

**ENVIRONMENTAL MONITORING AT  
ARGONNE NATIONAL LABORATORY**

**ANNUAL REPORT FOR 1980**

**by**

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



**ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS**

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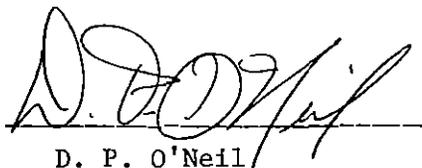
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ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1980 are presented and discussed. To evaluate the effect of Argonne operations on the environment, measurements were made for a variety of radionuclides in air, surface water, soil, grass, bottom sediment, and foodstuffs; for a variety of chemical constituents in air, surface water, and Argonne effluent water; and of the environmental penetrating radiation dose. Sample collections and measurements were made at the site boundary and off the Argonne site for comparison purposes. Some on-site measurements were made to aid in the interpretation of the boundary and off-site data. The results of the program are interpreted in terms of the sources and origin of the radioactive and chemical substances (natural, fallout, Argonne, and other) and are compared with applicable environmental quality standards. The potential radiation dose to off-site population groups is also estimated.

I. INTRODUCTION

A. General

This report is prepared to provide the U. S. Department of Energy (DOE) and the public with information on the levels of radioactive, chemical, and biological pollutants in the environment of Argonne National Laboratory (ANL) and on the amounts, if any, added to the environment as a result of Argonne operations. The report follows the guidelines given in DOE Manual Chapter 0513.<sup>(1)</sup> The Laboratory conducts a continuous environmental monitoring program on and near the Argonne site whose primary purpose is to determine the magnitude, origin, and identity of radioactive or potentially toxic chemical and biological substances in the environment. Of special interest is the detection of any such materials released to the environment by Argonne. One important function of the program is to verify the adequacy of Argonne's pollution controls.

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

The principal nuclear facilities at the Laboratory are a 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 60-inch cyclotron; several other charged particle accelerators (principally of the Van de Graaff type); a large fast neutron source, IPNS (Intense Pulsed Neutron Generator); cobalt-60 irradiation sources; chemical and metallurgical plutonium laboratories; and several hot cells and laboratories designed for work with multicurie quantities of the actinide elements. Two major facilities, a 12.5 GeV proton accelerator (ZGS, the Zero Gradient Synchrotron) and a 5 MW heavy water-enriched uranium reactor (CP-5) were not in operation during 1980 and are awaiting decontamination and decommissioning.

#### 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 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 the River, the Chicago Sanitary and Ship Canal, and the Illinois and Michigan Canal. Their presence extends the uninhabited area about 1.6 km (1 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.

### 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

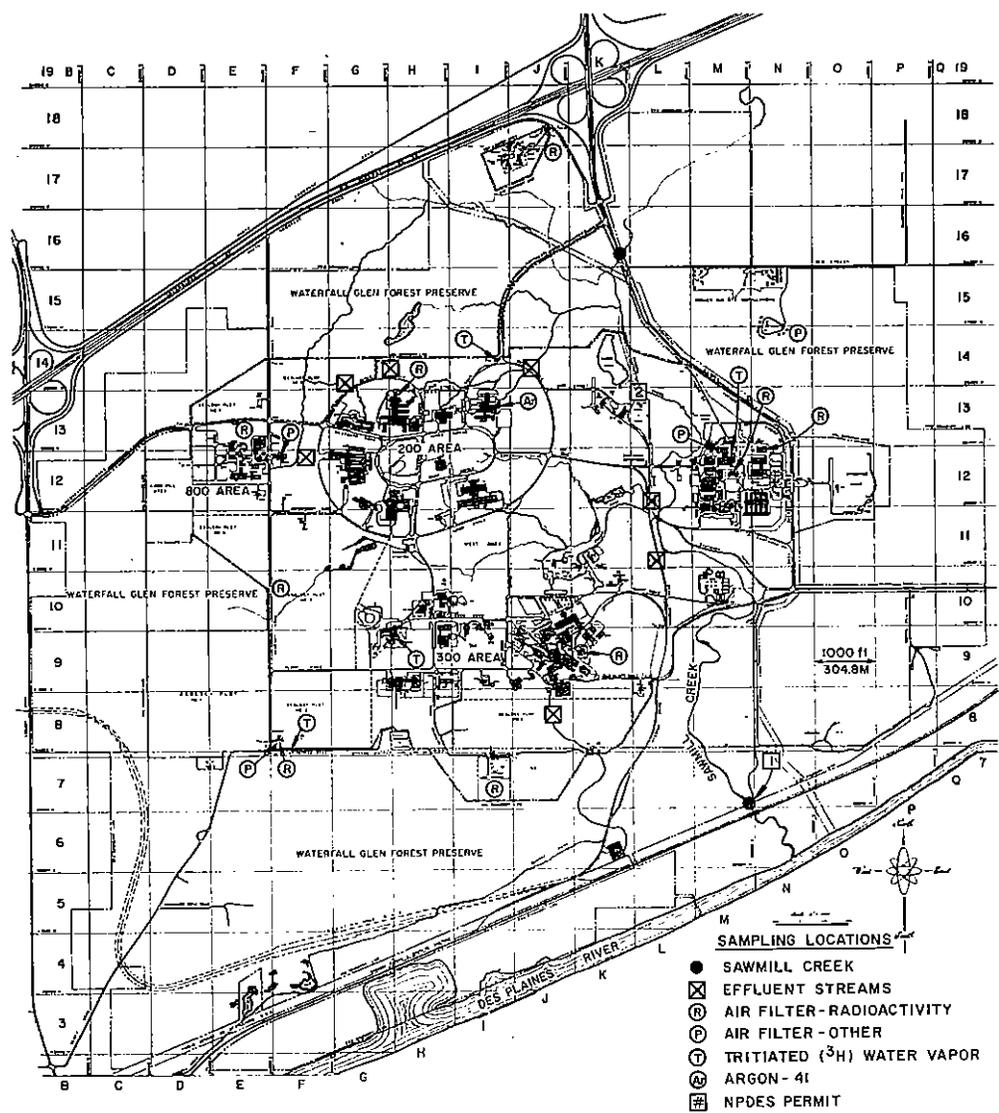


Fig. 1. Sampling Locations at Argonne National Laboratory

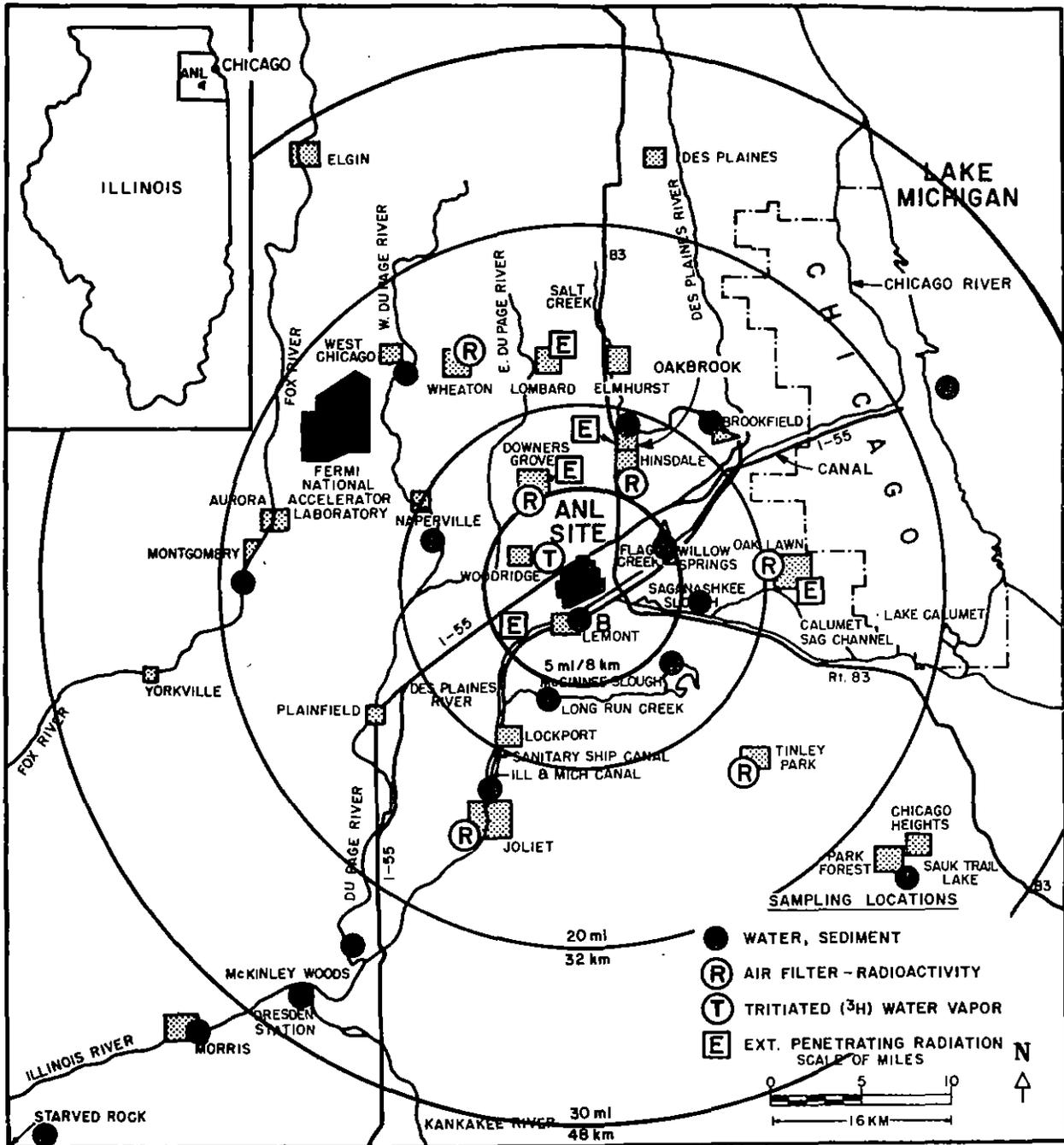


Fig. 2. Sampling Locations Near Argonne National Laboratory

on the site from 1950 to 1964 is available,<sup>(2)</sup> and provides an adequate sample of the climate. Similar data have been collected since 1964.

The most important meteorological parameters for the purposes of this report are wind direction, wind speed, temperature, and precipitation. The average monthly and annual wind roses on the Argonne site are shown in Figure 3. The wind roses are polar coordinate plots in which the lengths of the radii represent the percentage frequency of wind speeds in classes of 2.01-6 m/s (4.5-13.4 mph), 6.01-10 m/s (13.4-22.4 mph), and greater than 10.01 m/s (22.4 mph). The 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 January, the westerly component is predominant. The length and direction of this vector shows that, of the total wind observations, about 9% were in the 2.01-6 m/s range, about 4% were in the 6.01-10 m/s range, about 4% were greater than 10.01 m/s; and about 17% of the observations were from the direction between 258.75° and 281.25° (west). The number in the center represents the percent of observations of wind speed less than 2 m/s in all directions.

The roses show that the principal wind directions were from the northwest, west, and southwest, but seasonal variations are apparent. The winds were primarily westerly in January and February. In the spring, the northeast lake breezes are evident. The warm southerly winds, brought up by the high pressure areas in the southeastern U. S., began in June and continued for the remainder of the summer. The winds are sufficiently variable so that monitoring for airborne releases must be carried out in all directions from the site.

The precipitation and temperature data at Argonne for 1980 are shown in Table 1. Above-normal precipitation occurred in August and September, while the first half of the year was below normal. Temperatures were fairly close to normal most of the year.

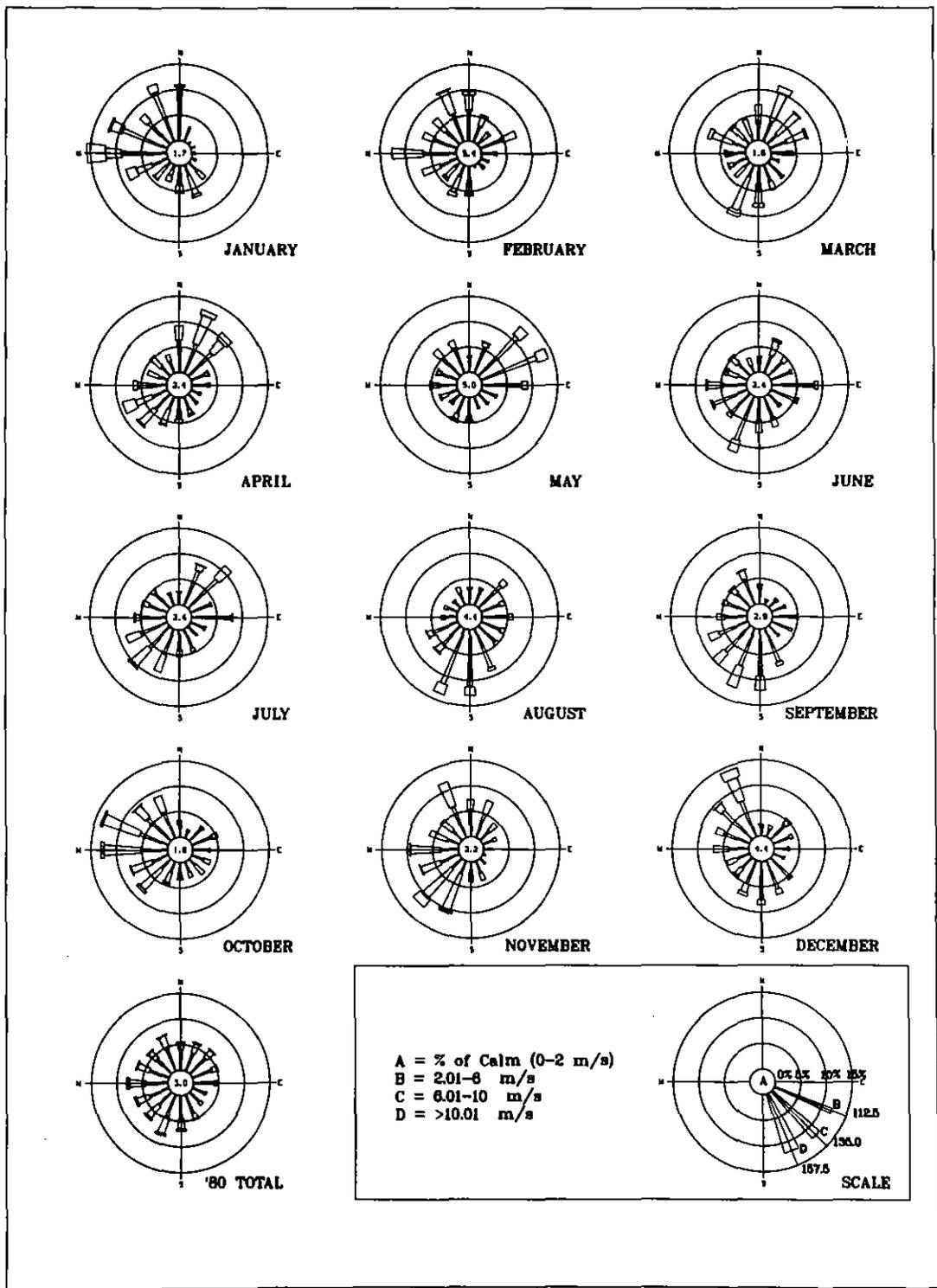


Fig. 3. Monthly and Annual Wind Roses at Argonne National Laboratory, 1980

TABLE 1

## ANL Weather Summary, 1980

Month	Precipitation (cm)		Temperature (°C)	
	Amount	Average <sup>*</sup>	Monthly Average	Average <sup>**</sup>
January	2.46	4.88	-5.4	-4.2
February	4.01	4.57	-6.0	-2.7
March	4.55	6.93	0.5	2.5
April	6.35	7.98	8.1	8.8
May	7.14	8.81	15.6	14.6
June	8.08	9.47	18.9	20.2
July	7.67	8.66	23.9	23.2
August	21.39	8.10	22.8	22.5
September	16.54	8.08	17.3	18.7
October	8.76	6.55	8.4	12.5
November	1.80	5.88	3.9	4.7
December	6.68	5.30	-2.8	-1.5

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

\*\* Average temperature, 1971-1977, U. S. Weather Bureau, Chicago, Illinois.

#### D. Population

The area around Argonne has exhibited a large population growth in the past 20 years. Large areas of farmland have been converted into housing. A directional and annular 80-km (50-mile) population distribution for the area, which is used for the population dose calculations later in this report, is shown in Table 2.<sup>(3)</sup> The distribution, centered on the CP-5 reactor, was obtained by modifying a similar distribution based on the 1970 U. S. Census and prepared by the Regional and Urban Studies Department at Oak Ridge National Laboratory. For distances within 8 km of the center, the updated values are largely based on quarter-section population data supplied by the Northeastern Illinois Planning Commission (NIPC). Some adjustments were made on the basis of local observations. At distances beyond 8 km, but within the

TABLE 2

## Incremental Population Data in the Vicinity of ANL, 1978

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

NIPC area, township forecasts were used, allocating population to each sector in proportion to the area of the township within the sector. The city of Chicago population was allocated in proportion to the area of each sector. Outside of the six-county area around Chicago included in the NIPC program, the Northwestern Indiana Regional Planning Commission provided quarter-section estimates for Lake and Porter Counties and other Illinois Counties provided estimates of the population in their respective areas. Population counts were usually based on locally adjusted estimates made by the State of Illinois Bureau of the Budget and on special interim U. S. censuses for several municipalities. Included within the 80-km radius are portions of Lake and Porter Counties, Indiana, portions of Kankakee, Grundy, LaSalle, DeKalb, McHenry, and Lake Counties in Illinois, and all of the DuPage, Will, Cook, Kendall, and Kane Counties in Illinois.

#### E. Water and Land Use

The principal stream that drains the site is Sawmill Creek. This Creek was formerly an intermittent stream, responding in flow rate largely to precipitation runoff. It now carries effluent water continuously from a municipal sewage treatment plant (Marion Brook Treatment Plant) located a few kilometers north of the site, which has an operating capacity of about 11.4 megaliters (3 million gallons) per day. In addition, the residential development in the area has resulted in the collection and channeling of additional runoff water into the Creek. Treated sanitary and laboratory waste water from Argonne are combined and discharged into Sawmill Creek at location 7M in Figure 1. This effluent averaged 2.7 megaliters (0.75 million gallons) per day, while the water flow in the Creek upstream of the waste-water outfall averaged about 38 megaliters (10 million gallons) per day during 1980. The combined Argonne effluent consisted of 70% laboratory waste water and 30% sanitary waste water.

The effluent channels from the 200 Area buildings and cooling towers are part of the effluent sampled at 12L in Figure 1. The 12L effluent also drains the Freund ponds (11K and 12K) and enters Sawmill Creek in the same coordinate. The stream that drains water from the ZGS complex is sampled at 8J, crosses the Laboratory boundary at 7K, and normally soaks into the ground south of the site. The cooling tower effluents are monitored on a regular weekly basis at

locations 8J and 12L. The other channels (11L, 12F, 14H, and 14J in Figure 1) are sampled monthly if they contain sufficient water.

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 receives Chicago Metropolitan Sanitary District effluent water, is used for industrial transportation and some recreational boating. Near Joliet, the River and Canal are combined into one waterway, which continues until it joins the Kankakee River to form the Illinois River about 48 km (30 miles) southwest of Argonne. The Dresden Nuclear Power Station complex is located at the confluence of the Kankakee, Des Plaines, and Illinois Rivers. This Station uses water from the Kankakee for cooling, and discharges the water into the Illinois River. The first use of water for drinking is an indirect one at Alton, on the Mississippi River about 710 km (370 miles) downstream from Argonne, where water is used to replenish groundwater supplies by infiltration. In the vicinity of the Laboratory, only subsurface water (from both shallow and deep aquifers) and Lake Michigan water are used for drinking purposes.

The principal recreational area near Argonne is Waterfall Glen Forest Preserve, which surrounds the site as described in Section I.B. and is shown in Figure 1. Most of this area has received little use thus far. It was available in 1980 for hiking, skiing, and equestrian sports, and its development and increased utilization by the public is expected. Very approximate estimates of usage are 600 individuals per day on weekends and 500 individuals for group educational purposes during 1980. The average stay is about two hours. Sawmill Creek flows through the south portion of the Preserve on its way to the Des Plaines River. This region of the Preserve (formerly named Rocky Glen) was used principally for picnicking, hiking, and overnight camping by youth groups, but has been closed for rehabilitation since June 6, 1976. East and southeast of Argonne and the Des Plaines River are located several large forestpreserves of the Cook County Forest Preserve District. The preserves include the two sloughs shown in Figure 2, McGinnis and

Saganashkee, as well as other smaller lakes. These areas are used for picnicking, boating, fishing, and hiking. A small park located in the eastern portion of the Argonne site (12-0 in Figure 1) is for the use of Argonne and Department of Energy employees only.

The most recently available information on milk and principal agricultural production in a 10,600 sq km (4,100 sq mi) area around Argonne is shown by County in Table 3. These estimates were obtained from the Illinois Crop Reporting Service of the State Department of Agriculture. For corn and soybeans, the data are for 1980; for milk, wheat, and oats, the data are for 1979.

TABLE 3

## Agricultural Production Near ANL, 1979, 1980

County	Milk		Corn	Soybeans	Wheat	Oats
	No. of Cows	Million Pounds				
DuPage	200	2.1	1.9	0.59	0.078	0.038
Cook	300	3.1	1.6	0.72	0.082	0.076
Will	3,000	31.3	13.3	4.7	0.29	0.33
Kane	6,800	70.9	13.6	2.8	0.05	0.35
Kendall	500	5.2	10.8	2.4	0.076	0.11
Grundy	600	6.3	10.6	2.9	0.058	0.075
Lake	1,600	16.7	2.3	0.83	0.27	0.13

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

## II. SUMMARY

This is one in a series of annual reports prepared to provide the U. S. Department of Energy (DOE) and the public with information on the level of radioactive and chemical pollutants in the environment and on the amounts of such substances, if any, added to the environment as a result of Argonne operations. The previous report in this series is ANL-80-29. Included in this report are the results of measurements obtained in 1980 for a number of radionuclides in air, surface water, soil, grass, bottom sediment, and food-stuffs; for a variety of chemical constituents in air and water, and for the external penetrating radiation dose.

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

The plutonium-239,240 concentrations in air averaged  $6 \times 10^{-18}$   $\mu\text{Ci/ml}$ ,\* about one half of the 1979 value, and were nearly identical at all sampling locations. The monthly variations showed a "spring maximum" in stratospheric fallout of plutonium similar to that observed for beryllium-7 (a naturally-occurring nuclide) and several fission products. The results indicated that the airborne plutonium was from atmospheric nuclear test detonations and there was no evidence that any of the plutonium originated at Argonne. The average plutonium concentration was equivalent to 0.0006% of the CG.\*\*

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\* The radioactivity units are described in Section III.

\*\* Unless otherwise indicated, the hazard due to a given concentration of a radioactive nuclide is assessed in this report by comparison with the Concentration Guides (CG) and annual dose limits, or Radiation Protection Standards, for uncontrolled areas specified by the U. S. Department of Energy in Chapter 0524 of the DOE Manual.<sup>(4)</sup> The pertinent CGs are listed in the Appendix, Section IV.B. Comparison with other standards is given where appropriate. The modified dose assessment methods introduced recently by the International Commission on Radiological Protection have not been used in this report for the reasons given in Section III.A.

The major airborne radionuclides released from the Laboratory were hydrogen-3 (as tritiated water vapor), argon-41, and krypton-85. The amount of hydrogen-3 and argon-41 released during 1980 were several orders of magnitude lower than in previous years when the CP-5 reactor was operating. The cumulative dose from these three nuclides at the site boundary in the predominant east to northeast direction toward which the wind is blowing was less than 0.001 mrem/yr, calculated from an atmospheric dispersion model. The calculated dose to the closest full-time resident, who is located about 0.5 km northeast of the site boundary, was less than 0.0005 mrem/yr. These releases constitute an insignificant addition to the dose received from the natural background radiation.

Argonne waste water is discharged into Sawmill Creek, and this stream was sampled above and below the site to evaluate the effect of Argonne operations on its radioactive content. The nuclides (for which analyses were made) added to the Creek in the waste water, and the percent of CG of their average Creek concentrations, were hydrogen-3, 0.008%; strontium-90, 0.12%; neptunium-237, 0.00033%; plutonium-239,240, 0.00033%; americium-241, 0.00014%; and curium-244 and/or californium-249, 0.00003%. Although Sawmill Creek is not a source of potable water, the dose to an individual using water at these concentrations as his sole source of drinking water would be less than 0.7 mrem/yr.

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

Plutonium concentrations in soil showed the same general range and average at the site perimeter as in the past years. The average plutonium-239,240 content of the top 5 cm of soil was  $1.0 \times 10^{-3}$   $\mu\text{Ci}/\text{m}^2$  at the site perimeter and  $0.6 \times 10^{-3}$   $\mu\text{Ci}/\text{m}^2$  off the site. The corresponding plutonium-238 averages were  $0.05 \times 10^{-3}$   $\mu\text{Ci}/\text{m}^2$  and  $0.04 \times 10^{-3}$   $\mu\text{Ci}/\text{m}^2$ , respectively. The plutonium content of grass was similar to that found in previous years and was about a factor of  $10^4$  less than soil from the same locations. The results were within the range reported by other laboratories for fallout from test detonations, and the plutonium found in soil and grass is attributed to

this source. The plutonium content of samples from beds of streams and ponds ranged from  $1 \times 10^{-9}$   $\mu\text{Ci/g}$  to  $44 \times 10^{-9}$   $\mu\text{Ci/g}$  of plutonium-239,240, a range found in previous years to be normal for fallout plutonium in such materials.

Milk from a dairy farm near the Laboratory was analyzed for several fission products, including hydrogen-3. Hydrogen-3 concentrations averaged  $126 \times 10^{-9}$   $\mu\text{Ci/ml}$ . The strontium-90 concentration ( $3.3 \times 10^{-9}$   $\mu\text{Ci/ml}$ ) decreased by 10% compared to 1979, while the cesium-137 concentration ( $1.2 \times 10^{-9}$   $\mu\text{Ci/ml}$ ) decreased by over a factor of two.

Measurements of penetrating radiation were made at several locations at the site boundary and off the site. The off-site results averaged 90 mrem/yr with a standard deviation of 7 mrem/yr, which is in the normal range for the area. At three locations at the site boundary, above-normal readings were recorded that were attributable to Argonne operations. At two locations at the south fence the dose rates averaged, respectively, about 560 mrem/yr above normal (grid 7I in Figure 1) and 5-10 mrem/yr above normal (grid 8H) as a result of radiation from an on-site temporary storage facility for radioactive waste. About 300 m (0.2 mile) south of the fence, the measured dose rate dropped to 98 mrem/yr, which is within the normal range. Along the north side of the site, the dose at the fence at location 14I was about 30 mrem/yr above normal due to radiation from cobalt-60 sources in Building 202. Since all of these locations are unoccupied, there are no individuals receiving these measured doses. The calculated outdoor dose rate from these sources to the residents closest to the south boundary, about 1.6 km (1 mile) from the fence line, was about 0.06 mrem/yr; similarly the dose rate to the residents closest to the north boundary, about 0.75 km (0.5 mile) from the fence, was about 0.006 mrem/yr. Thus, doses to individuals near the site from these sources will not exceed 0.01% of the 500 mrem/yr limit.

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

The performance of the Argonne combined sanitary and laboratory wastewater effluent has been evaluated by comparing results to a National Pollution

Discharge Elimination System (NPDES) Permit. While occasional samples exceeded permit conditions, corrective action kept the discharge in general compliance.

Average concentrations of each of the other chemical constituents measured in this effluent were at or below State of Illinois standards. The concentration of mercury averaged 66% of the State standard and exceeded this value 15% of the time (i.e., in 15% of the samples). No other constituent in this effluent exceeded the State standard at any time.

The concentrations of hexavalent chromium from six of the seven cooling tower effluent channels used for blowdown were below analytical detection limits. The concentration in the effluent channel at 8J was below the detection limit after April, 1980. Hexavalent chromium levels were low because chromium compounds were replaced as corrosion inhibitors in the cooling towers. Zinc levels in the 12L effluent were consistently low, but levels at 8J increased in the period September-December, 1980, for reasons that are as yet unclear.

The average values in Sawmill Creek for dissolved oxygen and chemical constituents, except ammonia nitrogen, dissolved solids, copper, and iron were within the State of Illinois Water Quality standards. The average ammonia nitrogen level above the Argonne waste-water outfall was 1.6 times the State standard, and exceeded this standard in 65% of the samples. The average level below the outfall was 1.4 times the State standard and exceeded this value in 56% of the samples. The average concentration of dissolved solids above the Argonne waste-water outfall exceeded the State standard by 10%, but below the outfall it was equal to the standard. The elevated levels of ammonia nitrogen and total dissolved solids were clearly due to contamination upstream of the Laboratory. The copper levels in 1980 below the outfall exceeded the State standard in 62% of the samples and averaged 1.7 times the standard. The copper in the Argonne effluent was insufficient to account for these high results and studies indicated bottom sediment contamination to be a source. Individual values for silver, mercury, and iron exceeded State standards from 2% to 56% of the time. Hexavalent chromium levels were not in excess of the State standard at any 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.

Determination of total suspended particulates (TSP) and trace metals in air was continued. The trace metal studies included iron, copper, lead, zinc, bromine, and strontium. Results were similar to last year. Values for the 24-hour and continuous TSP samples did not exceed the primary or secondary standards. The results for the 24-hour sample were higher than the comparable continuous sample by about 10%.

Polynuclear aromatic compounds were measured using the same samples collected for the continuous TSP studies. A useful inventory of background changes and levels of these compounds is now available to compare to results obtained when coal burning begins.

## III. MONITORING RESULTS

A. Radiological

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

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

picture of the true average concentration at locations where the concentrations not only varied greatly, but were at times not detectable. Penetrating radiation measurements are reported in units of millirem (mrem) per year and population dose in man-rem.

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

The measured concentration or radiation dose is compared with appropriate standards as a means of assessing the hazard. The standards used in this report are usually the Concentration Guides (CGs) and annual dose limits (Radiation Protection Standards) for uncontrolled areas given in DOE Manual Chapter 0524.<sup>(4)</sup> The pertinent CGs as well as the detection limits are given in the Appendix, Section IV.B. Although the CGs apply to concentrations above natural levels, the percent of CG is sometimes given in this report for activities that are primarily of natural origin for comparative purposes. Such values are enclosed in parentheses to indicate this. Where other standards are used, their source is identified in the text.

The new dose assessment and evaluation procedures and systems recommended by the International Commission on Radiological Protection (ICRP) in their reports ICRP-26,<sup>(5)</sup> *et seq*, are not used in this report for the following reasons. These recommendations have not yet been officially adopted by DOE, although it is fully expected that they will be used by DOE and all regulatory agents in the near future. The annual whole body dose limit recommended in ICRP-26 for both occupational and public exposure has not changed from their

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

## 1. Air

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

The total alpha and beta activities in the individual weekly samples are summarized in Table 4. These measurements were made in low-background gas-flow proportional counters, and the counting efficiencies used to convert counting rates to disintegration rates were those measured for radon decay products on filter paper. The average concentrations of a number of gamma-ray emitters, as determined by gamma-ray spectrometry performed on composite weekly samples are given in Table 5. The gamma-ray detector is a shielded 74 cm<sup>3</sup> lithium-drifted germanium diode, calibrated for each gamma-ray emitting nuclides listed in Table 5.

The alpha activities, principally due to naturally-occurring nuclides,

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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.

TABLE 4  
 TOTAL ALPHA AND BETA ACTIVITIES IN AIR-FILTER SAMPLES, 1980\*  
 (CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

MONTH	LOCATION	NO. OF SAMPLES	ALPHA ACTIVITY			BETA ACTIVITY		
			AV.	MIN.	MAX.	AV.	MIN.	MAX.
JANUARY	OFF-SITE PERIMETER	18	1.9	0.7	3.6	27	19	40
		42	1.7	0.6	3.0	28	9	44
FEBRUARY	OFF-SITE PERIMETER	20	2.2	1.3	4.6	27	14	42
		35	1.7	0.8	3.2	27	16	39
MARCH	OFF-SITE PERIMETER	20	2.1	1.2	4.9	24	14	49
		41	1.4	0.7	2.6	21	13	30
APRIL	OFF-SITE PERIMETER	19	1.7	0.4	4.9	16	4	28
		36	1.4	0.5	3.1	18	4	36
MAY	OFF-SITE PERIMETER	21	2.2	0.9	3.6	24	14	39
		42	2.1	0.9	4.8	23	12	39
JUNE	OFF-SITE PERIMETER	23	2.1	1.1	6.6	22	9	33
		38	2.1	0.9	3.0	22	14	35
JULY	OFF-SITE PERIMETER	17	2.4	1.0	4.3	28	17	40
		39	1.9	0.6	3.6	26	13	37
AUGUST	OFF-SITE PERIMETER	16	1.7	0.3	4.0	22	13	35
		38	1.6	0.7	3.6	23	13	49
SEPTEMBER	OFF-SITE PERIMETER	20	1.7	0.7	3.6	22	16	30
		40	1.6	0.7	4.6	21	9	30
OCTOBER	OFF-SITE PERIMETER	20	1.9	1.0	3.1	22	16	37
		39	1.9	0.9	4.3	21	10	34
NOVEMBER	OFF-SITE PERIMETER	18	1.9	0.3	3.0	47	1	86
		35	2.2	0.8	4.0	55	39	80
DECEMBER	OFF-SITE PERIMETER	19	2.9	1.3	6.5	95	47	242
		34	2.3	1.3	3.7	83	48	126
ANNUAL SUMMARY	OFF-SITE PERIMETER	231	2.1 ± 0.05	0.3	6.6	31 ± 2.8	1	242
		459	1.8 ± 0.03	0.5	4.8	31 ± 1.8	4	126
% CG	OFF-SITE PERIMETER	-	(0.002)	(0.0003)	(0.007)	0.031	0.001	0.242
% CG	PERIMETER	-	(0.002)	(0.0005)	(0.005)	0.031	0.004	0.126

\* 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.

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NOTE: (1E-15 = 10<sup>-15</sup>)

TABLE 5  
 GAMMA-RAY ACTIVITY IN AIR-FILTER SAMPLES, 1980  
 (CONCENTRATIONS IN 1E-15 MICROCURIES/ML)

MONTH	LOCATION	BE7	RU106-RH106	SB125	CS137	CE144
JANUARY	OFF-SITE PERIMETER	74	0.6	0.2	0.3	0.6
		76	0.5	0.2	0.3	0.5
FEBRUARY	OFF-SITE PERIMETER	78	0.5	0.2	0.3	0.5
		74	0.3	0.2	0.3	0.4
MARCH	OFF-SITE PERIMETER	94	0.6	0.2	0.4	0.6
		97	0.5	0.3	0.4	0.5
APRIL	OFF-SITE PERIMETER	70	0.6	0.2	0.4	0.6
		82	0.5	0.2	0.4	0.6
MAY	OFF-SITE PERIMETER	121	1.0	0.4	0.9	1.2
		112	0.6	0.4	0.7	1.1
JUNE	OFF-SITE PERIMETER	113	0.8	0.3	0.7	0.9
		123	0.7	0.3	0.7	0.8
JULY	OFF-SITE PERIMETER	93	0.5	0.2	0.6	0.6
		102	0.4	0.2	0.5	0.5
AUGUST	OFF-SITE PERIMETER	77	0.6	0.2	0.5	0.2
		92	0.4	0.3	0.3	0.4
SEPTEMBER	OFF-SITE PERIMETER	79	0.4	0.2	0.2	0.3
		73	0.3	< 0.1	0.2	0.2
OCTOBER	OFF-SITE PERIMETER	77	0.3	< 0.1	0.3	0.2
		77	0.2	0.1	0.2	0.3
NOVEMBER	OFF-SITE PERIMETER	71	0.7	< 0.1	0.2	0.8
		73	0.5	0.2	0.2	0.7
DECEMBER	OFF-SITE PERIMETER	68	1.3	0.1	0.3	2.3
		71	1.2	0.1	0.3	2.6
ANNUAL SUMMARY	OFF-SITE PERIMETER	84 ± 10	0.7 ± 0.2	0.2 ± 0.1	0.4 ± 0.1	0.7 ± 0.3
		88 ± 10	0.5 ± 0.2	0.2 ± 0.1	0.4 ± 0.1	0.7 ± 0.4
% CG (X 1E-3)	OFF-SITE PERIMETER	(0.21)	0.3	0.02	0.09	0.4
		(0.22)	0.3	0.02	0.08	0.4

averaged the same as the past several years and were in their normal range. The average beta activity for the year,  $31 \times 10^{-15}$   $\mu\text{Ci/ml}$ , was only slightly lower than the 1979 average. No evidence of a "spring maximum" is apparent in the total beta activity whereas significant increases in the beta activity occurred in November and December due to the October 16, 1980, atmospheric nuclear test of the People's Republic of China. The more sensitive gamma-ray spectrometric measurements, presented in Table 5, for the longer-lived fission products indicated a normal spring increase in stratospheric fallout, as did the naturally-occurring beryllium-7. The annual averages of the radionuclides in Table 5 are a factor of two to three lower than the 1979 averages. This decrease is offset by the presence of short half-life fission and activation products in November and December from the Chinese test. These results are collected in Table 6. First evidence of the test was in air samples collected during the first week in November. The decrease in air concentrations since mid-November was due to radioactive decay and removal from the air by precipitation and dry deposition. Most of the gamma-ray activity in air-filter samples was due to beryllium-7, produced in the stratosphere by cosmic-ray interactions.

Samples for radiochemical analyses were collected at perimeter locations 12N and 7I (Fig. 1) and off the site in Downers Grove (Fig. 2). Collections were made on a polystyrene filter medium. The total air volume filtered for the monthly samples was about  $25,000 \text{ m}^3$ . Samples were ignited at  $600^\circ\text{C}$  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  $1000^\circ\text{C}$ .

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

TABLE 6

Short Half-Life Fallout Nuclides in Air-Filter Samples, 1980  
(Concentrations in  $10^{-15}$   $\mu\text{Ci/ml}$ )

Sampling Period	Location	$^{95}\text{Zr}$	$^{95}\text{Nb}$	$^{103}\text{Ru}$	$^{131}\text{I}$	$^{140}\text{Ba}$ - $^{140}\text{La}$	$^{141}\text{Ce}$	$^{237}\text{U}$
1/1/80-10/31/80	Off-Site	< 0.1	< 0.1	< 0.1	< 5	< 0.1	< 0.1	< 0.1
	Perimeter	< 0.1	< 0.1	< 0.1	< 5	< 0.1	< 0.1	< 0.1
11/3-7/80	Off-Site	1.0	0.3	1.2	10	2.1	1.2	2.2
	Perimeter	0.9	0.4	1.1	10	3.0	1.1	2.7
11/10-14/80	Off-Site	3.7	2.2	3.1	13	8.5	3.3	3.5
	Perimeter	2.6	1.5	3.9	15	10.0	3.6	5.2
11/17-21/80	Off-Site	2.7	1.8	3.4	15	6.4	3.1	2.3
	Perimeter	2.9	2.0	3.2	10	6.5	3.3	2.9
11/24-28/80	Off-Site	3.0	2.5	3.5	9	5.3	3.7	1.2
	Perimeter	2.4	2.1	3.5	7	4.1	3.4	1.5
12/1-5/80	Off-Site	4.9	4.7	5.7	9	6.6	5.5	1.0
	Perimeter	3.2	2.9	4.5	6	5.0	4.1	1.0
12/8-12/80	Off-Site	4.0	3.9	4.4	5	5.2	4.1	0.6
	Perimeter	6.5	6.3	7.2	5	7.4	6.9	1.0
12/15-19/80	Off-Site	5.6	6.1	5.0	5	3.9	4.9	0.4
	Perimeter	7.9	8.0	8.9	5	6.0	7.7	0.8
12/22-26/80	Off-Site	8.5	9.9	8.3	< 5	4.1	6.7	< 0.1
	Perimeter	8.3	9.8	7.1	< 5	5.5	5.9	0.2
12/29/80-1/2/81	Off-Site	7.5	9.2	7.4	5	2.7	6.1	< 0.1
	Perimeter	7.8	9.2	7.6	5	3.1	5.8	< 0.1

isotope is also included. The results are given in Table 7.

The average plutonium-239 concentrations decreased at all locations to about a half of the 1979 values. The annual average strontium-90 concentrations decreased by about the same rate as the plutonium-239, while the shorter-lived strontium-89 was only detectable during November and December. Figure 4 shows the monthly plutonium-239 air concentrations for the past eight years. The arrows at the bottom of the figure indicate the approximate dates of atmospheric nuclear tests.

The long-lived radionuclides produced in nuclear detonations, plutonium-239, strontium-90, and cesium-137, showed similar monthly variations, and a spring peak is evident, indicating their stratospheric origin.

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

The thorium and uranium concentrations are in the same range found during the past several years and are considered to be of natural origin. The percent of CG for the averages is included for completeness; the values are placed in parentheses since the concentrations are considered to be background levels. The amounts of thorium and uranium in a sample were proportional to the mass of material collected on the filter paper, and the bulk of these elements in the air was due to resuspension of soil. In contrast, the amount of plutonium in the air samples contributed by soil, if the resuspended soil has the same plutonium concentration as the first centimeter on the ground, ranged from about 1.6% in April to 10% in March of the total plutonium in the samples.

With the termination of the CP-5 reactor operation (Building 330, location 9H) on September 30, 1979, the principal source of tritiated water vapor and argon-41 in air was eliminated. The only measurable source of argon-41 was from the Janus reactor (Building 202, location 13I). Argon-41 was collected in the exhaust stack of this reactor by filling an evacuated "Marinelli-type" container with air and measuring the argon-41 concentration by gamma-ray spectrometry. The total amount released during 1980 was about 0.8 Ci.

TABLE 7

STRONTIUM, THORIUM, URANIUM AND PLUTONIUM CONCENTRATIONS IN AIR-FILTER SAMPLES, 1980  
(CONCENTRATIONS IN 1E-18 MICROCURIES/ML)

MONTH	LOCATION <sup>1</sup>										<sup>2</sup>	
		SR-89	SR-90	TH-228	TH-230	TH-232	U-234	U-235	U-238	PU-238	PU-239 <sup>3</sup>	
JANUARY	7I	< 100	120 ± 30	12 ± 2	12 ± 1	10 ± 1	23 ± 2	0.7 ± 0.3	19 ± 2	< 0.3	5.6 ± 1.1	
	12N	< 100	100 ± 30	11 ± 2	20 ± 1	13 ± 2	19 ± 2	0.4 ± 0.4	16 ± 2	< 0.3	3.6 ± 0.7	
	OFF-SITE	< 100	100 ± 20	10 ± 2	15 ± 2	10 ± 2	13 ± 1	0.4 ± 0.3	12 ± 1	< 0.3	4.0 ± 0.7	
FEBRUARY	7I	< 100	130 ± 10	11 ± 1	15 ± 2	11 ± 1	22 ± 2	0.5 ± 0.3	19 ± 2	< 0.3	5.1 ± 0.8	
	12N	< 100	130 ± 10	5 ± 2	8 ± 1	5 ± 1	21 ± 2	0.5 ± 0.4	16 ± 2	< 0.3	3.9 ± 0.7	
	OFF-SITE	< 100	140 ± 10	21 ± 3	25 ± 3	22 ± 4	26 ± 2	0.8 ± 0.5	28 ± 2	< 0.3	3.9 ± 0.7	
MARCH	7I	< 100	190 ± 50	4 ± 2	7 ± 1	4 ± 1	12 ± 2	0.5 ± 0.6	9 ± 1	< 0.3	5.5 ± 0.8	
	12N	< 100	160 ± 10	8 ± 2	13 ± 1	7 ± 1	26 ± 2	1.0 ± 0.4	21 ± 2	< 0.3	4.7 ± 0.8	
	OFF-SITE	< 100	170 ± 20	7 ± 3	6 ± 1	6 ± 1	12 ± 2	0.3 ± 0.3	11 ± 1	< 0.3	4.2 ± 0.6	
APRIL	7I	< 100	350 ± 40	8 ± 2	11 ± 1	8 ± 1	14 ± 2	0.1 ± 0.3	12 ± 2	< 0.3	6.9 ± 0.9	
	12N	< 100	210 ± 30	6 ± 1	9 ± 1	6 ± 1	11 ± 2	0.3 ± 0.5	10 ± 2	< 0.3	6.0 ± 0.8	
	OFF-SITE	< 100	170 ± 30	4 ± 1	4 ± 1	4 ± 1	6 ± 2	0.5 ± 0.3	7 ± 1	< 0.3	7.3 ± 1.3	
MAY	7I	< 100	450 ± 30	16 ± 1	27 ± 2	17 ± 1	31 ± 2	0.3 ± 0.4	26 ± 1	0.8 ± 0.3	11 ± 0.9	
	12N	< 100	400 ± 20	20 ± 2	25 ± 2	17 ± 1	31 ± 1	0.6 ± 0.2	27 ± 1	0.6 ± 0.3	12 ± 0.9	
	OFF-SITE	< 100	440 ± 20	11 ± 1	14 ± 1	10 ± 1	14 ± 1	0.5 ± 0.2	13 ± 1	0.4 ± 0.3	9.8 ± 0.8	
JUNE	7I	< 100	370 ± 70	26 ± 2	13 ± 1	39 ± 3	28 ± 2	0.6 ± 0.4	21 ± 1	0.5 ± 0.3	10 ± 1.0	
	12N	< 100	440 ± 20	5 ± 1	10 ± 1	8 ± 1	28 ± 3	0.8 ± 1.0	26 ± 2	< 0.3	11 ± 1.1	
	OFF-SITE	< 100	330 ± 20	8 ± 1	10 ± 1	9 ± 1	23 ± 4	0.7 ± 0.8	19 ± 3	< 0.3	8.9 ± 1.2	
JULY	7I	< 100	250 ± 30	7 ± 1	11 ± 1	7 ± 1	20 ± 2	0.8 ± 0.5	21 ± 2	< 0.3	7.2 ± 1.8	
	12N	< 100	260 ± 10	7 ± 1	11 ± 2	8 ± 1	19 ± 2	0.4 ± 0.4	21 ± 2	< 0.3	8.7 ± 2.0	
	OFF-SITE	< 100	230 ± 10	7 ± 1	9 ± 1	7 ± 1	20 ± 1	0.6 ± 0.3	20 ± 1	< 0.3	6.4 ± 1.9	
AUGUST	7I	< 100	140 ± 50	3 ± 1	4 ± 1	3 ± 1	19 ± 1	0.5 ± 0.2	17 ± 1	0.4 ± 0.3	3.0 ± 0.5	
	12N	< 100	120 ± 30	9 ± 1	13 ± 2	9 ± 1	24 ± 2	0.6 ± 0.3	21 ± 1	< 0.3	5.1 ± 0.7	
	OFF-SITE	< 100	90 ± 30	2 ± 1	3 ± 1	2 ± 1	13 ± 1	0.2 ± 0.2	13 ± 1	< 0.3	3.1 ± 0.5	
SEPTEMBER	7I	< 100	200 ± 10	5 ± 1	6 ± 1	4 ± 1	18 ± 2	0.7 ± 0.6	17 ± 2	0.3 ± 0.3	4.5 ± 0.8	
	12N	< 100	120 ± 40	5 ± 1	7 ± 1	5 ± 1	22 ± 2	0.2 ± 0.3	22 ± 2	0.4 ± 0.3	7.7 ± 0.4	
	OFF-SITE	< 100	100 ± 10	1 ± 2	1 ± 1	1 ± 1	25 ± 2	0.7 ± 0.5	18 ± 2	0.8 ± 0.7	4.5 ± 0.8	
OCTOBER	7I	< 100	130 ± 20	13 ± 3	14 ± 2	13 ± 2	21 ± 2	0.6 ± 0.3	18 ± 1	< 0.3	6.3 ± 1.4	
	12N	< 100	100 ± 40	10 ± 2	13 ± 2	10 ± 1	18 ± 2	0.3 ± 0.3	17 ± 1	< 0.3	6.5 ± 1.8	
	OFF-SITE	< 100	110 ± 40	3 ± 1	5 ± 1	3 ± 1	9 ± 1	0.3 ± 0.2	8 ± 1	< 0.3	3.9 ± 1.0	
NOVEMBER	7I	1880 ± 190	120 ± 80	6 ± 2	14 ± 2	6 ± 1	9 ± 2	0.4 ± 0.3	12 ± 1	< 0.3	4.5 ± 0.7	
	12N	2290 ± 160	90 ± 70	7 ± 2	13 ± 1	7 ± 1	17 ± 2	0.3 ± 0.3	14 ± 1	< 0.3	4.0 ± 1.5	
	OFF-SITE	1570 ± 100	50 ± 40	2 ± 3	3 ± 1	2 ± 1	5 ± 1	0.1 ± 0.3	5 ± 1	< 0.3	4.4 ± 0.8	
DECEMBER	7I	4150 ± 150	140 ± 60	8 ± 2	13 ± 2	8 ± 1	14 ± 2	0.6 ± 0.3	15 ± 2	< 0.3	5.7 ± 1.1	
	12N	5190 ± 160	140 ± 60	10 ± 3	15 ± 2	10 ± 1	18 ± 2	0.5 ± 0.3	16 ± 2	< 0.3	4.1 ± 1.4	
	OFF-SITE	3720 ± 250	140 ± 90	5 ± 4	8 ± 1	5 ± 1	14 ± 2	0.5 ± 0.4	11 ± 2	< 0.3	5.2 ± 1.3	
ANNUAL SUMMARY	7I	< 580	220 ± 70	10 ± 4	12 ± 4	11 ± 6	19 ± 4	0.5 ± 0.1	17 ± 3	< 0.4	6 ± 1	
	OFF-SITE	< 520	170 ± 70	7 ± 3	9 ± 4	7 ± 3	15 ± 4	0.5 ± 0.1	14 ± 4	< 0.3	5 ± 1	
%CG (X 1E-3)	7I	< 0.06	0.11	(4.9)	(4.0)	(1.1)	(0.48)	(0.01)	(0.34)	< 0.04	0.6	
	12N	< 0.07	0.08	(4.3)	(4.4)	(0.9)	(0.53)	(0.01)	(0.38)	< 0.03	0.6	
	OFF-SITE	< 0.05	0.08	(3.4)	(2.8)	(0.7)	(0.37)	(0.01)	(0.27)	< 0.03	0.5	

<sup>1</sup> PERIMETER LOCATIONS ARE GIVEN IN TERMS OF THE GRID COORDINATES IN FIGURE 1.

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

<sup>3</sup> PLUTONIUM-240 IS INCLUDED (SEE TEXT).

PLUTONIUM-239, 240 AIR CONCENTRATION

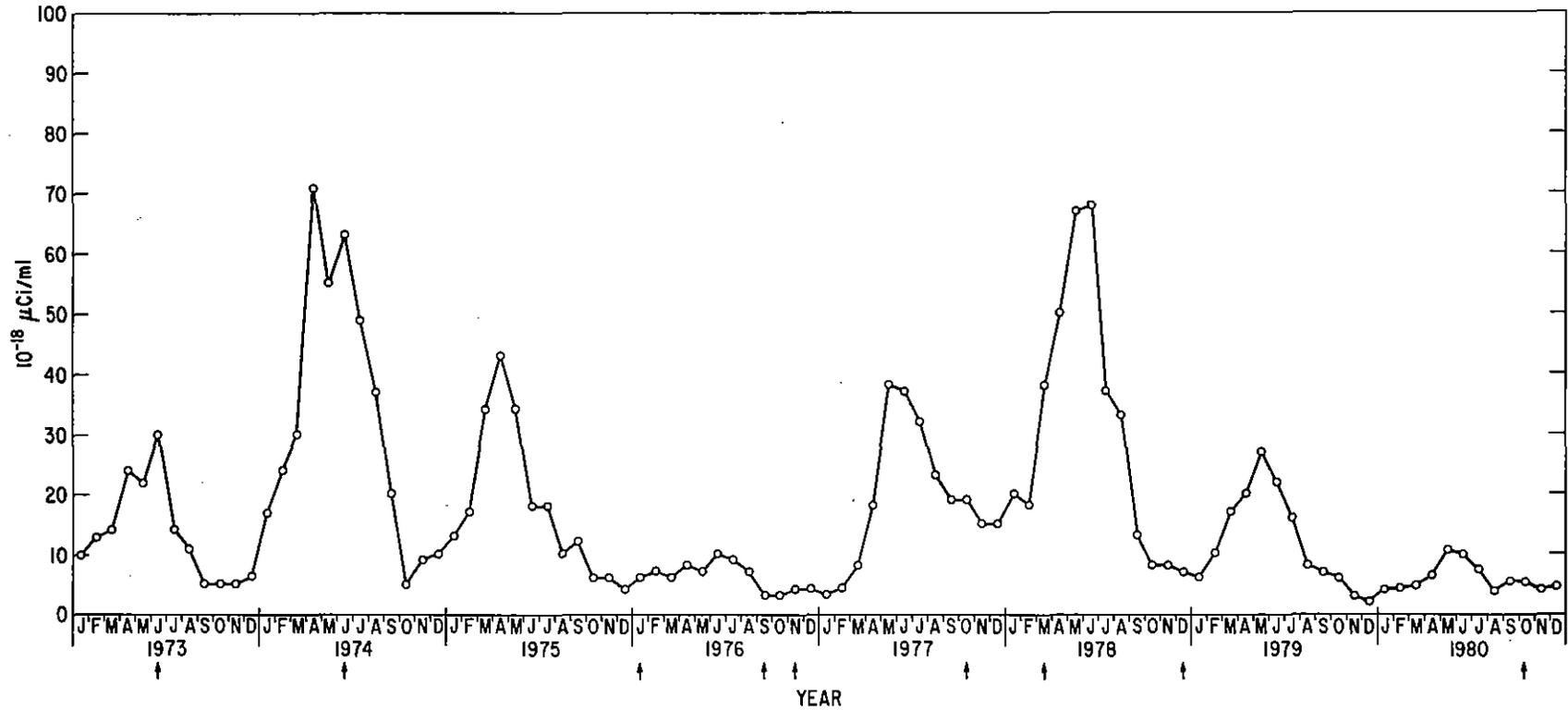


Fig. 4. Plutonium-239, 240 Air Concentrations, 1973-80

Although the CP-5 reactor is no longer operating, small amounts of residual tritiated water vapor continued to be emitted from the facility. Measurements of hydrogen-3 (as tritiated water vapor) in the exhaust stack of the CP-5 reactor indicated that 9 Ci were released during 1980. Collections were continued at the three perimeter and one off-site location. The results are given in Table 8. The 12M location (1900 m east-northeast of the CP-5 reactor) may be considered a perimeter sample for CP-5. Hydrogen-3 water vapor in air was collected by adsorption on silica gel and was measured by counting the desorbed water in a liquid scintillation counter. The average hydrogen-3 concentration of all perimeter locations was five times lower than in 1979 and not significantly different than the off-site concentration. The off-site concentrations measured about 10 km (6.2 miles) northwest of the Laboratory were similar to the 1979 results. If the off-site concentrations are subtracted from the concentrations at the perimeter locations to obtain the contribution from the Laboratory, the net result is within the error of the measurement, indicating dilution of the releases from CP-5 to ambient levels at the perimeter.

The only other major effluent released was krypton-85 from Building 212 (location 12I), estimated to be about 5 Ci during 1980. Several other fission products were also released in millicurie or smaller amounts. Since the CP-5 reactor no longer operates, the concentration of radionuclides released to the air by the Laboratory has been reduced by several orders of magnitude.

## 2. Surface Water

Total (nonvolatile) alpha and beta activities were determined by counting the residue remaining after evaporation of the water, and applying counting efficiency corrections determined for uranium-233 (for alpha activity) and thallium-204 (for beta activity) to obtain disintegration rates. Hydrogen-3 was determined by liquid scintillation counting of a separate sample, and this activity does not appear in the total nonvolatile beta activity. Uranium was determined fluorophotometrically, and the results calculated in terms of activity with the assumption that the isotopic composition was that of natural uranium. Analyses for other radionuclides were performed by specific radiochemical separations followed by appropriate counting. One liter aliquots were used for all analyses except hydrogen-3 and the transuranium nuclides. Hydrogen-3 analyses were performed by counting 10 ml in a gel system.

TABLE 8

Hydrogen-3 Perimeter and Off-Site Concentrations, 1980  
(Concentrations in  $10^{-12}$   $\mu\text{Ci/ml}$ )

Month	Location *	No. of Samples	Avg.	Min.	Max.
January	8F	8	0.5	0.2	1.3
	14I	8	0.9	0.3	1.5
	12M	2	1.0	0.1	1.8
	Off-Site	2	0.2	0.1	0.3
February	8F	8	0.8	0.3	1.7
	14I	8	0.7	0.3	1.3
	12M	2	0.7	0.7	0.7
	Off-Site	2	0.4	0.1	0.6
March	8F	9	1.0	0.3	4.0
	14I	9	1.0	0.4	1.5
	12M	2	1.0	0.6	1.3
	Off-Site	2	0.3	< 0.1	0.6
April	8F	9	2.0	0.3	5.3
	14I	9	1.0	0.2	2.3
	12M	2	0.9	0.6	1.3
	Off-Site	2	0.5	0.4	0.5
May	8F	9	3.4	0.7	10
	14I	9	1.6	0.8	2.7
	12M	2	1.5	0.8	2.2
	Off-Site	2	1.1	0.3	1.8
June	8F	8	2.6	1.4	4.2
	14I	8	2.0	1.5	2.7
	12M	2	2.5	1.4	3.6
	Off-Site	2	1.6	1.0	2.2
July	8F	9	4.5	2.5	12
	14I	9	2.8	1.5	6.6
	12M	2	3.1	2.9	3.2
	Off-Site	2	2.3	2.2	2.4
August	8F	9	3.4	1.7	5.5
	14I	9	3.1	1.9	5.2
	12M	2	2.6	2.0	3.2
	Off-Site	2	1.8	1.4	2.3
September	8F	9	2.2	0.6	5.3
	14I	9	1.7	0.5	2.7
	12M	2	2.6	1.9	3.3
	Off-Site	2	1.9	1.4	2.4
October	8F	8	3.3	1.0	9.8
	14I	8	1.3	0.3	2.6
	12M	2	3.7	2.6	4.9
	Off-Site	2	3.0	2.9	3.1
November	8F	9	1.5	0.2	2.5
	14I	9	1.6	0.7	2.7
	12M	2	1.3	1.2	1.5
	Off-Site	2	1.7	0.8	2.6
December	8F	8	1.5	0.3	2.9
	14I	8	1.4	0.5	3.0
	12M	2	1.3	1.2	1.4
	Off-Site	2	1.4	1.3	1.6
Annual Summary	8F	103	2.2 $\pm$ 0.7	0.2	12
	14I	103	1.6 $\pm$ 0.4	0.2	6.6
	12M	24	1.9 $\pm$ 0.6	0.1	4.9
	Off-Site	24	1.4 $\pm$ 0.5	< 0.1	3.1
Percent CG	8F	-	0.0011	0.00010	0.0060
	14I	-	0.0008	0.00010	0.0033
	12M	-	0.0010	0.00005	0.0025
	Off-Site	-	0.0007	< 0.00005	0.0016

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

Analyses for transuranium nuclides were performed on 10 or 50-liter samples by chemical separation methods followed by alpha spectrometry.<sup>(6,7)</sup> Plutonium-236 was used to determine the yields of plutonium and neptunium, which were separated together. A group separation of a fraction containing the transplutonium elements was monitored for recovery with americium-243 tracer.

Argonne waste water is discharged into Sawmill Creek, a small stream that runs through the Laboratory grounds, drains surface water from much of the site, and flows into the Des Plaines River about 500 m (0.3 mile) downstream from the waste-water outfall. Sawmill Creek was sampled upstream from the Argonne site and downstream from the waste-water outfall to determine if radioactivity was added to the stream by Argonne waste water or from surface drainage. The sampling locations are shown in Figure 1. Below the waste-water outfall, daily samples were collected by a continuous sampler, which operated about 97% of the year. When the continuous sampling device was not functioning, a grab sample was collected each working day. Equal portions of the daily samples collected each week were combined and analyzed to obtain an average weekly concentration. Above the site, samples were usually collected once a month and were analyzed for the same radionuclides as the below-outfall samples.

Annual summaries of the results obtained for Sawmill Creek are given in Table 9. Comparison of the results, and 95% confidence limits of the averages, for the two sampling locations shows that the nuclides whose presence in Creek water can be attributed to Argonne operations were hydrogen-3, neptunium-237, plutonium-239, americium-241, and occasionally strontium-90, plutonium-238, curium-242 and/or californium-252, and curium-244 and/or californium-249. The percentage of individual samples containing activity attributable to Argonne was 80% for hydrogen-3; 98% for neptunium-237; 100% for plutonium-239; and 80% for americium-241. The concentrations of all these nuclides were low compared to the CGs. The principal radionuclide added to the Creek by Argonne waste water, in terms of concentration, was hydrogen-3. Its average net concentration (equivalent to 0.004% of the CG) was the lowest since such measurements were made. The decrease in the hydrogen-3 is evidently related to the cessation of the operation of the CP-5 reactor. The total concentration, regardless of source, must be used in assessing the hazard of a radionuclide not naturally present, so the percent CG in the table was calculated on this basis.

TABLE 9  
RADIOISOTOPES IN SAWMILL CREEK, 1980

TYPE OF ACTIVITY	LOCATION *	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/ML)			AVG.	PERCENT CG	
			AVG.	MIN.	MAX.		MIN.	MAX.
ALPHA (NONVOLATILE)	16K	12	1.4 ± 0.1	0.2	2.7	(0.043)	(0.007)	(0.089)
	7H	254	1.9 ± 0.3	0.6	7.3	(0.065)	(0.020)	(0.242)
BETA (NONVOLATILE)	16K	12	20 ± 1	11	27	(0.65)	(0.38)	(0.90)
	7H	254	17 ± 1	11	30	(0.57)	(0.36)	(0.99)
HYDROGEN-3	16K	12	132 ± 3	< 100	205	0.0044	< 0.003	0.0068
	7H	254	250 ± 36	< 100	734	0.0083	< 0.003	0.0244
STRONTIUM-89	16K	12	-	-	< 2	-	-	< 0.067
	7H	254	-	-	< 2	-	-	< 0.067
STRONTIUM-90	16K	12	< 0.29	< 0.25	0.49	< 0.095	< 0.08	0.16
	7H	254	0.35 ± 0.04	< 0.25	0.82	0.117	< 0.08	0.27
IODINE-131	16K	12	< 1.1	< 1	1.6	< 0.37	< 0.33	0.55
	7H	254	< 1.6	< 1	18.2	< 0.52	< 0.33	6.06
BARIUM-140	16K	12	-	-	< 2	-	-	< 0.007
	7H	136	-	-	< 2	-	-	< 0.007
URANIUM ** (NATURAL)	16K	12	1.8 ± 0.1	1.2	2.4	(0.0044)	(0.0030)	(0.0061)
	7H	254	1.8 ± 0.1	1.1	2.8	(0.0044)	(0.0028)	(0.0069)
NEPTUNIUM-237	16K	11	-	-	< 0.001	-	-	< 0.000033
	7H	254	0.0100 ± 0.0050	< 0.0010	0.11	0.00033	< 0.00003	0.0036
PLUTONIUM-238	16K	11	-	-	< 0.001	-	-	< 0.00002
	7H	254	0.0062 ± 0.0023	< 0.001	0.051	0.00012	< 0.00002	0.0010
PLUTONIUM-239	16K	11	< 0.00055	< 0.0005	0.00083	< 0.000011	< 0.000010	0.000017
	7H	254	0.0115 ± 0.0028	0.0013	0.0425	0.00023	0.000026	0.0008
AMERICIUM-241	16K	12	-	-	< 0.001	-	-	< 0.000025
	7H	254	0.0057 ± 0.0021	< 0.001	0.043	0.00014	< 0.000025	0.0011
CURIUM-242 AND/OR CALIFORNIUM-252	16K	12	-	-	< 0.001	-	-	< 0.000005
	7H	254	< 0.0011	< 0.001	0.0028	< 0.000006	< 0.000005	0.000014
CURIUM-244 AND/OR CALIFORNIUM-249	16K	12	-	-	< 0.001	-	-	< 0.000014
	7H	254	0.0022 ± 0.0005	< 0.001	0.0076	0.000031	< 0.000014	0.00011

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

\*\* URANIUM CONCENTRATIONS IN UNITS OF MICROGRAMS/L CAN BE OBTAINED BY MULTIPLYING THE CONCENTRATION GIVEN BY 1.48 X 10<sup>9</sup>. THE AVERAGE CONCENTRATION IN THE CREEK THEN BECOMES 2.7 MICROGRAMS/L.

The hydrogen-3 in the Creek above the site was similar in concentration to levels found away from the Laboratory site and is characteristic of the current ambient levels in surface water. During 1980, the hydrogen-3 content of 21 other lakes and streams ranged from  $< 100 \times 10^{-9}$   $\mu\text{Ci/ml}$  to  $290 \times 10^{-9}$   $\mu\text{Ci/ml}$  and averaged  $150 \times 10^{-9}$   $\mu\text{Ci/ml}$ .

The total radioactive effluent discharged to the Creek in Argonne waste water can be estimated from the average net concentrations and the volume of water carried by the Creek. These totals are 1.6 Ci of hydrogen-3, 0.8 mCi of strontium-90, 0.15 mCi of plutonium-239, 0.06 mCi of americium-241, 0.07 mCi of neptunium-237, and  $< 0.05$  mCi of curium and californium nuclides.

Since Sawmill Creek empties into the Des Plaines River, which in turn flows into the Illinois River, the radioactivity in the latter two streams is important in assessing the contribution of Argonne waste water to the environmental radioactivity. The Des Plaines River was sampled twice a month below, and monthly above, the mouth of Sawmill Creek to determine if the radioactivity in the Creek had any effect on the activity in the River. Annual summaries of the results obtained for these two locations are given in Table 10. The average nonvolatile alpha, beta, and uranium concentrations in the River were very similar to past averages and remain in the normal range. Results were quite similar above and below the Creek for all radionuclides since the activity in Sawmill Creek was reduced by dilution so that it was not detectable as such in the Des Plaines River. The average nonvolatile alpha and beta activities,  $1.2 \times 10^{-9}$   $\mu\text{Ci/ml}$  and  $7 \times 10^{-9}$   $\mu\text{Ci/ml}$ , respectively, of 21 off-site surface water samples collected this year (excluding the Des Plaines River) were similar to the levels found in previous years.

Detectable concentrations of iodine-131 were found in three samples collected at location 16K in Sawmill Creek, eight samples collected at location 7M in Sawmill Creek, three samples at location A, upstream of ANL in the Des Plaines River, and four samples at location B, downstream of ANL in the Des Plaines River. For each detectable upstream sample, in either Sawmill Creek or the Des Plaines River, iodine-131 was also detected downstream, but not in both the Creek and River at the same time. It appears that iodine-131 in the Creek and the River is not from the same source. The detectable concentrations of iodine-131 did not correlate with the atmospheric nuclear test. It is speculated that the sources may be a medical facility or a sewage

TABLE 10  
RADIONUCLIDES IN DESPLAINES RIVER WATER, 1980

TYPE OF ACTIVITY	LOCATION *	NO. OF SAMPLES	CONCENTRATION (1E-09 MICROCURIES/ML)			AVG.	PERCENT CG	
			AVG.	MIN.	MAX.		MIN.	MAX.
ALPHA (NONVOLATILE)	A	12	1.4 ± 0.1	0.4	2.2	(0.047)	(0.012)	(0.074)
	B	23	1.5 ± 0.2	0.3	2.1	(0.048)	(0.010)	(0.071)
BETA (NONVOLATILE)	A	12	13 ± 1	8	18	(0.42)	(0.27)	(0.59)
	B	23	12 ± 2	7	22	(0.40)	(0.23)	(0.72)
HYDROGEN-3	A	12	124 ± 4	< 100	208	0.0041	< 0.003	0.0069
	B	23	< 145	< 100	437	< 0.0048	< 0.003	0.015
STRONTIUM-89	A	12	-	-	< 2	-	-	< 0.067
	B	23	-	-	< 2	-	-	< 0.067
STRONTIUM-90	A	12	0.41 ± 0.01	0.27	0.59	0.14	0.09	0.20
	B	23	0.38 ± 0.03	< 0.25	0.49	0.13	< 0.08	0.16
IODINE-131	A	12	< 1.5	< 1	3.5	< 0.49	< 0.33	1.18
	B	23	< 1.2	< 1	2.6	< 0.39	< 0.33	0.88
BARIUM-140	A	12	-	-	< 2	-	-	< 0.007
	B	12	-	-	< 2	-	-	< 0.007
URANIUM (NATURAL) **	A	12	1.5 ± 0.1	0.7	2.1	(0.0038)	(0.0017)	(0.0054)
	B	23	1.5 ± 0.1	0.7	2.3	(0.0038)	(0.0017)	(0.0057)
NEPTUNIUM-237	A	12	-	-	< 0.001	-	-	< 0.000033
	B	12	-	-	< 0.001	-	-	< 0.000033
PLUTONIUM-238	A	12	-	-	< 0.001	-	-	< 0.00002
	B	12	-	-	< 0.001	-	-	< 0.00002
PLUTONIUM-239	A	12	< 0.00068	< 0.0005	0.0013	< 0.000014	< 0.000010	0.000026
	B	12	< 0.00064	< 0.0005	0.0016	< 0.000013	< 0.000010	0.000032
AMERICIUM-241	A	10	-	-	< 0.001	-	-	< 0.000025
	B	12	-	-	< 0.001	-	-	< 0.000025
CURIUM-242 AND/OR CALIFORNIUM-252	A	10	-	-	< 0.001	-	-	< 0.000005
	B	12	-	-	< 0.001	-	-	< 0.000005
CURIUM-244 AND/OR CALIFORNIUM-249	A	10	-	-	< 0.001	-	-	< 0.000014
	B	12	-	-	< 0.001	-	-	< 0.000014

\* LOCATION A, NEAR ROUTE 45, IS UPSTREAM AND LOCATION B, NEAR LEHONT, IS DOWNSTREAM FROM THE MOUTH OF SAWHILL CREEK. SEE FIGURE 2.

\*\* URANIUM CONCENTRATIONS IN UNITS OF MICROGRAMS/L CAN BE OBTAINED BY MULTIPLYING THE CONCENTRATION GIVEN BY  $1.48 \times 10^{-9}$ . THE AVERAGE CONCENTRATION IS 2.4 MICROGRAMS/L.

treatment plant that has processed excretions from an individual who has received a medical injection of iodine-131.

It is difficult to locate the source of the iodine-131 because it is detected so rarely. An attempt was made to determine if the Marion Brook Sewage Treatment Plant, located upstream of the Laboratory on Sawmill Creek, was a source. Daily water samples were collected from the effluent of the plant and analyzed for iodine-131. However, during the five weeks these special samples were collected, no iodine-131 was detected in these samples nor in the regular Sawmill Creek samples at locations 16K or 7M.

The radioactivity in samples of Illinois River water, shown in Table 11, was similar to those found in other bodies of water in the area and to the activities found previously at these same locations. No radioactivity originating at Argonne could be detected in the Des Plaines or Illinois Rivers.

### 3. Soil, Grass, and Bottom Sediment

The plutonium content of soil, grass, and bottom sediment was measured at the site perimeter and off the site. The object of the off-site sampling was to determine the deposition of plutonium from weapons testing for comparison with perimeter samples, and with results obtained by other organizations for samples collected at large distances from nuclear installations. This latter comparison is useful in determining if the soil activity near Argonne is normal. For this purpose, the site selection criteria and sample preparation techniques of the DOE Environmental Measurements Laboratory<sup>(8)</sup> were used. Sites were selected in several directions and at various distances from the Laboratory. Each site was selected on the basis that the soil appeared, or was known to have been, undisturbed for a number of years. Attempts were made to select open, level, grassy areas that were mowed at reasonable intervals. Public parks were selected when available.

Each soil sample consisted of five cores totaling 432 cm<sup>2</sup> in area by 5 cm deep. Through 1976, samples had been collected down to 30 cm to measure total plutonium deposition and as a result of five years of sample collection and analysis at this depth, the total plutonium deposited in the Argonne environment has been established. By reducing the sampling depth to 5 cm, the analysis should be more sensitive to changes in current deposition. The grass samples were obtained by collecting the grass from a 1 m<sup>2</sup> area in the

TABLE 11

Radionuclides in Illinois River Water, 1980  
(Concentrations in  $10^{-9}$   $\mu\text{Ci/ml}$ )

Location	Date Collected	Alpha*	Beta*	Hydrogen-3	Uranium** (natural)	Plutonium-239
McKinley Woods State Park	June 12	0.86	7.6	< 100	0.7	0.00028
Below Dresden Power Station	June 12	0.38	3.9	< 100	1.2	0.00032
Morris	June 12	2.37	6.2	124	1.2	-
Starved Rock State Park	June 12	1.71	3.9	119	1.2	-
McKinley Woods State Park	October 16	0.31	9.1	287	0.6	0.00026
Below Dresden Power Station	October 16	0.80	6.9	254	1.1	0.00014
Morris	October 16	0.94	8.2	149	0.8	-
Starved Rock State Park	October 16	0.49	3.9	209	0.8	-

\* Nonvolatile activity.

\*\* Uranium concentrations in units of  $\mu\text{g/l}$  can be obtained by multiplying the concentration by  $1.48 \times 10^9$ .

immediate vicinity of a soil sample. A grab sample technique was used to obtain bottom sediment. After drying, grinding, and mixing, 100 g portions of soil, bottom sediment, and grass were analyzed by the same method described in Section III.A. for air-filter residue. Results are given in terms of oven-dried soil, bottom sediment, or grass.

Soil samples were analyzed by gamma-ray spectrometry and radiochemically for plutonium. In contrast to past years, samples were not analyzed for uranium and thorium. The results (Table 12) show that the average and range of the perimeter concentrations of cesium-137 and plutonium are similar to both the perimeter and off-site results obtained in previous years, and are considered to be the current fallout concentrations of these nuclides in soil. The average 1980 off-site concentrations are about a factor of two lower than the perimeter concentrations in 1980 and the off-site concentrations in other years, and this difference is believed to reflect the variability in the distribution of fallout radionuclides in soil.

Composite monthly precipitation samples were analyzed for plutonium-239. The results are given in Table 13, along with results since 1973 for comparison. The total 1980 deposition by precipitation was similar to that of 1979 and was equivalent to 0.3% of the total plutonium previously deposited, which is reported to be  $2.2 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ .<sup>(9)</sup> The data in Table 13 is illustrated in Figure 5. The arrows at the bottom of the figure indicate the approximate dates of the Chinese atmospheric nuclear tests. Comparison of the results in Figure 5 with the plutonium air concentrations for the past eight years in Figure 4 shows excellent correlation.

The results of radioactivity measurements in grass are given in Table 14. The grass samples were washed before analysis to remove surface soil, which contains considerably more radioactivity per gram than grass. If the grass is analyzed without adequate washing, results are more variable and difficult to compare between locations. This problem apparently occurred in 1979. As a result, considerable attention was given to a more complete removal of surface soil from the 1980 grass samples. This seems to account for the lower radionuclide concentrations in both perimeter and off-site samples compared to previous years. No fission product gamma-ray emitters were detected in any grass samples. In terms of deposition, the plutonium-239 concentration was a factor of about  $10^4$  less in the grass than in the soil from the same

TABLE 12

## Radionuclides in Soil, 1980

Date Collected	Location	Cesium-137	Plutonium-238		Plutonium-239		$^{238}\text{Pu}/^{239}\text{Pu}$
		$10^{-6}$ $\mu\text{Ci/g}$	$10^{-9}$ $\mu\text{Ci/g}$	$10^{-3}$ $\mu\text{Ci/m}^2$	$10^{-9}$ $\mu\text{Ci/g}$	$10^{-3}$ $\mu\text{Ci/m}^2$	
<u>Perimeter*</u>							
June 20	4EF	$1.1 \pm 0.1$	$0.9 \pm 0.2$	$0.042 \pm 0.007$	$19.7 \pm 0.6$	$0.92 \pm 0.03$	0.046
June 20	7EF	$1.2 \pm 0.1$	$1.2 \pm 0.2$	$0.038 \pm 0.006$	$23.4 \pm 0.7$	$0.76 \pm 0.02$	0.051
June 20	10P	$1.2 \pm 0.1$	$0.7 \pm 0.1$	$0.032 \pm 0.006$	$20.1 \pm 0.9$	$0.91 \pm 0.04$	0.036
June 20	13/14N	$1.4 \pm 0.1$	$1.8 \pm 0.3$	$0.076 \pm 0.012$	$40.2 \pm 1.8$	$1.69 \pm 0.08$	0.045
June 20	14E	$1.5 \pm 0.1$	$0.8 \pm 0.1$	$0.036 \pm 0.006$	$23.8 \pm 1.0$	$1.05 \pm 0.04$	0.034
October 20	7H	$1.0 \pm 0.1$	$1.3 \pm 0.3$	$0.055 \pm 0.011$	$21.4 \pm 1.0$	$0.89 \pm 0.04$	0.062
October 20	9/10N	$0.7 \pm 0.1$	$1.4 \pm 0.3$	$0.078 \pm 0.015$	$13.2 \pm 0.8$	$0.72 \pm 0.04$	0.11
October 20	13N	$1.2 \pm 0.1$	-	-	-	-	-
October 20	15H	$0.6 \pm 0.1$	-	-	-	-	-
October 20	13D	$1.2 \pm 0.1$	$1.4 \pm 0.3$	$0.059 \pm 0.012$	$23.8 \pm 1.2$	$1.00 \pm 0.05$	0.059
	Average	$1.1 \pm 0.2$	$1.2 \pm 0.3$	$0.052 \pm 0.013$	$23.2 \pm 5.4$	$0.99 \pm 0.21$	$0.055 \pm 0.017$
<u>Off-Site</u>							
June 12	Channahon, IL	$0.5 \pm 0.1$	$0.5 \pm 0.1$	$0.025 \pm 0.007$	$10.4 \pm 0.5$	$0.53 \pm 0.03$	0.046
June 12	Starved Rock State Park, IL	$0.4 \pm 0.1$	$0.7 \pm 0.2$	$0.042 \pm 0.014$	$7.4 \pm 0.4$	$0.47 \pm 0.03$	0.089
June 12	Morris, IL	$0.6 \pm 0.1$	$0.5 \pm 0.2$	$0.026 \pm 0.012$	$14.6 \pm 0.9$	$0.73 \pm 0.05$	0.036
June 17	Lemont, IL	$0.5 \pm 0.1$	$1.6 \pm 0.2$	$0.082 \pm 0.013$	$10.1 \pm 0.6$	$0.53 \pm 0.03$	0.155
June 17	McGinnis Slough, IL	$0.8 \pm 0.1$	$0.4 \pm 0.1$	$0.019 \pm 0.005$	$14.1 \pm 0.6$	$0.61 \pm 0.02$	0.031
June 17	Saganashkee Slough, IL	$0.8 \pm 0.1$	$0.2 \pm 0.1$	$0.010 \pm 0.005$	$12.9 \pm 0.5$	$0.59 \pm 0.02$	0.017
October 16	McKinley Woods State Park, IL	$0.3 \pm 0.1$	$0.4 \pm 0.5$	$0.023 \pm 0.008$	$9.7 \pm 0.8$	$0.54 \pm 0.04$	0.042
October 16	Dresden Lock and Dam, IL	$0.4 \pm 0.1$	$0.2 \pm 0.3$	$0.048 \pm 0.008$	$9.0 \pm 0.6$	$0.28 \pm 0.02$	0.047
October 17	Western Springs, IL	$0.5 \pm 0.1$	$0.2 \pm 0.4$	$0.013 \pm 0.002$	$10.3 \pm 0.9$	$0.67 \pm 0.06$	0.049
October 17	Brookfield, IL	$1.2 \pm 0.1$	$1.7 \pm 0.5$	$0.103 \pm 0.032$	$21.9 \pm 2.2$	$1.33 \pm 0.13$	0.077
	Average	$0.6 \pm 0.2$	$0.6 \pm 0.4$	$0.039 \pm 0.020$	$12.0 \pm 2.6$	$0.63 \pm 0.17$	$0.059 \pm 0.025$

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

TABLE 13

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

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

PLUTONIUM-239, 240 GROUND DEPOSITION

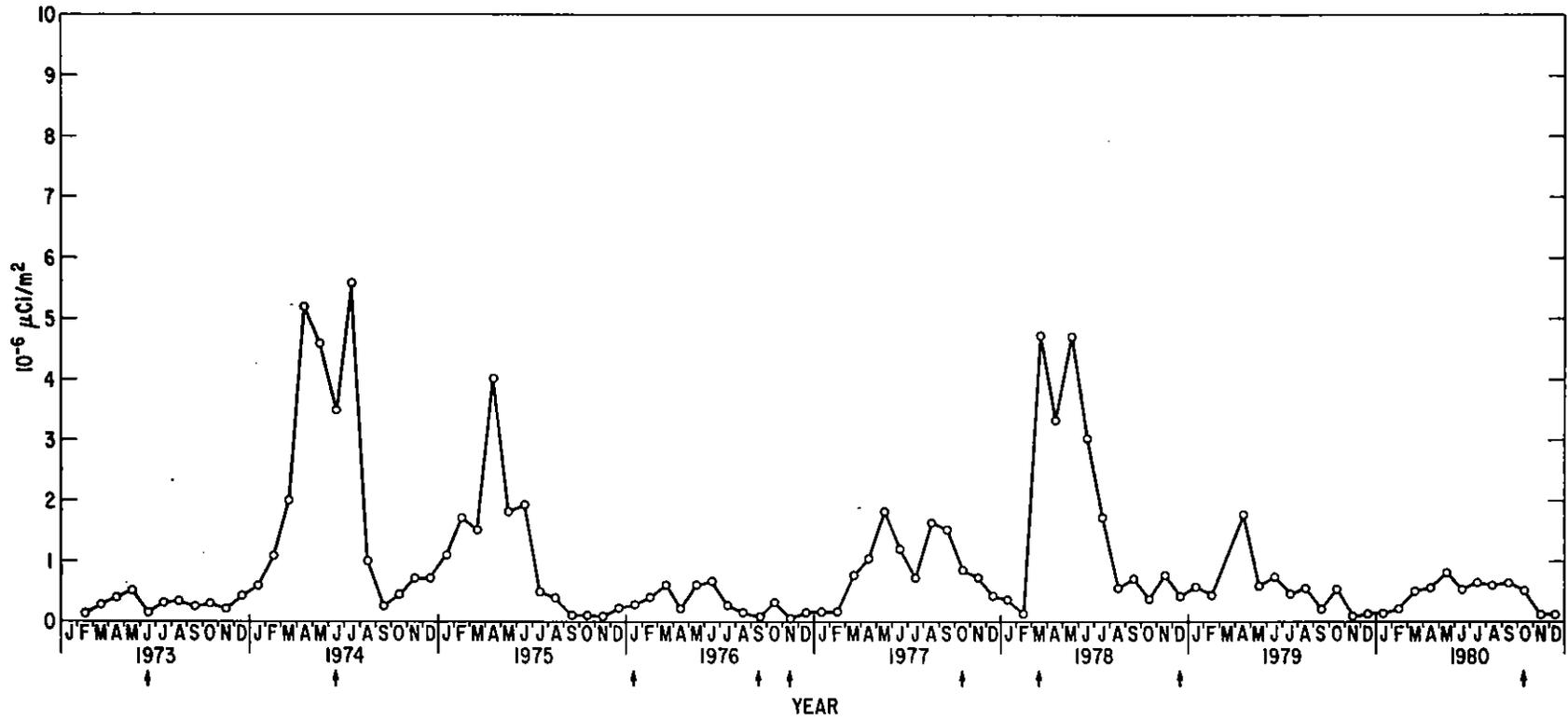


Fig. 5. Plutonium-239, 240 Surface Deposition, 1973-80

TABLE 14

## Radionuclides in Grass Samples, 1980

Date Collected	Location	Concentrations in $10^{-9}$ $\mu\text{Ci/g}$						$10^{-6}$ $\mu\text{Ci/m}^2$
		Thorium-228	Thorium-230	Thorium-232	Uranium (natural)	Plutonium-238	Plutonium-239	Deposited Plutonium-239
	<u>Perimeter</u> *							
June 20	4EF	$8.0 \pm 0.7$	$6.9 \pm 0.6$	$4.3 \pm 0.4$	$27 \pm 4$	< 0.1	$0.71 \pm 0.17$	$0.073 \pm 0.017$
June 20	7EF	$0.8 \pm 0.4$	$1.6 \pm 0.3$	$0.6 \pm 0.2$	$15 \pm 2$	< 0.1	$0.23 \pm 0.11$	$0.080 \pm 0.039$
June 20	10P	$3.1 \pm 0.4$	$5.2 \pm 0.5$	$2.9 \pm 0.4$	$29 \pm 2$	< 0.1	$0.73 \pm 0.15$	$0.165 \pm 0.035$
June 20	13/14N	$2.5 \pm 0.3$	$5.1 \pm 0.5$	$4.0 \pm 0.5$	$45 \pm 7$	< 0.1	$0.53 \pm 0.14$	$0.076 \pm 0.019$
June 20	14E	$3.6 \pm 0.4$	$7.8 \pm 0.6$	$6.3 \pm 0.7$	$32 \pm 4$	< 0.1	$0.73 \pm 0.15$	$0.156 \pm 0.033$
October 20	7M	$0.9 \pm 0.4$	$1.4 \pm 0.3$	$1.2 \pm 0.2$	-	< 0.1	$0.28 \pm 0.07$	$0.063 \pm 0.015$
October 20	9/10N	$3.2 \pm 0.3$	$3.4 \pm 0.6$	$2.0 \pm 0.4$	-	< 0.1	$0.36 \pm 0.14$	$0.079 \pm 0.030$
October 20	13N	$9.3 \pm 0.7$	$5.6 \pm 0.5$	$3.4 \pm 0.4$	-	< 0.1	$0.22 \pm 0.20$	$0.219 \pm 0.020$
October 20	15H	$2.8 \pm 0.3$	$4.0 \pm 0.8$	$2.5 \pm 0.6$	-	< 0.1	$0.45 \pm 0.12$	$0.101 \pm 0.028$
October 20	13D	$3.8 \pm 0.4$	$5.9 \pm 0.6$	$4.4 \pm 0.5$	-	-	-	-
	Average	$3.8 \pm 1.7$	$4.7 \pm 1.3$	$3.2 \pm 1.1$	$30 \pm 10$	< 0.1	$0.47 \pm 0.14$	$0.112 \pm 0.036$
	<u>Off-Site</u>							
June 12	Channahon, IL	$1.2 \pm 0.6$	$2.1 \pm 0.8$	$1.6 \pm 0.6$	$63 \pm 6$	< 0.1	$0.54 \pm 0.15$	$0.075 \pm 0.021$
June 12	Starved Rock State Park, IL	$1.6 \pm 0.2$	$4.2 \pm 0.5$	$2.7 \pm 0.4$	$40 \pm 4$	< 0.1	$0.39 \pm 0.15$	$0.036 \pm 0.014$
June 12	Morris, IL	$4.0 \pm 0.1$	$9.1 \pm 0.2$	$5.9 \pm 0.2$	$182 \pm 12$	< 0.1	$0.88 \pm 0.19$	$0.114 \pm 0.024$
June 17	Lemont, IL	$1.4 \pm 0.2$	$2.6 \pm 0.3$	$1.2 \pm 0.2$	$15 \pm 1$	< 0.1	$0.36 \pm 0.12$	$0.053 \pm 0.019$
June 17	McGinnis Slough, IL	$6.9 \pm 0.6$	$14.0 \pm 0.8$	$7.0 \pm 0.6$	$27 \pm 3$	< 0.1	$0.55 \pm 0.25$	$0.033 \pm 0.015$
June 17	Saganashkee Slough, IL	$2.6 \pm 0.4$	$6.0 \pm 0.7$	$3.4 \pm 0.5$	$37 \pm 4$	< 0.1	$0.28 \pm 0.17$	$0.021 \pm 0.013$
October 16	McKinley Woods State Park, IL	-	-	-	-	-	-	-
October 16	Dresden Lock and Dam, IL	-	-	-	-	-	-	-
October 17	Western Springs, IL	-	-	-	-	< 0.1	< 0.1	< 0.01
October 17	Brookfield, IL	$3.6 \pm 0.4$	$5.9 \pm 0.6$	$5.2 \pm 0.6$	-	< 0.1	$0.25 \pm 0.10$	$0.020 \pm 0.009$
	Average	$3.0 \pm 1.5$	$6.3 \pm 3.1$	$3.8 \pm 1.7$	$61 \pm 50$	< 0.1	$0.41 \pm 0.18$	$0.045 \pm 0.025$

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

location. Concentration ranges were the same at the perimeter and off-site, indicating no contribution from ANL operations.

Results of analysis of bottom sediment samples for gamma-ray emitters and plutonium are given in Table 15. Plutonium results vary widely between locations and are strongly dependent on the retentiveness of the bottom material. This is illustrated by the results from the sample collected from the bottom of the pond located at 15H. The composition of this sample is similar to that of off-site bottom sediment samples collected at similar ponds in past years.<sup>(10)</sup> The result at 15H is within the range of concentrations found at similar off-site ponds and has a  $^{238}\text{Pu}/^{239}\text{Pu}$  ratio normally found in fallout. The elevated plutonium-239 concentrations in the Sawmill Creek bottom sediment are residuals of previous Laboratory effluent releases.

#### 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  $1 \times 10^{-10}$   $\mu\text{Ci/ml}$  by an ion-exchange separation followed by beta counting. Cesium-137 was analyzed with a detection limit of  $5 \times 10^{-10}$   $\mu\text{Ci/ml}$  by an ion-exchange separation followed by gamma-ray spectrometry. The other nuclides were analyzed by the same methods used for water and with the same detection limits. The results are given in Table 16. The average strontium-90 concentration decreased by about 10% compared to 1979, while the average cesium-137 concentration decreased by a factor of two. These nuclides are fission products from nuclear tests and their presence in milk is not related to Argonne operations. No short-lived fission products, i.e., strontium-89, iodine-131, or barium-140, were detected in milk since the cows were being fed on stored feed during November and December, when fallout from the October 16, 1980, atmospheric test was found in the area. No garden vegetables were collected this year.

The concentrations given in Table 16 may be compared to the CGs for drinking water given in the Appendix, Part B. The drinking water CGs are based on an intake of 2.2 liters per day. The consumption of one liter of milk per day would result in an average intake of 0.48% of the strontium-90 and 0.003% of the cesium-137 Concentration Guides.

TABLE 15  
 Radionuclides in Bottom Sediment, 1980

Date Collected	Location	Concentrations in $10^{-6}$ $\mu\text{Ci/g}$		Concentrations in $10^{-9}$ $\mu\text{Ci/g}$	
		Cobalt-60	Cesium-137	Plutonium-238	Plutonium-239
	<u>Perimeter</u> *				
September 24	6N - Sawmill Creek and Des Plaines River	< 0.1	$0.3 \pm 0.1$	-	-
September 24	6N - 150 m Below Outfall	< 0.1	$0.1 \pm 0.1$	$1.2 \pm 0.2$	$13.7 \pm 0.8$
September 24	6N - 100 m Below Outfall	$0.1 \pm 0.1$	$0.1 \pm 0.1$	$1.4 \pm 0.2$	$9.2 \pm 0.6$
September 24	7M - 50 m Below Outfall	< 0.1	$0.1 \pm 0.1$	$7.3 \pm 0.5$	$44.4 \pm 1.2$
September 24	7M - At Outfall	$0.1 \pm 0.1$	$0.1 \pm 0.1$	$2.2 \pm 0.3$	$11.4 \pm 0.7$
September 24	7M - 20 m Above Outfall	< 0.1	$0.1 \pm 0.1$	$0.3 \pm 0.2$	$1.2 \pm 0.3$
October 20	Pond - 15H	< 0.1	$1.8 \pm 0.1$	$1.4 \pm 0.6$	$33.0 \pm 2.4$
	Average	< 0.1	$0.4 \pm 0.5$	$2.3 \pm 2.1$	$18.8 \pm 13.4$
	<u>Off-Site</u>				
June 12	Illinois River, McKinley Woods, IL	< 0.1	$0.1 \pm 0.1$	< 0.1	$0.6 \pm 0.2$
June 12	Illinois River, Dresden Lock & Dam, IL	$0.1 \pm 0.1$	$0.2 \pm 0.1$	$0.1 \pm 0.1$	$4.5 \pm 0.4$
June 17	Long Run Creek, Lemont, IL	< 0.1	$0.3 \pm 0.1$	< 0.1	$6.3 \pm 1.5$
June 17	McGinnis Slough, IL	< 0.1	$0.2 \pm 0.1$	$0.1 \pm 0.1$	$4.1 \pm 0.4$
October 16	DuPage River, Channahon, IL	< 0.1	$0.2 \pm 0.1$	$0.1 \pm 0.1$	$3.6 \pm 0.4$
October 17	55th Street Pond, Downers Grove, IL	< 0.1	$0.1 \pm 0.1$	< 0.1	$1.5 \pm 0.6$
	Average	< 0.1	$0.2 \pm 0.1$	< 0.1	$3.4 \pm 1.7$

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

TABLE 16

Radionuclides in Milk, 1980  
(Concentrations in  $10^{-9}$   $\mu\text{Ci/ml}$ )

Date Collected	Hydrogen-3	Strontium-89	Strontium-90	Iodine-131	Cesium-137	Barium-140
January 16	149	< 2	2.7	< 0.1	1.6	< 2
February 6	< 100	< 2	3.0	< 0.1	0.9	< 2
March 5	< 100	< 2	2.8	< 0.1	1.2	< 2
April 2	< 100	< 2	3.5	< 0.1	0.9	< 2
May 7	239	< 2	4.0	< 0.1	1.2	< 2
June 11	210	< 2	5.2	< 0.1	1.2	< 2
July 2	168	< 2	5.5	< 0.1	0.8	< 2
August 6	176	< 2	3.0	< 0.1	1.1	< 2
September 4	155	< 2	1.6	< 0.1	1.3	< 2
October 1	166	< 2	3.4	< 0.1	0.8	< 2
November 5	< 100	< 2	3.2	< 0.1	1.2	< 2
December 3	< 100	< 2	2.0	< 0.1	1.7	< 2
Average	126 $\pm$ 41	< 2	3.3 $\pm$ 0.7	< 0.1	1.2 $\pm$ 0.2	< 2

## 5. External Penetrating Radiation

Measurements were made with calcium fluoride (dysprosium activated) and lithium fluoride thermoluminescent dosimeter (TLD) chips. Each measurement was the average of three or four chips exposed in the same packet. The 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 any dose that might be 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. These locations are shown in Figure 2.

The results are summarized in Tables 17 and 18, and the site boundary and on-site readings are also shown in Figure 6. Measurements were made in six successive exposure periods that varied in length from 55 to 67 days, and in total covered the period from January 16, 1980, to January 12, 1981. The results for each period were calculated in terms of annual dose for ease in comparing measurements made for different elapsed times, and were weighted according to their exposure times in calculating the annual average at each location. The uncertainty given in the tables for an average is the 95% confidence limit calculated from the standard error.

The off-site results averaged  $90 \pm 8$  mrem/yr. The standard error of the mean of all 30 individual results was 3.0 mrem/yr. From 1973 to 1979, the corresponding averages and standard errors varied from  $89 \pm 7$  mrem/yr (1979) to  $100 \pm 2$  mrem/yr (1973). The seven-year average was 97 mrem/yr, with a standard deviation of 4 mrem/yr. Thus, the background radiation has been quite consistent and the 1980 average is within the range found previously. The variations from year-to-year have also been consistent at each location. The annual average at Downers Grove, Lemont, and Lombard have been within 5 mrem/yr of the average of all five off-site locations (usually within 2 mrem/yr). At Oak Lawn, the results have been from 6 to 11 mrem/yr below, and at Oakbrook, 6 to 11 mrem/yr above, the overall average. The differences between sampling periods were also relatively small compared to 1979, when

TABLE 17

## Environmental Penetrating Radiation at Off-Site Locations, 1980

Location	Dose Rate (mrem/year)						Average
	Period of Measurement						
	1/16-3/12	3/12-5/6	5/6-7/11	7/11-9/16	9/16-11/12	11/12-1/12	
Downers Grove	86	88	86	90	94	87	89 ± 3
Lemont	96	88	92	92	98	95	94 ± 4
Lombard	95	88	82	86	98	89	90 ± 6
Oakbrook	105	98	94	94	99	98	98 ± 4
Oak Lawn	85	78	77	74	75	83	79 ± 4
Average	93 ± 9	88 ± 8	86 ± 8	87 ± 9	93 ± 12	90 ± 7	90 ± 8

TABLE 18

## Environmental Penetrating Radiation at ANL, 1980

Location	Dose Rate (mrem/year)						Average
	1/16-3/12	3/12-5/6	Period of Measurement		9/16-11/12	11/12-1/12	
			5/6-7/11	7/11-9/16			
14L - Boundary	79	73	76	80	89	84	80 ± 5
14I - Boundary	130	121	121	116	119	123	122 ± 5
14G - Boundary	85	89	86	80	89	89	86 ± 4
13D - Boundary	68	62	64	62	70	71	66 ± 4
9/10EF - Boundary	83	81	79	80	96	81	83 ± 6
8H - Boundary	85	84	90	87	91	91	88 ± 3
8H - Boundary	-	97	98	98	101	98	98 ± 2
8H - Center, St. Patrick's Cemetery	-	100	101	103	-	101	101 ± 2
7I - Boundary	598	598	578	662	657	794	648 ± 79
6I - 200 m N of Quarry Road	94	97	96	97	102	99	98 ± 3
9L - Boundary	73	72	69	68	70	70	70 ± 2
9H - 50 m SE of CP-5	1960	1850	1690	1680	1890	1860	1820 ± 113
9G - 45 m W of CP-5	-	-	118	-	-	-	118
10H - 70 m NE of CP-5	-	147	133	-	-	-	140 ± 10
8H - 65 m S of 316	120	115	122	98	125	100	113 ± 12
8H - 200 m NW of Waste Storage Area (Heliport)	178	172	186	179	203	198	186 ± 12
7I - Center, Waste Storage Area	2150	1370	1330	4960	4880	7560	3710 ± 2500
10/11K - Lodging Facilities	75	74	70	71	71	76	73 ± 2
9J - Between ZGS Condenser and 386	150	151	153	121	-	-	144 ± 15
9J - Between 368 and 377	-	-	-	-	-	62	62
13J - 135 m E of 202	311	263	280	262	295	276	281 ± 19
12M - 30 m W of 55	66	60	55	54	62	61	60 ± 5
9I - 65 m NE of 350 230 m NE of 316	84	84	85	82	88	87	85 ± 2

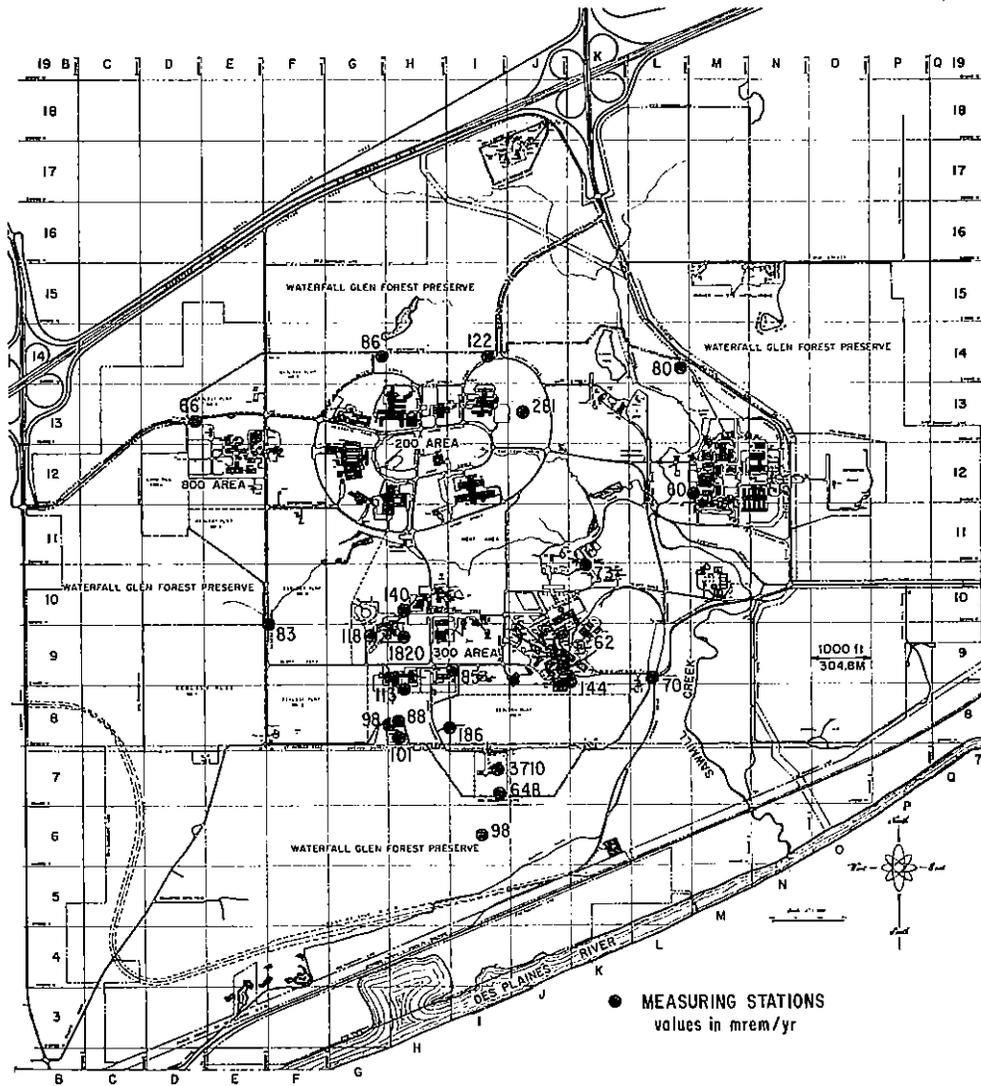


Fig. 6. Penetrating Radiation Measurements at the ANL Site, 1980

heavy snow cover resulted in lower doses during the winter.

If the off-site locations are an accurate sample of the radiation background in the area, then annual averages at the site boundary in the range of  $90 \pm 8$  mrem/yr may be considered normal with a 95% probability, or 95% of the boundary results should have averages in this range. To compare boundary results for individual sampling periods, the standard deviation of the mean of the 30 individual off-site results is useful. This value is 7.7 mrem/yr, so that individual results in the range of  $90 \pm 16$  mrem/yr may be considered normal with a 95% probability.

At two locations, 7I at the south boundary of the site and 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 560 mrem/yr; in previous years, this value has ranged from 440 mrem/yr in 1976 to 114 mrem/yr in 1977. The higher dose in 1980 was due to the fact that waste was retained for longer periods of time before shipment and disposal. The dose at the 7I boundary from the Waste Storage Facility is consistent with the dose at the center of the Facility; the two values vary in a parallel manner, and the changes with time reflect the amount of radioactive waste in storage and its gamma-ray intensity. At 14I, the average dose rate was about 32 mrem/yr above normal, about half of the 1979 rate. This dose is attributed to the use of cobalt-60 irradiation sources in Building 202.

Dose variations from period to period at the same location, which are at least in part statistical, and natural dose differences between locations make it difficult to determine with high certainty when site boundary doses are only a few mrem/yr above normal and due to Argonne operations. Three criteria are used to identify such locations: 1) the results for each sampling period are frequently above the off-site average for the same period; 2) the annual average at a location exceeds the off-site average plus the standard deviation of the average at the 95% confidence limit (98 mrem/yr); or 3) occasional results at a particular location are significantly above the normal value for that location, but do not exceed the off-site normal range. The last criterion could apply to results at a location such as 13D, where

dose rates have been consistently below the average. At this location, the ground contains considerable gravel, which has less radium than the usual clay soil, and this probably accounts for the low dose rates. Application of the first two criteria and the difficulties in interpreting small differences from natural background by TLD measurements are illustrated below.

The dose range at 8H is of interest, since it lies in St. Patrick's Cemetery, which was in use before Argonne was constructed, and which is open to visitors. The dose at the center of the Cemetery was 11 mrem/yr above the off-site average and 3 mrem/yr above the 95% upper confidence limit of the normal range (98 mrem/yr). Two sampling locations on the boundary line between ANL and the Cemetery (8H) were both in the normal range, both individually and in the annual average. However, they differ from each other by about 10 mrem/yr because of their elevations. The higher values were obtained at a location that has a direct line of sight to a radiation source. The other location does not. Based on these types of comparison, the dose at 8H is estimated to be 5 to 10 mrem/yr above normal for that location. Similar results were obtained in previous years.

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

## 6. Potential Radiation Dose Estimates

### a. Air and Waterborne Radionuclides

The radiation doses at the site boundary and off the site that could have been received by the public from radioactive materials leaving the site were calculated by two methods. Where measured radionuclide concentrations in water were available, conversion of concentration to dose was based on the ratio of environmental concentrations to the Concentration Guides given in Table 19. This table gives the annual radiation doses that would result from continuous exposure at the specified concentrations for those nuclides whose

TABLE 19

## Concentration-to-Dose Conversion Factors

Nuclide	Medium	Concentration* ( $\mu\text{Ci}/\text{ml}$ )	Dose* (rem)	Critical Organ
Americium-241	Water	$4 \times 10^{-6}$	1.5	Kidney
	Water	$5 \times 10^{-6}$	3	Bone
Argon-41	Air	$4 \times 10^{-8}$	0.5	Whole Body
Californium-249	Water	$4 \times 10^{-6}$	3	Bone
Californium-252	Water	$7 \times 10^{-6}$	1.5	GI (LLI)
	Water	$2 \times 10^{-5}$	3	Bone
Curium-242	Water	$2 \times 10^{-5}$	1.5	GI (LLI)
	Water	$2 \times 10^{-4}$	3	Bone
Curium-244	Water	$7 \times 10^{-6}$	3	Bone
Hydrogen-3 ( $\text{H}_2\text{O}$ )	Air	$2 \times 10^{-7}$	0.5	Whole Body
	Water	$3 \times 10^{-3}$	0.5	Whole Body
Iodine-131	Air	$1 \times 10^{-10}$	1.5	Thyroid
Krypton-85	Air	$3 \times 10^{-7}$	0.5	Whole Body
Neptunium-237	Water	$3 \times 10^{-6}$	3	Bone
Plutonium-238	Water	$5 \times 10^{-6}$	3	Bone
Plutonium-239	Water	$5 \times 10^{-6}$	3	Bone
Strontium-90	Water	$3 \times 10^{-7}$	3	Bone

\*The concentrations and doses are the Radiation Protection Standards (RPS) specified in DOE Manual Chapter 0524 for individuals in uncontrolled areas, except for americium-241 (bone), californium-252 (GI and bone), and curium-242 (bone). Since RPS values for these nuclide-organ combinations are not given in DOE Manual 0524, the concentrations used were one-tenth of the 168 hour occupational values specified by the ICRP.(13)

presence in the environment are attributable to Argonne. For argon-41, tritiated water vapor, and krypton-85 released from reactor stacks, doses were calculated from an atmospheric dispersion model which made use of a source term and meteorological data.

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

Argon-41, hydrogen-3 (in the form of tritiated water), and krypton-85 represent the major gaseous radioactive effluents released from the Laboratory. The concentrations and dose rates were calculated for these three nuclides by a computer program based on an atmospheric dispersion model.<sup>(11,12)</sup> The following parameters were used in the calculations:

- a) annual release rates: argon-41, 0.8 Ci; hydrogen-3, 9Ci; and krypton-85, 5.1 Ci.
- b) meteorological data: the wind velocity data shown in Figure 3.
- c) the usual parameters for stack height, building, wake, plume, momentum, temperatures, etc.

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

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

The population data in Table 2 was used to calculate the cumulative population dose from hydrogen-3, argon-41, and krypton-85. The results are given in Table 21, together with the natural external radiation dose. The natural radiation dose was that measured at the off-site TLD locations, and

it is assumed that this dose is representative of the entire area within an 80 km (50 miles) radius.

TABLE 20

Whole-Body Dose From Airborne Emissions, 1980  
(Millirems)

Nuclide	Pathway	Maximum Perimeter		Maximum Individual	
		Annual	50-Year	Annual	50-Year
Hydrogen-3	Inhalation	0.0004	0.0004	0.0002	0.0002
Argon-41	Submersion	0.0002	0.0002	0.0001	0.0001
Krypton-85	Submersion	0.0004	0.0004	0.0002	0.0002

TABLE 21

## 80 km Whole-Body Population Dose

Source	Man-Rems	
	Annual	50-Year
Hydrogen-3	0.03	0.03
Argon-41	0.004	0.004
Krypton-85	0.02	0.02
Natural	$7.15 \times 10^5$	-

The total argon-41 or krypton-85 released by the Laboratory during 1980 is so small to preclude their estimation by indirect methods such as has been done in the past for argon-41 with TLD measurements. Because the differences between the tritiated water vapor concentrations at the perimeter and off-site locations are within the error of the measurements, and the calculated perimeter concentration is very low, a correlation between measured and calculated concentrations is not possible.

The only location where radionuclides attributable to Argonne operations could be found in off-site water was Sawmill Creek below the waste-water

outfall. The nuclides added to Sawmill Creek by Argonne waste water, their net concentrations in the Creek, and the corresponding dose rates if water at these concentrations were used as the sole water supply by an individual are given in Table 22. The dose rates were all well below the standards for individuals in uncontrolled areas. It should be emphasized that Sawmill Creek is not used for drinking, swimming, or boating. Inspection of the area shows there are few fish in the stream, and they do not constitute a significant source of food for any individual.

TABLE 22

Radionuclide Concentrations and Dose Estimates  
for Sawmill Creek Water, 1980

Nuclide	Conc. (net avg.) $10^{-9}$ $\mu$ Ci/ml	Dose mrem/year	Percent of Standard
Hydrogen-3	118	0.020	0.004
Strontium-90	0.06	0.6	0.020
Neptunium-237	0.010	0.01	0.0003
Plutonium-238	0.0062	0.0037	0.00012
Plutonium-239	0.0110	0.0066	0.00022
Americium-241	0.0057	0.0021 (kidney) 0.0034 (bone)	0.00014 0.00011
Curium-244	0.0022	0.0009 (bone)	0.00003
Californium-249	0.0022	0.0017 (bone)	0.00006

As indicated in Table 9, occasional Creek samples (less than 10) contained traces of curium-242 and/or californium-252 nuclides, but the averages were only slightly greater than the detection limit. The annual dose due to an individual consuming water at these concentrations can be calculated as was done for those nuclides more commonly found in Creek water, but the method of averaging probably overestimates the true concentration. These annual doses range from  $2 \times 10^{-4}$  to  $6 \times 10^{-5}$  mrem/yr for these nuclides.

The EPA<sup>(14)</sup> has recently established drinking water standards based on a dose of 4 mrem/yr. The standards in Table 19 are based on doses of 500 mrem/yr to 3000 mrem/yr, depending on the critical organ. Except for strontium-90,

the approximate percent of the EPA standard can be obtained by multiplying the value in Table 22 by the ratio 500/4 for hydrogen-3 and 3000/4 or 1500/4 (depending on the critical organ) for the actinide elements. Thus, for hydrogen-3, the value becomes 0.5%. For strontium-90, the EPA, using a different model than reference (13), developed a standard of  $8 \times 10^{-9}$   $\mu\text{Ci/ml}$  to produce a dose of 4 mrem/yr and therefore the concentration in Table 22 would give 0.8% of that standard.

#### b. External Penetrating Radiation Dose

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

At location 7I, the fence-line dose from Argonne was about 560 mrem/yr. Approximately 300 m (0.3 mile) south of the fence line (grid 6I), the measured dose averaged  $98 \pm 3$  mrem/yr, at the upper end of the normal range. There are no individuals living in this area. The closest residents are about 1.6 km (1 mile) south of the fence line. At this distance, the calculated dose rate from the Waste Storage Area (based on exponential absorption of the radiation, a decrease in intensity with the square of the distance, and an increase in intensity with distance due to the buildup factor) is 0.02 mrem/yr, if the energy of the radiation was 0.66 MeV, and 0.06 mrem/yr, if the energy was 1.3 MeV. The energy spectrum of the radiation varies with the composition of the stored waste and is not known, so it is necessary to assume an energy to make the calculations. Since cesium-137 and cobalt-60 are common radio-nuclides, the energies of the gamma-rays from these nuclides were used in the calculations.

At St. Patrick's Cemetery (8H), the upper limit of the estimated dose attributable to Argonne was estimated at 5-10 mrem/yr. An individual spending an average of 1 hr/week at this location would receive an annual dose in the range of 0.003 mrem/yr to 0.006 mrem/yr.

In the area north of the site, where the fence-line radiation dose from the cobalt-60 sources in Building 202 was measured at about 30 mrem/yr, the nearest residents are 750 m (0.47 mile) to the north-northwest. The dose at

that location (calculated as described above) was about 0.006 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.<sup>(4)</sup> The latter criterion assumes that the maximum dose to individuals in the sample will not exceed the average by more than a factor of three. Thus, the external penetrating radiation doses to individuals living near the site will not exceed 0.01% of the 500 mrem/yr limit or 0.03% of the "suitable sample" limit. At the fence line, where higher doses were measured, the land is wooded and unoccupied.

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

#### c. Summary

The total dose received by off-site residents was combined from the separate pathways that contribute to this total: argon-41 and krypton-85 immersion dose, hydrogen-3 inhalation dose, and cobalt-60 external radiation dose. The highest dose was about 0.06 mrem/yr to individuals living south of the site if they were outdoors at that location during the entire year. The total annual population dose to the entire area within an 80 km (50 mile) radius is < 0.1 man-rem.

### B. Chemical and Biological Pollutants

#### 1. Air

The environmental air data has been obtained primarily to determine background levels of total suspended particulates (TSP), trace metals (TM), and polynuclear aromatic compounds (PNA) prior to the burning of coal at ANL as a replacement fuel for oil and gas. State and Federal levels for TSP and lead are listed in the Illinois EPA Annual Air Quality report for 1979.<sup>(15)</sup> and a summary appears in Table 23. Standards do not currently exist for other trace metals or PNAs primarily because background data is insufficient. This study establishes background levels for these materials for the current year at ANL and will be invaluable in estimating changes which may occur due to coal burning.

TABLE 23

Summary of National and Illinois  
Ambient Air Quality Standards

Pollutant	Time of Average	Primary Standard	Secondary Standard
Particulate Matter (TSP)	Annual Geometric Mean	75 $\mu\text{g}/\text{m}^3$	60 $\mu\text{g}/\text{m}^3$
	24 Hour	260 $\mu\text{g}/\text{m}^3$	150 $\mu\text{g}/\text{m}^3$
Lead (Pb)	Quarterly Arithmetic Mean	1.5 $\mu\text{g}/\text{m}^3$	Same as Primary

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

Studies of background levels of total suspended particulates were continued during 1980 at four locations situated to study effluents from the Argonne power plant at location 13K (Fig. 1). These samples were also analyzed for iron, copper, zinc, lead, bromine, and strontium using x-ray fluorescence spectrometry and for polynuclear aromatic compound content using liquid chromatography. These compounds are found in all fuels but particularly in coal effluents. The results reported herein represented a 12-month study before coal burning began.

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

Sample location 14N is in the Waterfall Glen Forest Preserve in an area with very low traffic volume. The sampler is located approximately 4 m (13 ft) above the ground and is frequently in a downwind direction of the Argonne power plant. Sample location 12F is west of the power plant and is approximately 3 m (10 ft) above ground. It was necessary to terminate this system

in September since the building upon which it was located was demolished. Sample location 8F is southwest of the power plant, at the Argonne meteorology station, and is 1.3 m (4 ft) above the ground. This sampler also monitors effluent from power plants located southwest of the Laboratory. Sample location 12M is southeast of the power plant and located about 6 m (20 ft) above ground. This is the nearest sampler to the plant. Beginning October 24, 1979, a 24-hour sample was collected every 6 days at location 12M and results are reported for all of 1980. The sample schedule used was that provided by the Illinois Environmental Protection Agency. On December 22, 1980, a second 24-hour sampler was installed at location 8F but insufficient data was obtained in 1980 to report.

Trace elements analyses were performed on the continuous samples collected for total suspended particulates by removing a one centimeter circle and counting it for 10-20 minutes using a cadmium-109 source to excite the x-rays. Studies have shown that distribution of the six elements across the filter is sufficiently homogenous so that counting this segment gives results representative of the entire filter.

The results are shown in Table 24. All values are listed as geometric means since the Federal standard for TSP is expressed in this way. Figure 7 is a log-normal distribution plot of the TSP data from location 12F. The fit of the data to the least-squares line indicates this distribution is a good description of the data. Results for continuous and 24-hr samples met both the primary and secondary standards. Results for lead were well below both primary and secondary standards.

The levels of total suspended particulates and trace elements reported herein are normal constituents of ambient air and represent background levels which will be useful for evaluating changes due to the planned coal burning.

Polynuclear aromatics studies were carried out throughout 1980. The same air particulate samples used for TSP determinations were analyzed. Monthly samples from each location were combined by cutting one-seventh of the sample from the middle of each weekly sample. Homogeneity studies using pyrene indicated uniform distribution.

The combined filters were extracted for eight hours with hexane in a Soxhlet extractor. Initially, the extract was evaporated using a Kuderna-Danish apparatus. For the last three months, the initial extract was diluted

TABLE 24

Environmental Air Data, 1980  
(Concentrations in  $\mu\text{g}/\text{ml}$ )

Location	Constituent	Geometric Mean $\pm$ S.D.	Arithmetic Mean
12F	TSP	53 $\pm$ 1.5	57
14N	TSP	53 $\pm$ 1.4	56
8F	TSP	46 $\pm$ 1.4	48
12M	TSP (Cont.)	49 $\pm$ 1.4	51
12M	TSP (24-hour)	54 $\pm$ 1.9	66
12F	Iron	1.0 $\pm$ 1.6	1.1
14N	Iron	1.0 $\pm$ 1.6	1.2
8F	Iron	0.91 $\pm$ 1.5	1.0
12M	Iron	0.95 $\pm$ 1.6	1.1
12F	Copper	0.30 $\pm$ 1.7	0.35
14N	Copper	0.18 $\pm$ 1.8	0.22
8F	Copper	0.18 $\pm$ 1.6	0.20
12M	Copper	0.17 $\pm$ 1.9	0.20
12F	Zinc	0.11 $\pm$ 2.4	0.17
14N	Zinc	0.07 $\pm$ 3.4	0.16
8F	Zinc	0.08 $\pm$ 2.8	0.13
12M	Zinc	0.08 $\pm$ 3.0	0.14
12F	Lead	0.25 $\pm$ 1.5	0.27
14N	Lead	0.30 $\pm$ 1.6	0.33
8F	Lead	0.22 $\pm$ 1.4	0.24
12M	Lead	0.27 $\pm$ 1.5	0.30
12F	Bromine	0.06 $\pm$ 1.6	0.06
14N	Bromine	0.08 $\pm$ 1.8	0.09
8F	Bromine	0.05 $\pm$ 1.9	0.07
12M	Bromine	0.06 $\pm$ 2.0	0.08
12F	Strontium	0.003 $\pm$ 2.6	0.005
14N	Strontium	0.005 $\pm$ 3.4	0.010
8F	Strontium	0.006 $\pm$ 3.3	0.012
12M	Strontium	0.003 $\pm$ 3.5	0.008

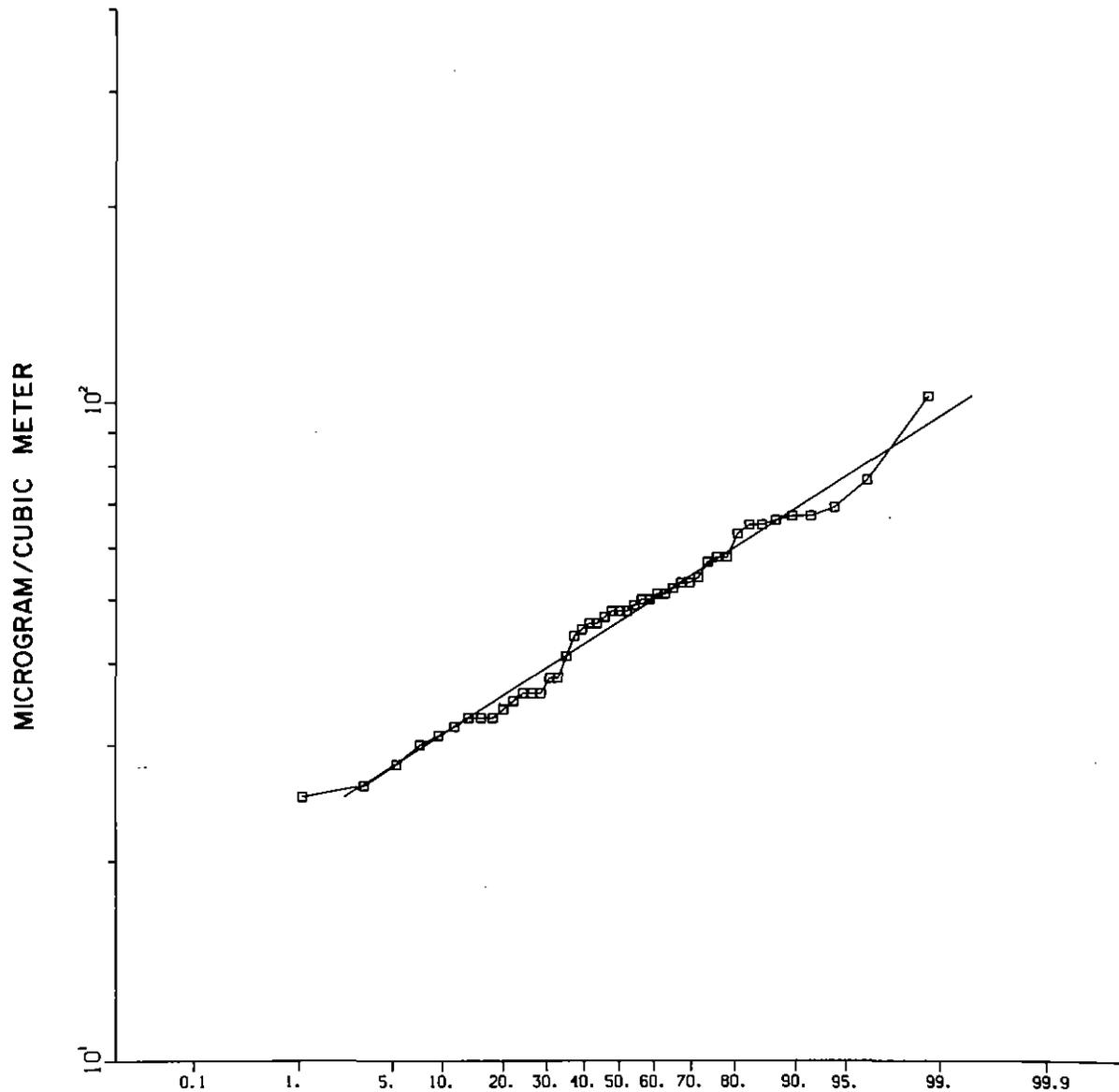


Fig. 7. Log-normal Distribution Plot of Total Suspended Particulates in Air, Location 12F, ANL, 1980. Geometric mean,  $45.9 \mu\text{g}/\text{m}^3$ ; geometric standard deviation,  $1.4 \mu\text{g}/\text{m}^3$ ; arithmetic mean,  $48.1 \mu\text{g}/\text{m}^3$ . The straight line is a least squares fit to the data points.

to volume and a larger aliquot taken for analysis. The results obtained are listed in Table 25.

Some problems exist in the analysis of Benzo- $\epsilon$ -pyrene and Benzo- $\alpha$ -pyrene. Isomerization or other changes can occur in air, in the extraction procedure, and in the actual chromatography. Results listed for these two compounds are the best obtained from interpretation of the data. Where they are both listed, both compounds were clearly present. Where only one is listed, the presence of the other was uncertain. The major objective of this study was to obtain a profile of these compounds for one year before coal burning began. Hence, subsequent data might reveal a different distribution between these two compounds.

The results for the other PNAs are straightforward and more simply interpreted. Penanthracene and pyrene were evident in all samples. There is a strong seasonal variation with all the compounds studied. Results are highest in the winter months and a minimum occurs in the July-September period. This indicates that the PNAs are probably generated by fuel usage. Meteorological conditions must also be considered to determine source location. There was no apparent quantitative difference between results obtained at the four locations during the same time period.

All of these compounds are present in ambient air as both particulate and vapor. The method of collection used is quantitative for particulates but the vapor efficiency is unknown. The literature for distribution between vapor and particulate is confusing, but it is believed that a large portion of the PNAs found in this study were particulate and/or vapor adsorbed on particulate. Studies on a limited basis will be conducted to verify this. It is of interest to note that this is one of the few studies to report data on a monthly basis at several nearby locations.

## 2. Water

The environmental water data contained in this section have been collected in an effort to ascertain Argonne's compliance with State of Illinois regulations on surface stream and effluent water quality, as well as to verify the adequacy of Argonne's effluent pollution controls. The appropriate standards are listed in the Illinois Pollution Control Board Rules and Regulations, Chapter 3.<sup>(16)</sup> Stream Quality Standards appear in Part II and Effluent Quality Standards appear in Part IV of that document.

TABLE 25  
POLYNUCLEAR AROMATIC HYDROCARBONS IN AIR, 1980  
(NANOGRAMS/CUBIC METER)

MONTH	LOCATION	N A P H T H A L E N E	F L U O R E N E	P H E N A N T H R E N E	A N T H R A C E N E	F L U O R A N T H E N E	P Y R E N E	T R I P H E N Y L E N E	C H R Y S E N E	B E N Z I A N T H R A C E N E	B E N Z O   E   P Y R E N E	B E N Z O   A   P Y R E N E
JANUARY	12F	0.20	0.42	<0.05	0.53	0.62	0.14	0.76	0.47	<0.10		
	12H	0.36	0.50	<0.05	0.58	0.66	0.15	0.82	0.68	0.57		
	14N	0.38	0.50	<0.05	0.56	0.72	0.18	0.93	0.86	0.42		
	8F	0.18	0.35	<0.05	0.32	0.48	0.13	0.68	0.36	0.26		
FEBRUARY	12F	<0.10	0.87	<0.05	1.15	1.03	0.31	1.69	1.15			1.51
	12H	<0.10	0.91	<0.05	1.50	3.16	0.25	1.70	1.10			1.28
	14N	<0.10	0.73	0.23	1.03	1.05	0.29	1.79	1.20			1.67
	8F	<0.10	0.78	<0.05	2.02	3.20	0.26	1.60	1.09			1.23
MARCH	12F	<0.10	0.39	<0.05	0.65	1.24	0.12	0.27	0.16			1.79
	12H	<0.10	0.52	<0.05	0.36	1.57	0.17	0.89	0.45			1.83
	14N	<0.10	0.47	<0.05	0.75	1.45	0.17	0.48				1.91
	8F	<0.10	0.33	<0.05	0.69	1.29	0.12	0.32	0.14			1.66
APRIL	12F	<0.10	0.28	<0.05	0.47	0.42	0.12	0.41	<0.10			1.01
	12H	<0.10	0.26	<0.05	0.50	0.45	0.11	0.57	0.40			1.07
	14N	<0.10	0.24	<0.05	0.50	0.42	0.15	0.45	<0.10			1.22
	8F	<0.10	0.25	<0.05	0.43	0.37	0.12	0.52	<0.10			1.11
MAY	12F	<0.10	0.50	0.08	0.54	1.11	0.13	0.77	<0.10			0.35
	12H	<0.10	0.55	0.08	0.51	1.27	0.12	0.87	<0.10			0.36
	14N	<0.10	0.55	0.08	0.61	1.46	0.13	0.89	<0.10			0.50
	8F	<0.10	0.49	0.08	0.56	1.16	0.12	0.75	<0.10			0.34
JUNE	12F	<0.10	0.36	0.06	0.36	0.42	0.14	0.44	<0.10	0.57		
	12H	<0.10	0.35	0.06	0.40	0.42	0.13	0.44	0.13	0.57	0.29	
	14N	<0.10	0.28	0.07	0.35	0.42	0.16	0.50	<0.10	0.70		
	8F	<0.10	0.32	<0.05	0.40	0.42	0.12	0.48	0.13	0.54		
JULY	12F	<0.10	0.36	<0.05	<0.20	0.25	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
	12H	<0.10	0.28	<0.05	<0.20	0.35	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
	14N	<0.10	0.18	<0.05	<0.20	0.28	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
	8F	<0.10	0.29	<0.05	<0.20	0.19	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
AUGUST	12F	<0.10	0.18	0.06	<0.20	0.15	<0.10	0.32	<0.10	0.19	<0.10	<0.10
	12H	0.11	0.24	0.07	0.25	0.26	<0.10	0.37	<0.10	0.24	<0.10	<0.10
	14N	<0.10	0.18	0.07	<0.20	0.13	<0.10	<0.10	<0.10	0.18	<0.10	<0.10
	8F	<0.10	0.18	0.07	<0.20	0.13	<0.10	<0.10	<0.10	0.20	<0.10	<0.10
SEPTEMBER	12H	<0.10	0.24	<0.05	0.49	0.31	<0.10	0.30		<0.10	<0.10	<0.10
	14N	<0.10	0.21	<0.05	0.53	0.31	<0.10	0.35		<0.10	<0.10	<0.10
	8F	<0.10	0.19	<0.05	0.32	0.15	<0.10	0.17		<0.10	<0.10	<0.10
OCTOBER	12H	0.45	0.29	0.54	<0.05	1.83	0.52	0.58	1.78	1.78		1.40
	14N	0.56	0.41	0.38	<0.05	1.35	0.31	0.56	<0.10	2.00		2.00
	8F	0.59	0.29	0.47	<0.05	1.62	0.32	0.41	1.32	1.32		1.20
NOVEMBER	12H	1.04	0.74	0.75	<0.05	0.73	0.58	0.61	1.41	0.25	0.64	0.40
	14N	1.28	0.89	0.87	<0.05	0.73	0.64	0.67	1.42	0.51	0.76	1.10
	8F	1.55	0.64	0.57	<0.05	0.45	0.44	0.52	1.21	0.37	0.58	0.56
DECEMBER	12H	1.59	0.80	1.30	<0.05	2.67	1.21	0.47	0.92	1.44	2.37	2.46
	14N	1.52	0.92	1.32	<0.05	2.93	1.20	0.59	1.10	1.69	2.96	3.24
	8F	1.56		1.08	<0.05	2.57	1.02	0.45	0.83	1.30	2.18	2.38

The testing procedures for water analysis have been described in previous reports in this series. The results of the measurement of chemical constituents are expressed as milligram (mg) or micrograms ( $\mu\text{g}$ ) per liter (l). Averages were calculated as described in Section III.A. Yearly averages are reported with a ( $\pm$ ) limit value. This value is the standard error at the 95% confidence limit and it is calculated from the standard deviation of the yearly average. Only when sample concentrations are random does this value approach the actual distribution occurring at the sampling location. In some instances, it appears that the measurements do represent a natural background concentration and the variation is representative of climatic conditions.

All of the results are compared to the appropriate State standards, which are listed in Table 26. Minimum detectable amounts are included for comparison. The detection limits for the atomic absorption methods represent twice the background variation, which is commonly used for this purpose. Detection limits for ion selective methods are those listed by the manufacturer. The lower limit of detection for x-ray fluorescence is based on the appropriate counting statistics.

As in the past, emphasis has been placed on three areas: Sawmill Creek, a tributary of the Des Plaines River, since this is the principal route for waste water leaving the Argonne site; the study of cooling tower blowdown effluents; and the control of mercury releases in the effluent. A separate section has been included this year to reflect results obtained at NPDES permit locations.

#### a. Effluent Water

The major discharge of waste water from Argonne operations is by way of the waste treatment plant. The water volume from this source was approximately 2.7 megaliters (0.75 million gallons) per day and in 1980 was comprised of 30% sanitary waste water and 70% water from laboratory operations. Laboratory waste is directed to a large holding tank (installed in December 1979) with an overflow which controls the flow at 300-400 gallons/minute. Unlike past years, when smaller holding tanks were discharged only when filled, the flow from this new system occurs over a 24-hour period. Since the sanitary system is fairly constant, the overall output is constant.

Effluents from the waste treatment plant were monitored on a continuous basis during the work week. A flow proportional 24-hour sample of the combined

TABLE 26

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

Constituent	State Standard		Analytical Detection Limit
	Stream	Effluent	
Ammonia Nitrogen (as N)	1.5	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	0.1
Arsenic	1.0	0.25	0.005
Barium	5.0	2.0	0.005
Cadmium	0.05	0.15	0.0004
Chromium(VI)	0.05	0.3	0.003
Chromium(III)	1.00	1.00	-
Copper	0.02	1.0	0.0008
Cyanide	0.025	0.025	0.020
Fluoride	1.4	15	0.02
Iron	1.0	2.0	0.05
Lead	0.1	0.1	0.002
Manganese	1.0	1.0	0.001
Mercury	0.0005	0.0005	0.0001
Nickel	1.0	1.0	0.003
pH	6.5-9.0	5.0-10.0	-
Selenium	1.0	1.0	0.005
Silver	0.005	0.1	0.0002
Sulfate	500	-	1.0
Temperature	-	-	0.1°C
Total Dissolved Solids	1000.	-	-
Zinc	1.0	1.0	0.01

sanitary and laboratory effluent was obtained each day and was analyzed for constituents of interest.

Blowdown water from buildings in the 200 Area is combined and the effluent from this combination was monitored at 12L. Sampling at this location was either once per week grab sampling or continuous monitoring. Grab samples were taken when mechanical difficulties precluded use of the continuous monitoring equipment. Monitoring continued on a once-per-month basis, when water was present, at locations 12F, 14G, 14H, and 14J to determine whether any residual chromium existed. Use of non-chromate corrosion inhibitors was instituted extensively during this period. Since these substitutes contain zinc, this element has been studied for the period of interest at locations 12L and 8J.

The results obtained for chemical constituents in the waste treatment plant effluent are shown in Table 27. All of the average concentrations were at or below the State standards. The average value for mercury was 66% of the State standard and 15% of the samples exceeded this value. Efforts are continuing to lower these levels. The level of hexavalent chromium was usually non-detectable. The levels of trivalent chromium are approaching what we consider to be natural levels.

Levels for arsenic, barium, beryllium, cadmium, fluoride, iron, lead, manganese, nickel, silver, and zinc are similar to past years' results and are probably a measurement of background levels. It is interesting to note that levels of zinc have not changed even though zinc compounds have been used to partially replace the chromate inhibitor. The average level of copper is about the same as last year's value.

Results obtained for the cooling tower effluents are shown in Table 28. Except for 8J, average chromium(VI) concentrations at all stations were below the detection limits. The cooling towers which provide blowdown for the 8J stream were switched in 1980 from zinc-chromate treatment to a combination of organic-zinc compounds. Zinc levels at 8J from September through December are elevated and the cause is being investigated. Zinc levels at the combined effluent, 12L, have remained very low and are believed to be natural. Beginning in April, chromium(VI) contamination could not be found in any of the effluents.

TABLE 27

## Chemical Constituents in Effluent From ANL Treatment Plant, 1980

Constituent	No. of Samples	Concentration ( $\mu\text{g}/\text{l}$ )			Percent of Standard (Avg.)	Percent Exceeding State Standard
		Avg.	Min.	Max.		
Arsenic	54	< 5	-	< 5	< 2.6	0
Barium	54	12 $\pm$ 1	5	37	0.6	0
Beryllium	12	0.06 $\pm$ 0.01	0.03	0.08	-	-
Cadmium	54	0.6 $\pm$ 0.1	< 0.2	1.5	0.43	0
Chromium(III)	54	5 $\pm$ 1	< 3	33	0.5	0
Chromium(VI)	54	< 5	< 5	20	< 1.2	0
Copper	54	35 $\pm$ 7	18	148	3.5	0
Fluoride	54	269 $\pm$ 13	194	430	1.8	0
Iron	54	258 $\pm$ 18	120	422	12.9	0
Lead	54	3 $\pm$ 0.4	< 2	9.6	3.1	0
Manganese	54	23 $\pm$ 2	9	48	2.3	0
Mercury	247	330 $\pm$ 40	50	2970	66.3	15
Nickel	54	13 $\pm$ 3	3	42	1.3	0
pH	247	-	6.3	7.8	-	0
Selenium	54	< 5	-	< 5	< 0.5	0
Silver	54	3 $\pm$ 0.3	1	5	2.6	0
Zinc	54	141 $\pm$ 20	60	362	14.0	0

TABLE 28

## Cooling Tower Effluents, 1980

Constituent	Location *	No. of Samples	Concentration ( $\mu\text{g}/\text{l}$ )			Percent of Standard (Avg.)	Percent Exceeding State Standard
			Avg.	Min.	Max.		
Chromium(VI)	8J	49	1193 $\pm$ 72	10	826	40	16
Zinc	8J	49	987 $\pm$ 370	108	7931	37	99
Chromium(VI)	11L	8	< 10	-	< 10	< 3	0
Silver	11L	8	1.4 $\pm$ 0.4	0.9	3.4	1.4	0
Chromium(III)	11L	8	4.9 $\pm$ 2.3	< 1	8.2	4.9	0
Chromium(VI)	12F	12	< 10	-	< 10	< 3	0
Chromium(VI)	12L	50	< 10	-	< 10	< 3	0
Zinc	12L	50	30.0 $\pm$ 6.0	< 10	158	3.0	0
Chromium(VI)	14G	12	< 10	-	< 10	< 3	0
Chromium(VI)	14H	12	< 10	-	< 10	< 3	0
Chromium(VI)	14J	12	< 10	-	< 10	< 3	0

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

b. National Pollution Discharge Elimination System (NPDES)

The Laboratory currently has two effluent points which are covered by NPDES permits. Discharge point 001 is located at the waste treatment plant and discharge point 002 is located at 13L and contains cooling water from the power plant. The monitoring data obtained as well as the permit conditions are listed in Tables 29 and 30. Sample type and frequency are also listed. Individual values occasionally exceeded the maximum levels and are so indicated in the tables.

c. 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 measurements 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)].

The data from these studies are in Table 31. The average level for ammonia nitrogen upstream is 1.6 times the State of Illinois standard and individual samples exceeded the standard 65% of the time. The downstream sample averaged 1.4 times the State standard and exceeded this standard 37% of the time. All of the samples obtained downstream that exceeded the standard were due to upstream contamination. The dissolved oxygen levels obtained during 1980 were all above the State minimum of 5 mg/l. The total dissolved solids above the outfall exceeded the State standard frequently, and increased the solids content of the downstream sample. Similar results for these constituents were obtained in 1979.

Samples to evaluate the effect of combined sanitary and laboratory waste on the concentrations of chemical constituents in Sawmill Creek were collected by a continuous sampler in the Creek five times per week. These were the same samples taken for radioactivity analyses. The results are summarized in Table 32.

Individual samples for arsenic, beryllium, barium, cadmium, chromium, cyanide, fluoride, lead, manganese, nickel, selenium, silver, and zinc did not exceed the State standards. The levels of mercury averaged 44% of the State limit and exceeded this limit 8% of the time. As in the past, high iron

TABLE 29

 ANL National Pollution Discharge Elimination  
 System Data Summary, Location 001 (7M),\* 1980

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.75	0.43	1.39	MGD	-	Cont.	N/A
	Permit Condition	0.84	-	-		-	Cont.	N/A
B.O.D.	Reported	2.8	-	8.7	mg/l	0	1/7	24 Hr.
	Permit Condition	10	-	15		-	1/7	24 Hr.
Suspended Solids	Reported	4.1	-	30	mg/l	1	2/7	24 Hr.
	Permit Condition	12	-	18		-	1/7	24 Hr.
Fecal Coliform Bacteria	Reported	2	-	22	Number/ 100 ml	0	1/7	Grab
	Permit Condition	200	-	400		-	1/7	Grab
Residual Chlorine	Reported	0.5	0.1	0.7	mg/l	2	1/7	Grab
	Permit Condition	-	0.2	0.75		-	1/7	Grab
Ammonia (N)								
Apr.-Oct.	Reported	0.8	-	2.8	mg/l	0	1/7	Grab
Nov.-Mar.		3.1	-	8.2		1	1/7	Grab
Apr.-Oct.	Permit Condition	2.5	-	4.0	mg/l	-	1/7	Grab
Nov.-Mar.		4.0	-	6.0		-	1/7	Grab
Nitrate (N)	Reported	5.3	-	9.0	mg/l	-	2/7	24 Hr.
	Permit Condition	-	-	-		-	1/30	Grab
Phosphorous (P)	Reported	1.2	-	2.1	mg/l	0	2/7	24 Hr.
	Permit Condition	-	-	-		-	1/30	Grab
pH	Reported	7.1	6.7	7.7	Units	0	2/7	24 Hr.
	Permit Condition	-	6.5	8.5		-	1/7	Grab

\*This location is given in terms of the grid coordinates in Figure 1.

TABLE 30

 ANL National Pollution Discharge Elimination  
 System Data Summary, Location 002 (13L),\* 1980

Parameter	Results Reported Permit Condition	Quantity or Conc.			Units	Number Exceeding	Frequency of Analysis	Type of Sample
		Avg.	Min.	Max.				
Flow	Reported	0.08	0.05	0.121	MGD	0	Cont.	N/A
	Permit Condition	0.1	-	-		-	Cont.	N/A
Suspended Solids	Reported	7.7	-	78	mg/l	1	1/7	Grab
	Permit Condition	12	-	15		-	1/7	Grab
Total Dissolved Solids	Reported	606	-	1330	mg/l	1	1/7	Grab
	Permit Condition	750	-	1200		-	1/7	Grab
Phosphorous (P)	Reported	0.2	-	0.6	mg/l	0	1/7	Grab
	Permit Condition	-	-	-		-	1/30	Grab
Temperature	Reported	65	-	80	°F	-	1/7	Grab
	Permit Condition	-	-	-		-	1/7	Grab
pH	Reported	7.7	6.6	9.1	Units	1	1/7	Grab
	Permit Condition	-	6.5	9.0		-	1/7	Grab

\*This location is given in terms of the grid coordinates in Figure 1.

TABLE 31

## Sawmill Creek - Effect of Sanitary Waste, 1980

Constituent	Location *	No. of Samples	Concentration (mg/l)			Percent of Standard	Percent Exceeding State Standard
			Avg.	Min.	Max.		
Ammonia Nitrogen	7M (up)	46	2.5 ± 0.5	0.1	7.6	164	65
	7M (down)	46	2.1 ± 0.4	0.1	5.3	137	56
Dissolved Oxygen	7M (up)	46	10.7 ± 0.6	6.9	14.4	-	-
	7M (down)	46	10.7 ± 0.6	7.2	14.8	-	-
pH	7M (up)	47	-	7.7	8.5	-	-
	7M (down)	47	-	7.6	8.4	-	-
Sulfate	7M (up)	12	139 ± 29	76	209	28	0
	7M (down)	12	140 ± 29	71	211	28	0
Temperature	7M (up)	46	14.2 ± 2.1	2.1	26.9	-	-
	7M (down)	46	14.6 ± 2.0	2.8	24.4	-	-
Total Dissolved Solids	7M (up)	45	1100 ± 106	583	1920	110	55
	7M (down)	45	1020 ± 83	577	1810	101	53

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

TABLE 32

## Chemical Constituents in Sawmill Creek, Location 7M,\* 1980

Constituent	No. of Samples	Concentration ( $\mu\text{g}/\text{l}$ )			Percent of Standard (Avg.)	Percent Exceeding State Standard
		Avg.	Min.	Max.		
Arsenic	53	< 5	-	< 5	0.5	0
Barium	53	52 $\pm$ 7	21	148	1.0	0
Beryllium	12	0.12 $\pm$ 0.04	0.03	0.30	-	-
Cadmium	53	0.9 $\pm$ 0.1	0.1	2.1	1.8	0
Chromium(III)	53	27 $\pm$ 9	3	167	2.7	0
Chromium(VI)	53	< 3	-	< 3	6	0
Copper	53	34 $\pm$ 7	7	134	172	62
Cyanide	47	< 20	-	< 20	< 80	0
Fluoride	53	369 $\pm$ 28	236	706	26.3	0
Iron	53	1840 $\pm$ 500	269	9000	184	57
Lead	53	15 $\pm$ 3	1	54	15.4	0
Manganese	53	194 $\pm$ 36	43	684	19	0
Mercury	254	222 $\pm$ 37	50	3190	44	8
Nickel	53	14 $\pm$ 3	3	57	1.4	0
pH	253	-	6.5	8.3	-	0
Selenium	53	< 5	-	< 5	0.5	0
Silver	53	2.1 $\pm$ 0.3	0.5	5.3	42.6	2
Zinc	53	104 $\pm$ 21	21	358	10.4	0

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

levels reflect high turbidity conditions and are not related to Argonne releases. The levels of copper exceeded State limits 54% of the time. A previous study of both upstream and downstream levels has shown that water entering above the sampling location contains about 10  $\mu\text{g}/\text{l}$  of copper as compared to the annual average of 34  $\mu\text{g}/\text{l}$  in the downstream sample. The average level of copper in the Argonne effluent, 35  $\mu\text{g}/\text{l}$ , after dilution with Creek water would produce a level of about 15  $\mu\text{g}/\text{l}$ . A study has shown that the elevated copper levels result from entrainment of stream bottom materials rather than from effluent contamination.

d. Des Plaines River

The effect of Sawmill Creek on the Des Plaines River was evaluated by collecting samples at Willow Springs (upstream of Argonne) and at Lemont (downstream of Argonne). These samples were analyzed for total mercury, hexavalent chromium, total iron, and total zinc. The results are in Table 33. Two of the samples had very high levels of suspended material, as evidenced by iron levels of 4.9 and 9.5 mg/l. In no case was there any indication that hexavalent chromium, mercury, or zinc levels were affected by the Argonne effluent.

TABLE 33

Chemical Constituents in the Des Plaines River, 1980

Constituent	Location *	No. of Samples	Concentration (mg/l or $\mu\text{g}/\text{l}$ )		
			Avg.	Min.	Max.
Chromium(VI)	A	12	< 0.01	-	< 0.01
	B	23	< 0.01	-	0.01
Iron	A	12	1.7 $\pm$ 1.6	0.4	9.5
	B	23	1.0 $\pm$ 0.4	< 0.1	4.9
Mercury **	A	12	< 0.1	0.24	0.2
	B	23	< 0.1	< 0.01	< 0.1
Zinc	A	12	0.06 $\pm$ 0.05	0.02	0.31
	B	23	0.04 $\pm$ 0.01	0.02	0.14

\* 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  $\mu\text{g}/\text{l}$ .

## IV. APPENDIX

A. References

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

1. Standards

All of the standards and detection limits for chemical constituents, and some of those for radionuclides and external radiation, are given in the main body of the report. In addition, in Table 34 are collected air and water environmental quality standards and detection limits (minimum detectable amounts) for all radionuclides, and for those materials, for which measurements were made. These standards are the Concentration Guides given in DOE Manual Chapter 0524,<sup>(4)</sup> 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.

TABLE 34

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

Nuclide or Activity	Concentration Guide		Detection Limit	
	Water	Air	Water	Air
Americium-241	$4 \times 10^{-6}$	$4 \times 10^{-12}$	$1 \times 10^{-12}$	$5 \times 10^{-19}$
Antimony-125	-	$9 \times 10^{-10}$	-	$5 \times 10^{-16}$
Argon-41	-	$4 \times 10^{-8}$	-	$1.5 \times 10^{-8}$
Barium-140	$3 \times 10^{-5}$	$1 \times 10^{-9}$	$2 \times 10^{-9}$	$5 \times 10^{-16}$
Beryllium-7	-	$4 \times 10^{-8}$	-	$5 \times 10^{-15}$
Californium-249	$4 \times 10^{-6}$	-	$1 \times 10^{-12}$	-
Californium-252	$7 \times 10^{-6}$	-	$1 \times 10^{-12}$	-
Cerium-141	-	$5 \times 10^{-9}$	-	$5 \times 10^{-16}$
Cerium-144	-	$2 \times 10^{-10}$	-	$1 \times 10^{-15}$
Cesium-137	$2 \times 10^{-5}$	$5 \times 10^{-10}$	-	$5 \times 10^{-16}$
Cobalt-60	-	$3 \times 10^{-10}$	-	$1 \times 10^{-16}$
Curium-242	$2 \times 10^{-5}$	-	$1 \times 10^{-12}$	-
Curium-244	$7 \times 10^{-6}$	-	$1 \times 10^{-12}$	-
Hydrogen-3	$3 \times 10^{-3}$	$2 \times 10^{-7}$	$1 \times 10^{-7}$	$1 \times 10^{-13}$
Iodine-131	$3 \times 10^{-7}$	$1 \times 10^{-10}$	$3 \times 10^{-9}$	$5 \times 10^{-15}$
Krypton-85	-	$3 \times 10^{-7}$	-	-
Neptunium-237	$3 \times 10^{-6}$	-	$1 \times 10^{-12}$	-
Plutonium-238	$5 \times 10^{-6}$	$1 \times 10^{-12}$	$1 \times 10^{-12}$	$1 \times 10^{-19}$
Plutonium-239	$5 \times 10^{-6}$	$1 \times 10^{-12}$	$5 \times 10^{-13}$	$1 \times 10^{-19}$
Ruthenium-103	-	$3 \times 10^{-9}$	-	$5 \times 10^{-16}$
Ruthenium-106	-	$2 \times 10^{-10}$	-	$1 \times 10^{-15}$
Strontium-89	$3 \times 10^{-6}$	$1 \times 10^{-9}$	$2 \times 10^{-9}$	$1 \times 10^{-16}$
Strontium-90	$3 \times 10^{-7}$	$2 \times 10^{-10}$	$2.5 \times 10^{-10}$	$1 \times 10^{-17}$
Thorium-228	-	$2 \times 10^{-13}$	-	$1 \times 10^{-18}$
Thorium-230	-	$3 \times 10^{-13}$	-	$1 \times 10^{-18}$
Thorium-232	-	$1 \times 10^{-12}$	-	$1 \times 10^{-18}$
Uranium-234	-	$4 \times 10^{-12}$	-	$1 \times 10^{-19}$
Uranium-235	-	$4 \times 10^{-12}$	-	$1 \times 10^{-19}$
Uranium-238	-	$5 \times 10^{-12}$	-	$1 \times 10^{-19}$
Uranium - natural *	$4 \times 10^{-5}$	$4 \times 10^{-12}$	$2 \times 10^{-10}$	$2 \times 10^{-17}$
Zirconium-95	-	$1 \times 10^{-9}$	-	$5 \times 10^{-16}$
Alpha **	$3 \times 10^{-6}$	$1 \times 10^{-10}$	$2 \times 10^{-10}$	$2 \times 10^{-16}$
Beta **	$1 \times 10^{-7}$	$1 \times 10^{-13}$	$1 \times 10^{-9}$	$5 \times 10^{-16}$

\* Concentration Guides converted from the "special curie" used in DOEM 0524 to the standard curie.

\*\* The Concentration Guides for unknown mixtures depend, within the range given, on whether certain radionuclides are known to be present in concentrations less than 0.1 of their CGs, and the sum of the fraction of the CGs for all such nuclides is less than 0.25. For most total alpha and beta results given in this report, the largest uncontrolled CG is applicable.

For water, the standard selected was for the soluble form of the radionuclide; for air, the standard for the insoluble form was selected (except for iodine-131, for which the soluble form was chosen as a more conservative standard).

## 2. Detection Limits

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

## 3. Quality Assurance Program

### a. Radiochemical Analysis and Radioactivity Measurements

All nuclear instrumentation is calibrated with standard sources obtained from the U. S. National Bureau of Standards (NBS), if possible. If NBS standards were not available for particular nuclides, standards from the Amersham-Searle Co. were used. The equipment is usually checked on a daily basis with secondary counting standards to insure proper operation. Samples are periodically analyzed in duplicate or with the addition of known amounts of a radionuclide to check precision and accuracy. In addition, standard and intercomparison samples distributed by the DOE Environmental Measurements Laboratory (EML), the Quality Assurance Branch of the U. S. Environmental Protection Agency (EPA-QA) at Las Vegas, and the International Atomic Energy Agency (IAEA) are analyzed regularly. The EPA-QA intercomparison program consists of analyzing a variety of samples, at intervals selected by the participant, to which known amounts of various radionuclides have been added by the EPA laboratory. A summary of all the EPA-QA samples analyzed in this laboratory in 1980 is shown in Table 35. The DOE Environmental Measurements Laboratory Quality Assurance Program (DOE-EML-QAP) is a semi-annual distribution of four or five different sample matrices containing various combinations of radionuclides.<sup>(17)</sup> Results of our participation in this program during 1980 are given in Table 36. In the table, the comparison is made between the EML value, which is the result of replicate determinations by that Laboratory, and the value obtained in our laboratory. More than 95% of all the intercomparison samples received were analyzed for the radionuclides for which results were requested. To assist in judging the quality of the results, typical errors for our analyses are

TABLE 35

## Summary of EPA-QA Samples, 1980

Type of Sample	Analysis	Number Analyzed	Avg. Difference From Added
Air Filter	Total Alpha	1	53%
	Total Beta	1	15%
	Strontium-90	1	7%
	Cesium-137	1	5%
Milk	Potassium-40	2	6%
	Strontium-89	1	11%
	Strontium-90	1	4%
	Iodine-131	1	13%
	Cesium-137	2	13%
Water	Total Alpha	5	50%
	Total Beta	5	16%
	Hydrogen-3	1	2%
	Chromium-51	1	4%
	Cobalt-60	3	7%
	Zinc-65	2	6%
	Strontium-89	3	13%
	Strontium-90	2	6%
	Ruthenium-106	2	4%
	Iodine-131	1	9%
	Cesium-134	3	20%
	Cesium-137	3	3%
	Radium-226	2	20%
	Radium-228	2	19%
	Total Uranium	1	2%
Plutonium-239	3	22%	

TABLE 36

## Summary of DOE-EML-QAP Samples, 1980

Nuclide	Average Difference From EML Value				
	Air Filters	Water	Soil	Tissue	Vegetation
Hydrogen-3	-	11% (2)	-	-	-
Beryllium-7	16% (2)	-	-	-	-
Sodium-22	-	7% (1)	-	-	-
Potassium-40	-	-	9% (2)	3% (2)	3% (2)
Chromium-51	-	1% (1)	-	-	-
Manganese-54	3% (1)	-	-	-	-
Cobalt-57	-	10% (1)	-	-	-
Cobalt-60	3% (2)	4% (2)	4% (1)	8% (2)	9% (2)
Zinc-65	-	-	-	-	-
Strontium-89	-	5% (2)	-	-	-
Strontium-90	11% (2)	2% (1)	5% (2)	13% (2)	16% (2)
Zirconium-95	0% (1)	-	-	-	-
Antimony-125	46% (1)	-	-	-	-
Cesium-134	7% (1)	13% (1)	-	-	-
Cesium-137	13% (1)	5% (2)	6% (2)	6% (2)	12% (2)
Cerium-141	9% (1)	-	-	-	-
Cerium-144	10% (2)	-	-	-	-
Radium-226	-	-	9% (2)	-	-
Thorium-228	-	-	10% (2)	-	-
Uranium-234	5% (1)	4% (1)	-	-	-
Uranium-238	3% (1)	3% (1)	-	-	-
Plutonium-238	-	17% (1)	21% (2)	27% (2)	-
Plutonium-239	5% (2)	27% (1)	13% (2)	27% (2)	1% (2)
Americium-241	22% (2)	16% (2)	-	-	-

The figure in parentheses is the number of samples.

2-50%, the error in the EML results is 1-30% (depending on the nuclide and the amount present), and the error in the added amount in the EPA-QA samples is 2-5% (our estimate).

b. Penetrating Radiation

Our laboratory participated in the Fifth International Comparison of Environmental Dosimeters conducted in 1980 by the DOE Environmental Measurements Laboratory and the School of Public Health, University of Texas (Houston), but the results are not yet available. The results for the Fourth such study in 1979, as reported in ANL-80-29,<sup>(18)</sup> are as follows: Participants supplied three sets of each type of dosimeter they wished to evaluate. One set was given a three-month field exposure to natural radiation in Houston; one set was exposed in the laboratory to a known (but unrevealed) quantity of radiation approximately equivalent to a three-month natural field exposure; and one set was exposed to a known quantity of radiation in a higher range. The latter two exposures were to a radium-226 source. After exposure, the dosimeters were returned to the participant for measurement. The natural radiation field exposure was measured by an EML high-pressure ion chamber for comparison. The results, in mR, were:

	<u>Field Exposure</u>	<u>Laboratory Exposure</u>	
		<u>Low</u>	<u>High</u>
School of Public Health	14.1 ± 0.7	12.2 ± 1.2	45.8 ± 4.6
ANL Value			
calcium fluoride	12.9 ± 2.4	11.7 ± 2.0	44.1 ± 5.9
lithium fluoride	14.6 ± 2.7	10.7 ± 2.7	43.7 ± 2.9

The uncertainties listed are the standard deviations as estimated by the School of Public Health for their values and by us for our results. Our standard deviations are relatively high because substantial corrections were required for irradiation of the dosimeters during storage and during air shipment. All results agree with the standard or accepted values within the measurement error. The most important comparison is for the field exposures. Here the differences between the ion-chamber result and our results were 1.2 mR ± 2.5 mR (8.5% ± 17.7%) for calcium fluoride and 0.5 mR ± 2.8 mR (3.6% ± 20%) for lithium fluoride.

c. Chemical Analysis

With each set of atomic absorption analyses, standard amounts of trace

metals were analyzed in blank solutions at concentrations corresponding to 50 and 100% of the current State standards. Recoveries were determined by comparing these results to results obtained by analyzing stream and effluent samples to which identical concentrations were added. Average recoveries ranged from about 85-100%, with a standard deviation of about 10%. Recovery studies were also performed for ammonia nitrogen, fluoride, and hexavalent chromium analyses, and similar results were obtained. All trace metals in air were referenced to Urban Air Particulate SRM 1648 from the Bureau of Standards.

d. Sampling, Sample Storage, etc.

Many factors enter into an overall quality assurance program other than the analytical quality control discussed above. Representative sampling is of prime importance. The continuous water sampler in Sawmill Creek provides a representative sample for a critical sampling location since the concentration of pollutants in the waste water may vary appreciably during each 24-hour period.

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

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

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