

**ENVIRONMENTAL MONITORING AT
ARGONNE NATIONAL LABORATORY
ANNUAL REPORT FOR 1974**

by

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



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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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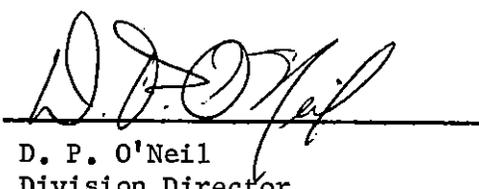
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ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1974 are presented and discussed. To evaluate the effect of Argonne operations on the environment, measurements were made for a variety of radionuclides in air, surface water, Argonne effluent water, soil, grass, benthos, and milk; for a variety of chemical constituents in surface and Argonne effluent water; and of the environmental penetrating radiation dose. Sample collections and measurements were made at the site boundary and off the Argonne site for comparison purposes. Some on-site measurements were made to aid in the interpretation of the boundary and off-site data. The results of the program are interpreted in terms of the sources and origin of the radioactive and chemical substances (natural, fallout, Argonne, and other) and are compared with accepted environmental quality standards. The potential radiation dose to off-site population groups is also estimated.

I. INTRODUCTION

A. General

This report is prepared to provide the AEC/ERDA and the public with information on the level of radioactive and non-radioactive pollutants in the environment of Argonne National Laboratory and on the amounts of pollutants, if any, added to the environment as a result of Argonne operations. The report follows the guidelines given in AEC/ERDA Manual Chapter 0513.⁽¹⁾ The Laboratory conducts a continuous environmental monitoring program on and near the Argonne site whose primary purpose is to determine the magnitude, origin, and identity of any radioactive or potentially toxic chemical substances present in the environment. Of special interest is the detection of any such material released to the environment by Argonne. One important

function of the program is to verify the adequacy of Argonne's effluent pollution controls.

Argonne is a multi-disciplinary research and development laboratory with several principal objectives. It carries out a broad program of research activities in biology, chemistry, physics, and mathematics, it serves as an important center for the design and development of nuclear reactors, and carries out studies on environmental pollution and energy resource problems. The reactor effort is devoted largely to development of the fast-breeder power reactor. Some of the energy-related problems are improvements in the utilization of coal for power production, the development of electric batteries for vehicles and off-peak energy storage, and studies on solar energy. Environmental research studies include a Great Lakes radioecology program, which is mainly concerned with the effects of effluents from nuclear and fossil power plants on Lake Michigan, and studies on the dispersion and behavior of airborne pollutants under various meteorological conditions. Almost all of the work at the Laboratory is of an unclassified nature.

The principal nuclear facilities at the Laboratory are a 5 MW heavy-water cooled and moderated general-purpose research reactor (CP-5) fueled with fully-enriched uranium; a 200 kW light-water cooled and moderated biological research reactor (JANUS) fueled with fully-enriched uranium; one critical assembly, or zero power reactor (ZPR-9), that is fueled at various times with plutonium, uranium, or a combination of the two; the Argonne Thermal Source Reactor (ATSR), a 10 kW research reactor fueled with enriched uranium; a 12.5 GeV proton accelerator, the Zero Gradient Synchrotron (ZGS); a 60-inch cyclotron; several other charged particle accelerators; plutonium chemical and metallurgical laboratories; and several hot cells and laboratories designed for work with irradiated fuel elements and with multicurie quantities of the actinide elements.

B. Description of Site

Argonne National Laboratory (Illinois site) occupies the central 1,700 acres of a 3,740-acre tract in Du Page County, 27 miles southwest of downtown Chicago, and 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 2,040-

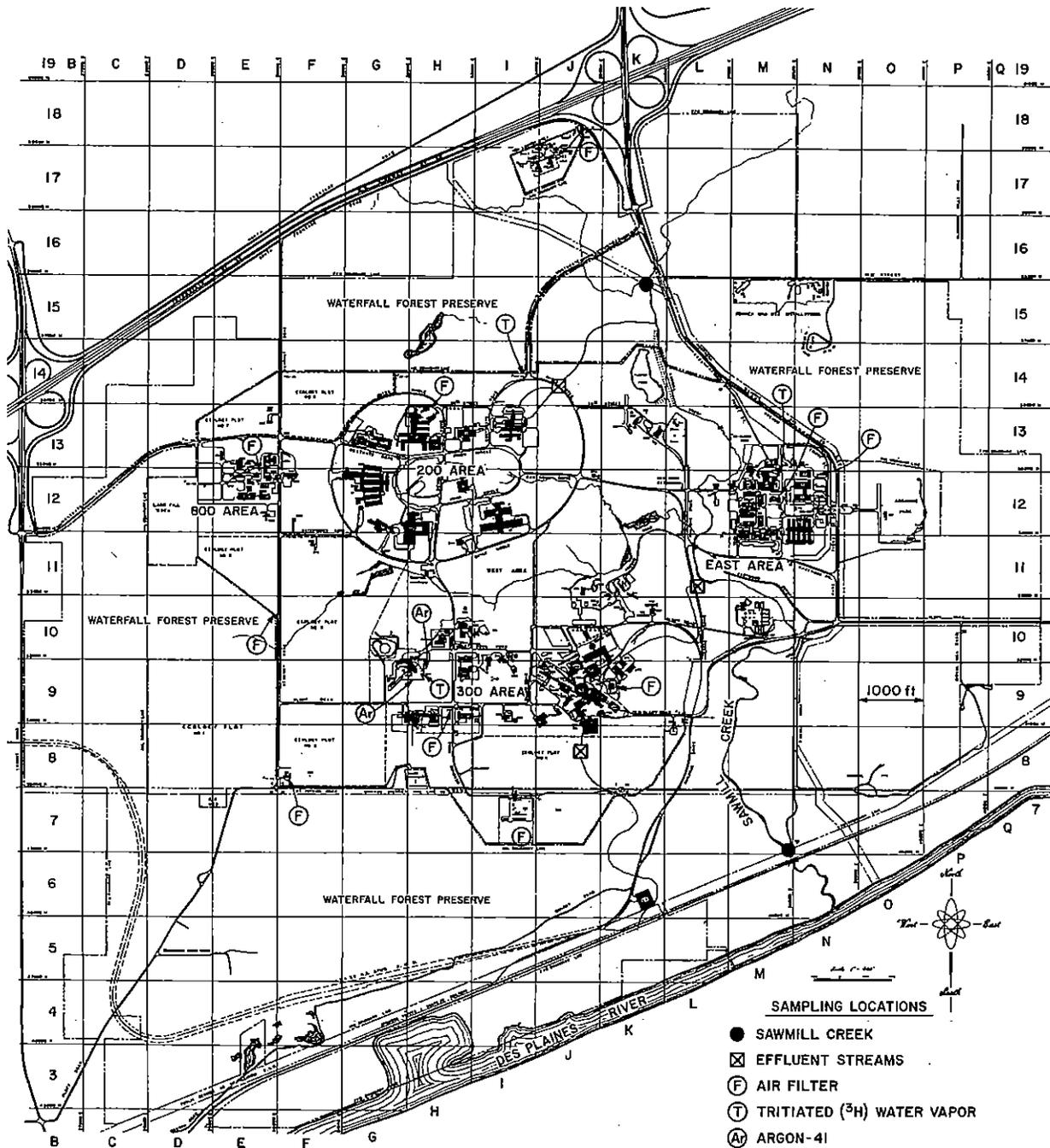


Fig. 1. Sampling Locations at Argonne National Laboratory

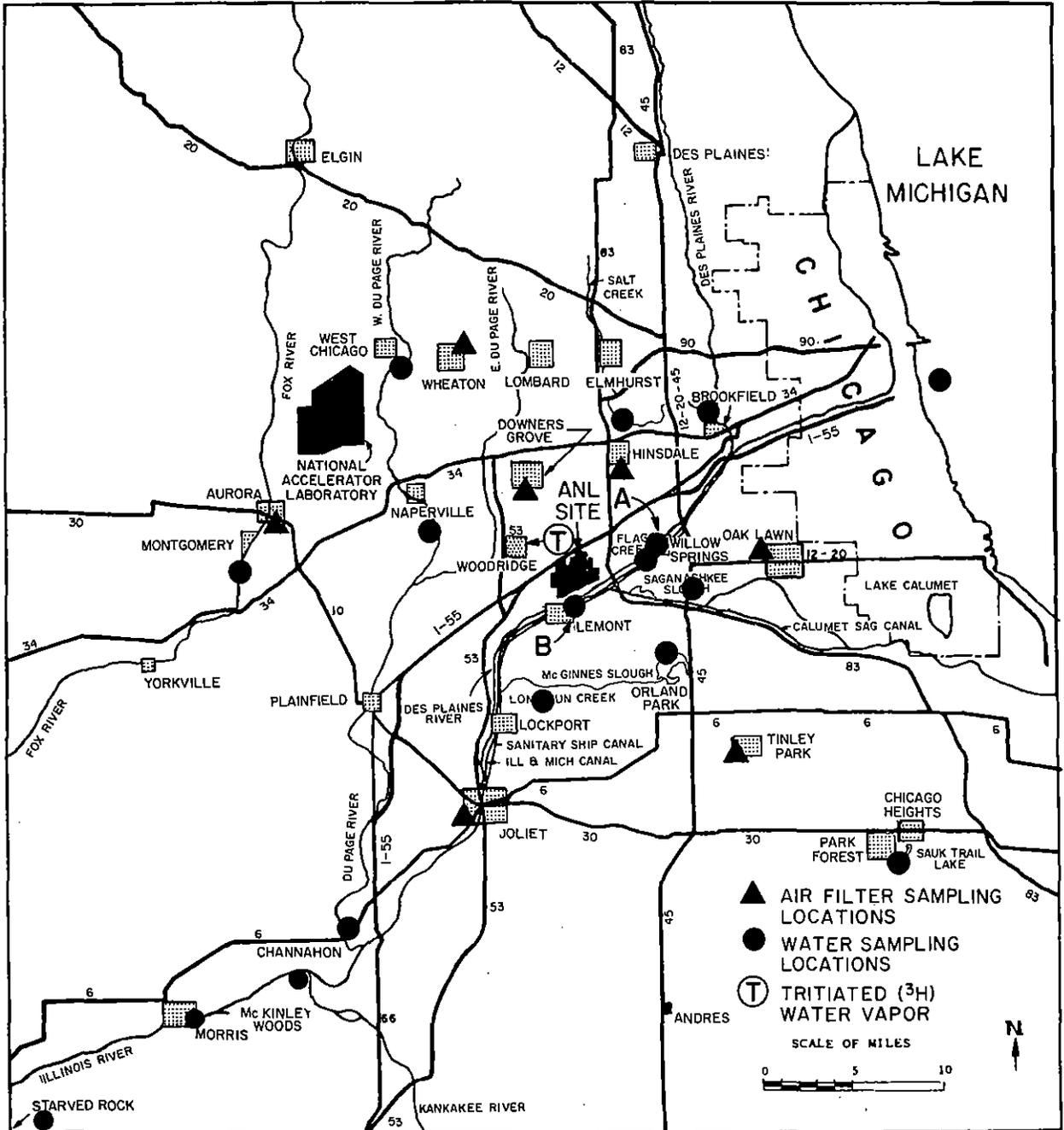


Fig. 2. Sampling Locations near Argonne National Laboratory

acre area surrounding the site was formerly Argonne property, but was deeded to the Du Page County Forest Preserve District in 1973 for their use as a public recreational area, nature preserve, and demonstration forest. The new site boundary is essentially the same as the site security fence referred to in earlier reports and shown on earlier maps.⁽²⁾

The terrain is gently rolling, partially-wooded, former prairie and farmland. The grounds contain a number of ponds and small streams, the principal one being Sawmill Creek, which runs through the site in a southerly direction and enters the Des Plaines River about 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 site. This river flows southwest until it joins the Kankakee River about 30 miles southwest of the Laboratory to form the Illinois River.

The largest topographical feature is the Des Plaines River channel, about one mile wide. This channel contains both the River and the Chicago Sanitary and Ship Canal. Their presence extends the uninhabited area about one mile south of the site. The elevation of the channel surface is 578 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 650 feet above sea level at the top. The land then slopes gradually upward reaching the average site elevation of 725 feet above sea level at 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 50-60 feet. The remaining portion of the site changes in elevation by no more than 25 feet in a distance of 500 horizontal 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 best described as a lake-moderated upper Mississippi Valley climate. A summary of the meteorological data collected on the site from 1950 to 1964 is available.⁽³⁾ Similar data have been collected since 1964, but have not been tabulated and published. The 15-year

summary, however, gives an adequate sample of the climate, and the following information is taken from this reference. The average monthly and annual wind roses are shown in Figure 3. The wind roses are essentially polar coordinate plots where the lengths of the radii represent percentage frequency of wind speeds in classes of 4 to 12 mph, 4 to 24 mph, and greater than 4 mph. The direction of the radii represents the direction from which the wind blows. For example, on the 15-year summary rose for January in Figure 3 the three points plotted on the spoke extending horizontally to the left represent the wind speed distribution when the wind is blowing from the ten-degree sector, 266 to 275 degrees. These points indicate that about 3.8 percent of the wind observations in January were from the west in the speed interval 4 to 12 mph, about 5.1 percent in the speed interval 4 to 24 mph, and about 5.2 percent in the speed interval greater than 4 mph. In the figure the curves for the latter two intervals are almost identical, and are not distinguishable on the drawing. The number 12.67 in the center of the rose represents the percent of observations of wind speed less than 4 mph in all directions.

The roses show that the predominant winds are from the southwest and west on the average, but seasonal variations are apparent. The winds are primarily westerly in January and February. In the spring, the northeast lake breeze becomes evident and continues through much of the summer. The warm southwesterly winds, brought up by the high pressure areas in the southeastern U. S., begin later in the spring, and become dominant in the fall. The winds are sufficiently variable so that monitoring for airborne releases must be carried out in all directions from the site. The daily average wind speed at 19 feet above ground level varied from 5 mph in August to 10 mph in March. Gusts exceeded 50 mph about once a year. The mean and median wind speeds for the 15-year period were 7.6 and 7.0 mph, respectively.

The air temperature data show a typical seasonal variation. Daily temperatures average from the low 20's in the winter to the 70's in the summer. The extreme temperatures recorded were -20°F and 100°F .

The annual precipitation ranged between 20 and 43 inches and averaged 31.5 inches during the 1950-1964 period. Precipitation is heaviest in the spring and summer, when moist air is carried up from the Gulf of Mexico, and is lowest during the winter months. The monthly average varied from 1.3 inches in December to 4.3 inches in July.

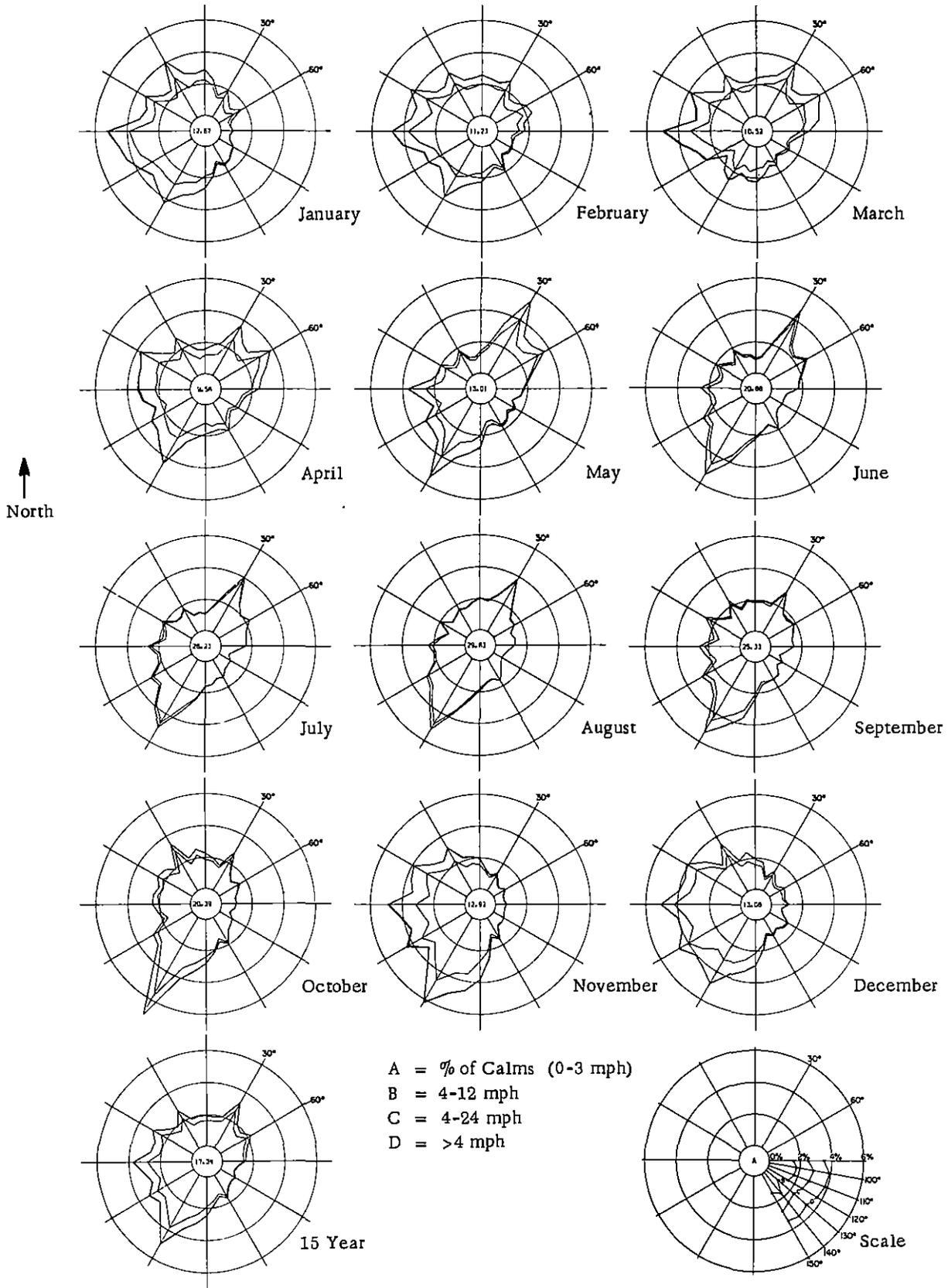


Fig. 3. Wind Roses Based on All Hourly Observations. January 1950-December 1964. 19-foot Level.

D. Population

The area around Argonne has exhibited a large growth in suburban, ex-urban, and rural housing. Large areas of farmland have been converted into housing. Two housing developments border the northeast portion of the new Forest Preserve that was formerly part of the site: Wellington Woods, an apartment house complex with 4,500 residents, and Timberlake, a development of single family homes with 2,500 residents at the time of the 1970 census.

The directional 50-mi population distribution for the area with Argonne as the center is shown in Table 1. The table was prepared from the 1970 U. S. census figures by the Environmental Information System Office at Oak Ridge National Laboratory. Some of the figures in the table for the first four miles have been changed, based on inspection of the actual area and of maps, since they did not appear to be current. The 1970 census of communities and townships in the area, and a map showing their relation to the Argonne site, are given in the preceding report in the series.⁽⁴⁾ Only the 50-mi population distribution is included in this report, since it is used for the population dose calculations later in this report.

E. Land and Water Use

The principal stream that drains the site is Sawmill Creek. This Creek was formerly an intermittent stream, responding in flow rate largely to precipitation runoff. It now carries effluent water continuously from a municipal sewage treatment plant located a few miles north of the site and operating at about 2 million gallons per day. In addition, the residential development in the area has resulted in the collection and channeling of additional runoff water in the Creek. Treated sanitary and laboratory waste water from Argonne is discharged into Sawmill Creek at location 7M in Figure 1. Water from this source varies between about 0.5 and 1 million gallons per day. In 1974, the water flow in the Creek averaged about 7.7 million gallons per day above the Argonne waste-water outfall; Argonne waste-water effluent averaged 0.74 million gallons per day. Three streams that carry blowdown discharge water from cooling towers are considered effluents since they leave the site boundary. These are shown in Figure 1. One stream from the 200 Area leaves the site at location 14J and enters the Creek at 15K; one from the 300 Area enters Sawmill Creek at location 11L (the Creek crosses the site

TABLE 1

Incremental Population Data in the Vicinity of ANL, 1970

Distance, Miles	0 - 1	1 - 2	2 - 3	3 - 4	4 - 5	5-10	10-20	20-30	30-40	40-50
<u>Direction</u>										
N	0	120	0	2592	310	33929	181886	257828	93833	185403
NNE	0	120	361	3420	4423	37082	336097	538710	112647	0
NE	0	120	3021	1175	1148	28825	803173	1071375	0	0
ENE	0	120	0	0	668	27658	737736	333866	0	0
E	0	120	0	15	0	26235	493591	252869	13483	23013
ESE	0	0	0	0	0	9044	197110	273190	290476	40540
SE	0	0	0	0	0	9606	39025	99748	20498	8965
SSE	0	0	377	0	0	924	10503	6437	13373	18151
S	0	200	1100	1542	0	2621	12007	1224	26042	38362
SSW	0	200	1848	3047	0	16232	104383	7940	14396	7063
SW	0	130	0	0	0	10260	24359	4470	13718	7223
WSW	0	130	0	462	0	5535	4201	1795	5560	9992
W	0	130	0	3387	4955	951	38397	16101	16475	7371
WNW	0	170	0	0	0	18162	82536	7700	3981	49352
NW	0	170	0	0	9311	11431	36959	63029	13843	11456
NNW	0	170	1424	1186	3341	33270	92276	110962	85107	63205
Total	0	1900	8131	16826	24156	271765	3194239	3047244	723432	470096
Cumulative Total	0	1900	10031	26857	51013	322778	3517017	6564261	7287693	7757789

boundary shortly thereafter); another stream from the 300 Area crosses the site boundary at location 7K and soaks into the ground south of the site.

Sawmill Creek and the Des Plaines River above Joliet (about 13 miles southwest of Argonne) receive very little recreational or industrial use. A very few people fish in these waters downstream from Argonne, and some duck hunting takes place on the Des Plaines River. Water from the Chicago Sanitary and Ship Canal is used for some industrial purposes, such as hydroelectric generators and condensers, and for irrigation at the state prison near Joliet. It is also used as secondary cooling water by Argonne. The Canal, which carries Chicago effluent water, is used for industrial transportation and some recreational boating. Near Joliet, the River and Canal are combined into one waterway, which continues until it forms the Illinois River about 30 miles southwest of Argonne. The Dresden Nuclear Power Station complex is located at the confluence of the Kankakee, Des Plaines, and Illinois Rivers. The plant uses water from the Kankakee for cooling, and discharges the water into the Illinois River. The first use of water for drinking is an indirect one at Peoria, on the Illinois River about 140 miles downstream from Argonne, where River water is used to replenish ground water supplies by infiltration. In the vicinity of the Laboratory, only subsurface water (from all aquifers, shallow and deep) is used for drinking purposes. Lake Michigan water is used by communities closer to the Lake.

The principal recreational area near Argonne is Waterfall Glen Forest Preserve, which surrounds the site as described earlier (Figure 1). Most of this area has received little use thus far. It was available for hiking and skiing in 1974. Its development and increased utilization by the public is expected. Sawmill Creek flows through the south portion of the Preserve on its way to the Des Plaines River. This region of the Preserve (formerly named Rocky Glen) is used principally for picnicking, hiking, and overnight camping by youth groups. During 1973, the latest year for which data is now available, approximately 7,800 individuals used the Preserve for camping and approximately 15,000 for picnics. East and southeast of Argonne and the Des Plaines River are located several large forest preserves of the Cook County Forest Preserve District. The preserves include the two sloughs shown in Figure 2, McGinnis and Saganashkee, as well as other smaller lakes. These areas are used for picnicking, boating, fishing, and hiking. A small park

located in the eastern part of the Argonne site (12-0 in Figure 1) is for the use of Argonne employees only.

II. SUMMARY

This is one in a series of reports prepared to provide the AEC/ERDA and the public with information on the level of radioactive and chemical pollutants in the environment and on the amounts of such substances, if any, added to the environment as a result of Argonne operations. The previous report in this series is ANL-8078. Included in this report are the results of measurements obtained in 1974 for a number of radionuclides in air, surface water, soil, grass, benthic materials, and milk; for a variety of chemical constituents in water; and for the external penetrating radiation dose. Since the primary means of dispersal of pollutants are by air and water, the environmental monitoring program has concentrated on these media.

Total alpha and beta activities, beryllium-7 (produced principally by cosmic-rays), fission and activation products, plutonium, thorium, and uranium were measured in air-filter samples at the site perimeter and off the site. All of the off-site and perimeter samples contained only activities from natural sources and nuclear test detonations. The detectable activities from nuclear tests ranged from 0.00002% of the Concentration Guide (CG)* for cerium-141 to 1.3% of the CG for mixed beta activity. The average beta activity, 1.3×10^{-13} $\mu\text{Ci/ml}$,** was about three times higher than during 1973, as a result of fallout from the Chinese atmospheric nuclear tests of June 26, 1973, and June 17, 1974.

The plutonium-239 concentrations in air averaged, respectively, 34×10^{-18} and 27×10^{-18} $\mu\text{Ci/ml}$ at the site perimeter and off the site, about three times the 1973 values, and well within the range reported by the AEC/ERDA New York Health and Safety Laboratory for plutonium from weapons

*The hazard due to a given concentration of a radioactive nuclide is usually 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. Atomic Energy Commission (now U. S. Energy Research and Development Administration) in Chapter 0524 of the AEC/ERDA Manual. The pertinent CG's are listed in the Appendix, Section IV.B.

**The radioactivity units are described in Section III.

tests. The monthly variations indicated a "spring maximum" in stratospheric fallout similar to that observed in the total beta and gamma-ray activities. The results indicate that the airborne plutonium was from nuclear tests and there is no evidence that any of the plutonium originated at Argonne. The averaged plutonium-239 concentration was equivalent to 0.003% of the CG.

Argon-41 and hydrogen-3 represent the major airborne radioactivity released from the Laboratory. The argon-41 concentration and corresponding radiation dose at the site boundary was less than the detection limit (50% of the CG) of the present sampling system, but was calculated from an atmospheric dispersion model to be 5×10^{-10} $\mu\text{Ci/ml}$ and 6.1 mrem/yr in the predominant north-northwest wind direction. These values are 1.2% of the CG and 1.2% of the non-occupational dose limit to individuals (500 mrem/yr) for uncontrolled areas. The calculated dose at 2.1 km NNE, where the closest full-time residents live, is 3.5 mrem/yr. These values are consistent with penetrating radiation dose measurements made at the site perimeter. The measured hydrogen-3 concentration at the site perimeter averaged about 8×10^{-12} $\mu\text{Ci/ml}$, which is 0.004% of the CG and about three times greater than the off-site concentration. The corresponding dose is 0.02 mrem/yr.

Argonne waste water is discharged into Sawmill Creek, and this stream was sampled above and below the site to evaluate the effect of Argonne operations on its radioactive content. The nuclides (for which analyses were made) added to the Creek in the waste water, and the percent CG of their average concentration, were hydrogen-3, 0.06%; strontium-90, 0.4%; neptunium-237, 0.006%; plutonium-239, 0.0004%; and americium-241, 0.0002%. The resulting dose to an individual using water at these concentrations as his sole source of drinking water would be about 11.5 mrem/yr. These nuclides are also present in fallout, and about 6 mrem/yr of the total dose would result from strontium-90 produced in weapons testing.

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

Plutonium concentrations in soil showed the same general range and average at the site perimeter and off the site. The average deposition of

plutonium-239 was $1.6 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ at the site perimeter and $2.7 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ off the site. The corresponding plutonium-238 averages were $0.10 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ and $0.15 \times 10^{-3} \mu\text{Ci}/\text{m}^2$. The plutonium content of grass was similar to that found in previous years and was about a factor of 10^4 less than soil from the same location. The results are within the range reported by other workers for fallout from test detonations and the plutonium found in soil and grass is attributed to this source. The plutonium content of samples from the beds of a number of streams near the site was measured. The stream beds contained from 1×10^{-9} to $25 \times 10^{-9} \mu\text{Ci}/\text{g}$ of plutonium-239, a range found in previous years to be normal for fallout plutonium in such materials. The concentrations of uranium, thorium, and some gamma-ray emitters measured in soil, plant, and benthic materials were normal.

Milk from a dairy farm near the Laboratory was collected monthly and analyzed for several fission products and hydrogen-3. The short-lived fission products, iodine-131 and barium-140, were not detected. Hydrogen-3 concentrations ranged from $< 200 \times 10^{-9} \mu\text{Ci}/\text{ml}$ (the detection limit) to $320 \times 10^{-9} \mu\text{Ci}/\text{ml}$, similar to the range found in surface water. Positive strontium-89 concentrations were measured in June and July. The strontium-90 and cesium-137 concentrations increased slightly compared to 1973. The consumption of one liter of milk per day at the average concentrations would have resulted in a total dose of about 1% of the annual limit, principally from strontium-90. These radionuclides are present in milk due to fallout, and are not related to Argonne operations.

Measurements of penetrating radiation were made at several locations at the site boundary and off the site. The off-site readings averaged 99 mrem/yr, with a standard deviation of 8.5 mrem/yr, very similar to the previous years' averages, and normal for the area. The readings at the site boundary were in the normal range except for two locations along the south boundary. At location 7I (Figure 1) the total dose was 283 mrem/yr, 184 mrem/yr above normal, as a result of radiation from an on-site temporary storage facility for radioactive waste. At location 8H, the average value, 110 ± 14 mrem/yr, was in the normal range, but some of the individual readings were slightly elevated. An upper limit for the Argonne-contributed dose at this location is about 11 mrem/yr. In this case several sources contributed to the above-normal values -- the waste storage facility and radiation-producing equipment

in buildings at location 9GH. The elevated doses at 7I and 8H were 37% and 2% of the non-occupational standard for critical individuals, about one-half of the corresponding 1973 results. These locations were very rarely occupied, and there were no individuals at the south boundary receiving these doses. The calculated dose rate to the nearest residents, about one mile south of the boundary, was about 0.02 mrem/yr, 0.004% of the standard for individuals in uncontrolled areas and well within the 5 mrem/yr "as low as practicable guidelines".

The levels of chemical constituents in the Argonne waste water and in Sawmill Creek were measured and results were compared to the standards adopted by the State of Illinois in 1972.

The oxygen balance in the Argonne waste water effluent was within acceptable levels, as were total coliform values. Average concentrations of each of the chemical constituents in the effluent were at or below the State of Illinois limits. The average level of total mercury equalled the State limit, and represents a reduction of three to four times from the previous two years. This reduction was a result of improved controls instituted in 1974. Improved analytical sensitivity for lead, cadmium, and silver has resulted in these elements being measured at the very low natural levels for lead and cadmium, and for silver at the very restrictive State limit.

The average values in Sawmill Creek for oxygen balance and chemical constituents were below the State of Illinois limits. This marks the first time in several years that levels of ammonia nitrogen and dissolved oxygen have consistently been in compliance with these regulations, and represents improved waste water treatment upstream of the Laboratory. The mercury and hexavalent chromium average levels were about 75% of the 1973 averages.

III. MONITORING RESULTS

A. Radioactivity

The radioactivity of the environment was determined by measuring the concentrations of radioactive nuclides in naturally-occurring materials and by measuring the external penetrating radiation dose. Sample collections and measurements were made principally at the site perimeter and off the site for comparison purposes. Some on-site results are also reported when they are useful in interpreting perimeter and off-site results. Since radioactivity is usually spread by air and water, the sample collection program has concentrated on these media. In addition, soil, plants, milk, 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 (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 the bulk 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 positive value. The averages that are obtained by using these two methods under the conditions indicated are believed to give an adequate picture of the true average activity at locations where the activity not only varied greatly, but was at times not detectable.

Average values are usually accompanied by a plus-or-minus (\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 error in the average of repeated measurements on the same or identical samples. Since many of the variations observed

in environmental radioactivity are not random but occur for specific reasons (e.g., nuclear testing), samples collected from the same location at different times are not replicates. The more random the variation in activity at a particular location, the closer the confidence limits will represent the actual distribution of values at that location. The averages and confidence limits should be interpreted with this in mind. When a \pm 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. Unless otherwise specified, the standards used in this report are the Concentration Guides (CG) and annual dose limits (Radiation Protection Standards) given in AEC/ERDA Manual Chapter 0524.⁽⁵⁾ The pertinent CGs as well as the detection limits are given in the Appendix, Section IV.B. Although the CGs apply to concentrations above natural levels, the percent of CG is sometimes given in this report for activities that are primarily of natural origin for comparative purposes. Such values are enclosed in parentheses to indicate this.

1. Air

The radioactivity of particulate matter was determined by collecting and analyzing air-filter samples. The sampling locations are shown in Figures 1 and 2. Separate collections were made for radiochemical analyses and for alpha, beta, and gamma counting. The latter measurements were made on samples collected continuously on asbestos-cellulose filter paper at eight locations at the Argonne site perimeter* and at five locations off the site. At one location the filter paper was changed daily; at all other locations the filter papers were changed at weekly intervals. The daily samples record short-term changes in radioactivity, while the weekly samples are used to compare perimeter and off-site activities. Measurements are 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

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

indicate radioactivity released by Argonne if the differences are greater than the error in sampling and counting. Such results require investigation to determine the cause of the difference. The error is between 5 and 20% for most results, but approaches 100% at the detection limit.

The total alpha and beta activities in the individual weekly samples are summarized in Table 2. 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 3. The gamma-ray detector is a shielded 35 cm³ lithium-drifted germanium diode, calibrated for each gamma-ray emitting nuclide given in Table 3.

The total alpha and beta activities were very similar at the site perimeter and off the site and no significant differences between locations were found. The alpha activities, principally due to naturally-occurring nuclides, averaged the same as 1973 and were in their normal range. About 50% of the gamma-ray activity, and a smaller fraction of the beta activity, was due to beryllium-7, principally produced in the stratosphere by cosmic-ray interactions. The remaining activity was primarily fission and activation products from nuclear test detonations. The average beta activity for the year, 1.3×10^{-13} $\mu\text{Ci/ml}$, was almost four times higher than the 1973 average due to increased fallout from nuclear tests. The usual increase in the rate of stratospheric fallout in the spring is shown by the monthly variations in the beryllium-7 concentrations and intermediate half-life fission products (such as zirconium-95-niobium-95). These fission products can be attributed principally to the June 26, 1973, atmospheric nuclear test of the People's Republic of China. Beginning in July, short-lived fission products (barium-140-lanthanum-140 and iodine-131) were measured at all locations and their appearance correlates well with the Chinese test of June 17, 1974.

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

TABLE 2

Total Alpha and Beta Activities in Air-Filter Samples, 1974*

Month	Location	No. of Samples	Alpha Act. (10^{-15} $\mu\text{Ci/ml}$)			Beta Act. (10^{-12} $\mu\text{Ci/ml}$)		
			Av.	Min.	Max.	Av.	Min.	Max.
January	perimeter	35	1.7	0.9	3.6	0.056	0.042	0.076
	off-site	25	2.5	1.0	11.	0.058	0.033	0.087
February	perimeter	31	1.9	0.8	3.0	0.087	0.062	0.12
	off-site	20	2.2	1.2	4.3	0.093	0.068	0.13
March	perimeter	35	2.0	0.7	4.0	0.13	0.063	0.19
	off-site	19	2.2	0.9	3.7	0.13	0.055	0.20
April	perimeter	34	2.7	1.0	5.6	0.26	0.11	0.40
	off-site	22	2.7	1.3	5.0	0.26	0.11	0.39
May	perimeter	34	2.1	0.7	4.5	0.25	0.16	0.32
	off-site	22	1.9	0.8	3.2	0.25	0.20	0.31
June	perimeter	34	2.4	0.7	6.2	0.21	0.11	0.31
	off-site	17	2.0	1.0	3.1	0.23	0.089	0.34
July	perimeter	35	3.6	1.1	7.0	0.19	0.098	0.27
	off-site	22	3.1	1.4	6.5	0.19	0.076	0.30
August	perimeter	36	2.8	1.2	5.8	0.11	0.064	0.15
	off-site	21	2.6	1.3	5.7	0.12	0.085	0.28
September	perimeter	34	3.1	1.5	8.1	0.066	0.039	0.10
	off-site	22	2.1	0.9	4.0	0.076	0.024	0.44
October	perimeter	35	2.6	1.2	5.5	0.060	0.030	0.21
	off-site	22	2.4	1.3	4.4	0.052	0.032	0.078
November	perimeter	34	1.8	0.8	3.3	0.057	0.034	0.12
	off-site	16	1.7	0.6	3.4	0.053	0.039	0.091
December	perimeter	35	2.1	1.0	6.1	0.071	0.054	0.094
	off-site	19	2.1	1.1	4.9	0.069	0.048	0.12
Annual Summary	perimeter	412	2.4 \pm 0.3	0.7	8.1	0.13 \pm 0.05	0.030	0.40
	off-site	247	2.3 \pm 0.2	0.6	11.	0.13 \pm 0.05	0.024	0.44

*These results were obtained by measuring the samples four days after they were collected in order to avoid counting the natural radioactivity due to radon and thoron decay products. This activity is normally present in the air and disappears within four days by radioactive decay.

TABLE 3

Gamma-Ray Activity in Air-Filter Samples, 1974
(concentrations in 10^{-15} $\mu\text{Ci/ml}$)

Nuclide	Location	January	February	March	April	May	June	July	August	September	October	November	December	Annual Average
Antimony-125	perimeter	< 0.5	0.8	1.2	2.5	2.7	2.5	2.7	1.4	0.9	0.5	< 0.5	< 0.5	1.3 \pm 0.6
	off-site	< 0.5	1.0	1.5	3.3	2.6	3.2	2.0	1.1	0.6	0.5	< 0.5	< 0.5	1.4 \pm 0.7
Barium-140- Lanthanum-140	perimeter	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	3.1	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.7
	off-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	2.8	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.7
Beryllium-7	perimeter	58	96	94	105	85	82	124	76	78	70	67	63	83 \pm 11
	off-site	56	95	104	108	88	91	99	75	71	63	63	56	81 \pm 11
Cerium-141	perimeter	0.6	1.0	1.0	1.3	0.8	0.5	1.3	1.6	1.1	0.6	1.0	1.0	1.0 \pm 0.2
	off-site	0.6	1.0	1.1	1.4	0.8	0.5	1.2	1.4	0.9	0.6	0.9	0.9	0.9 \pm 0.2
Cerium-144	perimeter	3.7	10	18	43	48	43	42	19	10	6.1	6.4	6.6	21 \pm 10
	off-site	3.6	11	19	47	48	47	36	19	10	6.1	6.5	6.3	22 \pm 10
Cesium-137	perimeter	< 0.5	1.0	1.7	3.9	4.4	4.1	4.5	2.2	1.4	0.8	0.8	0.7	2.1 \pm 0.9
	off-site	0.6	1.0	1.6	4.3	4.5	5.1	3.9	2.3	1.3	0.8	0.8	0.6	2.2 \pm 1.0
Cobalt-60	perimeter	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
	off-site	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Iodine-131	perimeter	< 5	< 5	< 5	< 5	< 5	< 5	< 6	< 5	< 5	< 5	< 5	< 5	< 5
	off-site	< 5	< 5	< 5	< 5	< 5	< 5	< 6	< 5	< 5	< 5	< 5	< 5	< 5
Manganese-54	perimeter	< 0.1	< 0.1	< 0.1	0.4	0.4	0.4	0.3	< 0.1	0.1	< 0.1	< 0.1	< 0.1	< 0.2
	off-site	< 0.1	< 0.1	< 0.1	0.5	0.5	0.5	0.3	0.2	< 0.1	< 0.1	< 0.1	< 0.1	< 0.2
Ruthenium-103	perimeter	1.4	2.6	3.0	4.1	2.8	1.7	2.4	1.6	1.0	0.5	1.3	1.6	2.0 \pm 0.6
	off-site	1.4	2.6	3.0	4.4	2.9	1.8	2.7	1.5	0.9	0.6	1.2	1.6	2.0 \pm 0.6
Ruthenium-106- Rhodium-106	perimeter	2.5	5.4	10	24	26	23	24	10	6.1	3.4	3.3	3.0	12 \pm 6
	off-site	2.5	6.6	9.9	26	28	28	20	10	5.4	2.8	3.7	2.8	12 \pm 6
Zirconium-95- Niobium-95	perimeter	12	29	42	78	68	48	40	20	9.0	10	15	16	32 \pm 13
	off-site	13	30	44	85	70	52	34	14	8.4	12	10	17	32 \pm 15

The results obtained for the air-filter samples are further summarized in Table 4 in order to compare the average concentrations with the concentration guides. The percent CG for beryllium-7 and alpha activity are placed in parentheses since, as noted earlier, all or a large part of these activities are naturally-occurring, and the CGs apply to concentrations above natural background. However, regardless of source, all concentrations were well below the CGs.

Samples for radiochemical analysis were collected at locations 12N and 9H (Figure 1) and off the site in Downers Grove (Figure 2). Collections were made on a polystyrene filter medium. The total air volume filtered for the monthly samples was about 25,000 or 60,000 m³, depending on the sampler. Samples were ignited at 600°C to remove organic matter and prepared for analysis by vigorous treatment with hot hydrochloric, hydrofluoric, and nitric acids. This treatment has been found in our laboratory to solubilize plutonium that had been ignited at 1000°C. Plutonium and thorium were separated on an anion exchange column, electrodeposited, and their isotopic compositions determined by alpha spectrometry. Chemical recoveries were monitored by adding known amounts of plutonium-242 and thorium-234 tracers prior to ignition. Alpha spectrometry cannot distinguish between plutonium-239 and plutonium-240, and although in the remainder of the report, including the tables, only plutonium-239 is mentioned, it should be understood that the alpha activity due to the less abundant plutonium-240 isotope is also included.

An aliquot of the sample solution was also analyzed for uranium by a standard fluorophotometric procedure. The uranium concentrations are obtained in mass units by this technique. Since the uranium is believed to have the normal isotopic composition, the results were converted to activity units for comparison purposes by using the specific activity of natural uranium, 0.675×10^{-6} $\mu\text{Ci/g}$. In addition, the column effluent from the anion exchange separation was analyzed for radiostrontium by standard radiochemical procedures. The results are given in Table 5.

The average plutonium-239 concentrations increased at all locations by about a factor of three from 1973. The concentrations are similar to those reported by other investigators. The results of the ERAMS⁽⁶⁾ program for the first quarter of 1974 averaged 18.5×10^{-18} $\mu\text{Ci/ml}$ for plutonium-239, while our results averaged 20.9×10^{-18} $\mu\text{Ci/ml}$ for the same period. The plutonium-239

TABLE 4

Average Radioactivity in Air-Filter Samples, 1974
(concentrations in 10^{-12} $\mu\text{Ci/ml}$)

Activity	Detection [*] Limit	Concentration [*] Guide (CG)	Concentration	Percent CG
Antimony-125	0.0005	900	0.0014	0.00016
Barium-140- Lanthanum-140	0.0005	1000	< 0.0008	< 0.00008
Beryllium-7	0.005	40000	0.082	(0.0002)
Cerium-141	0.0005	5000	0.0010	0.00002
Cerium-144	0.001	200	0.022	0.011
Cesium-137	0.0005	500	0.0022	0.00044
Cobalt-60	0.0001	300	< 0.0001	< 0.00003
Iodine-131	0.005	100	< 0.005	< 0.005
Manganese-54	0.0001	1000	< 0.0002	< 0.00002
Ruthenium-103	0.0005	3000	0.002	0.00007
Ruthenium-106- Rhodium-106	0.001	200	0.012	0.0006
Zirconium-95- Niobium-95	0.0005	1000	0.032	0.0032
Alpha	0.0002	10	0.0024	(0.024)
Beta	0.0005	10	0.13	1.3

*The CGs are those given in AEC/ERDA Manual Chapter 0524 for uncontrolled areas. Additional information on the CGs and detection limits is given in the Appendix, Section IV.B.

TABLE 5
Plutonium, Thorium, Uranium, and Strontium Concentrations in Air-Filter Samples, 1974
(concentrations in 10^{-18} $\mu\text{Ci}/\text{ml}$)

Month	Location*	Plutonium-239 ¹	Plutonium-238	Thorium-232 ²	Thorium-230	Thorium-228	Uranium ³	Strontium-90	Strontium-89
January	12N	17 ± 1	0.4 ± 0.2	10 ± 1	24 ± 2	10 ± 1	66 ± 6	420 ± 160	2370 ± 510
	9H	11 ± 1	0.4 ± 0.1	25 ± 1	60 ± 2	21 ± 1	140 ± 17	-	-
	off-site	12 ± 1	0.4 ± 0.3	5 ± 1	12 ± 1	5 ± 1	41 ± 3	320 ± 50	2280 ± 150
February	12N	24 ± 2	0.6 ± 0.3	16 ± 1	35 ± 2	15 ± 1	98 ± 7	620 ± 400	4740 ± 1770
	9H	18 ± 1	0.6 ± 0.2	34 ± 1	70 ± 1	38 ± 1	310 ± 31	350 ± 130	8430 ± 820
	off-site	24 ± 2	0.9 ± 0.4	26 ± 2	52 ± 2	27 ± 2	150 ± 10	1020 ± 70	5460 ± 320
March	12N	30 ± 2	0.7 ± 0.3	10 ± 1	21 ± 1	11 ± 1	65 ± 4	1510 ± 150	6190 ± 930
	9H	26 ± 1	0.8 ± 0.2	20 ± 1	43 ± 1	24 ± 1	428 ± 86	1240 ± 160	6230 ± 1090
	off-site	26 ± 1	0.3 ± 0.2	8 ± 1	13 ± 1	8 ± 1	39 ± 4	1030 ± 200	5260 ± 1480
April	12N	71 ± 3	1.3 ± 0.1	20 ± 2	37 ± 2	19 ± 2	70 ± 6	4460 ± 290	12170 ± 1230
	9H	76 ± 2	1.7 ± 0.1	46 ± 2	82 ± 2	45 ± 2	146 ± 12	1760 ± 220	5190 ± 1050
	off-site	64 ± 4	1.4 ± 0.4	15 ± 2	28 ± 2	14 ± 2	44 ± 4	3920 ± 320	10310 ± 1390
May	12N	55 ± 2	1.4 ± 0.4	11 ± 1	25 ± 2	11 ± 1	-	4600 ± 120	9160 ± 370
	9H	56 ± 1	1.0 ± 0.1	22 ± 1	46 ± 2	30 ± 1	-	1730 ± 70	3770 ± 220
	off-site	39 ± 1	0.7 ± 0.2	8 ± 1	17 ± 1	9 ± 1	-	3920 ± 120	7360 ± 360
June	12N	63 ± 3	1.0 ± 0.4	16 ± 1	27 ± 2	16 ± 1	-	3570 ± 280	4760 ± 1600
	9H	83 ± 2	2.2 ± 0.3	36 ± 2	65 ± 2	42 ± 2	-	6600 ± 150	4080 ± 2120
	off-site	49 ± 3	0.8 ± 0.4	-	-	-	-	4020 ± 20	-
July	12N	49 ± 3	1.1 ± 0.5	31 ± 3	51 ± 4	31 ± 3	-	3890 ± 130	2800 ± 860
	9H	58 ± 2	1.6 ± 0.3	35 ± 3	62 ± 4	45 ± 4	-	6170 ± 280	6540 ± 2040
	off-site	41 ± 3	0.8 ± 0.5	-	-	-	-	3140 ± 100	2100 ± 1310
August	12N	37 ± 2	0.6 ± 0.3	14 ± 2	23 ± 2	14 ± 2	75 ± 10	1880 ± 270	3710 ± 630
	9H	38 ± 1	1.0 ± 0.2	28 ± 2	48 ± 3	27 ± 2	184 ± 12	3370 ± 130	6250 ± 390
	off-site	41 ± 4	0.4 ± 0.7	8 ± 2	15 ± 2	11 ± 2	27 ± 20	2890 ± 130	3750 ± 540
September	12N	20 ± 2	0.4 ± 0.3	10 ± 1	17 ± 1	10 ± 1	30 ± 18	1370 ± 120	2400 ± 350
	9H	25 ± 1	0.7 ± 0.2	24 ± 1	45 ± 2	27 ± 1	180 ± 16	1970 ± 380	5210 ± 1060
	off-site	9 ± 2	0.4 ± 0.5	4 ± 1	9 ± 1	6 ± 1	6 ± 8	1050 ± 40	1700 ± 120
October	12N	5 ± 2	< 0.1	9 ± 1	17 ± 1	9 ± 1	85 ± 5	550 ± 65	970 ± 190
	9H	13 ± 1	0.4 ± 0.2	24 ± 1	51 ± 1	34 ± 1	164 ± 9	-	-
	off-site	5 ± 1	0.3 ± 0.2	4 ± 1	9 ± 1	5 ± 1	64 ± 4	550 ± 55	1190 ± 150
November	12N	9 ± 1	0.6 ± 0.2	11 ± 1	24 ± 2	12 ± 1	105 ± 6	690 ± 30	2130 ± 100
	9H	19 ± 1	0.6 ± 0.2	12 ± 1	29 ± 1	13 ± 1	92 ± 5	770 ± 10	2990 ± 30
	off-site	7 ± 1	< 0.1	11 ± 1	20 ± 1	11 ± 1	73 ± 4	625 ± 135	1590 ± 440
December	12N	10 ± 1	0.6 ± 0.2	8 ± 1	16 ± 1	9 ± 1	72 ± 4	560 ± 65	3605 ± 190
	9H	11 ± 1	0.5 ± 0.2	12 ± 1	28 ± 1	11 ± 1	79 ± 4	635 ± 25	4720 ± 70
	off-site	8 ± 1	0.3 ± 0.1	7 ± 1	14 ± 1	7 ± 1	64 ± 4	375 ± 25	1955 ± 70
Monthly Average	12N	32 ± 13	0.7 ± 0.2	14 ± 4	26 ± 6	14 ± 4	74 ± 14	2010 ± 950	4580 ± 1860
	9H	36 ± 15	1.0 ± 0.3	26 ± 6	52 ± 9	30 ± 7	191 ± 75	2460 ± 1410	5340 ± 1000
	off-site	27 ± 11	0.6 ± 0.2	10 ± 4	19 ± 8	10 ± 4	56 ± 27	1900 ± 880	3900 ± 1750
Percent CG	12N	0.003	0.00007	(0.0001)	(0.009)	(0.007)	(0.002)	0.007	0.002
	9H	0.004	0.0001	(0.0003)	(0.017)	(0.015)	(0.005)	0.008	0.002
	off-site	0.003	0.00006	(0.0001)	(0.006)	(0.005)	(0.001)	0.006	0.001

¹Plutonium-240 is included (see text).

²On-site locations are given in terms of the grid coordinates in Figure 1.

³The concentrations in units of $\mu\text{g}/\text{m}^3$ can be obtained by multiplying the value in $\mu\text{Ci}/\text{ml}$ by 1.48×10^{12} for uranium and by 9×10^{12} for thorium-232. The amounts of the other thorium isotopes in mass units are negligible in comparison to thorium-232.

concentrations are also similar to those reported by the ERDA New York Health and Safety Laboratory fallout program^(7,8) for samples collected away from nuclear installations. The latter results ranged from 9 to 79×10^{-18} $\mu\text{Ci/ml}$ from January to June, 1974, at New York, N. Y. and Sterling, Va., and showed the same general spring increase observed here. Since the results in Table 5 are in this same range, it may be concluded that the airborne plutonium is due to fallout from nuclear tests.

The thorium and uranium concentrations in Table 5 are in the same range found during 1973 and are considered to be of natural origin. Similar uranium concentrations have been reported by other investigators.⁽⁶⁾ The percent of CG for the averages is included for completeness; the values are placed in parentheses since the concentrations are considered to be background levels. The amounts of thorium and uranium in a sample were directly related to the mass of material collected on the paper and the concentrations per unit mass collected are quite similar to those found in soil samples. This indicates that the bulk of these elements in the air is due to resuspension of soil. The amount of plutonium in the air sample contributed by soil, if the resuspended soil has the same plutonium concentration as the first 5 cm on the ground, is about 2% of the total plutonium in the air sample. During 1973, this percentage was 5-10%, which indicates increased plutonium fallout in 1974, and is consistent with the increased air concentration observed.

The monthly variation in the concentrations of both strontium-89 and strontium-90 correlate with the results for the other fission products in Table 3. The concentrations are considered to be due to fallout, primarily from the June 26, 1973, atmospheric nuclear test mentioned previously, and are consistent with levels reported by other sources.⁽⁷⁾ There is no indication that any strontium in the air samples originated from Argonne operations. The spring increase is evident only in the plutonium and strontium activities, indicating their stratospheric-fallout origin, and not in the thorium and uranium concentrations.

Air sampling for argon-41 and hydrogen-3 was conducted on-site near the CP-5 reactor (Building 330, 9H) because this reactor is the principal source of these nuclides at the Laboratory, and knowledge of their concentrations at this location aids in interpreting and validating the calculated argon-41

concentrations and the very low, but measurable, hydrogen-3 concentrations at the boundary. Argon-41 was collected by filling an evacuated "Marinelli-type" container with air. Samples were obtained twice during each week of reactor operation and measured by gamma-ray spectrometry. Each sampling consisted of two "grab" samples, one taken 50 m east of the reactor and one downwind from the reactor at a point favorable for detection of argon-41. The results are given in Table 6. Because of the timing and location of the sample collection, the actual average concentration in the vicinity of the reactor was less than the values in the table. The 1974 results were about 70% of the 1973 average and the same as 1972, probably due to the randomness of the sampling procedure. The average and maximum concentrations of 1.2×10^{-7} and 1.6×10^{-6} $\mu\text{Ci/ml}$, respectively, are 6% and 80% of the CG for controlled areas. In this case, comparison with the CGs for controlled areas is appropriate since the sampling is conducted on the site and the argon-41 originated in the CP-5 reactor.

Hydrogen-3 (tritiated) water vapor in air was collected by adsorption on silica gel and was measured by counting the water in a liquid scintillation counter. The results of continuous air sampling at a permanent station 50 m east of CP-5 are given in Table 7. As will be seen, normal or background hydrogen-3 concentrations in air during 1974 averaged about 2.7×10^{-12} $\mu\text{Ci/ml}$. Near the CP-5 reactor, the results averaged 1.9×10^{-10} $\mu\text{Ci/ml}$, which is about 70% less than in 1973, and 35% higher than in 1972. The argon-41 averages over the past three years varied in a similar manner.

Hydrogen-3 concentrations at several additional locations are given in Table 8. The 12M location (1900 m east-northeast of the CP-5 reactor) may be considered a perimeter sample for CP-5. The average hydrogen-3 concentrations at both perimeter locations were very close to those measured in 1973. The data show correlation with wind direction and indicate that relative to the reactor, dilution to the background level occurs before reaching the site boundary in directions other than that from which the wind is blowing. A time-weighted average directional correlation study of the 12M results show that when the wind was from the southwest and the hydrogen-3 concentration near the reactor was sufficiently high, measurable increases above the normal background level were observed. The average and maximum perimeter concentrations were equivalent to 0.004% and 0.033% of the CG, respectively.

TABLE 6

Argon-41 Concentrations in Air, 300 Area, 1974

Month	No. of Samples	Conc. (10^{-9} $\mu\text{Ci/ml}$)			Percent CG*		
		Av.	Min.	Max.	Av.	Min.	Max.
January	14	245	< 15	1000	12	< 0.8	50
February	14	205	< 15	970	10	< 0.8	48
March	16	135	< 15	1490	7	< 0.8	74
April	12	150	< 15	815	8	< 0.8	41
May	16	165	< 15	995	8	< 0.8	50
June	12	20	< 15	110	1	< 0.8	6
July	16	150	< 15	1620	8	< 0.8	81
August	-	-	-	-	-	-	-
September	4	45	< 15	105	2	< 0.8	5
October	20	30	< 15	320	2	< 0.8	16
November	10	130	< 15	375	6	< 0.8	19
December	18	50	< 15	205	2	< 0.8	10
Annual Summary	152	120 \pm 45	< 15	1620	6	< 0.8	81

*This is the CG for controlled areas. See text.

TABLE 7

Hydrogen-3 Concentrations in Air, 300 Area, 1974

Month	No. of Samples	Conc. (10^{-12} μ Ci/ml)			Percent CG*		
		Av.	Min.	Max.	Av.	Min.	Max.
January	9	135	44	280	0.068	0.022	0.14
February	8	175	12	545	0.087	0.006	0.27
March	9	160	3.0	500	0.080	0.002	0.25
April	9	190	25	1055	0.095	0.012	0.53
May	8	250	5.5	800	0.125	0.003	0.40
June	9	240	17	925	0.120	0.009	0.46
July	9	160	29	715	0.080	0.015	0.36
August	9	80	9.2	275	0.040	0.005	0.14
September	8	270	16	1520	0.135	0.008	0.76
October	9	100	9.1	350	0.050	0.005	0.17
November	9	245	8.9	920	0.123	0.004	0.46
December	9	265	20	690	0.133	0.010	0.35
Annual Summary	105	189 \pm 38	3.0	1520	0.095	0.002	0.76

*This is the percent CG for uncontrolled areas, which is used to allow easy comparison with perimeter and off-site concentrations. The percent CG for controlled areas is 25 times less than the values in the table.

TABLE 8

Hydrogen-3 Perimeter and Off-Site Concentrations, 1974

Month	Location	No. of Samples	Conc. (10^{-12} $\mu\text{Ci/ml}$)			Percent CG*		
			Av.	Min.	Max.	Av.	Min.	Max.
January	perimeter (12M)	9	9.6	1.6	22	0.0048	0.00082	0.011
	off-site	2	1.4	0.74	2.2	0.00072	0.00037	0.0011
February	perimeter (12M)	8	2.1	0.42	5.4	0.0010	0.00021	0.0027
	off-site	2	2.9	1.2	4.6	0.0014	0.00095	0.0023
March	perimeter (12M)	9	3.2	0.95	7.2	0.0016	0.00048	0.0036
	off-site	2	1.3	0.74	1.9	0.00066	0.00037	0.00094
April	perimeter (12M)	9	7.4	1.3	27	0.0037	0.00066	0.014
	off-site	2	2.1	1.8	2.4	0.0010	0.00090	0.0012
May	perimeter (12M)	8	6.1	2.3	11	0.0030	0.0012	0.0055
	off-site	2	3.4	2.2	4.7	0.0017	0.0011	0.0024
June	perimeter (12M)	9	15	3.4	65	0.0073	0.0017	0.033
	off-site	2	4.6	4.0	5.2	0.0023	0.0020	0.0026
July	perimeter (12M)	9	6.7	4.2	12	0.0033	0.0021	0.0060
	off-site	2	6.5	4.7	8.2	0.0032	0.0024	0.0041
August	perimeter (12M)	9	9.3	2.8	38	0.0046	0.0014	0.019
	off-site	2	5.0	4.0	6.0	0.0025	0.0020	0.0030
September	perimeter (12M)	8	5.1	2.3	11	0.0025	0.0011	0.0055
	off-site	2	< 1.1	< 0.1	2.0	< 0.00054	< 0.00005	0.0010
October	perimeter (14I)	8	6.7	1.2	24	0.0033	0.00062	0.012
	perimeter (12M)	1	-	-	2.5	-	-	0.0012
	off-site	2	1.5	0.92	2.1	0.00075	0.00046	0.0010
November	perimeter (14I)	9	7.5	1.2	24	0.0038	0.00062	0.012
	perimeter (12M)	2	9.1	1.0	17	0.0046	0.00050	0.0086
	off-site	2	2.1	2.1	2.1	0.0010	0.0010	0.0010
December	perimeter (14I)	9	9.7	1.3	21	0.0049	0.00064	0.011
	perimeter (12M)	2	3.9	2.4	5.5	0.0020	0.0012	0.0027
	off-site	2	0.84	0.50	1.2	0.00042	0.00025	0.00060
Annual Summary	perimeter (14I)	26	8.0 \pm 1.8	1.2	24	0.0040	0.00062	0.012
	perimeter (12M)	83	7.1 \pm 2.2	0.42	65	0.0036	0.00021	0.033
	off-site	24	2.7 \pm 1.1	< 0.1	8.2	0.0014	< 0.00005	0.0041

*This is the CG for uncontrolled areas.

The off-site concentrations, measured about 10 km northwest of the Laboratory, increased by about 50% over 1973. This is probably related to the previously discussed atmospheric nuclear tests. This background level of hydrogen-3 should be subtracted from the other concentrations in Tables 7 and 8 to obtain the Argonne contribution.

Argon-41 and hydrogen-3 (in the form of tritiated water) from the CP-5 reactor constitute the major portion of the gaseous radioactive effluent released from the Laboratory. During 1974, the total amount of argon-41 discharged from the reactor is estimated to be 4.3×10^4 Ci, based on a measured release rate of 1.46 Ci/MW-hr. Since the half-life of this nuclide is only 110 minutes, about 5% will decay before reaching the site boundary if the argon-41 moves with the average wind speed of 7.6 mph. Because the half-life is so short, it is appropriate and more meaningful to consider the concentration at various distances from the Laboratory in discussing argon-41 effluent discharges. By the computational method discussed in Section III.A.6, the average concentrations in the predominant wind direction (NNE) were 4.7×10^{-10} $\mu\text{Ci/ml}$ (1.1% of the CG and 0.4% of the average concentration near CP-5) at 1.5 km (the site boundary); 2.2×10^{-10} $\mu\text{Ci/ml}$ (0.6% of the CG) at 2.4 km; and 9×10^{-11} $\mu\text{Ci/ml}$ (0.2% of the CG) at 4 km. The total amount of hydrogen-3 (as tritiated water) discharged from the CP-5 reactor was 450 Ci, based on measurements made in the exhaust stack, and 60 Ci of hydrogen-3 as elemental hydrogen was released from Building 205.

Other airborne effluents were considerably lower. A small amount of argon-41 was released from the JANUS reactor, about 2 Ci in 1974. In addition, the total amount of other effluents is estimated to be less than 2.5 Ci/yr. The principal long-lived nuclide in this group was krypton-85, estimated to be 2.4 Ci; the other nuclides, in millicurie or smaller amounts, were radon-222 and various fission products. The release of iodine-131 (a nuclide of particular interest) in CP-5 exhaust air was estimated to be 0.015 Ci/yr, based on concentration measurements. The maximum concentration at 1.5 km, assuming no ground deposition, would be about 1×10^{-16} $\mu\text{Ci/ml}$, or $10^{-4}\%$ of the CG.

2. Surface Water

Total (nonvolatile) alpha and beta activities were determined by counting the residue remaining after evaporation of the water, and applying counting efficiency corrections determined for uranium-233 (for alpha activity) and thallium-204 (for beta activity), respectively, to obtain disintegration rates. Hydrogen-3 was determined by liquid scintillation counting of a separate sample, and this activity does not appear in the total beta activity. Uranium was determined fluorophotometrically, and the results calculated in terms of activity with the assumption that the isotopic composition was that of natural uranium. Analyses for other radionuclides were performed by specific radiochemical separations followed by appropriate counting. One liter aliquots were used for all analyses except hydrogen-3, plutonium, neptunium, and the transplutonium nuclides. Hydrogen-3 analyses were performed by counting 10 ml in a gel system. Plutonium and neptunium analyses were performed on 10 or 50-liter samples by a plutonium chemical separation method,⁽⁹⁾ modified to include neptunium, followed by alpha spectrometry. Plutonium-236 was used to determine the plutonium yield. A recent modification of the plutonium procedure allowed the group separation of a fraction containing the transplutonium elements.⁽¹⁰⁾ Americium-243 was added to determine chemical recovery, and individual nuclides were measured by alpha spectrometry.

Argonne waste water is discharged into Sawmill Creek, a small stream that runs through the Laboratory grounds, drains surface water from much of the site, and flows into the Des Plaines River about 500 meters (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 in Argonne waste water or from surface drainage. The sampling locations are shown in Figure 1. Below the waste-water outfall, the Creek was sampled five times a week. Since it was impractical to analyze all the samples for all the nuclides and elements desired, equal portions of the samples collected each week were combined and analyzed. The results obtained in this way represent the average concentrations in the weekly samples. Above the site, samples were usually collected once a month and were analyzed for the same radionuclides as the below-outfall samples.

Annual summaries of the results obtained for Sawmill Creek are given in Table 9. Comparison of the results, and 95% confidence limits of the averages, for the two sampling locations show that the only nuclides whose presence in Creek water can be attributed to Argonne operations were hydrogen-3, strontium-90, neptunium-237, plutonium-239, americium-241, and occasionally plutonium-238 and other transplutonium nuclides. The fraction of individual samples containing activity attributable to Argonne was 100% for hydrogen-3 and neptunium-237, 95% for plutonium-239, 75% for americium-241, and 50% for strontium-90. 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. The average Argonne contribution was 25% greater than last year, but amounted to only 0.07% of the CG, and the highest concentration in any single sample was equivalent to 1.6% of the CG. If the highest result, 4.7×10^{-5} $\mu\text{Ci/ml}$, is excluded, the average decreases by about 40%.

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 1974, the hydrogen-3 content of other lakes and streams ranged from $< 200 \times 10^{-9}$ $\mu\text{Ci/ml}$ to 335×10^{-9} $\mu\text{Ci/ml}$ and averaged $< 235 \times 10^{-9}$ $\mu\text{Ci/ml}$, identical to last year.

The highest contributor in terms of CG was strontium-90. Below the outfall the Argonne contribution was about 0.16% of the CG, while the fallout contribution was somewhat larger, about 0.2% of the CG.

There were positive iodine-131 results in the Creek at each location in samples collected at the same time. This activity could not be related to Laboratory operations or to recent nuclear tests. It is speculated that this activity may have been the result of a medical dose of iodine-131 to a resident living in the area served by the sewage treatment plant located above the Laboratory site. The daily iodine-131 excretion of 10 to 100 μCi that would be required to produce the observed concentration in the Creek has been found in this laboratory in excretions from individuals who have had recent medical injections of iodine-131. The decrease in concentration between above and below site was evidently due to dilution by Laboratory water.

The average total alpha, beta, and uranium activities were slightly higher above the site, indicating that at times Argonne waste water contained less of these materials than Creek water. The higher activities above the

TABLE 9

Radioactivity in Sawmill Creek Water, 1974

Type of Activity	Location *	No. of Samples	Concentration (10^{-9} $\mu\text{Ci}/\text{ml}$)			Percent of CG		
			Av.	Min.	Max.	Av.	Min.	Max.
Alpha (nonvolatile)	15K	12	1.9 ± 0.2	1.4	3.0	(0.063)	(0.047)	(0.10)
	7M	253	1.7 ± 0.1	0.9	3.3	(0.057)	(0.030)	(0.11)
Beta (nonvolatile)	15K	12	19 ± 4	9.8	32	(0.63)	(0.33)	(1.1)
	7M	253	16 ± 2	8.0	44	(0.53)	(0.27)	(1.5)
Hydrogen-3	15K	12	< 202	< 200	212	< 0.0067	< 0.0067	0.0071
	7M	253	2140 ± 1800	240	47,000	0.071	0.0080	1.6
Strontium-89	15K	12	-	-	< 2	-	-	< 0.07
	7M	253	-	-	< 2	-	-	< 0.07
Strontium-90	15K	12	0.62 ± 0.14	< 0.5	0.93	0.21	< 0.17	0.31
	7M	253	1.1 ± 0.4	< 0.5	11	0.37	< 0.17	3.7
Iodine-131	15K	12	< 4	< 3	11	< 1.3	< 1	3.7
	7M	253	< 4	< 3	7.9	< 1.3	< 1	2.6
Barium-140	15K	12	-	-	< 2	-	-	< 0.007
	7M	253	-	-	< 2	-	-	< 0.007
Uranium (natural) **	15K	12	2.0 ± 0.2	1.3	2.7	(0.0050)	(0.0032)	(0.0068)
	7M	253	1.9 ± 0.2	1.0	3.9	(0.0048)	(0.0025)	(0.0098)
Neptunium-237	15K	12	-	-	< 0.002	-	-	< 0.00007
	7M	253	0.19 ± 0.09	0.013	1.5	0.0063	0.00043	0.050
Plutonium-238	15K	12	-	-	< 0.002	-	-	< 0.00004
	7M	253	< 0.0028	< 0.002	0.016	< 0.000056	< 0.00004	0.00032
Plutonium-239	15K	11	< 0.001	< 0.0005	0.0040	< 0.00002	< 0.00001	0.00008
	7M	253	0.018 ± 0.008	< 0.0005	0.12	0.00036	< 0.00001	0.0024
Americium-241	15K	12	-	-	< 0.001	-	-	< 0.000025
	7M	253	0.006 ± 0.003	< 0.001	0.057	0.00015	< 0.000025	0.0014
Curium-242 and/or Californium-252	15K	12	-	-	< 0.001	-	-	< 0.000005
	7M	253	< 0.0013	< 0.001	0.010	< 0.0000065	< 0.000005	0.00005
Curium-244 and/or Californium-249	15K	12	-	-	< 0.001	-	-	< 0.000014
	7M	253	< 0.0036	< 0.001	0.035	< 0.000051	< 0.000014	0.0005

* Location 15K is upstream from the Argonne site and location 7M is downstream from the Argonne waste-water outfall. See Figure 1.

** Uranium concentrations in units of $\mu\text{g}/\text{l}$ can be obtained by multiplying the concentration given by 1.48×10^9 . The average concentration in the Creek then becomes $2.9 \mu\text{g}/\text{l}$.

site are probably due to the water added to the Creek by a large municipal sewage treatment plant. The large amount of dissolved solids added in the sewage water is accompanied by a small amount of radioactive materials, and increases the radioactivity in the Creek water.

In addition to the natural beta activity and that added by Argonne waste water at the outfall, beta activity from nuclear detonations was detected at both sampling locations. The normal nonvolatile beta activity is approximately 5×10^{-9} $\mu\text{Ci/ml}$ while the contribution from the upstream municipal sewage treatment plant is another 5×10^{-9} $\mu\text{Ci/ml}$. It is estimated that fallout activity added about 6×10^{-9} $\mu\text{Ci/ml}$ to the nonvolatile beta activity at both locations and that the Argonne contribution to the water below the outfall averaged about 1×10^{-9} $\mu\text{Ci/ml}$, equivalent to 0.03% of the CG. The Argonne contribution remained the same as 1973 levels, while the fallout contribution increased by about a factor of two. The total concentration, regardless of source, must be used in assessing the health hazard of a radionuclide not naturally present, and the percent of the CGs for all nuclides listed in Table 9 were low.

The total radioactive effluent discharged to the Creek in Argonne waste water can be estimated from the average concentrations and the volume of water carried by the Creek. These values are 25 Ci of hydrogen-3, 5.8 mCi of strontium-90 (0.5×10^{-9} $\mu\text{Ci/ml}$ is the average concentration attributed to Argonne), 2.2 mCi of neptunium-237, 0.2 mCi of plutonium-239, 0.07 mCi of americium-241, and < 0.07 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 usually 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 total alpha and beta activities, 1.5×10^{-9} $\mu\text{Ci/ml}$ and 11×10^{-9} $\mu\text{Ci/ml}$, respectively of 22 off-site surface water samples collected this year (excluding the Des Plaines River) are identical to the levels found in the Des Plaines River and is evidence that the River activity levels are normal. The activity in Sawmill Creek is evidently reduced by dilution so

TABLE 10

Radioactivity in Des Plaines River Water, 1974

Type of Activity	Location*	No. of Samples	Concentration (10^{-9} $\mu\text{Ci/ml}$)			Percent of CG		
			Av.	Min.	Max.	Av.	Min	Max.
Alpha (nonvolatile)	A	12	1.5 ± 0.2	1.1	2.3	(0.050)	(0.037)	(0.077)
	B	24	1.6 ± 0.2	0.9	2.8	(0.053)	(0.030)	(0.093)
Beta (nonvolatile)	A	12	11 ± 2	5.2	20	(0.37)	(0.17)	(0.67)
	B	24	11 ± 1	6.2	18	(0.37)	(0.21)	(0.60)
Hydrogen-3	A	12	< 210	< 200	250	< 0.0070	< 0.0067	0.0083
	B	24	< 208	< 200	240	< 0.0069	< 0.0067	0.0080
Strontium-89	A	12	-	-	< 2	-	-	< 0.007
	B	24	-	-	< 2	-	-	< 0.007
Strontium-90	A	12	0.86 ± 0.18	< 0.5	1.4	0.29	< 0.17	0.47
	B	24	0.82 ± 0.09	< 0.5	1.2	0.27	< 0.17	0.40
Iodine-131	A	12	-	-	< 3	-	-	< 1
	B	24	-	-	< 3	-	-	< 1
Barium-140	A	12	-	-	< 2	-	-	< 0.007
	B	24	-	-	< 2	-	-	< 0.007
Uranium (natural)**	A	12	1.3 ± 0.3	0.7	2.4	(0.0032)	(0.0018)	(0.0060)
	B	24	1.4 ± 0.2	0.8	2.6	(0.0035)	(0.0020)	(0.0065)
Neptunium-237	A	12	-	-	< 0.002	-	-	< 0.00007
	B	12	< 0.0025	< 0.002	0.0072	< 0.00008	< 0.00007	0.00024
Plutonium-238	A	11	-	-	< 0.002	-	-	< 0.00004
	B	11	-	-	< 0.002	-	-	< 0.00004
Plutonium-239	A	11	< 0.0008	< 0.0005	0.0021	< 0.000016	< 0.00001	0.000042
	B	11	< 0.0012	< 0.0005	0.0030	< 0.000024	< 0.00001	0.000060
Americium-241	A	12	-	-	< 0.001	-	-	< 0.000025
	B	12	-	-	< 0.001	-	-	< 0.000025
Curium-242 and/or Californium-252	A	12	-	-	< 0.001	-	-	< 0.000005
	B	12	-	-	< 0.001	-	-	< 0.000005
Curium-244 and/or Californium-249	A	12	-	-	< 0.001	-	-	< 0.000014
	B	12	-	-	< 0.001	-	-	< 0.000014

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

** Uranium concentrations in units of $\mu\text{g/l}$ can be obtained by multiplying the concentration given by 1.48×10^9 . The average concentration is $2.0 \mu\text{g/l}$.

that it is not detectable as such in the Des Plaines River. The natural non-volatile beta activity in the River was 5×10^{-9} $\mu\text{Ci/ml}$, and the excess, 6×10^{-9} $\mu\text{Ci/ml}$, was due to fallout.

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

3. Soil, Grass, and Benthic Materials

The plutonium content of soil, grass, and benthic materials was measured at the site perimeter and off the site. The object of the off-site sampling was to determine the total deposition of plutonium from weapons testing for comparison with perimeter samples, and with results obtained by other organizations for samples collected at large distances from nuclear installations. This latter comparison is useful in determining if the soil activity near Argonne was normal. For this purpose, the site selection criteria and sample preparation techniques used by the AEC (now ERDA) New York Health and Safety Laboratory⁽¹¹⁾ were used. Sites were selected in several directions and at various distances from the Laboratory. Each site was selected on the basis that it appeared, or was known to have been, undisturbed for a number of years. Attempts were made to select open, level, grassy areas that were mowed at reasonable intervals. Public parks were selected when available.

Each soil sample consisted of two cores totalling 173 cm^2 in area by 30 cm deep. The grass samples were obtained by collecting the grass from a 1 m^2 area in the immediate vicinity of a soil sample. A grab sample technique was used to obtain benthic materials. After drying, grinding, and mixing, 100 g portions of soil, benthos, and grass were analyzed by the same method described in Section III.A. for air-filter residues. Results are given in terms of oven-dried soil, benthos, or grass.

Comparison of the perimeter (Table 12) and off-site (Table 13) soil results shows that the same general range of concentrations exist in all areas for both plutonium isotopes, and it may be concluded that the plutonium in the perimeter samples resulted primarily from fallout of debris from nuclear detonations. Fallout deposition values found by other laboratories⁽¹²⁻¹⁴⁾ are in the same range as those reported here, about $2-3 \times 10^{-3}$ $\mu\text{Ci/m}^2$.

TABLE 11

Radioactivity in Illinois River Water, 1974
(concentrations in 10^{-9} $\mu\text{Ci/ml}$)

Location	Date Collected	Alpha *	Beta *	Hydrogen-3	Uranium **	Neptunium-237	Plutonium-239
Starved Rock State Park	May 14	1.6	6.7	210	1.1	-	-
Morris	May 14	1.7	11	285	1.1	-	-
Below Dresden Power Station	May 14	1.1	4.5	270	1.0	< 0.0005	0.00043
McKinley Woods State Park	October 8	1.7	16	205	1.0	< 0.0005	0.0023
Below Dresden Power Station	October 8	0.8	8.9	< 200	0.8	< 0.0005	0.00034
Morris	October 8	1.4	9.9	< 200	0.9	-	-
Starved Rock State Park	October 8	2.2	11	210	1.1	-	-

* Nonvolatile activity.

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

TABLE 12

Plutonium Content of Perimeter Soil, 1974

Date Collected	Location*	Plutonium-239		Plutonium-238		$^{238}\text{Pu}/^{239}\text{Pu}$
		10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	
June 4	7M	6.9 ± 0.8	2.05 ± 0.25	0.4 ± 0.2	0.10 ± 0.06	0.050
June 4	12O	4.3 ± 0.5	1.65 ± 0.20	0.8 ± 0.2	0.29 ± 0.09	0.18
June 4	14I	3.0 ± 0.4	1.27 ± 0.18	0.1 ± 0.1	0.05 ± 0.04	0.040
June 4	13D	5.0 ± 0.6	2.09 ± 0.23	0.3 ± 0.1	0.12 ± 0.06	0.056
June 5	12M	4.7 ± 0.6	1.82 ± 0.24	0.2 ± 0.2	0.08 ± 0.06	0.045
November 6	10M	1.8 ± 0.2	0.88 ± 0.10	0.2 ± 0.1	0.08 ± 0.04	0.090
November 6	10M	5.7 ± 0.4	2.30 ± 0.14	-	-	-
November 7	13I	2.2 ± 0.2	1.07 ± 0.10	-	-	-
November 7	13/14I	4.6 ± 0.3	2.09 ± 0.15	< 0.1	< 0.01	-
November 7	13I	2.6 ± 0.2	1.28 ± 0.12	0.1 ± 0.1	0.07 ± 0.03	0.053
Average		4.1 ± 1.0	1.65 ± 0.31	0.3 ± 0.2	0.10 ± 0.06	0.073

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

TABLE 13

Plutonium Content of Off-Site Soil, 1974

Date Collected	Location	Plutonium-239		Plutonium-238		$^{238}\text{Pu}/^{239}\text{Pu}$
		10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	
May 14	Starved Rock State Park, Ill.	4.7 ± 0.6	2.06 ± 0.25	0.2 ± 0.1	0.06 ± 0.05	0.032
May 14	Morris, Ill. (Continental Grain)	5.7 ± 0.6	2.05 ± 0.20	0.2 ± 0.1	0.06 ± 0.04	0.027
May 14	Morris, Ill. Stratton Park	20.1 ± 1.0	6.94 ± 0.33	1.2 ± 0.2	0.41 ± 0.08	0.059
May 14	Pioneer Park, Naperville, Ill.	7.1 ± 0.6	2.54 ± 0.23	0.4 ± 0.2	0.12 ± 0.05	0.049
May 21	McGinnis Slough	6.7 ± 0.8	2.17 ± 0.25	0.3 ± 0.2	0.11 ± 0.06	0.050
May 21	Saganashkee Slough	7.2 ± 0.7	1.96 ± 0.20	0.4 ± 0.2	0.11 ± 0.05	0.058
October 8	McKinley Woods State Park, Ill.	5.9 ± 0.4	2.10 ± 0.12	0.4 ± 0.1	0.16 ± 0.06	0.075
October 23	Bemis Woods, Hinsdale, Ill.	10.1 ± 0.4	3.45 ± 0.15	0.6 ± 0.1	0.21 ± 0.04	0.061
October 23	McCormick Woods, Brookfield, Ill.	4.8 ± 0.5	1.78 ± 0.20	0.4 ± 0.2	0.16 ± 0.07	0.088
November 7	Lemont, Ill.	4.0 ± 0.3	1.53 ± 0.10	0.2 ± 0.1	0.07 ± 0.02	0.044
	Average	7.6 ± 3.0	2.7 ± 1.0	0.4 ± 0.2	0.15 ± 0.07	0.054

Composite monthly precipitation samples were analyzed for plutonium-239. Concentrations, expressed in terms of ground deposition, ranged from $2.5 \times 10^{-7} \mu\text{Ci}/\text{m}^2$ to $5.6 \times 10^{-6} \mu\text{Ci}/\text{m}^2$ and averaged $2.1 \times 10^{-6} \mu\text{Ci}/\text{m}^2$. The total 1974 deposition by precipitation was seven times greater than 1973, a reflection of the increased fallout from recent atmospheric nuclear tests, and was equivalent to 1.4% of the amount previously deposited in the soil.

Samples collected in the fall of 1973 at Stratton State Park in Morris, Illinois, gave plutonium-239 concentrations almost three times fallout levels, elevated uranium, thorium-230, and radium-226 concentrations, but normal thorium-232 and -228 concentrations. Samples collected from this same location on May 14, 1974, gave similar results while other samples collected several blocks away in Morris, Illinois, gave normal results. The reasons for these high results at Stratton State Park are unknown at this time.

The thorium, uranium, and cesium-137 content of soil samples was also measured and the concentrations in the perimeter (Table 14) and off-site (Table 15) samples were similar. Except for the result at Stratton State Park in Morris, Illinois, these are expected levels of the naturally-occurring thorium and uranium activities and normal fallout concentrations of cesium-137. In terms of mass, the thorium concentrations were $3.4 \mu\text{g}/\text{g}$ and $3.0 \mu\text{g}/\text{g}$ at the perimeter and off the site, respectively, while the uranium concentrations were $3.1 \mu\text{g}/\text{g}$ at the perimeter and $5.3 \mu\text{g}/\text{g}$ off-site.

The results of radioactivity measurements in grass are given in Tables 16 and 17. The perimeter and off-site plutonium concentrations are similar to each other and to results of previous years. All the results, perimeter and off-site, were within the range expected and observed from fallout. In terms of deposition, the plutonium-239 concentration was a factor of about 10^4 less than in soil from the same location. Thorium, uranium, and the detectable gamma-ray emitting nuclides were similar at the perimeter and off-site indicating that their concentrations are due to naturally-occurring nuclides or fallout.

A study was made of the radioactivity content of the beds of streams and rivers off the Laboratory site. The results are collected in Table 18. Plutonium results vary widely between locations and are related to the retentiveness of the bottom material. Plutonium-239 concentrations up to about $30 \times 10^{-9} \mu\text{Ci}/\text{g}$ may be considered normal based on previous data.⁽⁴⁾ The thorium

TABLE 14

Thorium, Uranium, and Cesium-137 in Perimeter Soil, 1974
(concentrations in 10^{-6} $\mu\text{Ci/g}$)

Date Collected	Location*	Thorium-232	Thorium-230	Thorium-228	Uranium (natural)	Cesium-137
June 4	7M	-	-	-	0.90 ± 0.06	0.2 ± 0.1
June 4	12O	-	-	-	1.4 ± 0.1	0.4 ± 0.1
June 4	14I	0.42 ± 0.03	0.48 ± 0.03	0.54 ± 0.03	3.9 ± 0.4	0.3 ± 0.1
June 4	13D	0.48 ± 0.03	0.56 ± 0.03	0.56 ± 0.03	4.9 ± 0.4	0.3 ± 0.1
June 5	12M	-	-	-	2.1 ± 0.1	0.2 ± 0.1
November 6	10M	0.33 ± 0.04	0.58 ± 0.05	0.33 ± 0.04	1.7 ± 0.2	< 0.1
November 6	10M	-	-	-	1.1 ± 0.1	0.2 ± 0.1
November 7	13I	0.32 ± 0.03	0.46 ± 0.04	0.37 ± 0.03	1.9 ± 0.1	0.1 ± 0.1
November 7	13/14I	-	-	-	1.4 ± 0.1	0.1 ± 0.1
November 7	13I	0.36 ± 0.03	0.55 ± 0.03	0.46 ± 0.03	2.1 ± 0.2	0.1 ± 0.1
Average		0.38 ± 0.06	0.53 ± 0.05	0.45 ± 0.09	2.1 ± 0.8	0.2 ± 0.1

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

TABLE 15

Thorium, Uranium, and Cesium-137 in Off-Site Soil, 1974
(concentrations in 10^{-6} $\mu\text{Ci/g}$)

Date Collected	Location	Thorium-232	Thorium-230	Thorium-228	Uranium (natural)	Cesium-137
May 14	Starved Rock State Park, Ill.	0.30 ± 0.03	0.46 ± 0.04	0.38 ± 0.04	4.6 ± 0.2	0.4 ± 0.1
May 14	Morris, Ill. (Continental Grain)	0.26 ± 0.03	0.40 ± 0.03	0.27 ± 0.03	4.3 ± 0.2	0.5 ± 0.1
May 14	Morris, Ill., Stratton Park	-	-	-	8.3 ± 0.5	0.8 ± 0.1
May 14	Pioneer Park, Naperville, Ill.	-	-	-	4.6 ± 0.2	0.7 ± 0.1
May 21	McGinnis Slough	-	-	-	2.4 ± 0.1	0.4 ± 0.1
May 21	Saganashkee Slough	-	-	-	6.2 ± 0.3	0.4 ± 0.1
October 8	McKinley Woods State Park, Ill.	-	-	-	1.0 ± 0.1	0.3 ± 0.1
October 23	Bemis Woods, Hinsdale, Ill.	-	-	-	1.7 ± 0.1	0.7 ± 0.1
October 23	McCormick Woods, Brookfield, Ill.	0.42 ± 0.04	0.56 ± 0.04	0.46 ± 0.04	1.5 ± 0.1	0.2 ± 0.1
November 7	Lemont, Ill.	-	-	-	1.1 ± 0.1	0.2 ± 0.1
	Average	0.33 ± 0.10	0.47 ± 0.10	0.37 ± 0.11	3.6 ± 1.5	0.5 ± 0.1

TABLE 16

Plutonium Content of Plant Samples, 1974

Date Collected	Location	Plutonium-239		Plutonium-238		$^{238}\text{Pu}/^{239}\text{Pu}$
		$10^{-9} \mu\text{Ci/g}$	$10^{-6} \mu\text{Ci/m}^2$	$10^{-9} \mu\text{Ci/g}$	$10^{-6} \mu\text{Ci/m}^2$	
<u>Off-Site</u>						
May 14	Starved Rock State Park, Ill.	3.6 ± 0.4	0.18 ± 0.02	0.2 ± 0.1	0.01 ± 0.01	0.055
May 14	Morris, Ill., Stratton Park	1.0 ± 0.2	0.20 ± 0.03	< 0.1	-	-
May 14	Pioneer Park, Naperville, Ill.	4.4 ± 0.4	0.36 ± 0.03	0.2 ± 0.1	0.02 ± 0.01	0.050
May 21	McGinnis Slough	1.6 ± 0.2	0.13 ± 0.02	0.1 ± 0.1	0.01 ± 0.01	0.067
May 21	Saganashkee Slough	1.5 ± 0.2	0.08 ± 0.01	< 0.1	-	-
October 8	McKinley Woods State Park, Ill.	2.2 ± 0.2	0.29 ± 0.03	< 0.1	-	-
October 23	Bemis Woods, Hinsdale, Ill.	0.6 ± 0.1	0.14 ± 0.02	< 0.1	-	-
October 23	McCormick Woods, Brookfield, Ill.	1.4 ± 0.2	0.20 ± 0.03	0.2 ± 0.1	0.02 ± 0.01	0.13
	Average	2.0 ± 0.9	0.20 ± 0.06	0.1 ± 0.1	0.02 ± 0.01	0.076
<u>Perimeter*</u>						
June 4	120	0.6 ± 0.1	0.19 ± 0.04	< 0.1	-	-
June 4	9I	1.0 ± 0.1	0.22 ± 0.03	< 0.1	-	-
October 14	9I	1.4 ± 0.2	0.39 ± 0.05	0.1 ± 0.1	0.03 ± 0.02	0.086
November 6	10M	1.8 ± 0.2	0.22 ± 0.02	0.2 ± 0.1	0.02 ± 0.01	0.095
November 7	13/14I	1.6 ± 0.2	0.71 ± 0.08	< 0.1	-	-
	Average	1.3 ± 0.4	0.35 ± 0.19	0.1 ± 0.1	0.02 ± 0.01	0.090

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

TABLE 17

Radioactivity in Plant Samples, 1974
(concentrations in 10^{-6} $\mu\text{Ci/g}$)

Date Collected	Location	Thorium-232	Thorium-230	Thorium-228	Uranium (natural)	Cerium-144	Cesium-137	Zirconium-95-Niobium-95	Beryllium-7
<u>Off-Site</u>									
May 14	Starved Rock State Park, Ill.	0.016 \pm 0.002	0.035 \pm 0.002	0.014 \pm 0.001	-	-	-	-	-
May 14	Morris, Ill., Stratton Park	-	-	-	0.010 \pm 0.002	-	-	-	-
May 14	Pioneer Park, Naperville, Ill.	0.013 \pm 0.001	0.021 \pm 0.002	0.015 \pm 0.002	-	-	-	-	-
May 21	McGinnis Slough	-	-	-	0.027 \pm 0.003	-	-	-	-
May 21	Saganashkee Slough	0.002 \pm 0.001	0.002 \pm 0.001	0.002 \pm 0.001	-	-	-	-	-
October 8	McKinley Woods State Park, Ill.	0.005 \pm 0.001	0.009 \pm 0.001	0.015 \pm 0.002	0.029 \pm 0.002	0.8 \pm 0.2	0.1 \pm 0.1	0.5 \pm 0.2	4.0 \pm 0.9
October 23	Bemis Woods, Hinsdale, Ill.	0.010 \pm 0.001	0.020 \pm 0.001	0.020 \pm 0.001	0.044 \pm 0.003	0.7 \pm 0.1	0.1 \pm 0.1	0.7 \pm 0.1	4.4 \pm 0.4
October 23	McCormick Woods, Brookfield, Ill.	-	-	-	0.056 \pm 0.004	0.5 \pm 0.2	0.1 \pm 0.1	0.5 \pm 0.2	3.8 \pm 0.9
	Average	0.009 \pm 0.005	0.017 \pm 0.011	0.013 \pm 0.006	0.033 \pm 0.016	0.7 \pm 0.2	0.1 \pm 0.1	0.6 \pm 0.1	4.1 \pm 0.4
<u>Perimeter*</u>									
June 4	120	0.012 \pm 0.001	0.018 \pm 0.001	0.011 \pm 0.001	-	-	-	-	-
June 4	9I	-	-	-	0.012 \pm 0.002	-	-	-	-
October 14	9I	-	-	-	0.033 \pm 0.004	0.6 \pm 0.1	0.1 \pm 0.1	0.5 \pm 0.2	2.5 \pm 0.8
November 6	10M	0.007 \pm 0.001	0.012 \pm 0.001	0.008 \pm 0.001	0.032 \pm 0.003	0.6 \pm 0.1	0.1 \pm 0.1	1.4 \pm 0.1	3.5 \pm 0.5
November 7	13/14I	-	-	-	0.024 \pm 0.002	0.5 \pm 0.2	0.1 \pm 0.1	0.5 \pm 0.2	2.0 \pm 0.8
	Average	0.010 \pm 0.005	0.015 \pm 0.006	0.010 \pm 0.003	0.025 \pm 0.010	0.6 \pm 0.1	0.1 \pm 0.1	0.8 \pm 0.6	2.7 \pm 0.9

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

TABLE 18

Radioactivity in Benthic Material, 1974

Date Collected	Location	Concentration (10^{-9} $\mu\text{Ci/g}$)		Concentration (10^{-6} $\mu\text{Ci/g}$)			Uranium (natural)
		Plutonium-239	Plutonium-238	Thorium-232	Thorium-230	Thorium-228	
May 14	Illinois River, Morris, Ill.	1.0 ± 0.2	< 0.1	0.16 ± 0.004	0.21 ± 0.005	0.14 ± 0.004	3.5 ± 0.2
August 7	Des Plaines River, Willow Springs, Ill.	24.7 ± 0.7	1.3 ± 0.2	0.33 ± 0.03	0.51 ± 0.04	0.41 ± 0.04	1.5 ± 0.1
August 7	Des Plaines River, Lemont, Ill.	3.6 ± 0.2	0.2 ± 0.1	-	-	-	0.44 ± 0.04
August 7	Sawmill Creek, Above Site	6.1 ± 0.4	0.4 ± 0.1	0.32 ± 0.03	0.50 ± 0.04	0.33 ± 0.03	-
October 8	Des Plaines River, Romeoville, J	6.7 ± 0.3	0.4 ± 0.1	0.12 ± 0.02	0.19 ± 0.03	0.29 ± 0.04	0.88 ± 0.04
October 23	Long Run Creek, Lemont, Ill.	6.6 ± 0.3	0.4 ± 0.1	-	-	-	0.97 ± 0.05
	Average	8.1 ± 6.9	0.5 ± 0.4	0.23 ± 0.11	0.35 ± 0.18	0.29 ± 0.11	1.44 ± 1.1

and uranium concentrations were similar to soil. In terms of mass, the thorium concentrations were 2.1 $\mu\text{g/g}$ and the uranium concentrations were 2.1 $\mu\text{g/g}$.

4. Milk

Raw milk was collected monthly from a local dairy farm and analyzed for several radioactive nuclides by methods similar to those used for water. Iodine-131 and barium-140 were not present in concentrations greater than their detection limits of 20×10^{-9} $\mu\text{Ci/ml}$ and 2×10^{-9} $\mu\text{Ci/ml}$, respectively. The results are given in Table 19. The average hydrogen-3 concentration in milk is identical to the concentration of hydrogen-3 found in surface water samples away from the site. The average strontium-90 and cesium-137 concentrations increased slightly compared to 1973 and positive strontium-89 results occurred in June and July. These nuclides are fission products from nuclear tests and their presence in milk is not related to Argonne operations.

The concentrations given in Table 19 may be compared to the CGs for drinking water given in the Appendix, Part B. The drinking water CGs are based on an intake of 2.2 liters per day. The consumption of one liter of milk per day would result in an average intake of $< 0.003\%$ of the hydrogen-3, $< 0.03\%$ of the strontium-89, 1.0% of the strontium-90, and 0.016% of the cesium-137 Concentration Guides.

5. External Penetrating Radiation

Dose measurements were made with dysprosium-activated calcium fluoride thermoluminescent chips. Each measurement was the average of the readings of three to five chips exposed in the same package. The response of the chips was calibrated with a NBS standard radium-226 source, and the results calculated in terms of air dose. Measurements were made in six successive exposure periods that varied in length from 45 to 81 days, and averaged 61 days. Results for each period were calculated in terms of annual dose rate, for ease in comparing measurements made for different time intervals, and were weighted according to their exposure times in calculating the annual average for a location.

Measurements were made at a number of locations at the site boundary to determine the dose due to Argonne operations at the closest uncontrolled

TABLE 19

Radioactivity in Milk, 1974
(concentrations in 10^{-9} $\mu\text{Ci/ml}$)

Date Collected	Hydrogen-3	Strontium-89	Strontium-90	Cesium-137
January 2	< 200	< 2	3.8	< 5
February 6	< 200	< 2	7.0	< 5
March 15	< 200	< 2	2.3	< 5
April 3	< 200	< 2	6.7	7
May 1	< 200	< 2	5.5	7
June 5	< 200	3.1	14.8	12
July 3	< 200	3.1	15.3	19
August 7	260	< 2	11.6	8
September 4	320	< 2	5.0	10
October 2	205	< 2	6.1	5
November 6	245	< 2	0.6	5
December 4	< 200	< 2	3.3	< 5
Average	< 220	< 2.2	6.8 ± 2.7	6.9 ± 2.8

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 abnormal dose readings obtained at the boundary. Measurements were also made at five off-site locations for comparison purposes. The results are summarized in Tables 20 and 21, and the site boundary and on-site readings are also shown in Figure 4. The error given for an average is the 95% confidence limit calculated from the standard deviation (not from the standard error). This confidence limit is used so that individual perimeter results can be compared with normal values.

The off-site results averaged 99 mrem/yr with a standard deviation (for a single result) of 8.5 mrem/yr. The standard error of the mean was 1.6 mrem/yr. For 1972 and 1973, averages and standard deviations were 105 ± 5 mrem/yr and 100 ± 11 mrem/yr, respectively. Thus, the off-site readings have remained constant, within the observed statistical variations. When the 1974 off-site results are compared by measurement period and location, it is evident that real differences exist between some locations. The standard deviations at each location are consistently smaller than the standard deviations for the separate exposure periods. The latter six averages ranged from 93 to 104 mrem/yr and had a standard deviation of only 4 mrem/yr, which indicates the dose levels varied in a similar manner at each location from period to period. The differences between locations appear to be the result of local conditions. For example, at Oak Lawn, three dosimeter packets within 50 meters of each other gave dose rates of 82, 92, and 103 mrem/yr for the same time interval, 3/8 to 5/10, while at Downers Grove, no such difference was observed. It should be noted that one location at Oak Lawn, near the usual one that has been used for several years and whose results are given in the table, gives values very close to the off-site average.

The off-site results, without regard to location, show a nearly normal distribution, so that a result in the range of 99 ± 17 mrem/yr has a 95% probability of being normal. Only 2.5% of the measurements of a natural background radiation field should be higher than 116 mrem/yr and lower than 82 mrem/yr.

At only one location, 7I, at the south boundary of the site, were the dose measurements consistently above the normal range. The net above-normal

TABLE 20

Environmental Penetrating Radiation at Off-Site Locations, 1974

Location	Dose Rate (mrem/year)						Average
	Period of Measurement						
	1/3-3/8	3/8-5/10	5/10-7/30	7/30-10/4	10/4-11/18	11/18-1/3	
Downers Grove	98	100	97	106	93	91	98 ± 13
Lockport	104	111	102	102	93	99	101 ± 15
Lombard	102	103	101	97	89	101	99 ± 13
Oak Lawn	91	92	87	90	83	86	88 ± 9
Oakbrook	117	112	110	110	106	107	110 ± 9
Average	102 ± 24	104 ± 21	99 ± 21	101 ± 20	93 ± 21	97 ± 21	99 ± 17

TABLE 21

Environmental Penetrating Radiation at ANL, 1974

Location	Dose Rate (mrem/year)						Average
	Period of Measurement						
	1/3-3/8	3/8-5/10	5/10-7/30	7/30-10/4	10/4-11/18	11/18-1/3	
14L - Boundary	94	95	88	92	90	82	90 ± 12
14I - Boundary	109	103	105	108	105	101	105 ± 7
14H - Boundary	91	118	98	102	91	97	100 ± 25
13D - Boundary	-	85	77	75	74	73	77 ± 12
10EF - Boundary	-	112	-	-	-	-	-
9EF - Boundary	110	128	102	93	90	92	103 ± 36
8F - Boundary	108	98	103	110	97	92	101 ± 17
8H - Boundary	118	119	108	108	101	105	110 ± 18
7I - Boundary	297	310	296	314	245	236	282 ± 84
9L - Boundary	99	107	87	-	-	-	98 ± 30
11H - 500 m N of CP-5	118	106	104	-	-	-	109 ± 23
11F - 900 m NW of CP-5	99	-	-	-	-	-	-
9H - 50 m SE of CP-5	2040	2590	1040	1260	-	3180	2020 ± 2320
9H - 225 m S of CP-5, 50 m N of 316	183	177	-	-	-	-	180 ± 18
8H - 25 m S of 316	207	197	138	131	304	167	191 ± 158
8H - 200 m NW of Waste Storage Area (Heliport)	135	130	132	128	112	133	128 ± 21
7I - Center - Waste Storage Area	3250	4280	3780	3640	2690	3120	3460 ± 1400
9H - 50 m N of 350, 200 m ENE of 316	-	128	131	135	220	260	175 ± 153
8H - 200 m NE of Heliport, 200 m SW of 316	-	120	111	122	118	137	122 ± 24
8J - 400 m N of Waste Storage Area	-	110	87	100	-	-	99 ± 37
10J - Lodging Facilities	-	-	-	91	-	-	-
10I - 350 m W of CP-5	-	-	81	75	-	-	78 ± 18

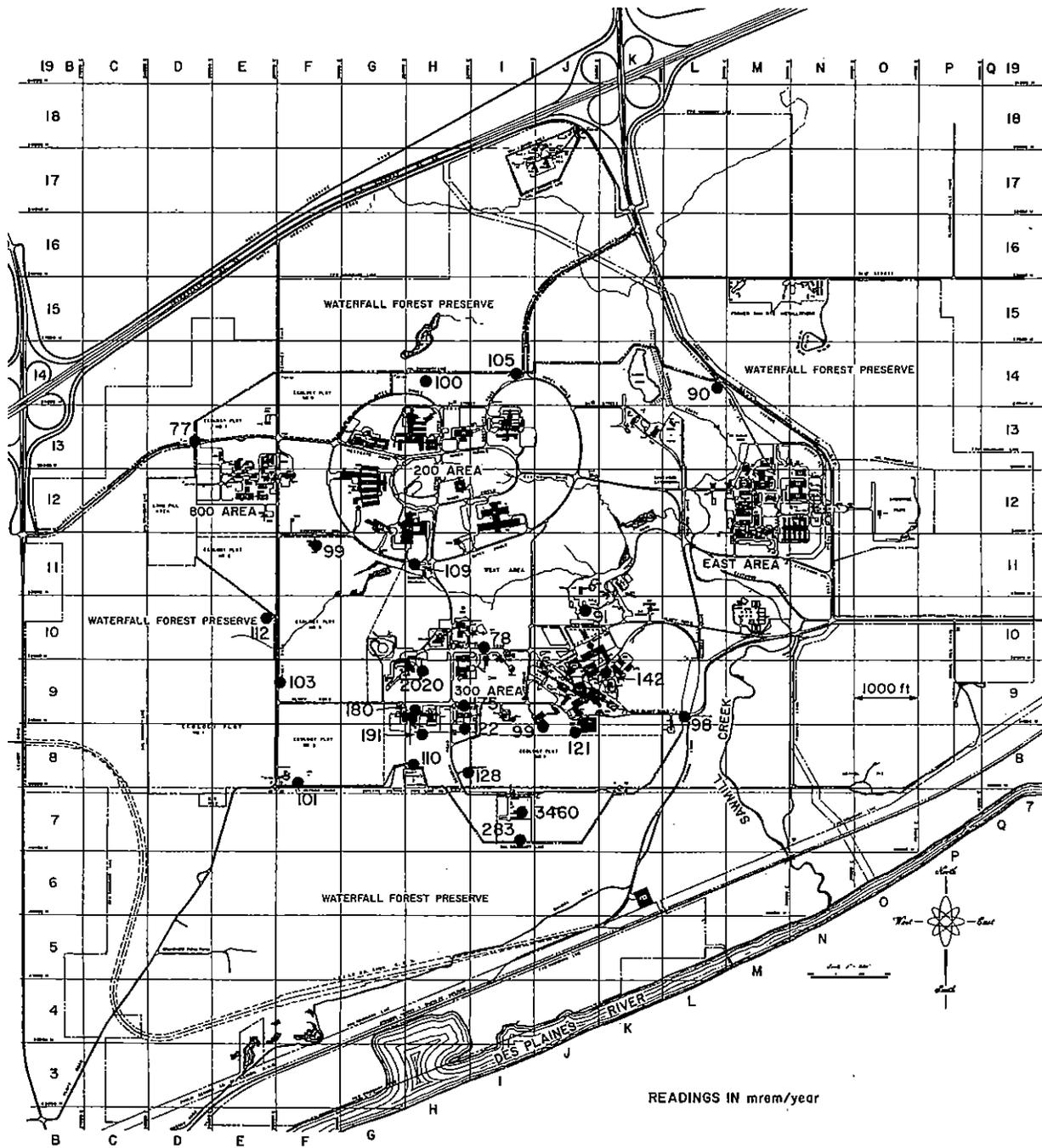


Fig. 4. Penetrating Radiation Measurements at Argonne National Laboratory

dose at this location, 184 mrem/yr, 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 excess dose rate at 7I decreased substantially in 1974; during the previous two years the average at the same location was about 320 mrem/yr above the off-site average. This change was also reflected in decreases at two other locations, 8H at the boundary and 8H at the Heliport, where slightly elevated dose levels have been found in the past, and attributed in part to radiation from the storage facility.

The dose at the 8H boundary site is of particular interest since it lies on the edge of St. Patrick's Cemetery, which was in use before Argonne was constructed, and which is open to visitors. The dose at this location averaged 110 ± 14 mrem/yr during 1974, which is in the range found off-site, as compared to 126 ± 23 mrem/yr during 1972 and 1973. Two of the readings at this location, 118 and 119 mrem/yr, were slightly outside the 95% confidence limits of the off-site average. A conservative or upper estimate of the dose at this location due to Argonne may be obtained by comparing the off-site and 8H readings for the same time period. These differences varied from 8 to 16 and averaged 11 mrem/yr higher at 8H. During 1973, a similar comparison gave a range and average of 20 to 30 and 25 mrem/yr, respectively.

At the south end of the Laboratory site, three possible sources of external radiation exist: direct radiation from the Waste Storage Facility, direct radiation from a tandem dynamitron and low-power reactors in Buildings 314 and 316, respectively (location 9GH), and argon-41 from the CP-5 reactor at location 9H. The contribution from CP-5 is considered minor, since dose readings in other directions from the reactor, where higher doses from argon-41 are expected because of wind direction and distance, were less than at 8H. By comparing results obtained at various distances from both the Storage Facility and Buildings 314 and 316, and from results obtained at locations between the Cemetery and both radiation sources, it may be concluded that both the Storage Facility and the equipment in Buildings 314 and 316 contributed to the dose at 8H.

The size of the dose variations from period to period, which are at least in part statistical, and the observed differences between locations make it difficult to determine with high certainty when site boundary doses

are only a few mrem/yr above normal and due to Argonne operations. Three criteria are used here to identify such locations: 1) the results for each sampling period are consistently above the off-site average for the same period; 2) the annual average at a location exceeds the off-site average (99 mrem/yr) plus twice the standard deviation of the average, 17 mrem/yr; or 3) occasional results at a particular location are significantly above the normal value at that location. The last criterion would be applied to a location such as 13D where the results have been consistently below the off-site average. At this location the ground contains considerable gravel, which contains less radium than the usual clay sub-soil, and this probably accounts for the low dose rates.

Location 14I warrants closer examination in the future on the basis of the first two criteria. The JANUS reactor is a possible contributor in this area. The abnormal results at 14H and 9EF during the second exposure period may be due to statistical fluctuations, since the dose rates were normal at other times. These areas will also be studied in more detail.

6. Potential Radiation Dose Estimates

a. Air and Water Borne Radioactivity

The radiation doses at the site boundary and off the site that could have been received by the public from radioactive materials leaving the site were calculated by two methods. Where measured radionuclide concentrations in air and water are available, the conversion of concentration to dose was based on the ratio of environmental concentrations to the Concentration Guides in AECM/ERDA Chapter 0524. The Guides, and the annual radiation dose that would result from continuous exposure at these concentrations, are given in Table 22 for those nuclides whose presence in the environment is attributable to Argonne. The dose values in the table are the Radiation Protection Standards for individuals in uncontrolled areas. The second method was used for argon-41, tritiated water, and iodine-131 released from reactor stacks. In these cases, doses were calculated from an atmospheric dispersion model making use of a source term and meteorological data.

The principal exposure pathway for radioactive substances released from Argonne is directly from air to man. A minor exposure route is from water to man by ingestion 140 miles downstream from Argonne (Section I.E.). Since the

TABLE 22

Concentration-to-Dose Conversion Factors

Nuclide	Medium	Concentration* ($\mu\text{Ci/ml}$)	Dose* (rem)	Critical Organ
Americium-241	Water	4×10^{-6}	1.5	Kidney
	Water	5×10^{-6}	3	Bone
Argon-41	Air	4×10^{-8}	0.5	Whole Body
Californium-249	Water	4×10^{-6}	3	Bone
Californium-252	Water	7×10^{-6}	1.5	GI (LLI)
	Water	2×10^{-5}	3	Bone
Curium-242	Water	2×10^{-5}	1.5	GI (LLI)
	Water	2×10^{-4}	3	Bone
Curium-244	Water	7×10^{-6}	3	Bone
Hydrogen-3 (H_2O)	Air	2×10^{-7}	0.5	Whole Body
	Water	3×10^{-3}	0.5	Whole Body
Iodine-131	Air	1×10^{-10}	1.5	Thyroid
Neptunium-237	Water	3×10^{-6}	3	Bone
Plutonium-238	Water	5×10^{-6}	3	Bone
Plutonium-239	Water	5×10^{-6}	3	Bone
Strontium-90	Water	3×10^{-7}	3	Bone

*The concentrations and doses are the Radiation Protection Standards specified in AEC/ERDA Manual Chapter 0524 for individuals in uncontrolled areas. (5)

dilution at this point is so great that the dose calculation is meaningless, a hypothetical calculation is made assuming Sawmill Creek water is ingested. No other exposure pathways are significant.

Argon-41 and hydrogen-3 (in the form of tritiated water) from the CP-5 reactor represent the major portion of the gaseous radioactive effluent released from the Laboratory. The concentrations and dose rates, as a function of distance from CP-5, were calculated for these two nuclides by a computer program based on an atmospheric dispersion model.⁽¹⁵⁾ The following parameters were used in the calculations:

- a) release rates (measured in the CP-5 exhaust stack): argon-41 1.46 Ci/MW-hr; hydrogen-3, 0.051 Ci/hr
- b) meteorological data: the 15-year average values given in ANL-7084⁽³⁾
- c) the usual parameters for stack height, building, wake, plume momentum, temperatures, etc.

The calculations were carried out to 50 miles (80 km). The argon-41 results for the first three miles (4.8 km) are given in Table 23. Doses were calculated for the mid-point of the annular interval. Thus, the dose for 0-1 mile average is the dose at 0.5 mile (0.8 km). The highest dose rates are in the N to ENE sectors. The closest full-time residents in this area are 1.3 miles from the reactor, where the dose (in the NNE direction) is 3.5 mrem/yr, less than 1% of the standard (500 mrem/yr) for individuals in uncontrolled areas and within the "as low as practical" guidelines of 5 mrem/yr proposed for light-water-cooled reactors. A small section of the most-used portion of the Waterfall Glen Forest Preserve, the former Rocky Glen Forest Preserve southeast of the site, begins about 1,500 m from CP-5, and in this direction the dose rate at 1,500 m is 3.2-3.7 mrem/yr.

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

TABLE 23

Argon-41 Radiation Dose From CP-5 Reactor
(millirem/year)

Sector	1500 m	Distance		
		0-1 mi	1-2 mi	2-3 mi
N	5.3	13.1	2.5	1.0
NNE	6.1	15.2	2.8	1.2
NE	4.8	11.9	2.3	0.8
ENE	5.1	12.8	2.3	1.0
E	4.0	10.1	1.8	0.8
ESE	3.8	9.5	1.8	0.7
SE	3.7	9.1	1.7	0.7
SSE	3.2	7.6	1.5	0.7
S	3.3	8.3	1.5	0.7
SSW	3.8	9.5	1.8	0.7
SW	3.3	8.1	1.5	0.7
WSW	2.8	7.0	1.3	0.5
W	2.2	5.3	1.0	0.3
WNW	2.3	5.8	1.2	0.5
NW	2.8	6.8	1.3	0.5
NNW	3.3	8.1	1.5	0.7

from a waste storage facility and a dynamitron accelerator in the vicinity, but in all other directions, including the predominant wind direction to the north-northeast, the dose rates were in the normal range found off-site, 99 ± 17 mrem/yr. Increases in excess of two standard deviations of the off-site average (i.e., greater than 116 mrem/yr) would have been recognized as abnormal, and on this basis the dose from argon-41 at the site boundary was less than about 15 mrem/yr. Thus, the calculated doses and those measured by TLD agree within the ability of the TLD system to detect above-normal doses.

The population data in Table 1 was used to calculate the population dose from argon-41. The results are given in Table 24, together with the average individual dose. For comparison, the table also gives the estimated natural external radiation dose, which was calculated with the assumption that the average off-site, outdoor, radiation dose measured by TLD applies to the entire area within a 50-mile radius. There are no full-time residents within 1 mile of the CP-5 exhaust stack. The maximum dose to full-time residents occurs in the 1-2 mile annulus in the NNE direction, where individuals would receive 5.4 mrem/yr if they were outdoors throughout the year at 1 mile and 1.5 mrem/yr, outdoors at 2 miles.

TABLE 24

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

Distance (miles)	Population	Dose			
		man-rem/year		Avg. mrem/year	
		Argon-41	Natural	Argon-41	Natural
0-1	0	0	0	0	0
1-2	1,900	3.3	190	1.74	100
2-3	8,131	5.8	813	0.72	100
0-50	7,757,789	156	7.76×10^5	0.020	100

The dose rates due to hydrogen-3 from CP-5, calculated in the same manner as for argon-41, are as follows. The maximum dose at 1,500 m, in the NNE direction, is 0.014 mrem/yr. The maximum individual exposure to full-time residents in the area occurs in the 1-2 mile annulus in the NNE direction,

where the annual dose is calculated to be 0.008 mrem/yr. A brief summary of the results is given in Table 25. A comparison of the doses calculated from the meteorological model with the measured data from Table 8 is given in Table 26. In both cases concentrations are converted to dose as described earlier. The agreement is very good in view of the large number of variables and parameters involved in obtaining both the calculated and measured values.

TABLE 25

Distance (miles)	Population	man-rem/year	Avg. mrem/year
0-1	0	0	0
1-2	1,900	0.0082	0.0043
2-3	8,131	0.0179	0.0022
0-50	7,757,789	1.37	0.00018

TABLE 26

Direction	Distance (km)	Calculated (mrem/yr)	Measured (mrem/yr)
NNE	1.5	0.014	0.013
ENE	1.9	0.0090	0.010

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

The only locations where radioactivity attributable to Argonne operations could be found in off-site water was Sawmill Creek below the waste-water out-fall. The concentrations of those nuclides added to Sawmill Creek by Argonne waste water, and the corresponding dose rates if water at these concentrations were used as the sole water supply by an individual are given in Table 27.

In the case of strontium-90 and hydrogen-3, about 50% and 95%, respectively, of these nuclides were contributed by Argonne, but the total concentration is included in the dose calculation since the total exposure is the figure of interest, regardless of source. For the other nuclides, essentially all of the activity may be attributed to Argonne. The dose rates were all well below the standards for individuals in uncontrolled areas. It should be emphasized that Sawmill Creek is not used for drinking or recreational purposes. There are very few fish in the stream, and they do not constitute a significant source of food for any individual.

TABLE 27

Radioactivity Concentrations and Dose Estimates
for Sawmill Creek Water, 1974

Nuclide	Conc. (avg.) 10^{-9} μ Ci/ml	Dose Rate mrem/year	Percent of Standard
Hydrogen-3	2140	0.36	0.07
Strontium-90	1.1	11	0.4
Neptunium-237	0.19	0.19	0.006
Plutonium-239	0.018	0.011	0.0004
Americium-241	0.006	0.0023 (kidney) 0.0036 (bone)	0.0002 0.0001

As indicated in Table 9, occasional Creek samples (less than 10%) contained traces of plutonium-238 and transplutonium elements, but the averages were only slightly greater than the detection limit. The annual dose due to consuming water at these concentrations can be calculated as was done for those nuclides more commonly found in Creek water, but it should be noted that the method of averaging probably exaggerates the true concentration. These annual doses are: 1.7×10^{-3} mrem/yr for plutonium-238 and from 1.5×10^{-3} to 2.0×10^{-4} mrem/yr for the transplutonium isotopes.

b. External Penetrating Radiation

The results of external penetrating radiation measurements are given in Section III.A.5. The only above-normal dose rates from Argonne operations

were found at the south boundary adjacent to the Waste Storage Facility. At this fence line the dose from Argonne is 184 mrem/yr. The dose at any distance can be calculated on the basis of 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. The closest residents to the boundary are about 1.6 km south, and at this distance the calculated dose rate is 0.0046 mrem/yr, if the energy of the radiation were 0.66 MeV, and 0.023 mrem/yr, if the energy were 1.3 MeV. The energy spectrum of the radiation is not known, so it is necessary to assume an energy to make the calculations. Since cesium-137 and cobalt-60 are common radionuclides, the energies of the gamma-rays from these nuclides were used in the calculations.

At the fence between the site boundary and the Cemetery (8H) the maximum dose attributable to Argonne is 11 mrem/yr. An individual spending 1 hour/week at this location would then receive an annual dose of 0.065 mrem from Argonne operations.

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. 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.⁽¹⁶⁾ The doses at the south fence (7J) are about 36% of the 500 mrem/yr limit to individuals, and 108% of the 170 mrem/yr limit to the "suitable sample". However, the area south of the site boundary is heavily wooded, and the land rises steeply from the Des Plaines River. As a result, the area is relatively inaccessible and no individuals frequent this location on a regular basis. The penetrating radiation dose to individuals from Argonne operations, either those visiting the Cemetery or living south of the site, is well within all proposed radiation protection standards, including the "as low as practicable limit" of 5 mrem/yr.

B. Chemical Pollutants

The nonradioactive environmental data contained in this report have been collected in an effort to ascertain Argonne compliance with State of Illinois regulations with regard to general use stream quality and effluent criteria.

The standards used are those adopted by the State of Illinois and approved by the Federal Government in 1972.⁽¹⁷⁾ The new regulations represent a comprehensive and well-documented statement with regard to the quality of the aquatic sector.

The concentrations of barium, chromium, copper, iron, nickel, and zinc were determined using conventional atomic absorption spectrophotometry. The concentrations of silver, cadmium, and lead were determined using a flameless technique. This consists of adding the sample directly to a tantalum ribbon and drying, ashing, and atomizing the desired elements under an argon atmosphere. Mercury was determined using cold atomic absorption spectrophotometry. Fluoride levels were determined using an ion selective electrode, and the pH was determined using conventional pH electrode measurements. Dissolved oxygen, ammonia nitrogen, and hexavalent chromium levels were determined using procedures described in Standard Methods.⁽¹⁸⁾ The levels of fecal coliform were determined using the membrane filter technique.⁽¹⁸⁾ The cyanide concentration was estimated with a Hach Chemical Co. cyanide test kit. Beryllium was determined fluorophotometrically as previously described.⁽¹⁹⁾

The results of the measurement of chemical constituents are expressed as milligrams per liter (mg/l) except as indicated. Averages were calculated as described in Section III.A. Yearly average values 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 average. Only when the sample concentrations are random does this value approach the actual distribution occurring at the sampling location. In some instances it appears that the measurements do represent a natural background value and the variation is representative of climatic conditions.

All of the results are compared to the appropriate State limits, which are listed in Table 28. Minimum detectable amounts are included for comparison. The detection limits for the atomic absorption methods represent twice the background variation, which is commonly used for this purpose. Detection limits for ion selective methods are those listed by the manufacturer, since they depend entirely on solubility considerations.

As in the past, the major emphasis has been placed on Sawmill Creek, which is tributary to the Des Plaines River, since this is the principal route for waste water leaving the Argonne site. However, a large effort was

TABLE 28

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

Constituent	Applicable Water Type		Detection Limit
	Stream	Effluent	
Ammonia Nitrogen (as N)	1.5	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	0.1
Barium (total)*	5.0	2.0	1.0
Cadmium (total)	0.05	0.15	0.0002
Chromium (total hexavalent)	0.05	0.3	0.006
Copper (total)	0.02	1.0	0.005
Cyanide	0.025	0.025	0.020
Fluoride	1.4	2.5	0.02
Iron (total)	1.0	2.0	0.08
Lead (total)	0.1	0.1	0.0015
Mercury (total)	0.0005	0.0005	0.0001
Nickel (total)	1.0	1.0	0.2
pH	6.5-9.0	5.0-10.0	-
Silver (total)	0.005	0.1	0.0002
Zinc (total)	1.0	1.0	0.01

* Total limits apply to samples not filtered before treatment. All other limits apply to filtered samples.

devoted to determining both the sources of and the source strengths of mercury and hexavalent chromium in liquid wastes, since past data had indicated that these were constituents of concern.

1. Sawmill Creek

The majority of samples collected were grab samples obtained in the early afternoon. For three months a continuous sampler was operative and a 24-hour composite was obtained. Since a different sampling technique is required, samples to be analyzed for dissolved oxygen and ammonia nitrogen were collected separately. A specially-designed collection bottle was utilized. Samples to be analyzed for fecal coliform were collected on a somewhat random basis with an effort to obtain five samples per month, which is the minimum State requirement for levels previously encountered in these waters.

The values obtained for the pertinent parameters are listed in Table 29. Other criteria not listed are dissolved oxygen and pH, since these levels are expressed somewhat differently. The State law provides that the oxygen levels of a stream shall not be less than 5 mg/l at any time. The oxygen levels above the outfall averaged 11.3 mg/l and ranged from a minimum of 7.2 to a maximum of 14.4 mg/l. The oxygen levels below the outfall averaged 10.6 mg/l and ranged from a minimum of 8.0 mg/l to a maximum of 12.7 mg/l. This indicates a significant improvement in the quality of the water entering the Laboratory site as compared to past years. The oxygen saturation of the water frequently exceeded 100%, indicating photosynthetic activity was of major importance.

The pH criteria give a range of 6.5-9 units and these results are not subject to averaging. The pH levels measured below the waste-water outfall were in the range from 7.7 to 8.6 and above the outfall in the range from 7.9 to 8.8, indicating no significant effect of the outfall water on the Creek.

The average ammonia nitrogen levels are, for the first time, below the State limit of 1.5 mg/l both above and below the Argonne outfall. The average level above the outfall was 1.1 mg/l and below the outfall was 1.0 mg/l. The general decrease in ammonia nitrogen levels is a further indication of improvement in the water quality of the Creek. Very low levels of ammonia nitrogen occurred when oxygen supersaturation was present and higher levels occurred when cold weather prevailed even though oxygen levels were adequate. This

TABLE 29

Chemical Constituents in Sawmill Creek, 1974

Constituent	Location *	No. of Samples	Concentration (mg/l)			Percent State Levels		
			Av.	Min.	Max.	Av.	Min.	Max.
Barium	7M	52	2.59 ± 0.19	1.24	4.45	51.8	24.8	89
Beryllium**	7M	12	0.114 ± 0.050	0.015	0.334	-	-	-
Cadmium**	7M	52	1.83 ± 0.29	0.4	4.4	3.7	0.8	8.8
Chromium(VI)	7M (up)	51	0.009 ± 0.002	0.006	0.035	18	12	70
	7M (down)	152	0.03 ± 0.01	0.006	0.426	60	12	852
Copper**	7M	52	11.1 ± 1.6	5	29	55.5	25	145
Cyanide	7M	37	-	-	< 0.02	-	-	< 80
Fluoride	7M	52	0.40 ± 0.04	0.21	0.69	28.6	15	49.3
Iron	7M	52	0.53 ± 0.10	0.11	1.47	53	11	147
Lead**	7M	52	17.2 ± 7.0	0.5	101	17.2	0.5	101
Mercury**	7M (up)	51	-	-	< 0.1	-	-	< 20
	7M (down)	152	0.16 ± 0.03	0.05	2.15	32	10	430
Nickel	7M	52	-	-	< 0.2	-	-	< 0.2
Silver**	7M	52	1.80 ± 0.46	0.1	9.2	36	2	184
Zinc	7M	52	0.14 ± 0.05	0.05	1.3	14	5	130

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

** All concentration values multiplied by 10^3 .

increase in ammonia nitrogen with decreasing temperature is commonly observed and is related to the very high temperature coefficient of solubility of ammonia in water.

The average levels for barium, beryllium, copper, fluoride, iron, nickel, and zinc during this report period remained essentially the same as for 1973. Levels for cadmium are included for the first time in several years and the value of 1.8 $\mu\text{g}/\text{l}$ is essentially the same as for previous report periods. The average value for lead is 50% higher than the level reported in 1972.⁽²⁾ The reason for this increase can be related to the initial results obtained by direct flameless analysis which tended to give high and variable results. The reasons for this were not defined, but an extraction step prior to analysis produced good precision and improved accuracy. Hence, the values from the major portion of the year are essentially the same as previously reported.

The large improvement in the sensitivity of the silver analysis provided positive results for the majority of the samples analyzed. The average value of 1.8 $\mu\text{g}/\text{l}$ is indicative of possible contamination from a film processing area. The waste from this source does not enter the Creek by way of the waste treatment plant, but in the stream at 11L. However, the average value was well below the State limit and this limit was exceeded only rarely.

The average mercury level for the year 1974 was approximately 40% of the State limit and represented a reduction from 1973 of about a factor of three. This reduction is the result of efforts to reduce effluent levels by suitable treatment before release. In addition to reducing the average levels, the maximum level in any one sample was about four times the State limit in 1974 as compared to 25 times the limit in 1973.

The average value for hexavalent chromium at 7M downstream was similar to the level found in 1973. The average upstream of the outfall at 7M was greatly reduced. This reduction is probably related to the fact that samples were obtained for 12 months in 1974 as contrasted with five months in 1973. These five months contained disproportionate numbers of times when cooling was required, and zinc chromate is used as an anti-oxidant in the cooling towers. Average levels at both sampling points are below the State limits.

Quality standards for beryllium do not exist, but its use at the

Laboratory and its toxicity make its measurement desirable. The levels obtained are considered to be from natural sources.

2. Effluent Water

The principal effluent from Argonne is water from the waste treatment plant which enters Sawmill Creek at 7M. This water is sampled by a proportional collector at the waste treatment facility immediately after treatment. The water actually enters the Creek some 1,500 meters downstream of this point. Samples are obtained on a 24-hour basis from Monday through Friday and they are combined into one sample, for all analyses except for mercury and hexavalent chromium. Analyses for these two elements are performed on individual samples. Additionally, the land between the site security fence and the former site boundary has been released to the Du Page County Forest Preserve District. The cooling tower blowdown channels at 14J, 11L, and 8J thus leave the controlled area and should be considered effluents. The latter usually does not reach the Des Plaines River, but instead soaks into a marsh. The water from 14J and 11L enters Sawmill Creek, and its contribution is assessed in the Creek samples. Grab samples were obtained weekly at 8J, 11L, and 14J, and they were analyzed for chromium immediately.

The results obtained in the waste treatment plant effluent samples (Table 30) are comparable to the Creek results with five exceptions, zinc, chromium, mercury, copper, and iron. The levels of zinc are about two times higher in the plant effluent than in the Creek and this is probably related to zinc chromate used in the cooling towers. The levels in the effluent averaged 25% of the State limit of 1.0 mg/l, but never exceeded it.

The average value for hexavalent chromium was somewhat higher than the Creek value but well below the effluent limit of 0.3 mg/l. The plant effluent was sampled for the entire year and the average value is about 20% less than last year. This report covers the whole of the cooling season, when chromium usage is maximum.

Sampling of the cooling tower blowdown channels covered all of the year. The cooling tower blowdown channel at 14J averaged 0.32 mg/l and exceeded the effluent limit of 0.3 mg/l about 40% of the time. This stream enters Sawmill Creek at a location above the Argonne outfall and its contribution to the overall level of hexavalent chromium is assessed by the Creek sample. Since

TABLE 30

Chemical Constituents in Effluent From ANL Treatment Plant, 1974

Constituent	No. of Samples	Concentration (mg/l)			Percent of Standard		
		Av.	Min.	Max.	Av.	Min.	Max.
Barium	56	1.74 ± 0.14	0.5	2.77	87	25	139
Beryllium*	12	0.066 ± 0.019	0.022	0.105	-	-	-
Cadmium*	56	1.66 ± 0.16	0.8	3.3	1.1	0.5	2.2
Chromium(VI)	253	0.038 ± 0.004	0.006	0.219	12.7	2.0	73
Copper*	56	55 ± 10	20	280	5.5	2.0	28
Fluoride	56	0.37 ± 0.03	0.21	0.62	14.8	8.4	24.8
Iron	56	0.13 ± 0.01	0.06	0.30	6.5	2.4	15.0
Lead*	56	13 ± 4	1	58	13.0	1.0	58.0
Mercury*	253	0.50 ± 0.05	0.13	4.77	100.0	26.0	954
Nickel	56	-	-	< 2	-	-	< 20
pH	253	7.51	7.03	8.10	-	-	-
Silver*	56	1.3 ± 0.4	0.1	6.0	1.3	0.1	6
Zinc	56	0.25 ± 0.02	0.15	0.62	25	15	62

*All concentration values multiplied by 10³.

the volume of water flowing in this stream is low with respect to Sawmill Creek flow, a significant increase in the levels of chromium in the Creek was not produced. The cooling tower blowdown at 8J averaged 0.25 mg/l, which is about a factor of four less than in 1973. This reduction was the result of a large scale diversion of blowdown water to the Creek from the ZGS cooling tower complex. The contribution to the Creek by this diversion was measured at the 7M upstream station. To assess the effect of this source as well as any others on the Des Plaines River, samples were obtained in the River at Willow Springs (upstream) monthly and at Lemont (downstream) bi-monthly. The average hexavalent chromium levels were less than 0.01 mg/l at both sites. These results are comparable to results obtained in previous years.

Waste water which could contain mercury was very carefully monitored before release. When excessive levels were found, the waste was treated until the mercury content was reduced to acceptable values. As a result of these controls, the average mercury level in the effluent water was reduced from about 2 $\mu\text{g/l}$ in 1973 to 0.5 $\mu\text{g/l}$ in 1974, which is the State limit. Additional studies are planned to further reduce mercury levels in the effluent.

The copper levels in the effluent were five times the levels found in the Creek, which could be due to the use of copper tubing in Laboratory systems. However, these values are well below the State of Illinois limit. Average levels of iron in Sawmill Creek were much higher than effluent values. This reflects the effect of suspension of solids during periods of precipitation and is unrelated to dissolved iron levels.

Fecal coliform studies were conducted in the Creek during all of 1974. The samples were analyzed using the membrane filter technique. Values obtained above the Argonne outfall ranged from < 2 coliforms/100 ml to 237 coliforms/100 ml. The results are expressed as the monthly geometric mean. Results in the outfall as it enters the Creek ranged from < 1 coliform/100 ml to 11 coliforms/100 ml. The State limit is 200 coliforms/100 ml.

The Argonne Sewage Treatment Plant effluent was sampled and analyzed by the Reclamation Control Laboratory of the Plant Operations Division. Twice weekly samples were analyzed for biochemical oxygen demand (B.O.D.), suspended solids, and ammonia nitrogen content. Each sample was a composite of eight separate grab samples taken approximately once per hour. The analyses were

performed as outlined in Standard Methods.⁽¹⁸⁾ Results are as shown in Table 31.

TABLE 31

Oxygen Balance and Coliform Measurements
in Argonne Effluent Water, 1974

Month	Concentration (mg/l)		
	B.O.D. ₅	Suspended Solids	Ammonia Nitrogen
January	1.8	1.00	1.94
February	2.4	6.62	2.26
March	1.5	1.85	1.47
April	1.6	7.50	0.05
May	1.1	7.1	1.55
June	1.1	3.5	0.46
July	0.8	1.75	1.19
August	1.0	1.56	0.46
September	0.9	1.43	0.55
October	1.5	3.00	1.18
November	1.2	4.25	1.32
December	1.2	0.40	1.42
State Limit	30.0	37.0	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)

The average value for ammonia nitrogen did not exceed the State limits at any time. The values for B.O.D. and suspended solids were also well below State regulations during the entire period.

IV. APPENDIX

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

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

All of the standards and detection limits for chemical constituents, and some of those for radionuclides, are given in the main body of the report. In addition, in Table 32 are collected the air and water environmental quality standards and detection limits (minimum detectable amounts) for all radionuclides, and for those materials, for which measurements were made. These standards are the Concentration Guides given in AEC/ERDA Manual Chapter 0524, and are used in this report to assess the hazard of a measured concentration of a radioactive nuclide. Chapter 0524 distinguishes two CGs, one for occupational exposure in controlled areas and one for uncontrolled areas, beyond the site boundary where individuals can be exposed, nonoccupationally, for 168 hours per week. Both CGs are given in the table for nuclides released by

TABLE 32

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

Nuclide or Activity	Concentration Guide		Detection Limit	
	Water	Air	Water	Air
Americium-241	4×10^{-6}	-	1×10^{-12}	-
Antimony-125	-	9×10^{-10}	-	5×10^{-16}
Argon-41	-	4×10^{-8} 2×10^{-6}	-	1.5×10^{-8}
Barium-140	3×10^{-5}	1×10^{-9}	2×10^{-9}	5×10^{-16}
Beryllium-7	-	4×10^{-8}	-	5×10^{-15}
Californium-249	4×10^{-6}	-	1×10^{-12}	-
Californium-252	2×10^{-5}	-	1×10^{-12}	-
Cerium-141	-	5×10^{-9}	-	5×10^{-16}
Cerium-144	-	2×10^{-10}	-	1×10^{-15}
Cesium-137	2×10^{-5}	5×10^{-10}	-	5×10^{-16}
Cobalt-60	-	3×10^{-10}	-	1×10^{-16}
Curium-242	2×10^{-5}	-	1×10^{-12}	-
Curium-244	7×10^{-6}	-	1×10^{-12}	-
Hydrogen-3	3×10^{-3}	2×10^{-7} 5×10^{-6}	2×10^{-7}	1×10^{-13}
Iodine-131	3×10^{-7}	1×10^{-10}	3×10^{-9}	5×10^{-15}
Manganese-54	-	1×10^{-9}	-	5×10^{-16}
Neptunium-237	3×10^{-6}	-	2×10^{-12}	-
Plutonium-238	5×10^{-6}	1×10^{-12}	2×10^{-12}	1×10^{-19}
Plutonium-239	5×10^{-6}	1×10^{-12}	5×10^{-13}	1×10^{-19}
Ruthenium-103	-	3×10^{-9}	-	5×10^{-16}
Ruthenium-106	-	2×10^{-10}	-	1×10^{-15}
Strontium-89	3×10^{-6}	3×10^{-10}	1×10^{-9}	1×10^{-16}
Strontium-90	3×10^{-7}	3×10^{-11}	2×10^{-10}	1×10^{-17}
Thorium-228	-	2×10^{-13}	-	1×10^{-18}
Thorium-230	-	3×10^{-13}	-	1×10^{-18}
Thorium-232	-	1×10^{-12}	-	1×10^{-18}
Uranium - natural	4×10^{-5}	4×10^{-12}	2×10^{-10}	2×10^{-17}
Zirconium-95	-	1×10^{-9}	-	5×10^{-16}
Alpha *	3×10^{-6}	1×10^{-10}	2×10^{-10}	2×10^{-16}
Beta *	1×10^{-7}	1×10^{-13}	1×10^{-9}	5×10^{-16}

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

Argonne and detected in air on the site, and the larger of the two values applies to controlled areas. Where only one is listed, the CG is for uncontrolled areas. For water the standard selected was for the soluble form of the radionuclide; for air the standard for the insoluble form was selected (except for iodine-131, for which the soluble form was selected as a more conservative standard).

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

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