continuously in air-filter samples at the site perimeter and off the site. All the off-site and perimeter samples contained only activities from natural sources and nuclear test detonations. Short-lived fission products were detected at all sampling locations in February and at the end of the year, and are attributed to fallout from the Chinese atmospheric nuclear tests of January 23, 1976, and September 26, 1976. No activity attributable to Argonne operations could be detected.

The plutonium-239,240 concentrations in air averaged, respectively, 6×10^{-10} and 5×10^{-18} μCi/ml* at the site perimeter and off the site, about one-third of the 1975 values. The monthly variations indicate a "spring maximum" in stratospheric fallout of plutonium similar to that observed in beryllium-7 (a naturally-occurring nuclide) and several fission products. The results indicate 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 0.0006% of the CG.**

---

*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. Energy Research and Development Administration in Chapter 0524 of the ERDA Manual.[4] The pertinent CGs are listed in the Appendix, Section IV.B.

Argon-41 and hydrogen-3 (as water vapor) represent the major airborne radioactivity released from the Laboratory. The argon-41 concentration and corresponding radiation dose at the site boundary was less than the detection limit (50% of the CG) of the present sampling system, but was calculated from an atmospheric dispersion model to be 4.6×10^{-10} μCi/ml and 5.9 mrem/yr in the predominant north-northeast wind direction. 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 2.1 km (1.3 mi) NE, where the closest full-time residents live, is 3.4 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 9×10^{-12} μCi/ml, which is 0.005% 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 concentrations, were hydrogen-3, 0.054%; strontium-90, < 0.18%; neptunium-237, 0.001%; plutonium-239,240, 0.0002%; americium-241, 0.0001%; and curium-244 and/or californium-249, < 0.00006%. 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 5.8 mrem/yr. About 95% of the total dose would result from strontium-90 produced in weapons testing.

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 soil was 1.8×10^{-3} μCi/m^2 at the site perimeter and 2.5×10^{-3} μCi/m^2 off the site. The corresponding plutonium-238 averages were 0.08×10^{-3} μCi/m^2 and 0.14×10^{-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 are within the range reported by other workers 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 contained from 1 x 10^{-9} to 8 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 some 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 ranged from < 200 x 10^{-9} μCi/ml (the detection limit) to 300 x 10^{-9} μCi/ml, similar to the range found in surface water. The strontium-90 and cesium-137 concentrations decreased compared to 1975. Iodine-131 and barium-140 were detected in milk late in the year and were due to fallout from the Chinese atmospheric test of September 26, 1976. The consumption of one liter of milk per day at the average concentrations would have resulted in a total dose of about 0.5% of the annual limit, principally from strontium-90. These radionuclides are present in milk due to fallout, and are not related to Argonne operations.

Measurements of penetrating radiation were made at several locations at the site boundary and off the site. The off-site readings averaged 90 mrem/yr, with a standard deviation of 9 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 440 mrem/yr above normal at location 7I in Figure 1 and 10-20 mrem/yr above normal at location 8H, as a result of radiation from an on-site temporary storage facility for radioactive waste. Along the north side of the site, the dose at the fence at location 14I was 300 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 mi) from the fence line, was about 0.05 mrem/yr; similarly, the dose rate to the residents closest to the north boundary, about 0.75 km (0.5 mi) from the fence was about 0.5 mrem/yr. These dose rates are about 0.01% to 0.1%, respectively, of the standard for individuals in uncontrolled areas.

Levels of chemical constituents and other water quality parameters were measured in Argonne waste and effluent water and in Sawmill Creek; the results were compared to the standards adopted by the State of Illinois. Concentrations of mercury, hexavalent chromium, iron, and zinc in the Des Plaines River were measured to determine whether any contribution from Argonne waste water could be seen.

Ammonia nitrogen and biochemical oxygen demand (B.O.D.) in the Argonne combined sanitary and laboratory waste-water effluent were within State of Illinois standards with the exception of ammonia nitrogen in December and B.O.D. in April. Average concentrations of each of the other chemical constituents measured in this effluent (Table 29, Section III.B.) were at or below these standards. The concentration of mercury averaged 86% of the State standard and exceeded this value 14% of the time (i.e., in 14% of the samples). No other constituent in this effluent exceeded the State standard at any time.

The average concentration of chromium in water from three of the six cooling tower effluent channels used for blowdown exceeded the State standard by varying amounts, while in the remaining three the concentrations were well below the standard.

The average values in Sawmill Creek for dissolved oxygen and chemical constituents, except ammonia nitrogen, were within the State of Illinois standards. The average ammonia nitrogen level above the Argonne waste-water outfall was 3.5 times the State standard, and exceeded this standard 66% of the time. The average level below the outfall was twice the State standard and exceeded this value in 60% of the samples. This indicates a degradation of water quality over last year, the source of which originates upstream of the Laboratory. On four 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, iron, mercury, and silver exceeded State standards from 4% to 14% of the time. Hexavalent chromium levels were in excess of the State standard 14% 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.

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

III. MONITORING RESULTS

A. Radioactivity

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 were 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 activity at locations where the activity not only varied greatly, but was 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 ERDA Manual Chapter 0524.[4] The pertinent CGs as well as the detection limits are given in the Appendix, Section IV.B. Although the CGs apply to concentrations above natural levels, the percent of CG is sometimes given in this report for activities that are primarily of natural origin for comparative purposes. Such values are enclosed in parentheses to indicate this.

1. Air

The radioactivity 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 counting. Such results require investigation to determine the cause of the difference.

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

The error is between 5 and 20% for most results, but approaches 100% at the detection limit.

The total alpha and beta activities in the individual weekly samples are summarized in Table 3. These measurements were made in low-background gas-flow proportional counters, and the counting efficiencies used to convert counting rates to disintegration rates were those measured for radon decay products on filter paper. The average concentrations of a number of gamma-ray emitters, as determined by gamma-ray spectrometry performed on composite weekly samples, are given in Table 4. The gamma-ray detector is a shielded 35 cm^3 lithium-drifted germanium diode, calibrated for each gamma-ray emitting nuclide in Table 4.

The alpha activities, principally due to naturally-occurring nuclides, averaged the same as 1975 and were in their normal range. About 85% of the gamma-ray activity, and a smaller fraction of the beta 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. The average beta activity for the year, 4.6 x 10^{-14} μCi/ml, was about 65% of the 1975 average. The decrease is principally due to the decay of intermediate half-life fission products (such as zirconium-95-niobium-95, ruthenium-106-rhodium-106, and cerium-144). These fission products are the residuals of atmospheric nuclear tests by the People's Republic of China on June 26, 1973, and June 17, 1974.

The customary spring maximum in stratospheric fallout was observed for several of the individual gamma-ray emitters (e.g., ^{7}Be), but was not noted in the total beta activity. The total beta activity would have been similar to the 1973 annual average of 3.5 x 10^{-14} μCi/ml except for the presence of short-lived fission products from recent atmospheric nuclear tests by the People's Republic of China. As shown in Table 4, these were present in February from an atmospheric test conducted on January 23, 1976, and during the last three months of the year, from a test on September 26, 1976. Although a 4 megaton atmospheric nuclear test occurred on November 17, 1976, no fallout was detected from this detonation.

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

TABLE 3

Total Alpha and Beta Activities in Air-Filter Samples, 1976*

Month	Location	No. of Samples	Alpha Act. (10^{-15} µCi/ml) Av.	Min.	Max.	Beta Act. (10^{-15} µCi/ml) Av.	Min.	Max.
January	perimeter	42	2.4	0.9	4.9	38	24	48
	off-site	22	2.7	1.0	5.5	38	30	48
February	perimeter	36	1.9	0.4	4.1	31	15	46
	off-site	20	2.2	1.0	6.1	30	14	37
March	perimeter	39	1.8	0.6	3.9	26	16	36
	off-site	22	2.2	0.7	3.7	26	11	35
April	perimeter	38	3.1	0.9	7.4	27	13	45
	off-site	16	2.3	1.2	3.8	27	16	36
May	perimeter	41	2.0	0.5	5.0	23	15	35
	off-site	20	2.7	1.0	8.1	24	14	42
June	perimeter	36	3.3	0.8	7.8	30	18	48
	off-site	20	2.6	1.0	6.5	28	15	47
July	perimeter	42	2.7	0.9	7.3	26	20	37
	off-site	18	2.2	1.4	4.0	26	13	32
August	perimeter	38	2.9	0.8	6.3	29	18	46
	off-site	20	2.0	0.8	3.4	28	17	48
September	perimeter	40	2.6	0.5	5.5	32	21	89
	off-site	20	2.5	0.6	6.1	32	9.9	81
October	perimeter	38	2.4	1.0	4.6	102	29	354
	off-site	19	2.9	0.9	8.7	98	4.3	228
November	perimeter	40	3.0	1.0	14.5	116	68	219
	off-site	20	2.7	1.5	3.7	117	83	171
December	perimeter	37	2.8	1.3	6.3	77	48	129
	off-site	20	3.3	1.6	5.1	78	48	146
Annual Summary	perimeter	467	2.6 ± 0.3	0.4	14.5	46 ± 19	15	354
	off-site	235	2.5 ± 0.2	0.6	8.7	46 ± 19	4.3	228
Percent CG	perimeter	-	(0.026)	(0.004)	(0.145)	0.46	0.15	3.5
	off-site	-	(0.025)	(0.006)	(0.087)	0.46	0.043	2.3

*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 4

Gamma-Ray Activity in Air-Filter Samples, 1976

(concentrations in 10^{-15} μCi/ml)

Month	Location	^{7}Be	^{54}Mn	^{60}Co	^{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	73	< 0.1	< 0.1	< 0.5	< 0.1	0.5	< 0.5	< 5	0.3	< 0.1	< 0.1	0.9
	off-site	70	< 0.1	< 0.1	< 0.5	< 0.1	0.7	< 0.5	< 5	0.3	< 0.1	< 0.1	1.0
February	perimeter	80	< 0.1	< 0.1	< 0.5	0.1	0.8	< 0.5	< 5	0.4	0.7	0.2	1.2
	off-site	80	< 0.1	< 0.1	< 0.5	0.2	0.6	< 0.5	< 5	0.3	0.6	0.2	1.1
March	perimeter	92	< 0.1	< 0.1	< 0.5	< 0.1	0.8	< 0.5	< 5	0.5	0.2	< 0.1	1.2
	off-site	81	< 0.1	< 0.1	< 0.5	< 0.1	0.7	< 0.5	< 5	0.6	< 0.1	< 0.1	1.3
April	perimeter	122	< 0.1	< 0.1	< 0.5	< 0.1	0.8	< 0.5	< 5	0.7	< 0.1	< 0.1	1.6
	off-site	128	< 0.1	< 0.1	< 0.5	< 0.1	1.1	< 0.5	< 5	0.7	< 0.1	< 0.1	2.3
May	perimeter	104	< 0.1	< 0.1	< 0.5	< 0.1	0.9	< 0.5	< 5	0.6	< 0.1	< 0.1	1.3
	off-site	105	< 0.1	< 0.1	< 0.5	< 0.1	0.7	< 0.5	< 5	0.6	< 0.1	< 0.1	1.5
June	perimeter	147	< 0.1	< 0.1	< 0.5	< 0.1	1.3	< 0.5	< 5	0.9	< 0.1	< 0.1	1.5
	off-site	150	< 0.1	< 0.1	< 0.5	< 0.1	1.2	< 0.5	< 5	0.9	< 0.1	< 0.1	1.4
July	perimeter	124	< 0.1	< 0.1	< 0.5	< 0.1	0.5	< 0.5	< 5	0.6	< 0.1	< 0.1	1.6
	off-site	123	< 0.1	< 0.1	< 0.5	< 0.1	< 0.5	< 0.5	< 5	0.6	< 0.1	< 0.1	1.0
August	perimeter	108	< 0.1	< 0.1	< 0.5	< 0.1	0.5	< 0.5	< 5	0.4	< 0.1	< 0.1	0.8
	off-site	100	< 0.1	< 0.1	< 0.5	< 0.1	0.7	< 0.5	< 5	0.4	< 0.1	< 0.1	0.9
September	perimeter	110	< 0.1	< 0.1	< 0.5	< 0.1	< 0.5	< 0.5	< 5	0.3	< 0.1	< 0.1	0.5
	off-site	103	< 0.1	< 0.1	< 0.5	< 0.1	< 0.5	< 0.5	< 5	0.3	< 0.1	< 0.1	0.4
October	perimeter	104	< 0.1	< 0.1	3.4	3.0	< 0.5	< 0.5	26	0.2	11	4.0	0.8
	off-site	66	< 0.1	< 0.1	3.1	2.1	< 0.5	< 0.5	22	0.2	9.0	2.8	0.8
November	perimeter	74	< 0.1	< 0.1	14	12	1.5	< 0.5	15	0.3	14	11	2.9
	off-site	66	< 0.1	< 0.1	11	10	1.3	< 0.5	13	0.4	13	8.7	2.0
December	perimeter	84	< 0.1	< 0.1	8.3	6.8	1.3	< 0.5	< 5	0.3	3.3	4.5	1.6
	off-site	74	< 0.1	< 0.1	8.3	6.1	0.9	< 0.5	< 5	0.4	3.1	4.3	1.8
Annual	perimeter	102 ± 14	< 0.1	< 0.1	< 2.5	< 1.9	0.8 ± 0.2	< 0.5	< 8	0.5 ± 0.1	< 2.5	< 1.7	1.3 ± 0.3
Summary	off-site	96 ± 17	< 0.1	< 0.1	< 2.3	< 1.7	0.7 ± 0.2	< 0.5	< 8	0.5 ± 0.1	< 2.3	< 1.4	1.3 ± 0.3
Percent	perimeter	(0.26)	< 0.02	< 0.03	< 0.25	< 0.063	0.40	< 0.06	< 8	0.10	< 0.25	< 0.034	0.65
CG (x 10^{-3})	off-site	(0.24)	< 0.02	< 0.03	< 0.23	< 0.057	0.35	< 0.06	< 8	0.10	< 0.23	< 0.028	0.65

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, electrodeposited, and their isotopic compositions determined by alpha spectrometry. Chemical recoveries were monitored by adding known amounts of plutonium-242 and thorium-234 tracers prior to ignition. Alpha spectrometry cannot distinguish between plutonium-239 and plutonium-240, and although in the remainder of the report, including the tables, only plutonium-239 is mentioned, it should be understood that the alpha activity due to the plutonium-240 isotope is also included.

The acidity of the column effluent from the anion exchange separation was adjusted to allow the extraction of uranium and of americium. The separated elements were electrodeposited and measured by alpha spectrometry. Chemical recoveries were monitored by adding known amounts of uranium-232 and americium-243 tracers prior to ignition. Following the extractions, the aqueous phase was analyzed for radiostrontium by standard radiochemical procedures. The results are given in Tables 5 and 6.

The average plutonium-239 concentrations decreased at all locations by about a factor of three from 1975 and are similar to those obtained by the ERDA New York Health and Safety Laboratory (HASL). Their results[5] for New York City during the first six months of the year ranged from 6×10^{-18} μCi/ml in February to 10×10^{-18} μCi/ml in May and showed the same monthly variations as those in the table.

The monthly variations in the concentrations of both strontium-89 and strontium-90 correlate with the results for the other fission products in Table 4. The concentrations are considered to be due to fallout from the atmospheric nuclear tests mentioned previously, and are consistent with strontium-90 levels reported by HASL[5] for the first six months of 1976. These

TABLE 5

Strontium and Plutonium Concentrations in Air-Filter Samples, 1976

(concentrations in 10^{-18} μCi/ml)

Month	Location[1]	Strontium-89	Strontium-90	Plutonium-238	Plutonium-239[2]
January	12N	< 100	240 ± 60	< 0.5	5.8 ± 0.9
	7I	< 100	390 ± 150	< 0.5	6.2 ± 0.6
	off-site	< 100	260 ± 20	< 0.5	6.0 ± 0.7
February	12N	350 ± 80	180 ± 40	< 0.5	7.3 ± 1.1
	7I	530 ± 90	220 ± 30	< 0.5	6.0 ± 0.9
	off-site	< 100	220 ± 130	< 0.5	5.4 ± 1.4
March	12N	< 100	280 ± 10	1.3 ± 0.5	5.9 ± 0.9
	7I	< 100	590 ± 120	0.6 ± 0.4	6.0 ± 0.8
	off-site	< 100	310 ± 60	0.9 ± 0.4	6.8 ± 0.9
April	12N	< 100	420 ± 80	1.0 ± 0.7	8.1 ± 1.3
	7I	< 100	500 ± 30	0.5 ± 0.4	8.4 ± 1.1
	off-site	< 100	330 ± 20	0.5 ± 0.4	7.3 ± 1.0
May	12N	< 100	370 ± 40	0.7 ± 0.3	7.0 ± 0.7
	7I	< 100	470 ± 70	0.5 ± 0.3	7.8 ± 0.8
	off-site	< 100	400 ± 60	1.0 ± 0.4	7.1 ± 0.8
June	12N	< 100	460 ± 90	0.5 ± 0.4	9.9 ± 1.0
	7I	< 100	570 ± 60	1.0 ± 0.5	9.8 ± 1.2
	off-site	< 100	500 ± 120	0.7 ± 0.4	9.7 ± 1.2
July	12N	< 100	390 ± 100	0.5 ± 0.3	9.2 ± 1.0
	7I	< 100	510 ± 110	1.2 ± 0.4	8.3 ± 1.0
	off-site	< 100	310 ± 80	0.5 ± 0.7	6.0 ± 1.4
August	12N	< 100	270 ± 60	< 0.5	7.1 ± 0.9
	7I	< 100	370 ± 90	< 0.5	7.1 ± 0.8
	off-site	< 100	240 ± 80	< 0.5	4.8 ± 0.7
September	12N	< 100	90 ± 30	< 0.5	2.8 ± 0.5
	7I	180 ± 60	130 ± 10	< 0.5	3.0 ± 0.6
	off-site	110 ± 90	90 ± 20	< 0.5	1.8 ± 0.5
October	12N	4400 ± 100	70 ± 30	< 0.5	3.4 ± 0.7
	7I	4670 ± 100	100 ± 40	< 0.5	3.8 ± 1.0
	off-site	3320 ± 130	120 ± 40	< 0.5	1.3 ± 0.6
November	12N	11200 ± 320	250 ± 120	< 0.5	3.6 ± 0.5
	7I	8860 ± 310	240 ± 120	< 0.5	3.0 ± 0.5
	off-site	7090 ± 280	200 ± 100	< 0.5	2.8 ± 0.6
December	12N	5590 ± 390	120 ± 170	< 0.5	3.5 ± 0.6
	7I	5710 ± 280	200 ± 110	< 0.5	3.4 ± 0.6
	off-site	4760 ± 500	180 ± 210	< 0.5	2.2 ± 0.4
Annual Summary	12N	< 1860	260 ± 80	< 0.7	6.1 ± 1.5
	7I	< 1720	360 ± 110	< 0.7	6.1 ± 1.5
	off-site	< 1340	260 ± 70	< 0.6	5.1 ± 1.6
Percent CG	12N	< 0.0002	0.00013	< 0.00007	0.00061
	7I	< 0.0002	0.00018	< 0.00007	0.00061
	off-site	< 0.0002	0.00013	< 0.00006	0.00051

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

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

TABLE 6

Thorium and Uranium Concentrations in Air-Filter Samples, 1976

(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	8 ± 3	19 ± 2	13 ± 2	29 ± 2	1.0 ± 0.3	28 ± 2
	7I	9 ± 2	16 ± 1	9 ± 1	26 ± 2	0.7 ± 0.3	26 ± 2
	off-site	14 ± 2	29 ± 2	20 ± 2	32 ± 2	1.4 ± 0.3	34 ± 2
February	12N	26 ± 2	32 ± 2	22 ± 2	-	0.8 ± 0.4	26 ± 2
	7I	8 ± 2	22 ± 2	14 ± 1	29 ± 2	0.8 ± 0.3	24 ± 1
	off-site	-	15 ± 4	7 ± 3	-	-	20 ± 5
March	12N	10 ± 1	16 ± 2	13 ± 1	31 ± 6	1.1 ± 0.2	30 ± 6
	7I	2 ± 1	7 ± 2	3 ± 1	22 ± 2	0.7 ± 0.3	24 ± 2
	off-site	4 ± 1	10 ± 1	4 ± 1	21 ± 2	1.0 ± 0.4	21 ± 2
April	12N	8 ± 2	18 ± 2	10 ± 1	-	1.6 ± 1.1	34 ± 2
	7I	12 ± 1	25 ± 2	12 ± 1	51 ± 3	1.9 ± 0.6	46 ± 3
	off-site	3 ± 2	10 ± 2	6 ± 1	20 ± 2	0.7 ± 0.5	19 ± 2
May	12N	19 ± 1	24 ± 2	19 ± 1	33 ± 2	1.6 ± 0.4	30 ± 2
	7I	18 ± 2	19 ± 2	17 ± 1	32 ± 2	1.4 ± 0.4	30 ± 2
	off-site	14 ± 2	15 ± 2	15 ± 1	23 ± 2	1.2 ± 0.4	21 ± 2
June	12N	18 ± 2	26 ± 2	18 ± 1	33 ± 2	1.0 ± 0.4	38 ± 2
	7I	17 ± 2	19 ± 2	14 ± 1	29 ± 2	1.1 ± 0.4	33 ± 2
	off-site	16 ± 2	22 ± 2	15 ± 1	25 ± 2	0.9 ± 0.4	30 ± 2
July	12N	38 ± 2	55 ± 2	32 ± 2	66 ± 3	2.6 ± 0.5	67 ± 3
	7I	12 ± 2	12 ± 2	10 ± 1	21 ± 2	0.7 ± 0.3	20 ± 2
	off-site	4 ± 2	5 ± 2	3 ± 1	13 ± 1	0.6 ± 0.3	14 ± 1
August	12N	12 ± 2	17 ± 2	12 ± 1	29 ± 2	1.2 ± 0.4	28 ± 2
	7I	10 ± 2	8 ± 1	6 ± 1	16 ± 2	0.8 ± 0.4	19 ± 2
	off-site	9 ± 1	3 ± 1	4 ± 1	12 ± 1	0.3 ± 0.3	10 ± 1
September	12N	10 ± 1	13 ± 1	10 ± 1	31 ± 2	0.7 ± 0.4	32 ± 2
	7I	9 ± 1	10 ± 1	8 ± 1	21 ± 2	0.8 ± 0.5	22 ± 2
	off-site	4 ± 1	6 ± 1	4 ± 1	12 ± 1	0.3 ± 0.3	13 ± 1
October	12N	13 ± 1	24 ± 2	13 ± 1	41 ± 3	1.3 ± 0.5	38 ± 3
	7I	8 ± 1	20 ± 2	11 ± 1	27 ± 2	1.0 ± 0.6	26 ± 2
	off-site	2 ± 1	8 ± 1	3 ± 1	6 ± 2	< 0.1	10 ± 2
November	12N	21 ± 2	28 ± 2	16 ± 1	-	-	-
	7I	8 ± 1	17 ± 1	9 ± 1	15 ± 2	0.5 ± 0.4	18 ± 2
	off-site	6 ± 1	12 ± 1	7 ± 1	8 ± 1	0.1 ± 0.2	12 ± 1
December	12N	14 ± 1	19 ± 1	13 ± 1	63 ± 3	2.2 ± 0.7	79 ± 4
	7I	12 ± 1	14 ± 1	11 ± 1	19 ± 1	0.7 ± 0.3	22 ± 1
	off-site	10 ± 1	10 ± 1	6 ± 1	12 ± 1	0.4 ± 0.2	13 ± 1
Annual Summary	12N	16 ± 5	24 ± 7	16 ± 4	40 ± 11	1.4 ± 0.4	39 ± 12
	7I	10 ± 3	16 ± 4	10 ± 2	26 ± 6	0.9 ± 0.2	26 ± 5
	off-site	8 ± 3	12 ± 5	8 ± 4	17 ± 5	0.6 ± 0.3	18 ± 5
Percent CG	12N	(0.0080)	(0.0080)	(0.0016)	(0.0010)	(0.00004)	(0.00078)
	7I	(0.0050)	(0.0053)	(0.0010)	(0.00065)	(0.00002)	(0.00052)
	off-site	(0.0040)	(0.0040)	(0.0008)	(0.00042)	(0.00002)	(0.00036)

[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 x 10^{12} for uranium-238 and by 9 x 10^{12} for thorium-232. The mass of the other thorium isotopes in comparison to thorium-232 and the other uranium isotopes in comparison to uranium-238 are negligible.

ranged from 0.23 x 10^{-15} μCi/ml in January to 0.45 x 10^{-15} μCi/ml in June. There is no indication that any radiostrontium in the air samples originated from Argonne operations. The spring increase is evident only in the plutonium and strontium activities, indicating their stratospheric-fallout origin, and not in the thorium and uranium concentrations.

The thorium and uranium concentrations in Table 6 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 directly related to the mass of material collected on the filter paper, and the bulk of these elements in the air is due to resuspension of soil. 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 6% in August to 22% in November of the total plutonium in the samples. The results of americium-241 measurements in air for the year were all less than the detection limit of 5 x 10^{-19} μCi/ml.

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 during reactor operation 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 7. 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 were about 25% less than 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 0.005% and 0.07% of the CG, respectively.

TABLE 7

Hydrogen-3 (Water Vapor) Concentrations in Air, 1976

Month	Location*	No. of Samples	Concentrations in 10^{-12} µCi/ml Av.	Min.	Max.
January	8F	9	5.8	0.35	43
	14I	9	5.5	2.0	11
	12M	2	14	8.2	19
	off-site	2	1.4	0.60	1.6
February	8F	8	12	0.92	50
	14I	8	22	1.0	88
	12M	2	7.4	4.4	10
	off-site	2	0.70	0.47	0.93
March	8F	9	8.4	< 0.1	50
	14I	9	5.4	< 0.1	18
	12M	2	1.3	0.99	1.7
	off-site	2	0.32	< 0.1	0.55
April	8F	9	17	0.31	52
	14I	9	6.9	0.56	26
	12M	1	-	-	18
	off-site	1	-	-	< 0.1
May	8F	8	20	1.8	29
	14I	8	1.3	0.10	3.0
	12M	2	0.72	0.37	1.1
	off-site	2	0.91	0.73	1.1
June	8F	9	23	4.3	131
	14I	8	7.3	3.1	22
	12M	2	11	2.9	19
	off-site	2	3.2	2.4	4.0
July	8F	9	20	1.4	85
	14I	9	9.9	2.6	27
	12M	2	4.9	3.4	6.4
	off-site	2	2.7	1.8	3.6
August	8F	9	14	2.2	52
	14I	9	11	3.2	22
	12M	2	2.6	2.4	2.7
	off-site	2	1.4	0.58	2.2
September	8F	8	8.1	0.10	25
	14I	8	15	0.82	46
	12M	2	5.8	1.7	9.9
	off-site	2	0.92	0.77	1.1
October	8F	9	16	1.9	50
	14I	9	6.2	4.2	10
	12M	2	11	7.0	14
	off-site	2	2.6	2.0	3.2
November	8F	9	1.5	0.40	4.7
	14I	9	3.7	0.41	11
	12M	2	0.87	0.63	1.1
	off-site	2	0.50	0.44	0.55
December	8F	9	0.56	0.12	1.2
	14I	9	10	1.0	30
	12M	2	2.9	1.9	4.0
	off-site	2	0.38	0.21	0.55
Annual Summary	8F	105	12 ± 5	< 0.1	131
	14I	104	8.7 ± 3.5	< 0.1	88
	12M	23	6.7 ± 3.6	0.37	19
	off-site	23	1.3 ± 0.7	< 0.1	4.0
Percent CG	8F	-	0.0060	< 0.00005	0.066
	14I	-	0.0044	< 0.00005	0.044
	12M	-	0.0034	0.00018	0.010
	off-site	-	0.00065	< 0.00005	0.002

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

The off-site concentrations, measured about 10 km (6.2 mi) northwest of the Laboratory, were similar to levels observed at this location in 1975. 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 1976, the total amount of argon-41 discharged from the reactor is estimated to be 4.1×10^4 Ci, based on a measured release rate of 1.42 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). Because the half-life is short, it is appropriate and more meaningful to consider the concentration at various distances from the Laboratory in discussing argon-41 effluent discharges. By the computational method discussed in Section III.A.6., the average concentrations in the predominant wind direction (NNE) were 4.6×10^{-10} μCi/ml (1.1% of the CG) at 1.5 km (0.93 mi) (the site boundary); 2.1×10^{-10} μCi/ml (0.6% of the CG) at 2.4 km (1.5 mi); and 9×10^{-11} μCi/ml (0.2% of the CG) at 4 km (2.5 mi). 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 2.9 Ci in 1976, was released from the Janus reactor (Building 202, location 13I). In addition, other effluents were krypton-85, estimated to be 7.2 Ci, elemental tritium gas, estimated to be 1.9 Ci, and carbon-11, estimated to be 1.3 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 mi), 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), respectively, 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 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,[6] 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.[7] Americium-243 was added to determine their 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 mi) downstream from the waste-water outfall. Sawmill Creek was sampled upstream from the Argonne site and downstream from the waste-water outfall to determine if radioactivity was added to the stream by Argonne waste water or from surface drainage. The sampling locations are shown in Figure 1. Below the waste-water outfall, samples were collected daily by a continuous sampler, which operated about 35% 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 8. Comparison of the results, and 95% confidence limits of the averages, for the two sampling locations show that the nuclides whose presence in Creek water can be attributed to Argonne operations were hydrogen-3, neptunium-237,

TABLE 8

Radionuclides in Sawmill Creek Water, 1976

Type of Activity	Location*	No. of Samples	Concentration (10^{-9} µCi/ml) Av.	Min.	Max.	Percent CG Av.	Min.	Max.
Alpha (nonvolatile)	16K	12	2.2 ± 0.4	1.1	3.3	(0.073)	(0.037)	(0.11)
	7M	253	1.4 ± 0.2	0.44	3.7	(0.047)	(0.015)	(0.12)
Beta (nonvolatile)	16K	12	22 ± 5	6.8	30	(0.73)	(0.23)	(1.0)
	7M	253	16 ± 2	7.5	27	(0.53)	(0.25)	(0.90)
Hydrogen-3	16K	11	< 205	< 200	230	< 0.0068	< 0.0067	0.008
	7M	253	1620 ± 440	< 200	6250	0.054	< 0.0067	0.21
Strontium-89	16K	12	-	-	< 2	-	-	< 0.07
	7M	253	-	-	< 2	-	-	< 0.07
Strontium-90	16K	12	< 0.51	< 0.5	0.58	< 0.17	< 0.17	0.19
	7M	253	< 0.55	< 0.5	0.88	< 0.18	< 0.17	0.29
Iodine-131	16K	12	< 3.4	< 3	7.4	< 1.1	< 1	2.5
	7M	253	< 3.3	< 3	14	< 1.1	< 1	4.7
Barium-140	16K	12	-	-	< 2	-	-	< 0.007
	7M	253	-	-	< 2	-	-	< 0.007
Uranium** (natural)	16K	12	2.4 ± 0.7	1.0	4.8	(0.0060)	(0.0025)	(0.012)
	7M	253	2.5 ± 0.3	1.2	5.9	(0.0062)	(0.0030)	(0.015)
Neptunium-237	16K	12	-	-	< 0.001	-	-	< 0.00003
	7M	253	0.031 ± 0.013	< 0.001	0.32	0.0010	< 0.00003	0.011
Plutonium-238	16K	12	-	-	< 0.002	-	-	< 0.00004
	7M	253	< 0.0055	< 0.002	0.089	0.00011	< 0.00004	0.0018
Plutonium-239	16K	12	-	-	< 0.0005	-	-	< 0.00001
	7M	253	0.010 ± 0.009	0.00066	0.250	0.00020	0.000012	0.0050
Americium-241	16K	11	-	-	< 0.001	-	-	< 0.000025
	7M	243	0.0051 ± 0.0036	< 0.001	0.092	0.00013	< 0.000025	0.0023
Curium-242 and/or Californium-252	16K	11	-	-	< 0.001	-	-	< 0.000005
	7M	243	< 0.0012	< 0.001	0.0037	< 0.000006	< 0.000005	0.000018
Curium-244 and/or Californium-249	16K	11	-	-	< 0.001	-	-	< 0.000014
	7M	243	< 0.0061	< 0.001	0.13	< 0.000087	< 0.000014	0.0019

*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.

plutonium-239, americium-241, curium-244 and/or californium-249, and occasionally strontium-90, iodine-131, plutonium-238, and curium-242 and/or californium-252. The percentage of individual samples containing activity attributable to Argonne was 100% for hydrogen-3, neptunium-237, and plutonium-239; 75% for americium-241; and 50% for curium-244 and/or californium-249. 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 both concentration and CG, was hydrogen-3. Its average concentration (equivalent to 0.054% of the CG) was similar to 1974 and earlier years, and about 12% of the 1975 average.

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 1976, the hydrogen-3 content of other lakes and streams ranged from $< 200 \times 10^{-9}$ μCi/ml to 445×10^{-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 2×10^{-9} μCi/ml to the nonvolatile beta activity at both locations and that the Argonne contribution to the water below the outfall averaged about 1×10^{-9} μCi/ml. The Argonne contribution remained the same as 1975 levels, while the fallout contribution decreased by about 30%. 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 14 Ci of hydrogen-3, 0.3 mCi of neptunium-237, 0.09 mCi of plutonium-239, 0.04 mCi of americium-241, and < 0.05 mCi of curium and californium nuclides.

Since Sawmill Creek empties into the Des Plaines River, which in turn flows into the Illinois River, the radioactivity in the latter two streams is important in assessing the contribution of Argonne waste water to the environmental radioactivity. The Des Plaines River was sampled twice a month below, and monthly above, the mouth of Sawmill Creek to determine if the radioactivity in the Creek had any effect on the activity in the River. Annual summaries of the results obtained for these two locations are given in Table 9. The average nonvolatile alpha and beta activities and uranium concentrations in the River are slightly higher than previous years and are attributed to below normal rainfall which resulted in River samples with a higher dissolved solids content and associated radioactive materials. The average nonvolatile alpha and beta activities, 1.1×10^{-9} μCi/ml and 10×10^{-9} μCi/ml, respectively, of 21 off-site surface water samples collected this year (excluding the Des Plaines River) are similar to the levels found in previous years. The activity in Sawmill Creek is usually reduced by dilution so that it is not detectable as such in the Des Plaines River. The natural nonvolatile beta activity was 8×10^{-9} μCi/ml, and the excess, 2×10^{-9} μCi/ml, was due to fallout.

The radioactivity in samples of Illinois River water, shown in Table 10, were similar to those found in other bodies of water in the area and to the activities found previously at these same locations. No radioactivity originating at Argonne could be detected in the Des Plaines or Illinois Rivers.

3. Soil, Grass, and Bottom Sediment

The plutonium content of soil, grass, and bottom sediment was measured at the site perimeter and off the site. The object of the off-site sampling was to determine the total 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

TABLE 9

Radionuclides in Des Plaines River Water, 1976

Type of Activity	Location*	No. of Samples	Concentration (10^{-9} µCi/ml) Av.	Min.	Max.	Percent CG Av.	Min.	Max.
Alpha (nonvolatile)	A	12	2.0 ± 0.5	1.3	4.3	(0.067)	(0.043)	(0.14)
	B	24	1.6 ± 0.3	0.74	3.8	(0.053)	(0.025)	(0.13)
Beta (nonvolatile)	A	12	14 ± 3	7.3	21	(0.47)	(0.24)	(0.70)
	B	24	15 ± 3	6.2	27	(0.50)	(0.21)	(0.90)
Hydrogen-3	A	9	-	-	< 200	-	-	< 0.007
	B	22	< 205	< 200	230	< 0.007	< 0.007	0.008
Strontium-89	A	12	-	-	< 2	-	-	< 0.07
	B	24	-	-	< 2	-	-	< 0.07
Strontium-90	A	12	< 0.55	< 0.5	0.77	< 0.18	< 0.17	0.26
	B	24	< 0.52	< 0.5	0.63	< 0.17	< 0.17	0.21
Iodine-131	A	12	-	-	< 3	-	-	< 1
	B	24	-	-	< 3	-	-	< 1
Barium-140	A	12	-	-	< 2	-	-	< 0.007
	B	24	-	-	< 2	-	-	< 0.007
Uranium** (natural)	A	12	1.8 ± 0.4	1.2	3.4	(0.0045)	(0.0030)	(0.0085)
	B	24	1.9 ± 0.3	0.91	3.3	(0.0048)	(0.0023)	(0.0082)
Neptunium-237	A	12	-	-	< 0.001	-	-	< 0.00003
	B	12	-	-	< 0.001	-	-	< 0.00003
Plutonium-238	A	12	-	-	< 0.002	-	-	< 0.00004
	B	12	-	-	< 0.002	-	-	< 0.00004
Plutonium-239	A	12	-	-	< 0.0005	-	-	< 0.00001
	B	12	-	-	< 0.0005	-	-	< 0.00001
Americium-241	A	11	-	-	< 0.001	-	-	< 0.000025
	B	12	-	-	< 0.001	-	-	< 0.000025
Curium-242 and/or Californium-252	A	11	-	-	< 0.001	-	-	< 0.000005
	B	12	-	-	< 0.001	-	-	< 0.000005
Curium-244 and/or Californium-249	A	11	-	-	< 0.001	-	-	< 0.000014
	B	12	-	-	< 0.001	-	-	< 0.000014

*Location A, near 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 2.7 µg/l.

TABLE 10

Radionuclides in Illinois River Water, 1976

(concentrations in 10^{-9} μCi/ml)

Location	Date Collected	Alpha*	Beta*	Hydrogen-3	Uranium**	Neptunium-237	Plutonium-239
McKinley Woods State Park	June 22	1.5	13	< 200	1.2	< 0.0005	0.00027
Below Dresden Power Station	June 22	0.91	5.4	< 200	1.2	< 0.0005	0.00090
Morris	June 22	1.8	7.2	< 200	1.0	-	-
Starved Rock State Park	June 22	1.4	7.0	< 200	1.1	-	-
McKinley Woods State Park	October 14	0.57	12	< 200	0.89	< 0.0005	0.00017
Below Dresden Power Station	October 14	0.73	8.6	< 200	0.64	< 0.0005	0.00018
Morris	October 14	0.95	8.2	< 200	0.47	-	-
Starved Rock State Park	October 14	1.1	9.4	< 200	0.87	-	-

*Nonvolatile activity.

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

preparation techniques used by the ERDA New York Health and Safety Laboratory[8] were used. Sites were selected in several directions and at various distances from the Laboratory. Each site was selected on the basis that the soil appeared, or was known to have been, undisturbed for a number of years. Attempts were made to select open, level, grassy areas that were mowed at reasonable intervals. Public parks were selected when available.

Each soil sample consisted of two cores totaling 173 cm^2 in area by 30 cm deep. 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 11 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. Fallout deposition values found by other laboratories[9] are in the same range as those reported here, about 2-3 x 10^{-3} $\mu Ci/m^2$.

Composite monthly precipitation samples were analyzed for plutonium-239. The results are given in Table 12 along with results from 1973, 1974, and 1975 for comparison. The total 1976 deposition by precipitation was a fourth of that of 1975, a reflection of the decreased fallout from atmospheric nuclear tests, and was equivalent to 0.2% of the total plutonium previously deposited, which is reported to be 2.2 x 10^{-3} $\mu Ci/m^2$.[10] The increase from 1973 to 1974 can be related to announced atmospheric nuclear tests.

The thorium, uranium, and cesium-137 content of soil samples was also measured and the concentrations in the perimeter and off-site samples (Table 13) were similar. These are expected levels of the naturally-occurring thorium and uranium activities and normal fallout concentrations of cesium-137. In terms of mass, the average thorium concentrations were 4.0 µg/g and 2.2 µg/g at the perimeter and off the site, respectively, while the uranium concentrations were 2.2 µg/g at the perimeter and 1.6 µg/g off-site.

The results of radioactivity measurements in grass are given in Tables 14 and 15. The grass samples were washed before analysis to remove surface

TABLE 11

Plutonium Content of Soil, 1976

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*					
July 22	10P	0.2 ± 0.1	0.08 ± 0.03	3.4 ± 0.3	1.5 ± 0.1	0.055
July 22	15H	0.1 ± 0.1	0.04 ± 0.02	2.3 ± 0.2	0.9 ± 0.1	0.047
July 22	12C	0.3 ± 0.1	0.13 ± 0.03	6.7 ± 0.4	2.6 ± 0.2	0.050
October 18	10N	0.2 ± 0.1	0.07 ± 0.03	1.9 ± 0.2	0.8 ± 0.1	0.089
October 18	14L	0.2 ± 0.1	0.08 ± 0.03	5.1 ± 0.3	2.0 ± 0.1	0.037
October 18	13D	0.3 ± 0.1	0.10 ± 0.02	7.3 ± 0.4	2.7 ± 0.1	0.037
	Average	0.2 ± 0.1	0.08 ± 0.03	4.4 ± 2.3	1.8 ± 0.8	0.052
	Off-Site					
June 22	McKinley Woods State Park, IL	0.3 ± 0.1	0.13 ± 0.05	6.5 ± 0.5	3.1 ± 0.3	0.044
June 23	McCormick Woods, Brookfield, IL	0.2 ± 0.1	0.10 ± 0.03	4.7 ± 0.4	1.9 ± 0.2	0.051
June 23	Bemis Woods, Hinsdale, IL	0.4 ± 0.1	0.12 ± 0.04	7.0 ± 0.6	2.2 ± 0.2	0.053
October 12	St. Joseph, Michigan	0.3 ± 0.1	0.14 ± 0.04	6.6 ± 0.5	2.7 ± 0.2	0.052
October 13	Willow Springs, IL	0.5 ± 0.1	0.18 ± 0.04	7.6 ± 0.5	2.9 ± 0.2	0.062
October 14	Dresden Lock and Dam, IL	0.6 ± 0.3	0.17 ± 0.10	5.2 ± 0.9	2.0 ± 0.3	0.089
	Average	0.4 ± 0.1	0.14 ± 0.03	6.3 ± 1.1	2.5 ± 0.5	0.058

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

TABLE 12

Total Plutonium-239,240 Deposition
(units of 10^{-6} μCi/m^2)

Month	1973	1974	1975	1976
January	-	0.60 ± 0.10	1.1 ± 0.18	0.25 ± 0.06
February	0.14 ± 0.04	1.10 ± 0.16	1.7 ± 0.19	0.38 ± 0.09
March	0.27 ± 0.07	2.0 ± 0.22	1.5 ± 0.17	0.58 ± 0.09
April	0.40 ± 0.07	5.2 ± 0.37	4.0 ± 0.43	0.21 ± 0.07
May	0.50 ± 0.06	4.6 ± 0.31	1.8 ± 0.18	0.58 ± 0.11
June	0.16 ± 0.04	3.5 ± 0.26	1.9 ± 0.18	0.65 ± 0.12
July	0.32 ± 0.10	5.6 ± 0.37	0.48 ± 0.10	0.26 ± 0.07
August	0.34 ± 0.14	1.0 ± 0.19	0.38 ± 0.09	0.15 ± 0.06
September	0.27 ± 0.17	0.25 ± 0.11	0.10 ± 0.06	0.06 ± 0.05
October	0.30 ± 0.12	0.45 ± 0.10	0.12 ± 0.08	0.30 ± 0.10
November	0.22 ± 0.10	0.73 ± 0.12	0.08 ± 0.07	0.05 ± 0.04
December	0.46 ± 0.12	0.71 ± 0.12	0.21 ± 0.08	0.12 ± 0.07
Average Monthly Deposition	0.31 ± 0.07	2.1 ± 1.2	1.1 ± 0.7	0.30 ± 0.13
Annual Deposition	3.38	25.74	13.37	3.59
Percent Added to Existing	0.2	1.2	0.6	0.2

TABLE 13

Cesium-137, Thorium, and Uranium in Soil, 1976

(concentrations in 10^{-6} μCi/g)

Date Collected	Location	Cesium-137	Thorium-228	Thorium-230	Thorium-232	Uranium (natural)
	Perimeter*					
July 22	10P	0.3 ± 0.1	0.21 ± 0.05	0.19 ± 0.04	0.21 ± 0.04	1.3 ± 0.1
July 22	15H	0.1 ± 0.1	0.46 ± 0.04	0.59 ± 0.05	0.49 ± 0.04	2.0 ± 0.1
July 22	12C	0.3 ± 0.1	0.55 ± 0.04	0.72 ± 0.05	0.48 ± 0.04	1.5 ± 0.1
October 18	10N	0.1 ± 0.1	0.65 ± 0.08	0.77 ± 0.07	0.65 ± 0.07	1.5 ± 0.1
October 18	14L	0.3 ± 0.1	0.41 ± 0.04	0.55 ± 0.05	0.43 ± 0.04	1.4 ± 0.1
October 18	13D	0.4 ± 0.1	0.38 ± 0.04	0.51 ± 0.05	0.39 ± 0.04	1.3 ± 0.1
	Average	0.2 ± 0.1	0.44 ± 0.15	0.56 ± 0.20	0.44 ± 0.14	1.5 ± 0.3
	Off-Site					
June 22	McKinley Woods State Park, IL	0.4 ± 0.1	0.18 ± 0.02	0.16 ± 0.02	0.16 ± 0.02	0.9 ± 0.1
June 23	McCormick Wocds, Brookfield, IL	0.3 ± 0.1	0.23 ± 0.02	0.27 ± 0.02	0.22 ± 0.02	1.2 ± 0.1
June 23	Bemis Woods, Hinsdale, IL	0.4 ± 0.1	0.23 ± 0.02	0.30 ± 0.02	0.18 ± 0.01	1.6 ± 0.1
October 12	St. Joseph, Michigan	0.4 ± 0.1	0.22 ± 0.03	0.20 ± 0.02	0.20 ± 0.02	0.3 ± 0.1
October 13	Willow Springs, IL	0.5 ± 0.2	-	-	-	1.0 ± 0.1
October 14	Dresden Lock and Dam, IL	0.4 ± 0.1	0.51 ± 0.03	0.64 ± 0.03	0.45 ± 0.03	1.6 ± 0.1
	Average	0.4 ± 0.1	0.27 ± 0.15	0.31 ± 0.22	0.24 ± 0.14	1.1 ± 0.5

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

TABLE 14

Plutonium Content of Grass Samples, 1976

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*				
July 22	10P	< 0.1	< 0.01	0.5 ± 0.2	0.08 ± 0.04
July 22	15H	< 0.1	< 0.01	0.1 ± 0.1	0.03 ± 0.02
July 22	12C	< 0.1	< 0.01	0.1 ± 0.1	0.05 ± 0.02
October 18	10N	< 0.1	< 0.01	0.2 ± 0.1	0.02 ± 0.01
October 18	14L	< 0.1	< 0.01	0.3 ± 0.1	0.03 ± 0.01
October 18	13D	< 0.1	< 0.01	0.3 ± 0.1	0.05 ± 0.02
	Average	< 0.1	< 0.01	0.2 ± 0.2	0.04 ± 0.02
	Off-Site				
June 22	McKinley Woods State Park, IL	< 0.1	< 0.01	0.2 ± 0.1	0.04 ± 0.01
June 23	McCormick Woods, Brookfield, IL	< 0.1	< 0.01	0.4 ± 0.1	0.05 ± 0.01
June 23	Bemis Woods, Hinsdale, IL	< 0.1	< 0.01	0.5 ± 0.1	0.07 ± 0.02
October 12	St. Joseph, Michigan	< 0.1	< 0.01	0.7 ± 0.1	0.08 ± 0.02
October 13	Willow Springs, IL	< 0.1	< 0.01	< 0.1	< 0.01
October 14	Dresden Lock and Dam, IL	< 0.1	< 0.01	0.4 ± 0.1	0.05 ± 0.01
	Average	< 0.1	< 0.01	0.4 ± 0.2	0.05 ± 0.02

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

TABLE 15

Radionuclides in Grass Samples, 1976

(concentrations in 10^{-6} μCi/g)

Date Collected	Location	Cesium-137	Thorium-228	Thorium-230	Thorium-232	Uranium (natural)
	Perimeter*					
July 22	10P	0.1 ± 0.1	0.004 ± 0.001	0.004 ± 0.001	0.003 ± 0.001	0.053 ± 0.005
July 22	15H	< 0.1	0.006 ± 0.001	0.006 ± 0.001	0.003 ± 0.001	0.029 ± 0.003
July 22	12C	< 0.1	0.002 ± 0.001	0.002 ± 0.001	0.001 ± 0.001	0.020 ± 0.002
October 18	10N	< 0.1	0.003 ± 0.001	0.006 ± 0.001	0.003 ± 0.001	0.017 ± 0.004
October 18	14L	< 0.1	0.008 ± 0.001	0.007 ± 0.001	0.004 ± 0.001	0.021 ± 0.002
October 18	13D	< 0.1	0.006 ± 0.001	0.007 ± 0.001	0.003 ± 0.001	0.014 ± 0.003
	Average	< 0.1	0.005 ± 0.002	0.005 ± 0.002	0.003 ± 0.001	0.026 ± 0.014
	Off-Site					
June 22	McKinley Woods State Park, IL	< 0.1	< 0.001	< 0.001	< 0.001	0.017 ± 0.002
June 23	McCormick Woods, Brookfield, IL	< 0.1	< 0.001	< 0.001	< 0.001	0.059 ± 0.005
June 23	Bemis Woods, Hinsdale, IL	< 0.1	< 0.001	< 0.001	< 0.001	0.014 ± 0.002
October 12	St. Joseph, Michigan	0.1 ± 0.1	0.007 ± 0.001	0.010 ± 0.001	0.007 ± 0.001	0.030 ± 0.004
October 13	Willow Springs, IL	< 0.1	< 0.001	< 0.001	< 0.001	0.027 ± 0.004
October 14	Dresden Lock and Dam, IL	0.1 ± 0.1	-	0.006 ± 0.001	0.004 ± 0.001	0.019 ± 0.003
	Average	< 0.1	< 0.002	< 0.003	< 0.002	0.028 ± 0.016

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

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.

Two bottom sediment samples were analyzed for thorium, uranium, and plutonium for comparison with previous measurements. The results are given in Table 16. Plutonium results vary widely between locations and are strongly dependent on the retentiveness of the bottom material. Plutonium concentrations up to about 30 x 10^{-9} µCi/g may be considered normal based on previous data.[3] The thorium and uranium concentrations were normal and similar to those found in soil. In terms of mass, the concentrations in Table 16 were 1.9 µg/g for thorium and 2.4 µg/g for uranium.

TABLE 16

Radionuclides in Bottom Sediment, 1976

Nuclide	Concentration (10^{-9} µCi/g) Des Plaines River, Brookfield, IL June 23	Flagg Creek Willow Springs, IL October 13
Thorium-228	240 ± 20	230 ± 30
Thorium-230	280 ± 20	410 ± 40
Thorium-232	180 ± 10	200 ± 30
Uranium (natural)	1600 ± 100	1700 ± 100
Plutonium-238	0.5 ± 0.1	0.1 ± 0.1
Plutonium-239	7.9 ± 0.5	1.1 ± 0.1

4. Foodstuffs

Raw milk was collected monthly from a local dairy farm south of Lemont and analyzed for several radioactive nuclides by methods similar to those used for water. Iodine-131 was analyzed with a detection limit of 1×10^{-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 17. The average hydrogen-3 concentration in milk is identical to the concentration of hydrogen-3 found in surface water samples away from the site. The average strontium-90 and cesium-137 concentrations decreased compared to 1975. These nuclides are fission products from nuclear tests and their presence in milk is not related to Argonne operations. Iodine-131 and barium-140 concentrations above the detection limit late in the year are attributed to the previously discussed atmospheric nuclear tests.

The concentrations given in Table 17 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 < 0.003% of the hydrogen-3, 0.5% of the strontium-90, and 0.005% of the cesium-137 Concentration Guides.

Several samples of garden vegetables grown 8-16 km (5-10 mi) from Argonne were obtained and analyzed for plutonium and uranium. No thorium or gamma-ray emitters were detected. The plutonium and uranium results are given in Table 18, and the concentrations are expressed in terms of air-dried weight. Compared to the results for grass, the plutonium-239 concentrations are a minimum of 4 times lower, while the uranium concentrations are 5 to 25 times lower. The uranium results are in good agreement with analyses of the same types of vegetables measured during 1975. The plutonium content, although extremely variable, covered about the same range of concentrations as in 1975. As in the case of milk, the radioactivity is unrelated to Argonne operations, but the information is valuable as background data.

TABLE 17

Radicnuclides in Milk, 1976

(concentrations in 10^{-9} µCi/ml)

Date Collected	Hydrogen-3	Strontium-89	Strontium-90	Iodine-131	Cesium-137	Barium-140-Lanthanum-140
January 14	< 200	< 2	3.2	< 0.1	3.5	< 2
February 4	< 200	< 2	2.5	< 0.1	3.5	< 2
March 3	< 200	< 2	2.4	< 0.1	3.7	< 2
April 7	< 200	< 2	3.3	< 0.1	2.4	< 2
May 5	< 200	< 2	2.4	< 0.1	3.2	< 2
June 2	< 200	< 2	6.3	< 0.1	3.3	< 2
July 7	< 200	< 2	7.1	< 0.1	2.2	< 2
August 4	201	< 2	2.5	< 0.1	1.3	< 2
September 1	379	< 2	2.8	< 0.1	0.6	< 2
October 6	< 200	< 2	3.6	< 0.1	1.2	< 2
November 3	< 200	< 2	3.6	3.3	1.9	< 2
December 1	< 200	< 2	1.4	< 0.1	1.7	2.5
Average	< 215	< 2	3.4 ± 1.0	< 0.4	2.4 ± 0.7	< 2.1

TABLE 18

Uranium and Plutonium-239 in Garden Vegetables, 1976

Food	Uranium (natural) (concentration 10^{-9} μCi/g)	Plutonium-239 (concentration 10^{-12} μCi/g)
Beets	3.2 ± 0.3	< 1
Beet Tops	7.3 ± 0.7	14 ± 20
Cabbage	5.0 ± 0.4	2 ± 1
Corn (kernels)	1.7 ± 0.3	47 ± 10
Tomatoes	1.1 ± 0.3	< 1

5. External Penetrating Radiation

Measurements were made with calcium fluoride (dysprosium activated) thermoluminescent dosimeter 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 19 and 20, 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 57 to 68 days, and in total covered the period from January 5, 1976, to January 4, 1977. 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.

The off-site results averaged 90 mrem/yr. The standard error of the mean of all 30 results was 3 mrem/yr. (The standard error of the average of the

TABLE 19

Environmental Penetrating Radiation at Off-Site Locations, 1976

Location	Dose Rate (mrem/year)						
	Period of Measurement						
	1/5-3/9	3/9-5/12	5/12-7/8	7/8-9/14	9/14-11/11	11/11-1/4	Average
Downers Grove	90	90	82	91	92	93	90 ± 5
Lockport	87	82	82	87	88	81	85 ± 4
Lombard	89	88	90	96	96	103	94 ± 7
Oak Lawn	87	81	72	80	81	84	81 ± 6
Oakbrook	94	100	92	107	104	107	101 ± 8
Average	89 ± 3	88 ± 9	84 ± 9	92 ± 12	92 ± 10	94 ± 13	90 ± 9

TABLE 20

Environmental Penetrating Radiation at ANL, 1976

Location	Dose Rate (mrem/year)						
	Period of Measurement						
	1/5-3/9	3/9-5/12	5/12-7/8	7/8-9/14	9/14-11/11	11/11-1/4	Average
14L - Boundary	85	90	80	94	92	97	90 ± 7
14I - Boundary	-	376	374	397	-	397	386 ± 18
14G - Boundary	94	100	89	97	102	100	97 ± 5
13D - Boundary	77	74	-	68	68	70	71 ± 5
9/10EF - Boundary	101	91	96	93	101	100	97 ± 4
8H - Boundary	125	116	103	100	120	107	112 ± 10
8H - Center, St. Patrick's Cemetery	127	113	108	110	120	115	116 ± 7
7I - Boundary	542	444	-	524	625	511	528 ± 75
6I - 200 m N of Quarry Road	-	98	97	106	117	118	107 ± 12
9L - Boundary	-	88	79	88	88	92	87 ± 5
9H - 50 m SE of CP-5	2650	2260	2230	2310	2670	2400	2420 ± 200
8H - 50 m S of 316	1390	527	177	198	159	197	455 ± 485
9H - 23 m E of 316	2990	3410	561	671	198	516	1440 ± 1420
9I - 45 m NE of 350 210 m NE of 316	179	141	98	100	115	115	125 ± 31
8H - 200 m NW of Waste Storage Area (Heliport)	161	154	139	156	176	145	155 ± 13
7I - Center, Waste Storage Area	5780	5270	7250	8510	9290	6410	7080 ± 1570
11J - Lodging Facilities	107	94	-	-	-	-	101 ± 28
10/11K - Lodging Facilities	96	87	73	83	85	82	84 ± 7
9J - Between ZGS Condenser and 386	81	105	75	66	83	69	80 ± 14
14I - 210 m N of 202	-	-	738	827	877	852	823 ± 84
13J - 140 m NE of 202	1090	1750	1560	1920	1820	1890	1670 ± 310
11G - 30 m SW of Cyclotron	94	-	-	-	-	-	(94)
12M - 30 m W of 55	142	138	-	118	159	121	136 ± 19
14/15N - Former Nike Site (Exterior)	91	99	-	-	-	-	95 ± 17

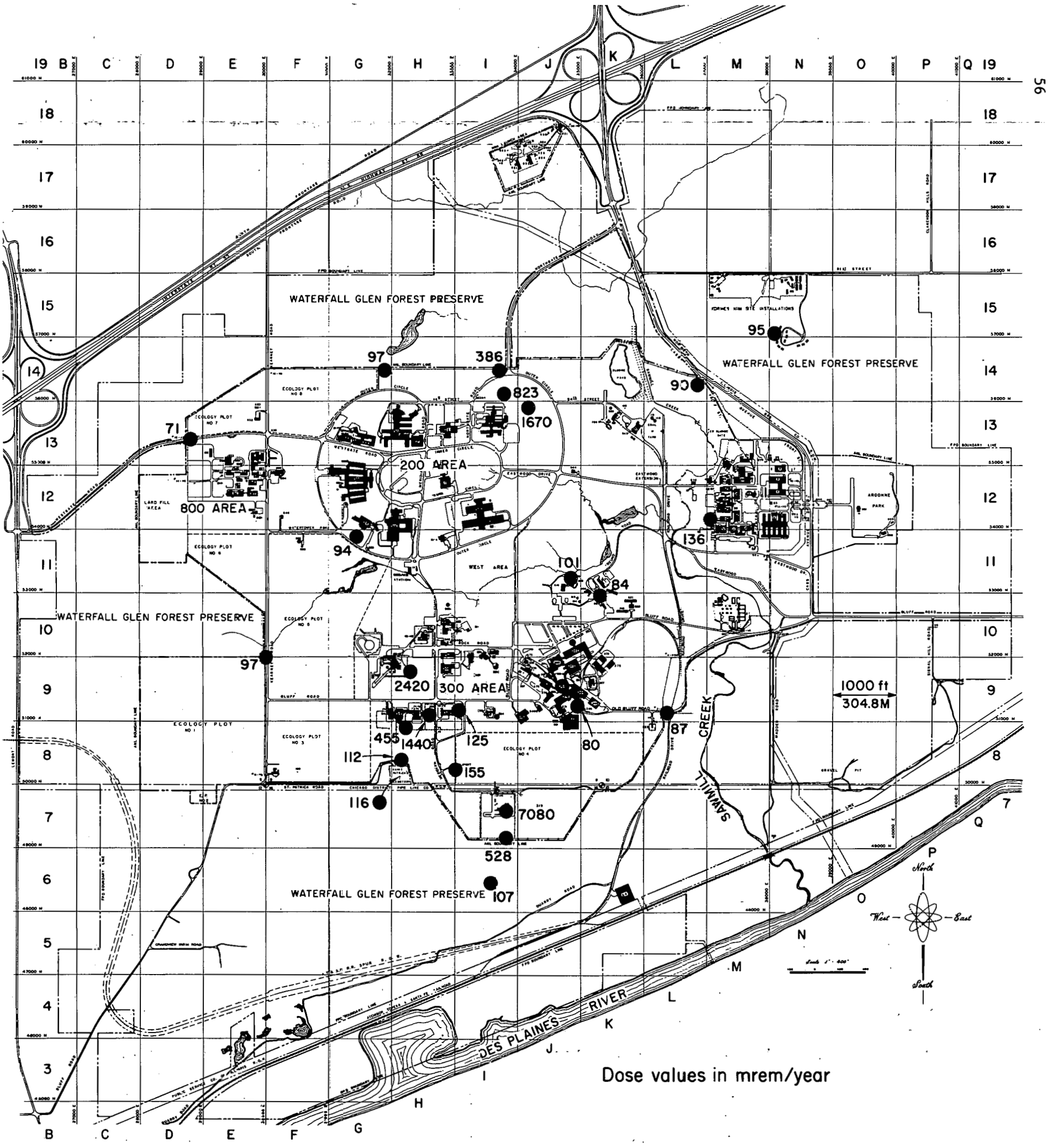

Fig. 6. Penetrating Radiation Measurements at the ANL Site

five separate locations was 9 mrem/yr.) For 1972, 1973, 1974, and 1975, corresponding averages and standard errors were 105 ± 1.5 mrem/yr, 100 ± 2 mrem/yr, 99 ± 2 mrem/yr, and 94 ± 1 mrem/yr, respectively. Thus, while averages for consecutive years have agreed within statistical variations at the 95% confidence level, there has been a small but steady decrease averaging about 3 mrem/yr for the past five years. 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 8 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 75 to 109 mrem/yr may be considered normal with a 95% probability, or 95% of the boundary results should have averages in this range. To compare boundary results for individual sampling periods, the standard deviation of the 30 individual off-site results is useful. This value is 8.5 mrem/yr, so that single boundary results in the range of 90 ± 17 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 440 mrem/yr. The excess dose rate at 7I in 1976 was about twice that found in 1974 and 1975, but about the same as in 1972 and 1973. At 14I the average dose rate was about 300 mrem/yr above normal. This dose is attributed to the increased use during 1976 of cobalt-60 irradiation sources in Building 202.

Dose variations from period to period at the same location, which are at least in part statistical, and 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 here 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 (90 mrem/yr) plus twice the standard error in the average, 9 mrem/yr; or 3) occasional results at a particular location are significantly above the normal value for that location, although such results were still in the off-site normal range. The last criterion would apply to a location such as 13D, where the results have been consistently below the off-site 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 the 8H boundary location is of interest, since it lies on the edge of St. Patrick's Cemetery, which was in use before Argonne was constructed, and which is open to visitors. The dose at this location was about 22 mrem/yr above the off-site average but within the upper limit (118 mrem/yr) of the normal range. Results for some individual periods were above the expected range. A conservative upper limit for the excess dose at 8H is 10-20 mrem/yr, based on this type of comparison. Similar results were obtained in 1974 and 1975.

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, respectively (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 is 4 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 Radioactivity

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 21. 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 mi) downstream from Argonne (Section I.E.), but the dilution of Sawmill Creek water at this point is so great that the dose calculation is meaningless. No other exposure pathways are significant.

Argon-41 and 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.[11] The following parameters were used in the calculations:

a) release rates (measured in the CP-5 exhaust stack); argon-41, 1.42 Ci/MW-hr; hydrogen-3, 0.041 Ci/hr
b) meteorological data: the 15-year average values given in ANL-7084[2]
c) the usual parameters for stack height, building, wake, plume momentum, temperatures, etc.

The calculations were carried out to 80 km (50 mi). The argon-41 results for the first 4.8 km (3 mi) are given in Table 22. Doses were calculated for the mid-point of the annular interval. Thus, the dose for 0-1.6 km (0-1 mi) average is the dose at 0.8 km (0.5 mi). The highest dose rates are in the N to ENE sectors. The closest full-time residents in this area are 2.1 km (1.3 mi) from the reactor, where the dose (in the NNE direction) is 3.4 mrem/yr,

TABLE 21

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 ERDA 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 ERDAM 0524, the concentrations used were one-tenth of the 168 hour occupational values specified by the ICRP.[12]

TABLE 22

Argon-41 Radiation Dose From CP-5 Reactor, 1976

(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	5.2	12.8	2.4	1.0
NNE	5.9	14.8	2.7	1.2
NE	4.7	11.6	2.2	0.8
ENE	5.0	12.5	2.2	1.0
E	3.9	9.9	1.7	0.8
ESE	3.7	9.2	1.7	0.7
SE	3.6	8.8	1.6	0.7
SSE	3.1	7.4	1.4	0.7
S	3.2	8.1	1.4	0.7
SSW	3.7	9.2	1.7	0.7
SW	3.2	7.9	1.4	0.7
WSW	2.7	6.8	1.2	0.5
W	2.1	5.2	1.0	0.3
WNW	2.2	5.7	1.2	0.5
NW	2.7	6.6	1.2	0.5
NNW	3.2	7.9	1.4	0.7
Average	3.6	9.0	1.6	0.7

less than 1% of the standard (500 mrem/yr) for individuals in uncontrolled areas. The value in Table 22 that corresponds most closely to this dose and distance is 2.7 mrem/yr at 2.4 km, the mid-point of the 1.6-3.2 km interval. The dose varies greatly with distance in the first several kilometers. Thus, in the NNE direction individuals would receive 5.4 mrem/yr if they were outdoors throughout the year at 1.6 km and 1.5 mrem/yr if they were outdoors at 3.2 km. A small section of the most-used portion of the Waterfall Glen Forest Preserve, the former Rocky Glen Forest Preserve southeast of the site, begins about 1.5 km (0.93 mi) from CP-5, and in this direction the dose rate at 1.5 km (0.93 mi) is 3.1-3.6 mrem/yr.

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 direction to the north-northeast, the dose rates at the site boundary were in the normal range found off-site. Increases in excess of two standard deviations of the off-site average (i.e., greater than 90 + 17 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 15-20 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 1 was used to calculate the population dose from argon-41. The results are given in Table 23, 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 mi) 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 mi), 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 mi) annulus in the NNE direction, where the annual dose is calculated to be 0.007 mrem/yr. A summary

TABLE 23

Argon-41 Average Individual and Population Dose From CP-5 Reactor, 1976

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	90	5.8	324
3.2-4.8	17,405	0.7	90	12.2	1566
0-80	8,118,740	0.019	90	154	7.31×10^5

of the results is given in Table 24. A comparison of the doses calculated from the meteorological model with the measured data from Table 7 is given in Table 25. 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 24

Hydrogen-3 Average Individual and Population Dose From CP-5 Reactor, 1976

Distance (km)	Population	Avg. mrem/year	Man-rem/year
0-1.6	0	-	-
1.6-3.2	3,595	0.0033	0.012
3.2-4.8	17,405	0.0018	0.031
0-80	8,118,740	0.00014	1.15

TABLE 25

Comparison of Calculated and Measured Hydrogen-3 Dose Rates, 1976

Direction	Distance (km)	Calculated (mrem/yr)	Measured (mrem/yr)
NNE	1.5	0.011	0.018
ENE	1.9	0.007	0.014
SW	0.45	0.032	0.027

The iodine-131 released from CP-5 would result in an individual dose of 0.002 mrem/yr at 1.5 km (0.93 mi) in the NNE sector and a population dose of about 0.001 man-rem/yr in the 1.6-3.2 km (1-2 mi) 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 concentrations of those nuclides added to Sawmill Creek by Argonne waste water, and the corresponding dose rates if water at these concentrations were used as the sole water supply by an individual are given in Table 26. In the case of strontium-90, a small fraction 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 or recreational purposes. Inspection of the area shows there are few fish in the stream, and they do not constitute a significent source of food for any individual.

TABLE 26

Radionuclide Concentrations and Dose Estimates
for Sawmill Creek Water, 1976

Nuclide	Conc. (avg.) 10^{-9} µCi/ml	Dose Rate mrem/year	Percent of Standard
Hydrogen-3	1.6×10^3	0.27	0.054
Strontium-90	< 0.55	< 5.5	< 0.18
Neptunium-237	0.031	0.031	0.0010
Plutonium-239	0.010	0.0060	0.002
Americium-241	0.0051	0.0019 (kidney) 0.0031 (bone)	0.00013 0.00010
Curium-244 and/or Californium-249	< 0.0061	< 0.0026 or < 0.0046	< 0.00009 or < 0.00015

As indicated in Table 8, 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: 3×10^{-3} mrem/yr for plutonium-238 and from 2.6×10^{-4} to 1.8×10^{-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 south boundary adjacent to St. Patrick's Cemetery (8H), and at the north boundary near Building 202 (14I). The results are discussed in Section III.A.5.

At location 7I, the fence-line dose from Argonne was about 440 mrem/yr. Approximately 300 m (0.2 mi) south of the fence line the measured dose had decreased to about 15 mrem/yr above normal. There are no individuals living in this area. The closest residents are about 1.6 km (1 mi) south of the fence line. At this distance the calculated dose rate (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 0.011 mrem/yr, if the energy of the radiation was 0.66 MeV, and 0.055 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 20 mrem/yr. An individual spending an average of 1 hr/week at this location would receive an annual dose of 0.12 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 300 mrem/yr, the nearest residents are 750 m (0.47 mi) to the north-northwest. The dose at that location (calculated as described above) was 0.51 mrem/yr.

The applicable Radiation Protection Standards for whole body external radiation dose to the general population is a maximum of 500 mrem/yr to

critical individuals, or if individual doses are not known, 170 mrem/yr to a suitable sample of the exposed population.[4] The latter criterion assumes that the maximum dose to individuals in the sample will not exceed the average by more than a factor of three. Thus, the doses to individuals living near the site will not exceed 0.1% of the 500 mrem/yr limit or 0.3% of the "suitable sample" limit. At the fence line where higher doses were measured, the land is wooded and unoccupied.

B. Chemical Pollutants

The nonradioactive environmental 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.[13] Stream Quality standards appear in Part II and Effluent Quality standards appear in Part IV of this document.

The analytical techniques were as follows. The concentrations of chromium, copper, iron, nickel, and zinc were determined using conventional atomic absorption spectrophotometry. Barium concentrations were determined using an acetylene-nitrous oxide flame. The concentrations of silver, cadmium, and lead were determined using a flameless technique as previously described.[14] Mercury was determined using cold atomic absorption spectrophotometry. Fluoride levels were determined using an ion selective electrode, and the pH was determined using conventional pH electrode measurements. Dissolved oxygen, ammonia nitrogen, and hexavalent chromium levels were determined using procedures described in Standard Methods.[15] The levels of fecal coliform were determined using the membrane filter technique.[15] The cyanide concentration was estimated with a Hach Chemical Co. cyanide test kit. Beryllium was determined fluorophotometrically as previously described.[16] The quality assurance studies performed are described in Appendix IV.B.

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 average values 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 value and the variation is representative of climatic conditions.

All of the results are compared to the appropriate State standards, which are listed in Table 27. 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, which is a tributary of the Des Plaines River, since this is the principal route for waste water leaving the Argonne site. An expanded effort was devoted to studying cooling tower blowdown effluents and a continuing emphasis was placed on the control of mercury release in the effluent.

1. 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 is normally comprised of about equal parts of sanitary waste water and water from laboratory operations. This year, however, the laboratory waste exceeded the sanitary waste by as much as a factor of two from October to December (see Figure 5). 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. Hence, during some periods of collection, the water samples contain a higher proportion of laboratory waste than at other times.

The performance of the sanitary waste treatment plant was monitored by the Reclamation Control Laboratory of the Plant Operations 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

TABLE 27

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
Cyanide	0.025	0.025	0.020
Fluoride	1.4	15*	0.02
Iron	1.0	2.0	0.08
Lead	0.1	0.1	0.0015
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

*Revised and adopted in 1976 by the Illinois Pollution Control Board.

separate grab samples taken approximately once per hour. All analyses were performed as outlined in Standard Methods.[15]

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

There are, in addition, six effluent channels from cooling water operations which were monitored on a once-per-week schedule by grab sampling. These channels carry blowdown water from various cooling towers. 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 28. The December average exceeded the State of Illinois standard for ammonia nitrogen. In addition, the five-day biochemical oxygen demand for the latter two weeks in April exceeded the State standard. Levels before and after these samples were well below the standard.

The results obtained for chemical constituents in the waste treatment plant effluent are shown in Table 29. All of the average concentrations were below the State standards. The average concentration of mercury was reduced from 94% of the State standard in 1975 to 86% in 1976, and the frequency with which this level was exceeded was more than halved, from 29% to 14%. This reduction was the result of additional treatment in January, 1976, and would have been more effective had not two inadvertant releases occurred. Steps have been taken to prevent such releases in the future. The average levels of hexavalent chromium, while well below the State standards, were higher than in 1975.

Measured concentrations of barium were very much lower than in the past. This was due to improvements in the analytical technique, using the hotter nitrous oxide-acetylene flame, which considerably increased sensitivity and decreased the interference from molecular calcium absorption. Levels for beryllium, cadmium, fluoride, iron, lead, nickel, silver, and zinc remained essentially the same as in the past. The levels of copper were reduced by approximately 50% from 1975. This can be attributed to replacement of the

TABLE 28

Performance of Sanitary Waste Treatment Plant, 1976

Month	Concentration (mg/l) B.O.D.$_5$	Ammonia Nitrogen	Suspended Solids
January	1.4	3.11	4.50
February	9.2	3.12	6.75
March	5.5	1.74	2.67
April	> 16	1.04	3.11
May	1.9	0.27	4.00
June	1.2	0.18	3.75
July	1.6	0.16	1.56
August	1.2	0.17	3.50
September	0.9	0.06	1.56
October	4.5	2.35	3.56
November	3.8	2.87	0.86
December	3.0	5.18	4.75
State Limit	10	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	12

TABLE 29

Chemical Constituents in Effluent From ANL Treatment Plant, 1976

Constituent	No. of Samples	Concentration (mg/l or µg/l) Avg.	Min.	Max.	Percent of Standard (Avg.)	Percent Exceeding State Standard
Barium	47	< 0.1	-	-	< 5	0
Beryllium*	12	0.064 ± 0.004	0.02	0.259	-	-
Cadmium*	55	1.3 ± 0.25	0.2	4.8	0.9	0
Chromium(VI)	253	0.064 ± 0.006	< 0.01	0.253	21	0
Chromium(III)	55	0.01 ± 0.006	< 0.01	0.032	1.4	0
Copper*	55	19.1 ± 3.1	7	56	1.9	0
Fluoride	55	0.37 ± 0.03	0.27	1.07	2.5	0
Iron	55	0.12 ± 0.01	0.06	0.27	6.6	0
Lead*	55	2.7 ± 0.6	< 2	8.8	2.7	0
Mercury*	253	0.43 ± 0.12	< 0.1	12.03	86	14
Nickel	42	< 0.05	-	-	< 5	0
pH	253	-	6.80	8.80	-	0
Silver*	55	1.3 ± 0.2	0.1	3.7	1.3	0
Zinc	55	0.19 ± 0.04	0.03	0.36	19	0

*Concentrations in µg/l.

proportional sampling system, which contained some copper components, by an all plastic system.

Results obtained for the cooling tower effluents are shown in Table 30. The sampling site 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 exceeded the State standard for hexavalent chromium 52% of the time. The average concentration was 260% of the State standard. Average values for trivalent chromium and for silver at this location were well below State standards. The chromium (III) standard was never exceeded, and the silver standard was exceeded only in two samples. The average value for hexavalent chromium at 8J exceeded the State standard by about 80%. The average value for hexavalent chromium at 14J was 0.38 mg/l, which is about 30% above the State standard; 35% of the values at this location exceeded the State standard.

Samples were collected for hexavalent chromium analyses at three additional effluent points, 12L, 14G, and 14H, which were not sampled in 1975. The sampling site at 14H was the only one where appreciable chromium was found, and this average was well below the State standard. Future plans call for redirecting the effluents containing high chromium levels into other channels, which should reduce the levels substantially.

2. 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 ft) upstream of the Argonne outfall [7M (up)] and 60 m (200 ft) downstream of the outfall [7M (down)]. Additionally, samples to be examined for fecal coliform were collected at least five times per month using the 7M (up) location, but downstream the sample was collected immediately in front of the outfall grating to minimize contamination from the receiving stream. Once per month a sample was obtained as the water enters the site (16K), which is downstream of the Marion Brook Treatment Plant.

TABLE 30

Cooling Tower Effluents, 1976

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	0.53 ± 0.20	< 0.01	3.19	177	48
Chromium(VI)	11L	46	0.78 ± 0.26	< 0.01	3.36	260	52
Chromium(III)	11L	46	0.26 ± 0.12	< 0.01	1.50	32	9
Silver*	11L	46	8.4 ± 5.4	0.6	125	8.4	2
Chromium(VI)	12L	46	< 0.01	< 0.01	0.02	< 3.0	0
Chromium(VI)	14G	44	< 0.01	< 0.01	0.22	< 3.0	0
Chromium(VI)	14H	44	0.10 ± 0.06	< 0.01	1.13	33	9
Chromium(VI)	14J	52	0.38 ± 0.14	< 0.01	2.1	127	35

*Concentrations in µg/l.

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

The data from these studies are in Tables 31 and 32. The average levels for ammonia nitrogen upstream are 3.5 times the State of Illinois standard and individual samples exceeded this value 66% of the time. The downstream sample averaged twice the State standard and exceeded this value 60% of the time. All of the samples obtained downstream that exceeded the standard could be shown to be due to upstream contamination. These levels represent a severe degradation of water quality from the recent past and are, in part, attributable to severe cold weather, since ammonia is appreciably more soluble at lower temperatures. The dissolved oxygen levels obtained during 1976 averaged 104% and 105% saturation for 7M (up) and 7M (down) samples. The fact that values exceeded 100% saturation is due primarily to photosynthetic activity.

The fecal coliform standards require 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 and June and there were eight instances when the 400 organisms/100 ml standard was exceeded. Samples obtained at 16K exceeded the 400 organisms/100 ml standard 54% of the time. 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 four 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. The total dissolved solids above the outfall exceeded the State standard frequently, and increased the solids content of the downstream samples.

The average results for the chemical constituents in Sawmill Creek are given in Table 32. Individual samples for hexavalent chromium at 7M (down) exceeded the State standard 14% of the time, while the standard was not exceeded in samples upstream of the outfall. The latter samples contain contributions from effluents 11L, 12L, 14J, and 14G, which were described earlier. Individual samples exceeded the State standard for mercury 8% of the time at the 7M (down) location. The State standard for silver was exceeded in 14% of the samples. The likely source is the 11L effluent channel which

TABLE 31

Sawmill Creek - Effect of Sanitary Waste, 1976

Constituent	Location	No. of Samples	Concentration (mg/l) Avg.	Min.	Max.	Percent of Standard (Avg.)	Percent Exceeding State Standard
Ammonia N	7M (up)	50	5.25 ± 1.52	< 0.1	16.67	350	66
	7M (down)	50	3.09 ± 0.82	< 0.1	11.11	206	60
Dissolved Oxygen	7M (up)	50	104%*	-	-	-	-
	7M (down)	50	105%	-	-	-	-
Total Dissolved Solids	7M (up)	50	1299 ± 98	572	1897	130	74
	7M (down)	50	897 ± 44	511	1339	90	24
			Organisms/100 ml**				
Fecal Coliform	7M (up)	40	80			40	
	7M (down)	50	7			3.5	
	16K	10	108			54	

*Percent saturation is computed by comparing the value obtained at the temperature measured to the oxygen equilibrium value of water at that temperature.

**Average of monthly geometric mean.

TABLE 32

Chemical Constituents in Sawmill Creek, 1976

Constituent	Location*	No. of Samples	Concentration (mg/l or µg/l) Avg.	Min.	Max.	Percent of Standard (Avg.)	Percent Exceeding State Standard
Barium	7M	42	< 0.1	-	-	< 2	0
Beryllium**	7M	12	0.073 ± 0.017	0.022	0.120	-	-
Cadmium**	7M	50	2.68 ± 0.5	0.5	9.8	5	0
Chromium(VI)	7M (up)	50	0.01 ± 0.001	< 0.01	0.02	20	0
	7M (down)	145	0.029 ± 0.004	< 0.01	0.140	58	14
Chromium(III)	7M	145	0.019 ± 0.011	< 0.01	0.83	2	0
Copper**	7M	50	17.5 ± 7.9	5	208	88	10
Cyanide	7M	50	< 0.02	-	-	< 80	0
Fluoride	7M	50	0.51 ± 0.03	0.32	0.83	36	0
Iron	7M	50	0.46 ± 0.11	0.15	2.65	46	4
Lead**	7M	50	4.2 ± 1.0	1.5	19.5	4	0
Mercury**	7M (up)	50	< 0.1	< 0.1	0.1	< 20	0
	7M (down)	145	0.38 ± 0.33	< 0.1	23.71	76	8
Nickel	7M	50	< 0.05	-	-	< 5	0
pH	7M	145	-	7.35	8.82	-	0
Silver**	7M	50	3.0 ± 0.8	< 0.2	13.5	60	14
Zinc	7M	50	0.12 ± 0.04	0.03	1.0	12	0

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

** entrations in µg/l.

contains photographic waste. The iron standard was exceeded in 4% 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 29, the iron source must be in the stream area since effluent levels for iron were much lower. Individual samples exceeded the standard for copper 10% of the time, and it is believed that the copper is contributed largely by plumbing. Levels for other constituents did not exceed State standards in any individual sample. The annual average did not exceed the State standard for any of these constituents.

3. Des Plaines River

The effect of Sawmill Creek on the Des Plaines River was evaluated by collecting samples at Willow Springs (upstream of Argonne) and at Lemont (downstream of Argonne). These samples were analyzed for total mercury, hexavalent chromium, total iron, and total zinc. The results are in Table 33. Two of the samples had very high levels of suspended material, as evidenced by iron levels of 4.1 and 6.5 mg/l.

TABLE 33

Chemical Constituents in the Des Plaines River, 1976

Constituent	Location*	No. of Samples	Concentration (mg/l or µg/l) Avg.	Min.	Max.
Chromium (IV)	A	12	< 0.01	< 0.01	< 0.01
	B	24	< 0.01	< 0.01	< 0.01
Iron	A	12	2.04	0.34	6.50
	B	24	1.29	0.21	4.08
Mercury**	A	12	< 0.1	< 0.1	0.20
	B	24	< 0.1	< 0.1	< 0.01
Zinc	A	12	0.07	0.03	0.22
	B	24	0.05	0.03	0.13

*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.

In no case was there any indication that hexavalent chromium or mercury levels were affected by the Argonne effluent. Total iron and zinc are analyzed by this laboratory in an effort to "normalize" the water samples since both elements are naturally occurring in reasonably constant amounts.

IV. APPENDIX

A. References

(1) U. S. Energy Research and Development Administration, *Effluent and Environmental Monitoring and Reporting*, ERDA Manual Chapter 0513 and Appendix 0513 (1974).

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

(3) J. Sedlet, N. W. Golchert, and T. L. Duffy, "Environmental Monitoring at Argonne National Laboratory, Annual Report for 1973," *U.S.A.E.C. Report ANL-8078* (March, 1974).

(4) U. S. Energy Research and Development Administration, *Standards for Radiation Protection*, ERDA Manual Chapter 0524 and Appendix 0524 (1975).

(5) E. P. Hardy, Jr., U.S.E.R.D.A. Health and Safety Laboratory, New York, NY, Private Communication, February, 1977.

(6) N. W. Golchert and J. Sedlet, "Radiochemical Determination of Plutonium in Environmental Water Samples," *Radiochem. Radioanal. Letters, 12*, 215 (1972).

(7) M. H. Campbell (ed.), "Proceedings of the Symposium on High-Level Radioactive Waste Management," *Advances in Chemistry, No. 153* (1976).

(8) J. H. Harley (ed.), "HASL Procedure Manual," *U.S.A.E.C. Report HASL-300* (1972).

(9) E. P. Hardy, Jr. and P. W. Krey, "Determining the Accumulated Deposit Radionuclides by Soil Sampling and Analysis," in Proceedings of an Environmental Plutonium Symposium, Los Alamos Scientific Laboratory, August 4-5, 1971, *U.S.A.E.C. Report LA-4756* (December, 1971).

(10) E. P. Hardy, P. W. Krey, and H. L. Volchok, "Global Inventory and Distribution of Fallout Plutonium," *Nature, 241*, 444 (1973).

(11) N. A. Frigerio, K. F. Eckerman, and R. S. Stowe, "Argonne Radiological Impact Program (ARIP)," *U.S.A.E.C. Report ANL/ES-26* (1973).

(12) International Commission on Radiological Protection, *Report of Committee II on Permissible Dose for Internal Radiation, ICRP Publication 2 (1959)*, and *ICRP Publication 6 (1962)*; Pergamon Press, New York

(13) Illinois Pollution Control Board, *Rules and Regulations*, Chapter 3, 1972 (Chicago).

(14) N. W. Golchert, T. L. Duffy, and J. Sedlet, "Environmental Monitoring at Argonne National Laboratory, Annual Report for 1975, *U.S.E.R.D.A. Report ANL-76-29* (March, 1976).

(15) *Standard Methods for the Examination of Water and Waste Water*, 13th Edition, American Public Health Association, Washington, D. C., 1971.

(16) T. L. Duffy, R. Kasper, and J. A. Wronski, "Environmental Water Studies at Argonne National Laboratory, Jan. 1968-Dec. 1969," Argonne National Laboratory Internal Report. (Available from the authors.)

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 are given in the main body of the report. In addition, in Table 34 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 ERDA 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).

TABLE 34

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 ERDAM 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.

2. Detection Limits

The detection limits were chosen so that the error at the 95% confidence level is equal to the detection limit. The error in a result decreases with increasing concentration. At twice the detection limit, the error is about 50% of the measured value, and at 10 times the detection limit, the error is about 10%.

3. Quality Assurance

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 ERDA Health and Safety Laboratory (HASL), 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; and the laboratory participated in the Second International Intercomparison of Environmental Dosimeters sponsored by HASL and the University of Texas. Typical results obtained the intercomparison programs are shown in Table 35, which is a summary of all the EPA-QA samples analyzed in this laboratory in 1976. To judge the differences between the added and measured amounts, typical errors for our analyses are about 10% and the error in the amount added 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 samples to which identical concentrations were added. Average recoveries ranged from about 85-100%, with a standard deviation of about 10%.

TABLE 35

Summary of EPA-QA Samples, 1976

Type of Sample	Analysis	Number Analyzed	Avg. Difference From Added
Air Filter	Total Alpha	4	14%
	Total Beta	4	6%
	Strontium-90	1	21%
	Cesium-137	4	22%
Water	Total Alpha	5	11%
	Total Beta	5	8%
	Hydrogen-3	2	6%
	Chromium-51	1	7%
	Cobalt-60	3	3%
	Zinc-65	4	2%
	Ruthenium-106	4	19%
	Cesium-134	4	9%
	Cesium-137	4	2%
Milk	Potassium-40	2	4%
	Strontium-89	2	11%
	Strontium-90	2	6%
	Iodine-131	2	16%
	Cesium-137	2	11%
	Barium-140	1	9%

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).

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 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 consituent sought. For example, samples for trace radionuclide analysis are acidified immediately after collection to prevent hydrolytic loss of metal ions, but aliquots for radioiodine and tritium analyses were withdrawn first, since trace iodine is unstable in acid solution.

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