

**ENVIRONMENTAL MONITORING AT
ARGONNE NATIONAL LABORATORY**

ANNUAL REPORT FOR 1973

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



U of C-AUA-USAEC

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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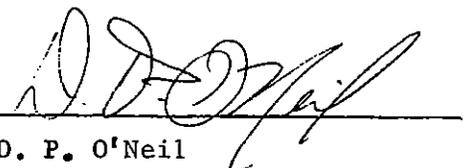
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ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1973 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 both on and off the Argonne site for comparison purposes. 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.

I. INTRODUCTION

A. General

This report is prepared to provide the AEC 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 format prescribed in AEC 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 substance 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 basic research activities, 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. Most of the basic research is concerned with the application of radiation as a tool in the physical and life sciences. The reactor effort is devoted largely to development of the liquid-metal-cooled fast-breeder power reactor. Some of the energy-related problems are the removal of sulfur dioxide and other pollutants from the combustion products of coal and the development of electric batteries for vehicles. 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; a 12.5 GeV proton accelerator, the Zero Gradient Synchrotron (ZGS); two critical assemblies, or zero power reactors (ZPR-6 and -9), that are fueled at various times with plutonium, uranium, or a combination of the two; a 60-inch cyclotron; several Van de Graaff accelerators; a fuel fabrication facility designed for plutonium usage; 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-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.^(2,3)

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 1 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 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

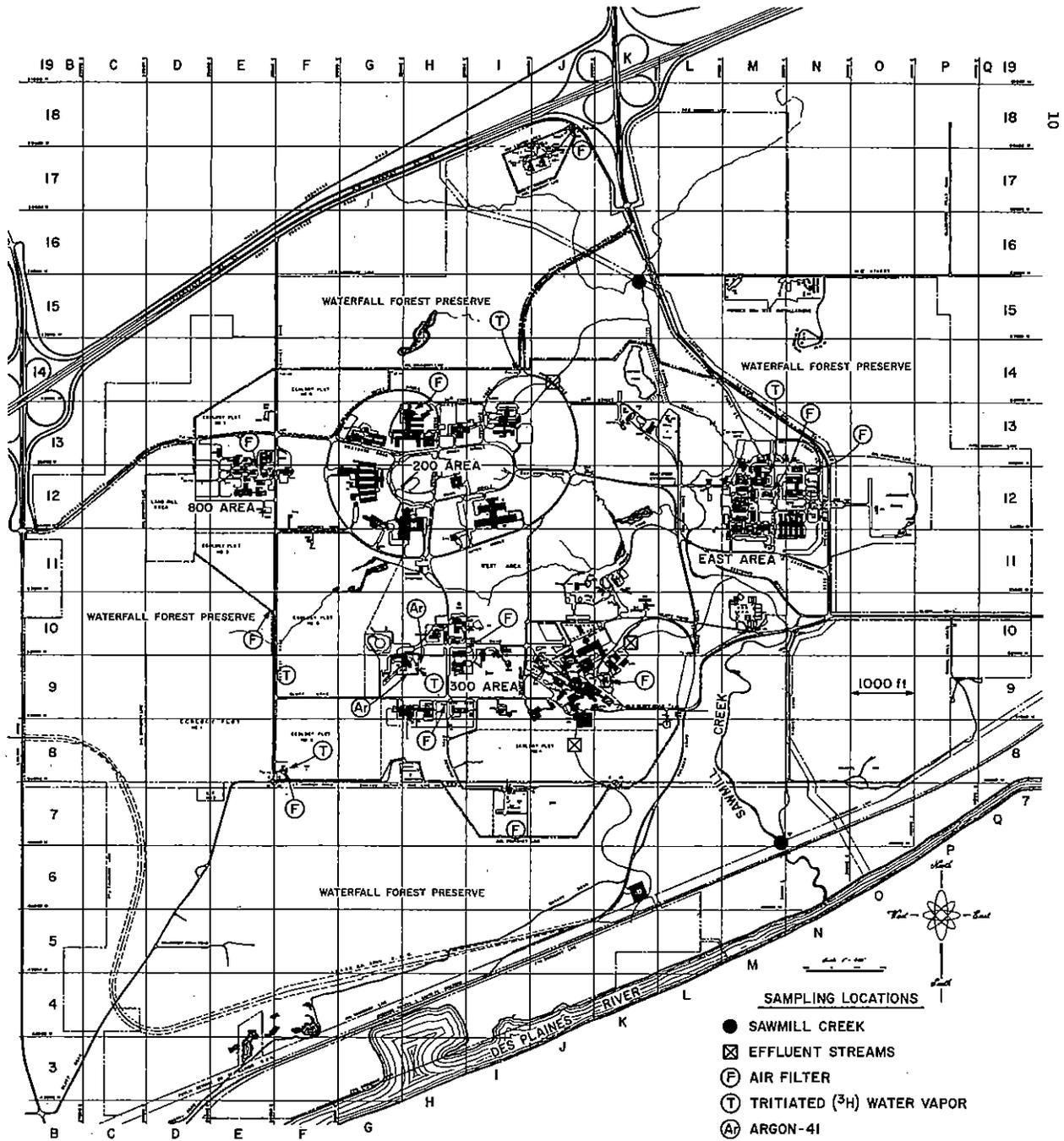


Fig. 1. Sampling Locations on the Site of Argonne National Laboratory

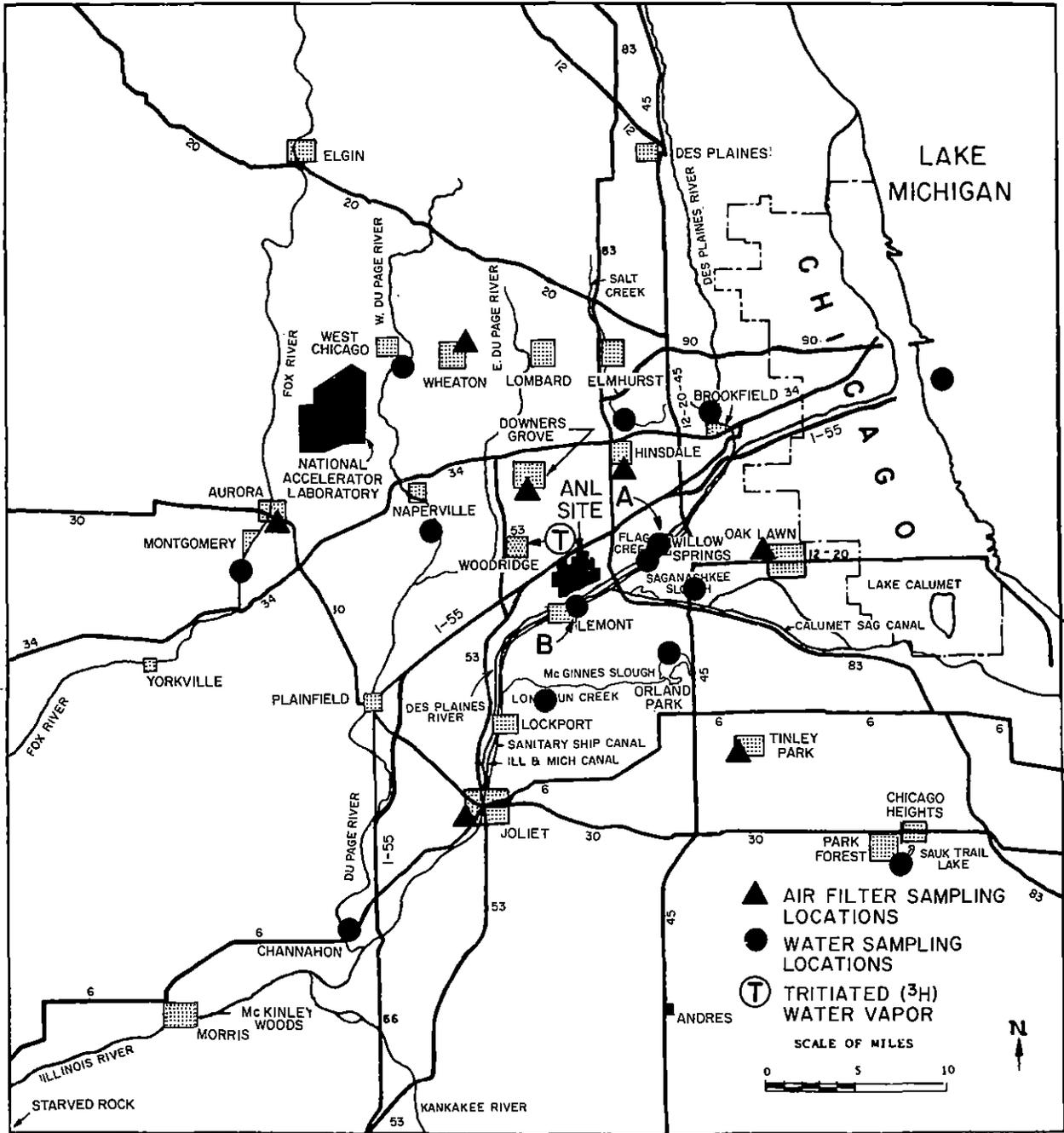


Fig. 2. Sampling Locations near Argonne National Laboratory

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.⁽⁴⁾ 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 15-year 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. 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

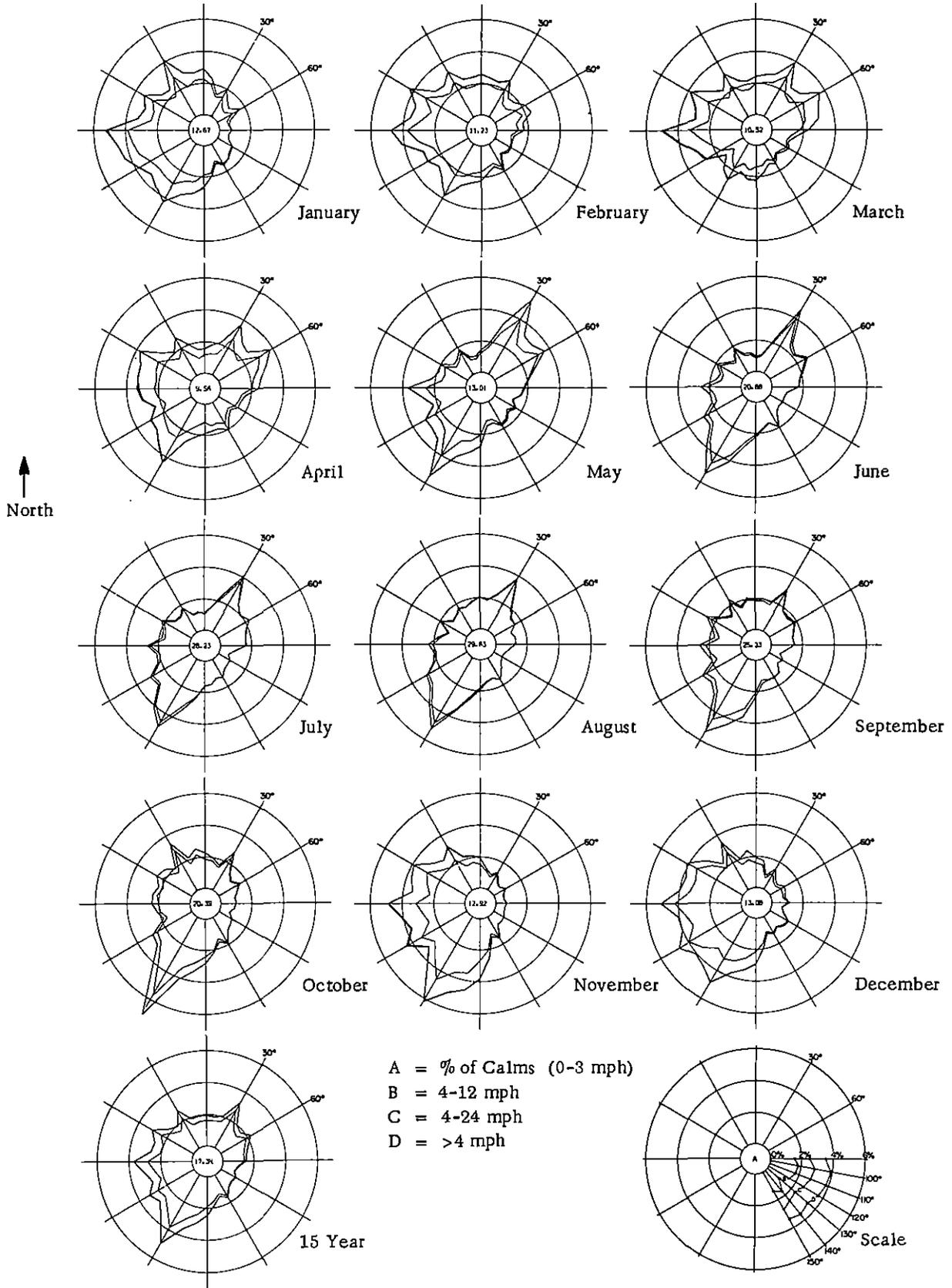


Fig. 3. Wind Roses Based on All Hourly Observations. January 1950-December 1964. 19-foot Level

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.

D. Population

The area around Argonne has exhibited a large growth in suburban, exurban, and rural housing. Large areas of farmland have been converted into housing. The population of communities and townships in the area are shown in Tables 1 and 2. Their relationship to the Argonne site is shown in Figure 4. The dotted township lines in the figure are at six-mile intervals. Two housing developments border the northeast portion of the new Forest Preserve that was formerly part of the site: Rosewood Trace, 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.

A population rose for the area is shown in Table 3. The table was prepared from the 1970 U. S. census figures by the Environmental Information System Office at Oak Ridge National Laboratory. The figures give the population in annular segments at various site distances in a 22.5° sector centered on the direction listed. The zero point for the data is the Argonne Meteorology Tower located in the southwest portion of coordinate 8F in Figure 1. The tower position is 41.70° north latitude and 87.98° west longitude. Some of the figures in the table for the first 4 miles have been changed, based on inspection of the actual area and of maps, since they did not appear to be current.

TABLE 1

Population of Communities Near
Argonne National Laboratory

(Based on 1960 and 1970 Census)

COMMUNITY	POPULATION		COMMUNITY	POPULATION	
	1960	1970		1960	1970
ADDISON	6741	25230	MATTESON	3225	4753
ALSIP	3770	10804	MAYWOOD	27330	30152
BEDFORD PARK	737	1506	MC COOK	441	441
BELLWOOD	20729	22108	MELROSE PARK	22291	23471
BERKELEY	5792	6130	MIDLOTHIAN	6605	16050
BERWYN	54224	52274	MOKENA	1332	1623
BLUE ISLAND	19618	22986	NAPERVILLE	12933	22426
BOLLINGBROOK	--	6476	NEW LENOX	1750	2817
BRIDGEVIEW	7334	17079	NORTHLAKE	12318	13644
BROOKFIELD	20429	20273	NORTH RIVERSIDE	7989	10373
BURR RIDGE	299	4853	OAK BROOK	324	4099
CAROL STREAM	836	3755	OAK BROOK TERRACE	1121	1122
CHICAGO	3550404	3322855	OAK FOREST	3724	15919
CHICAGO RIDGE	5748	9086	OAKLAWN	27471	61637
CICERO	69130	66695	OAK PARK	61093	61745
CLARENDON HILLS	5885	6715	OLYMPIA FIELDS	1503	3473
COUNTRY CLUB HILLS	3421	6827	ORLAND PARK	2592	6444
COUNTRYSIDE	2393	2886	PALOS HEIGHTS	3775	8083
CREST HILL	5887	16806	PALOS HILLS	3766	5643
CRESTWOOD	1213	5550	PALOS PARK	2169	3265
DARIEN	--	8200	PARK FOREST	29993	30388
DOWNERS GROVE	21154	31509	PLAINFIELD	2183	2900
ELMHURST	36991	46325	POSEN	4517	6742
ELMWOOD PARK	23866	26088	RICHTON PARK	933	2531
EVERGREEN PARK	24178	25476	RIVER GROVE	8464	11239
FLOSSMOOR	4624	7559	RIVERSIDE	9750	10373
FOREST PARK	14452	15391	ROBBINS	7511	9410
FOREST VIEW	1042	1042	ROCKDALE	1272	2021
FRANKFORT	1135	2294	ROMEOVILLE	3574	12580
FRANKLIN PARK	18322	20654	ROSEWOOD TRACE	--	4500
GLENDALE	173	18976	SHOREWOOD	358	1765
GLEN ELLYN	15972	21672	STICKNEY	6239	6665
HAZELCREST	6205	10245	STONE PARK	3038	4233
HICKORY HILLS	2707	7776	SUMMIT	10374	17800
HILLSIDE	7794	8581	TIMBERLAKE	--	2500
HINSDALE	12859	13704	TINLEY PARK	6392	12253
HODGKINS	1126	2265	VILLA PARK	20391	25653
HOMETOWN	7479	6708	WARRENVILLE	1470	3134
INDIAN HEAD PARK	385	485	WESTCHESTER	18092	20190
JOLIET	66780	78623	WEST CHICAGO	6854	9942
JUSTICE	2803	9371	WESTERN SPRINGS	10838	12522
LA GRANGE	15285	18189	WESTMONT	5997	8480
LA GRANGE PARK	13793	15402	WHEATON	24312	30910
LEMONT	3397	5018	WILLOW BROOK	157	4790
LISLE	4219	6635	WILLOW SPRINGS	2348	3344
LOCKPORT	7560	9621	WINFIELD	1575	4277
LOMBARD	22561	36528	WOODRIDGE	542	10907
LYONS	9936	11124	WORTH	8196	11831
MARKHAM	11704	15303	YORK CENTER	--	1704

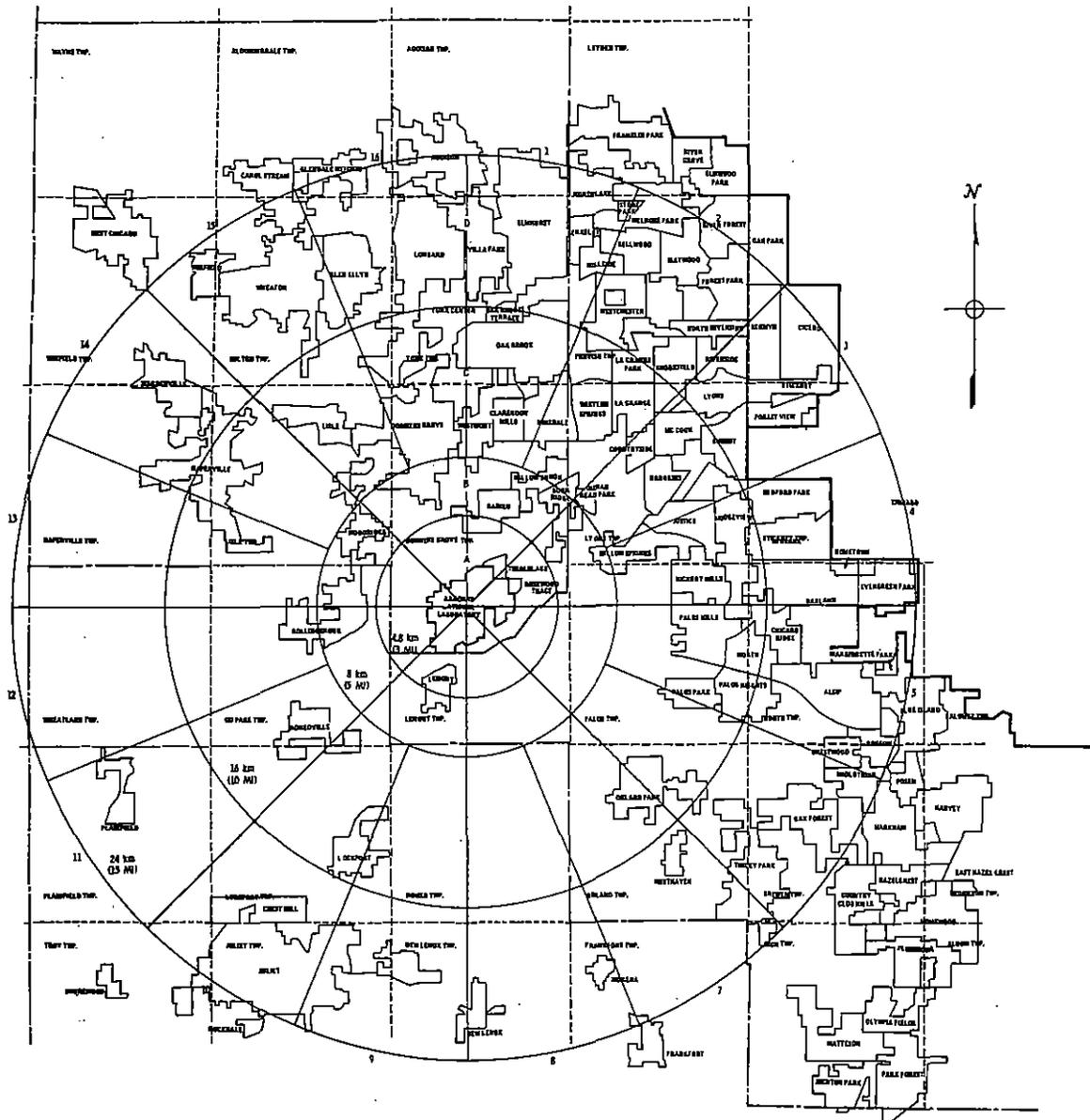


TABLE 2

Population Density by Township

Township	Population 1970 Census	Area sq. miles	Persons per sq. mile
Addison	72,280	36	2,008
Berwyn	52,502	3.25	16,154
Bloom	94,757	36	2,632
Bloomington	36,654	36	1,018
Bremen	93,906	36	2,609
Calumet	23,614	3.86	6,118
Cicero	67,058	5.71	11,743
Downers Grove	92,899	48	1,935
Du Page	20,037	36	556
Frankfort	9,633	36	268
Homer	6,686	36	186
Joliet	96,001	36	2,666
Lemont	8,314	21	395
Lisle	49,061	36	1,363
Lockport	33,354	36	926
Lyons	100,898	36.7	2,749
Milton	75,750	36	2,104
Naperville	13,028	36	362
New Lenox	10,049	36	279
Orland	15,028	36	417
Palos	33,100	35	946
Plainfield	11,028	36	306
Proviso	172,760	30	5,758
Rich	44,801	36	1,244
Riverside	18,475	5.7	3,241
Stickney	41,752	18.1	2,307
Thornton	187,863	36	5,218
Troy	11,568	36	321
Wheatland	1,794	36	50
Winfield	23,001	36	638
Worth	155,834	37	4,211
York	123,724	36	3,437

TABLE 3

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	10	120	0	0	0	10260	24359	4470	13718	7223
WSW	10	120	0	462	0	5535	4201	1795	5560	9992
W	10	120	0	3387	4955	951	38397	16101	16475	7371
WNW	50	120	0	0	0	18162	82536	7700	3981	49352
NW	50	120	0	0	9311	11431	36959	63029	13843	11456
NNW	50	120	1424	1186	3341	33270	92276	110962	85107	63205
Total	180	1720	8131	16826	24156	271765	3194239	3047244	723432	470096
<u>Cumulative</u>										
Total	180	1900	10031	26857	51013	322778	3517017	6564261	7287693	7757789

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 1973, the water flow in the Creek averaged about 8.1 million gallons per day above the Argonne waste-water outfall; Argonne waste-water effluent averaged 0.77 million gallons per day. With the change in the ANL boundary line, three streams that carry blowdown discharge water from the cooling towers have become effluent streams since they now 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; a second 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 streams 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 in 1973 remained the Rocky Glen Forest Preserve (now part of the larger Waterfall Glen Forest Preserve), bordering the site at the southeast corner (Figure 1), and part of the Du Page County Forest Preserve District. The new portion of the forest preserve has received little use thus far. It was available for hiking and skiing in 1973. Its development and increased utilization by the public should begin soon. Sawmill Creek flows through Rocky Glen on its way to the Des Plaines River. This Preserve is used principally for picnicking, hiking, and overnight camping by youth groups. During 1973, 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.

The principal agricultural land use and production in a 3,700 sq. mi. area around the Laboratory is shown by County in Table 4. These data are estimates obtained by the Illinois Crop Reporting Service of the State Department of Agriculture. The corn and soybean estimates are for 1973; the figures for the other crops are 1972 estimates; and the cow inventory was made on January 1, 1973.

TABLE 4

Annual Farm Production Near ANL

County	Corn		Soybeans		Oats		Wheat		Hay		Milk	Beef
	Acres	Bushels	Acres	Bushels	Acres	Bushels	Acres	Bushels	Acres	Bushels	Cows*	Cattle
Cook	17,400	1,432,000	25,300	657,100	2,900	190,800	1,600	67,600	6,700	20,400	1,000	1,100
Du Page	17,500	1,728,300	22,000	637,300	2,200	131,400	1,300	59,100	3,200	9,800	400	400
Kane	99,000	9,777,000	79,500	2,467,900	10,800	710,500	2,000	95,100	17,700	62,600	9,700	3,000
Kendall	78,600	7,681,500	67,800	2,031,700	4,100	253,100	900	40,900	4,200	13,200	900	1,900
Lake	17,900	1,436,300	22,000	483,400	4,200	255,000	3,800	160,600	9,100	24,200	2,900	2,200
Will	140,400	11,121,300	135,900	3,393,700	10,400	578,900	6,100	282,500	14,800	26,600	5,700	4,030

*The annual milk production per cow is estimated to be 10,300 pounds.

II. SUMMARY

This is one in a series of reports prepared to provide the AEC 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-8007. Included in this report are the results of measurements obtained in 1973 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 neutron activation products, plutonium, thorium, and uranium were measured in air-filter samples on and off the site. All of the off-site samples and most of the on-site samples contained only activities from natural sources and nuclear test detonations. The exceptions were the April sample collected near the northeast site boundary, which contained about five times the usual uranium concentration, the result of a small uranium release in the area; and samples collected throughout the year in the central portion of the site, which contained small quantities of barium-140-lanthanum-140, believed to originate in the CP-5 reactor. The barium-140 concentration was equivalent to 0.00012% of the CG* and the elevated uranium concentration in April to 0.02% of the CG. Averaged over the year, the uranium concentration was 0.003% of the CG. Barium-140 from CP-5 was not detected near the site boundary. The detectable activities from nuclear test detonations ranged from 0.001% of the CG for cerium-141 and ruthenium-106 to 0.4% of the CG for mixed beta activity. The average beta activity for the year, 3.6×10^{-14} $\mu\text{Ci/ml}$, was about one-third of that

*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 for uncontrolled areas specified by the U. S. Atomic Energy Commission in Chapter 0524 of the U.S.A.E.C. Manual. These standards are listed in the Appendix, Section V.B.

during 1972, as a result of an overall decrease in fallout from nuclear tests. Fallout from the Chinese test of June 26, was evident in samples collected from July to the end of the year.

The plutonium-239 concentrations in air averaged, respectively, 13×10^{-18} and 10×10^{-18} $\mu\text{Ci/ml}$ on and off the site, about one-half of last year's values and well within the range reported by the AEC New York Health and Safety Laboratory for plutonium from weapons tests. The monthly variations indicated a "spring maximum" similar to that observed in the stratospheric fallout of other radionuclides. The results are consistent with the interpretation that the airborne plutonium is from nuclear tests; there is no evidence that any of the plutonium originated at Argonne. The average plutonium-239 concentration is equivalent to 0.001% of the CG.

Argon-41 and hydrogen-3 represent the major airborne radioactivity released from the Laboratory. The concentration, and corresponding dose rate, of argon-41 at the site boundary in the predominant north-northeast wind direction, calculated from an atmospheric dispersion model, are 3×10^{-10} $\mu\text{Ci/ml}$ and 3.7 mrem/year, respectively, equivalent to 0.75% of the maximum non-occupational dose limit to individuals (500 mrem/year). These values are consistent with penetrating radiation dose measurements made at the site boundary, which yielded an upper limit of 20 mrem/year of argon-41. The dose rate from hydrogen-3 at the site boundary, based on measured hydrogen-3 concentrations, ranged from 0.012 to 0.024 mrem/year, depending on direction; and from 0.006 to 0.014 mrem/year based on meteorological calculations. The agreement is considered good in view of the many variables involved. These dose rates are equivalent to about 0.004% of the annual limit.

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) that were added to the Creek in the waste water were hydrogen-3, neptunium-237, and plutonium-239. The average concentrations of these nuclides in the Creek were equivalent to these percentages of the CGs: 0.06% for hydrogen-3, 0.006% for neptunium-237, and 0.0001% for plutonium-239.

In terms of the CG, the most abundant non-natural nuclide for which analyses were made was strontium-90, present from nuclear tests at an average concentration equal to 0.3% of its CG.

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 in 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 on and off the site. The average plutonium-239 concentrations were $2.7 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ on-site and $2.6 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ off-site; the corresponding plutonium-238 averages were $0.17 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ and $0.22 \times 10^{-3} \mu\text{Ci}/\text{m}^2$. The plutonium-239 content of grass was similar to that found in previous years both on and off the site, 0.1×10^{-6} to $0.3 \times 10^{-6} \mu\text{Ci}/\text{m}^2$, a factor of about $1-2 \times 10^4$ 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 and lagoons on the site was measured. Stream beds contained from 4×10^{-9} to $32 \times 10^{-9} \mu\text{Ci}/\text{g}$ of plutonium-239, a range found in previous years to be normal for fallout plutonium in such materials. Samples from two lagoons contained $62 \times 10^{-9} \mu\text{Ci}/\text{g}$ and $147 \times 10^{-9} \mu\text{Ci}/\text{g}$, respectively. These higher accumulations are believed to have occurred because the slow flow of water through the lagoons, compared to the streams, allows for greater removal of the plutonium from the water.

Milk from a dairy farm near the Laboratory was collected monthly and analyzed for several fission products and hydrogen-3. Short-lived fission products were not detected. Hydrogen-3 concentrations ranged from $< 200 \times 10^{-9} \mu\text{Ci}/\text{ml}$ (the detection limit) to $270 \times 10^{-9} \mu\text{Ci}/\text{ml}$, similar to the range found in surface water. The strontium-90 concentrations were similar to those in 1972, while the cesium-137 concentrations decreased by about a factor of two. The consumption of one liter of milk per day at the average

concentrations would have resulted in an intake of about $< 0.003\%$ of the hydrogen-3, 0.85% of the strontium-90, and $< 0.014\%$ of the cesium-137 Concentration Guides. These radionuclides are present in milk primarily from fallout.

Penetrating radiation measurements were made with thermoluminescent dosimeters on and off the site. The off-site readings averaged 100 mrem/year with a standard deviation of 11 mrem/year, similar to the 1972 average of 105 ± 5 mrem/year, and considered normal for this area. The readings at the site boundary were in the normal range except for three locations along the south boundary (Figure 1). At locations 7I and 7J the dose rates were approximately 320 and 70 mrem/year, respectively, above normal as a result of radiation from an on-site temporary storage facility for radioactive waste. At 8H the dose was about 25 mrem/year above average values, and at this location, contributions to the elevated readings were made by two radiation sources - the waste storage facility and a Van de Graaff generator and other radiation sources in buildings at location 9G/H. The elevated dose rates at these locations, 7I, 7J, and 8H are, respectively, 64%, 14%, and 5% of the 500 mrem/year whole body non-occupational exposure standard for critical individuals. However, these locations are very rarely occupied and there are no individuals at these locations receiving these doses.

The calculated dose rate from these sources to the nearest residents, about 1 mile south of the boundary, was 0.008 mrem/year, if the radiation energy were 0.66 MeV (cesium-137) and 0.04 mrem/year, if the energy were 1.33 MeV (cobalt-60). Such dose rates are considerably less than 1% of the standard for individuals in uncontrolled areas, and well within the 5 mrem/year "as low as practicable guidelines" for light-water-cooled power reactors.

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

Deoxygenating wastes in Argonne effluent water were all within acceptable limits with the exception of ammonia nitrogen levels in February, when

the average was about 1.5 times the State standard of 4 mg/l. Total coliform levels exceeded the State limit in May and July due to construction and changes in the Argonne waste treatment plant. However, corresponding fecal coliform levels in Sawmill Creek for these months were below the limit.

Levels of total mercury in the effluent were about three times the State limit and were very similar to 1972. A study was conducted during the course of the year to isolate and eliminate the source of the contamination. This was accomplished in the latter part of the year and results for December were lower than previous months. All of the other constituents were below State limits with the exception of barium. The effluent barium level was essentially the same as the stream level but a more stringent effluent limit has been adopted. In many cases, natural levels exceed this limit.

Although some individual samples exceeded the State limit for mercury and hexavalent chromium, the average values for all of the chemical constituents in Sawmill Creek were at or below the State limits with the exception of ammonia nitrogen. The levels of ammonia nitrogen found upstream of the ANL outfall are consistently higher than those found below the outfall. The levels encountered this year are significantly lower than those found in the past, reflecting improved waste treatment upstream.

III. ENVIRONMENTAL 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 both on and off the site for comparison purposes. 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. 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.

Where environmental quality standards have been established, the measured concentration or radiation dose is compared with the standard as a means of assessing its hazard. Unless otherwise specified, the standards used in this report are the Concentration Guides and annual dose limits given in AEC Manual Chapter 0524.⁽⁵⁾ These values as well as the minimum detectable amounts are given in the Appendix, Part 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.

The calculation of dose from radionuclide concentrations in air and water is based on the ratio of the concentrations to the appropriate Concentration Guides in AECM 0524. For the nuclides discussed in this report, the Guides and the annual radiation doses that would result from continuous exposure at these concentrations are given in the Appendix, Part B. The dose values are the Radiation Protection Standards for individuals in uncontrolled areas. These standards are 10% of the occupational dose permitted by the ICRP,⁽⁶⁾ and the Concentration Guides are, generally, 10% of the ICRP maximum permissible concentration for 168 hours per week exposure. The only exception in the table is strontium-90. For this nuclide, the ICRP concentration is 4×10^{-6} $\mu\text{Ci/ml}$, while the 0524 Concentration Guide is 7.5% of this value, or 3×10^{-7} $\mu\text{Ci/ml}$.

A. Radioactivity in 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 at nine locations on the Argonne site and at six locations off the site with an asbestos-cellulose filter paper. At one location on the site 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 on-site and off-site activities. The on-site results are included because the comparison between on- and off-site concentrations is necessary in evaluating and establishing the normal environmental concentration. If only off-site concentrations were reported, their normality or origin could not be evaluated. Higher activities on the site may indicate radioactivity released by Argonne if the differences are greater than the error in sampling and counting, and 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 5. 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 6. The gamma-ray measurements were made with a spectrometer system that utilized a shielded 35 cm³ lithium-drifted germanium diode as the detector. This detector was calibrated for each gamma-ray emitting nuclide given in Table 6.

The total alpha and beta activities were very similar on 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 1972 and were in their normal range. About 90% of the gamma-ray

TABLE 5

Total Alpha and Beta Activities in Air-Filter Samples, 1973*

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	on-site	35	2.0	0.8	7.4	0.038	0.021	0.12
	off-site	22	2.2	0.8	4.8	0.034	0.015	0.063
February	on-site	31	2.1	0.9	4.1	0.034	0.026	0.046
	off-site	22	2.4	1.0	7.5	0.034	0.025	0.043
March	on-site	36	1.8	0.7	3.6	0.024	0.010	0.038
	off-site	25	2.0	1.0	3.2	0.023	0.009	0.032
April	on-site	35	1.8	0.6	3.2	0.029	0.015	0.041
	off-site	23	2.2	1.2	5.8	0.026	0.014	0.040
May	on-site	31	3.1	0.6	8.8	0.034	0.016	0.055
	off-site	23	2.4	0.8	3.8	0.032	0.016	0.044
June	on-site	35	2.9	0.2	5.2	0.031	0.021	0.043
	off-site	22	2.5	1.6	4.8	0.030	0.024	0.042
July	on-site	37	3.3	1.1	9.1	0.043	0.021	0.070
	off-site	23	2.6	1.5	4.3	0.044	0.026	0.066
August	on-site	35	3.4	1.8	5.6	0.045	0.032	0.070
	off-site	22	3.0	0.9	4.8	0.046	0.008	0.068
September	on-site	34	2.3	1.1	5.0	0.030	0.020	0.040
	off-site	18	2.2	0.5	5.9	0.031	0.006	0.040
October	on-site	35	2.9	1.3	8.3	0.042	0.028	0.076
	off-site	20	2.7	1.2	5.0	0.040	0.015	0.061
November	on-site	34	2.6	0.7	6.3	0.038	0.021	0.051
	off-site	20	2.4	1.0	6.7	0.037	0.024	0.046
December	on-site	37	2.1	0.6	5.1	0.040	0.028	0.062
	off-site	18	2.7	1.4	9.1	0.039	0.023	0.059
Annual Summary	on-site	415	2.5 \pm 0.3	0.2	9.1	0.036 \pm 0.004	0.010	0.12
	off-site	258	2.4 \pm 0.2	0.5	9.1	0.035 \pm 0.004	0.006	0.068

*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 6

Gamma-Ray Activity in Air-Filter Samples, 1973
(concentrations in 10^{-15} $\mu\text{Ci}/\text{ml}$)

Nuclide	Location	January	February	March	April	May	June	July	August	September	October	November	December	Annual Average
Antimony-125	on-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.6	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
	off-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Barium-140- Lanthanum-140	on-site	1.3	0.8	0.7	0.8	0.7	0.6	3.0	1.9	1.4	1.3	0.7	0.6	1.2 ± 0.4
	off-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	2.4	1.0	< 0.5	< 0.5	< 0.5	< 0.5	< 0.7
Beryllium-7	on-site	73	99	75	90	119	120	119	130	94	89	65	59	94 ± 14
	off-site	82	90	72	97	115	119	121	128	85	86	63	53	93 ± 14
Cerium-141	on-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	0.8	1.2	0.8	0.9	0.6	0.5	< 0.7
	off-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	1.0	1.2	0.7	1.0	0.6	0.5	< 0.7
Cerium-144	on-site	2	2	2	2	3	2	2	1	1	1	2	2	2 ± 1
	off-site	2	2	2	2	3	2	2	1	1	1	1	2	2 ± 1
Cesium-137	on-site	0.6	0.8	0.9	1.0	1.9	1.5	1.1	0.6	< 0.5	< 0.5	< 0.5	< 0.5	0.8 ± 0.3
	off-site	0.6	0.8	1.0	1.1	1.6	1.4	1.1	0.7	< 0.5	< 0.5	< 0.5	0.6	0.8 ± 0.3
Cobalt-60	on-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
	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	on-site	< 5	< 5	< 5	< 5	< 5	< 5	5	< 5	< 5	< 5	< 5	< 5	< 5
	off-site	< 5	< 5	< 5	< 5	< 5	< 5	5	< 5	< 5	< 5	< 5	< 5	< 5
Manganese-54	on-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
	off-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Ruthenium-103	on-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	1.9	1.6	1.1	1.4	1.2	1.1	< 0.9
	off-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	1.4	1.7	1.1	1.3	1.1	0.9	< 0.9
Ruthenium-106- Rhodium-106	on-site	2	2	2	2	3	2	2	2	1	1	1	2	2 ± 1
	off-site	2	2	2	3	3	2	2	2	1	1	2	1	2 ± 1
Zirconium-95- Niobium-95	on-site	0.9	0.8	0.5	0.5	< 0.5	< 0.5	1.1	2.4	2.3	5.1	5.0	7.3	2 ± 2
	off-site	0.8	0.7	< 0.5	< 0.5	< 0.5	< 0.5	1.2	2.5	2.4	4.7	5.1	6.5	2 ± 2

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 is primarily fission and activation products from nuclear test detonations. The average beta activity for the year, 3.6×10^{-14} $\mu\text{Ci/ml}$, was about three times lower than in 1972 and ten times lower than in 1971, the result of a decrease in the fallout from nuclear detonations. The usual increase in the rate of stratospheric fallout in the spring is shown by the monthly variations in the beryllium-7 concentrations. The corresponding increase in the concentrations of long-lived fission products (cerium-144, cesium-137, and ruthenium-106) is less obvious because their stratospheric concentrations have been depleted in recent years. The stratospheric fission product content was apparently too small this year for the "spring maximum" to be evident in the total beta activity results.

The detection limits for several of the gamma-emitting activities have been improved substantially during the past year. As a result, barium-140-lanthanum-140 were detected in on-site samples only in the 300 Area in months when the off-site concentrations were below the detection limit. These nuclides were not found in the samples collected near the site boundary, so the concentrations immediately off-site were also less than the detection limit. The average on-site concentration of barium-140 was only 0.00012% of the CG for uncontrolled areas. Air samples collected in the CP-5 exhaust stack (Building 330, location 9H) indicate that the noble gas parent, xenon-140, was the probable source of these fission products in the air-filter samples.

Except for barium-140-lanthanum-140, the similarity of the airborne alpha, beta, and gamma activities on and off the site indicates that these activities originated in a widespread source - fallout from nuclear test detonations and naturally-occurring materials - and not in a localized source such as Argonne.

The most significant effect upon the airborne radioactivity occurred as a result of a Chinese atmospheric nuclear test on June 26, 1973. The presence of the intermediate half-life fission products, barium-140-lanthanum-140, cerium-141, iodine-131, ruthenium-103, and zirconium-95-

niobium-95, beginning in July both on and off the site can be attributed to this event. A small effect on the total beta activities can be seen in July and August. The sustained increase in beryllium-7 concentrations into July and August, past its normal spring peak, may be due to its production in the June 26 detonation. The results obtained for the air-filter samples are further summarized in Table 7 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, primarily plutonium, were collected in the East Area (12N in Figure 1), in the 300 Area (10H or 9H), and off the site in Downers Grove (Figure 2). Collections were made on a polystyrene filter medium at flow rates of 35 or 80 m³/hr, depending on the sampler, and combined on a monthly basis for analysis. The total air volume filtered for each sample was about 25,000 and 60,000 m³, respectively. Samples were ignited at 600°C to remove organic matter and prepared for plutonium 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. The plutonium was separated from nitric acid solution on an anion-exchange column, electroplated, and its composition determined by alpha spectrometry. 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. The chemical recovery was determined by adding a known amount of plutonium-236 tracer to the sample before ignition.

The chemical separation procedure also yields a separated thorium fraction, and this fraction was also electrodeposited and measured in an alpha spectrometer to determine the contribution from the various thorium isotopes. To obtain the thorium chemical recovery, thorium-234 was added prior to ignition. An aliquot of the sample solution was also analyzed for

TABLE 7

Average Radioactivity in Air-Filter Samples, 1973
(concentrations in 10^{-12} $\mu\text{Ci/ml}$)

Activity	Detection* Limit	Concentration* Guide (CG)	Concentration	Percent CG
Antimony-125	0.0005	900	< 0.0005	< 0.00005
Barium-140- Lanthanum-140	0.0005	1000	< 0.0007	< 0.00007
Beryllium-7	0.005	40000	0.094	(0.0002)
Cerium-141	0.0005	5000	< 0.0007	< 0.00004
Cerium-144	0.001	200	0.002	0.001
Cesium-137	0.0005	500	0.0008	0.0002
Cobalt-60	0.0001	300	< 0.0001	< 0.00003
Iodine-131	0.005	100	< 0.005	< 0.005
Manganese-54	0.0005	1000	< 0.0005	< 0.00005
Ruthenium-103	0.0005	3000	< 0.0009	< 0.00003
Ruthenium-106- Rhodium-106	0.001	200	0.002	0.001
Zirconium-95- Niobium-95	0.0005	1000	0.002	0.0002
Alpha	0.0002	10	0.0025	(0.025)
Beta	0.0005	10	0.036	0.36

*The CGs are those given in U.S.A.E.C. Manual Chapter 0524 for uncontrolled areas. Additional information on the CGs and detection limits is given in the Appendix, Section V.B.

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} $\mu\text{Ci}/\mu\text{g}$. In addition, the load solution from the anion exchange separation was analyzed for radiostrontium by standard radiochemical procedures. The results are given in Table 8.

The plutonium concentrations have decreased by about a factor of two from 1972. There is evidence in the monthly variations to indicate a "spring maximum" in stratospheric fallout similar to that noted for beryllium-7, but which could not be discerned in the airborne total beta activity. In this case, however, the maximum appears at a more normal time, May and June. The concentrations are similar to those reported by the HASL fallout program^(7,8) for samples collected away from nuclear installations. For example, during the first six months of 1973, monthly plutonium-239 concentrations in air ranged from 11×10^{-18} $\mu\text{Ci}/\text{ml}$ to 59×10^{-18} $\mu\text{Ci}/\text{ml}$ at four U. S. locations in the HASL sampling network. Since the results on and near the Argonne site are in this same range, it may be concluded that the concentrations in Table 8 are the result of fallout.

During some months the on-site concentration was higher than that off-site, although the annual averages were fairly similar. These samples contain appreciable quantities of surface dirt, and the amount of resuspended soil collected by the filter is a factor in determining the plutonium concentration. The amount of solids per cubic meter of air collected by the off-site sample is, on the average, one-half of that collected by the on-site samples, and this may account for some of the difference in plutonium concentration. In agreement with this observation are the results for December, the only month in which more solids were collected in the off-site sample and the only month in which the off-site concentration was higher. The amount of plutonium in the air sample contributed by soil, if the resuspended soil has the same plutonium concentration as the first two inches on the ground, is only 5-10% of the total plutonium in the air sample.

TABLE 8

Plutonium, Thorium, Uranium, and Strontium Concentrations in Air-Filter Samples, 1973
(concentrations in 10^{-18} $\mu\text{Ci/ml}$)

Month	Location*	Plutonium-239	Plutonium-238	Thorium-232**	Thorium-230	Thorium-228	Uranium**	Strontium-90	Strontium-89
January	12N	9.5 ± 1.6	0.9 ± 0.9	28 ± 4	54 ± 11	35 ± 5	140 ± 25	-	-
	10H	11 ± 1	0.6 ± 0.4	27 ± 2	50 ± 3	30 ± 2	145 ± 15	470 ± 50	-
February	12N	12 ± 1	1.1 ± 0.5	-	-	-	54 ± 7	590 ± 60	-
	9H	10 ± 1	1.2 ± 0.4	-	-	-	130 ± 20	680 ± 20	-
	Off-Site	13 ± 3	-	10 ± 2	10 ± 2	21 ± 3	84 ± 9	-	-
March	12N	14 ± 1	1.0 ± 0.7	14 ± 3	14 ± 4	18 ± 4	77 ± 6	570 ± 240	-
	9H	13 ± 1	1.0 ± 0.3	23 ± 2	34 ± 3	31 ± 3	135 ± 30	790 ± 50	-
	Off-Site	9.4 ± 0.9	0.6 ± 0.4	5 ± 2	8 ± 2	5 ± 2	38 ± 7	580 ± 50	-
April	12N	24 ± 4	< 1	7 ± 5	18 ± 5	24 ± 6	660 ± 45	-	-
	9H	18 ± 1	1.9 ± 0.3	-	-	-	110 ± 25	-	-
	Off-Site	15 ± 1	0.7 ± 0.2	2 ± 2	5 ± 3	8 ± 4	44 ± 4	-	-
May	12N	22 ± 2	2.0 ± 0.5	14 ± 2	27 ± 3	17 ± 3	130 ± 6	-	-
	9H	20 ± 1	2.8 ± 0.5	18 ± 1	31 ± 2	23 ± 2	68 ± 9	-	-
	Off-Site	18 ± 1	1.4 ± 0.4	5 ± 1	9 ± 2	9 ± 2	38 ± 7	-	-
June	12N	30 ± 2	2.4 ± 0.7	5 ± 1	10 ± 2	4 ± 1	125 ± 10	-	-
	9H	15 ± 1	1.3 ± 0.4	2 ± 1	10 ± 2	6 ± 1	89 ± 4	-	-
	Off-Site	17 ± 1	1.6 ± 0.7	4 ± 4	11 ± 5	9 ± 7	48 ± 4	-	-
July	12N	14 ± 1	0.6 ± 0.6	16 ± 4	31 ± 5	27 ± 5	120 ± 15	720 ± 140	-
	9H	16 ± 1	1.8 ± 0.4	28 ± 3	53 ± 5	27 ± 4	155 ± 20	-	-
	Off-Site	11 ± 1	< 0.1	6 ± 2	10 ± 3	5 ± 3	22 ± 2	-	-
August	12N	11 ± 1	0.8 ± 0.5	12 ± 3	24 ± 4	12 ± 4	79 ± 10	500 ± 70	1360 ± 960
	9H	10 ± 1	0.8 ± 0.4	28 ± 4	60 ± 6	22 ± 4	115 ± 10	530 ± 60	1480 ± 940
	Off-Site	7.4 ± 1.0	< 0.1	6 ± 2	12 ± 3	6 ± 3	34 ± 3	530 ± 70	1240 ± 970
September	12N	5.0 ± 0.4	< 0.1	12 ± 3	31 ± 4	11 ± 4	67 ± 9	230 ± 30	1650 ± 350
	9H	-	-	21 ± 4	39 ± 5	17 ± 4	87 ± 9	320 ± 120	2260 ± 830
	Off-Site	2.5 ± 0.3	< 0.1	3 ± 1	5 ± 1	2 ± 1	26 ± 4	170 ± 20	630 ± 150
October	12N	5.3 ± 0.7	< 0.1	16 ± 2	41 ± 4	16 ± 2	67 ± 6	210 ± 40	1710 ± 180
	9H	5.0 ± 0.6	< 0.1	30 ± 3	62 ± 4	22 ± 3	105 ± 5	-	-
	Off-Site	3.3 ± 0.6	< 0.1	8 ± 1	19 ± 2	13 ± 2	24 ± 3	160 ± 30	1210 ± 130
November	12N	5.4 ± 0.9	< 0.1	12 ± 4	34 ± 6	20 ± 5	51 ± 4	220 ± 80	1250 ± 620
	9H	3.9 ± 0.4	< 0.1	16 ± 2	39 ± 2	13 ± 2	75 ± 5	-	-
	Off-Site	2.8 ± 0.5	< 0.1	8 ± 1	20 ± 2	14 ± 2	21 ± 2	120 ± 110	1040 ± 320
December	12N	6.5 ± 0.7	< 0.1	27 ± 4	75 ± 6	23 ± 4	53 ± 8	260 ± 60	1380 ± 160
	9H	9.8 ± 0.7	< 0.1	20 ± 2	42 ± 2	19 ± 2	110 ± 8	160 ± 30	3770 ± 90
	Off-Site	9.7 ± 0.8	< 0.1	12 ± 2	24 ± 2	12 ± 2	68 ± 5	240 ± 30	1200 ± 80
Monthly Average	12N	13 ± 5	0.71 ± 0.47	15 ± 4	33 ± 11	19 ± 5	135 ± 97	-	-
	9H/10H	12 ± 3	1.0 ± 0.53	21 ± 5	42 ± 10	21 ± 5	110 ± 16	-	-
	Off-Site	9.9 ± 3.3	0.46 ± 0.38	6 ± 2	12 ± 4	9 ± 3	41 ± 12	-	-
Percent CG	12N	0.001	0.0001	(0.002)	(0.011)	(0.010)	(0.003)	-	-
	9H/10H	0.001	0.0001	(0.002)	(0.014)	(0.010)	(0.003)	-	-
	Off-Site	0.001	0.00005	(0.001)	(0.004)	(0.005)	(0.001)	-	-

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

** The concentrations in units of $\mu\text{g/m}^3$ can be obtained by multiplying the value in $\mu\text{Ci/ml}$ 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.

The thorium and uranium concentrations (with the exception of the uranium in April) given in Table 8 are in the same range found during 1972 and are considered to be of natural origin. Similar uranium concentrations have been reported by other investigators.⁽⁹⁾ 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 are directly related to the mass of the material collected on the paper. Also, the concentrations per unit mass collected are quite similar to that found in soil samples. This is evidence to indicate that the bulk of these elements in the air is due to resuspension of soil.

The previously noted exception, the April uranium result at location 12N, is high due to a small fire on April 5, in a barrel containing uranium scrap outside of Building 55 at location 12M. The 12N sample location was about 400 yards directly downwind of the fire. The uranium concentration, 6.6×10^{-16} $\mu\text{Ci/ml}$, is still low compared to the exposure standards, 0.02% of the CG for uncontrolled areas and 0.006% of the CG for controlled areas. If this single high result is excluded from the average, the uranium concentration at location 12N would be 87×10^{-18} $\mu\text{Ci/ml}$. Since the fire, all uranium scrap stored in barrels in this area has been removed.

The strontium concentrations are given in Table 8. The strontium-90 concentrations are considered to be due to fallout from nuclear tests and are consistent with levels reported by other sources.⁽⁷⁾ The positive strontium-89 results in the second half of the year are due to the previously mentioned atmospheric nuclear test of June 26, 1973. No averages are included because of the incomplete data. There is no indication that any strontium in the air samples originated from Argonne operations.

Air sampling for argon-41, a beta emitter with a 1.8 hr half-life that is produced in an operating reactor by neutron irradiation of the stable argon in air, was conducted near the CP-5 reactor (Building 330, 9H), the principal source of this nuclide at the Laboratory. Samples were collected by filling an evacuated "Marinelli-type" container once a day, twice during each week of reactor operation, and the argon-41 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 9. 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. This year's results are 50% higher than last year's, probably because of the random sampling procedure. The average and maximum concentrations of 1.8×10^{-7} and 1.8×10^{-6} $\mu\text{Ci/ml}$, respectively, are about 4.5 and 45 times the CG for uncontrolled areas and 0.09 and 0.9 times 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 was monitored in air on and off the site because substantial amounts are in use at the Laboratory. Water vapor was removed from the air by adsorption on silica gel and the hydrogen-3 measured by counting the water in a liquid scintillation counter. Hydrogen-3 is produced continuously in an operating reactor by several methods. In CP-5, the largest source is by neutron irradiation of the heavy water used for cooling and neutron moderating. Air was sampled continuously for tritiated water vapor at a permanent station 50 m east of the CP-5 reactor. The results, given in Table 10, were all positive although the concentrations were well below the CG. The uncontrolled area CG is used in Table 10 to allow easy comparison with data from other locations to be discussed later. As in the case of argon-41 near the reactor, application of the CG for controlled areas is appropriate. The percent of CG for controlled areas is 25 times less than the values in Table 10. As will be seen, normal or background hydrogen-3 concentrations in air during 1973 averaged about 1.9×10^{-12} $\mu\text{Ci/ml}$. Near the CP-5 reactor, the results averaged 2.7×10^{-10} $\mu\text{Ci/ml}$, which is almost a factor of two higher than 1972. This increase is primarily due to a single unusually high result in October. If this result is excluded, the annual average becomes 1.8×10^{-10} $\mu\text{Ci/ml}$, not significantly different than 1972. The elevated hydrogen-3 in the air at the time this sample was collected may have been released during the movement of some reactor materials in the vicinity. In any case, this event appears to be a limited area, short term release that was still well below the CG.

TABLE 9

Argon-41 Concentrations in Air, 300 Area, 1973

Month	No. of Samples	Conc. (10^{-9} $\mu\text{Ci/ml}$)			Percent CG*		
		Av.	Min.	Max.	Av.	Min.	Max.
January	14	96	< 15	570	5	< 0.8	28
February	14	170	< 15	760	8	< 0.8	38
March	16	89	< 15	930	4	< 0.8	46
April	12	140	< 15	790	7	< 0.8	39
May	20	260	< 15	1780	13	< 0.8	89
June	12	240	< 15	1100	12	< 0.8	55
July	14	160	< 15	940	8	< 0.8	47
August	16	160	< 15	710	8	< 0.8	36
September	12	310	< 15	1080	16	< 0.8	54
October	18	100	< 15	640	5	< 0.8	32
November	16	280	< 15	1080	14	< 0.8	54
December	13	130	< 15	430	6	< 0.8	22
Annual Summary	177	178 \pm 44	< 15	1780	9	< 0.8	89

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

TABLE 10

Hydrogen-3 Concentrations in Air, 300 Area, 1973

Month	No. of Samples	Conc. (10^{-12} $\mu\text{Ci/ml}$)			Percent CG*		
		Av.	Min.	Max.	Av.	Min.	Max.
January	9	100	22	235	0.049	0.011	0.12
February	8	60	5.8	220	0.030	0.0029	0.12
March	9	90	7.0	325	0.046	0.0035	0.16
April	8	105	21	270	0.052	0.010	0.14
May	9	195	31	660	0.098	0.015	0.33
June	9	340	48	1340	0.17	0.024	0.67
July	9	220	13	455	0.11	0.0067	0.23
August	8	145	60	220	0.073	0.030	0.11
September	9	190	4.6	905	0.096	0.0023	0.45
October	9	1330	92	9660	0.67	0.046	4.8
November	8	260	11	610	0.13	0.0053	0.30
December	9	150	6.2	635	0.076	0.0031	0.32
Annual Summary	104	265 \pm 200	4.6	9660	0.13	0.0023	4.8

* This is the CG for uncontrolled areas.

Hydrogen-3 concentrations at several additional locations are given in Table 11. The results at the site boundary 650 m directly west (9EF), 850 m southwest (8F), and 1,600 m north-northeast (14I) of CP-5 are lower by almost a factor of two from 1972. The data show good correlation with wind direction and indicate that dilution to the background level occurs before reaching the boundary line in directions other than that from which the wind is blowing. The average and maximum concentrations were equivalent to 0.005% and 0.01% of the CG, respectively. The average boundary line concentration corresponds to a dose of 0.020 mrem/year, about 75% from Argonne operations and the remainder from background.

Samples were also collected once a month in the East Area (12M), 1,900 m east-northeast of the CP-5 reactor. Higher concentrations in the East Area compared to off-site concentrations may be attributed to CP-5. Adequate correlation exists to 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. In fact, the number of times when these conditions occurred during sample collection was relatively high this year, and as a result, the annual average in the East Area is a factor of two higher than the previous year.

The off-site concentrations, measured about 10 km northwest of the Laboratory, decreased about 25% from last year. This background level of hydrogen-3 is due primarily to nuclear test detonations with some contribution from the natural production of hydrogen-3 in the upper atmosphere by cosmic-rays, and should be subtracted from the other concentrations in Tables 10 and 11 to obtain the Argonne contribution.

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, and are the only nuclides whose presence in air at the ANL site boundary can unequivocally be attributed to Argonne operations. The concentrations and dose rates, as a function of distance from CP-5, were calculated for these two nuclides by a computer program (ASSMRD) based on an atmospheric dispersion model.^(10,11) The following parameters were used in the calculations:

TABLE 11

Hydrogen-3 Concentrations On and Near the ANL Site, 1973

Month	Location	No. of Samples	Conc. (10^{-12} $\mu\text{Ci/ml}$)			Percent CG*		
			Av.	Min.	Max.	Av.	Min.	Max.
January	West Fence (9EF)	9	4.0	0.14	18	0.0020	0.00007	0.0088
	East Area	1	-	-	2.7	-	-	0.0014
	Off-Site	1	-	-	0.17	-	-	0.00009
February	West Fence (9EF)	8	1.7	0.33	3.7	0.00083	0.00016	0.0018
	East Area	1	-	-	5.4	-	-	0.0027
	Off-Site	1	-	-	0.38	-	-	0.00019
March	West Fence (9EF)	9	2.4	1.0	6.3	0.0012	0.00050	0.0032
	East Area	1	-	-	7.4	-	-	0.0037
	Off-Site	1	-	-	0.70	-	-	0.00035
April	West Fence (9EF)	8	15	0.43	63	0.0073	0.00022	0.031
	East Area	1	-	-	3.5	-	-	0.0017
	Off-Site	1	-	-	1.2	-	-	0.00058
May	West Fence (9EF)	9	9.4	0.49	34	0.0047	0.00024	0.017
	East Area	1	-	-	10	-	-	0.0050
	Off-Site	1	-	-	1.9	-	-	0.00095
June	South Fence (8F)	8	9.6	2.8	30	0.0048	0.0014	0.015
	East Area	1	-	-	5.0	-	-	0.0025
	Off-Site	1	-	-	3.0	-	-	0.0015
July	South Fence (8F)	9	13	5.8	43	0.0065	0.0029	0.021
	East Area	1	-	-	5.2	-	-	0.0026
	Off-Site	1	-	-	6.4	-	-	0.0032
August	South Fence (8F)	8	11	4.9	26	0.0055	0.0025	0.013
	East Area	1	-	-	14	-	-	0.0070
	Off-Site	1	-	-	2.4	-	-	0.0012
September	South Fence (8F)	9	12	2.2	47	0.0062	0.0011	0.0024
	East Area	1	-	-	5.3	-	-	0.0026
	Off-Site	1	-	-	3.0	-	-	0.0015
October	North Fence (14I)	9	9.7	1.9	28	0.0048	0.00096	0.014
	East Area	1	-	-	35	-	-	0.017
	Off-Site	1	-	-	1.8	-	-	0.0009
November	North Fence (14I)	8	7.2	0.96	20	0.0036	0.00048	0.010
	East Area	1	-	-	13	-	-	0.0065
	Off-Site	1	-	-	0.81	-	-	0.0004
December	North Fence (14I)	9	2.7	0.59	6.7	0.0014	0.00030	0.0034
	East Area	1	-	-	18	-	-	0.0090
	Off-Site	1	-	-	0.54	-	-	0.0003
Annual Summary	Fence Line	103	8.1 \pm 2.6	0.14	63	0.0040	0.00007	0.031
	East Area	12	10. \pm 5.	2.7	35	0.0050	0.0014	0.017
	Off-Site	12	1.9 \pm 1.0	0.17	6.4	0.00095	0.00009	0.003

*This is the CG for uncontrolled areas.

- a) release rates: argon-41, 72.3 Ci/day; hydrogen-3, 1 Ci/day
- b) meteorological data: the 15-year average values given in ANL-7084⁽⁴⁾ for a height of 5.8 m (considered to be most representative of the situation at CP-5)
- c) stack height: 15 m
- d) the usual parameters for building, wake, plume momentum, temperatures, etc.

The calculations were carried out to 50 miles. The results for the first three miles (4.8 km) are given in Table 12. Doses were calculated at 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 rate at 1,500 m is in the NNE sector, 3.7 mrem/year, and in all directions the dose is less than 5 mrem/year, 1% of the standard for individuals in uncontrolled areas. The 1,500 m distance is within the site boundary in the NNW, NE, WNE, and NW directions. A small portion of the former Rocky Glen Forest Preserve is within 1,500 m of CP-5, and in this direction the dose rate at 1,500 m is 2.4 mrem/year. The average dose in the 0-1 mile area is 5.6 mrem/year.

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.E. The measurements made south of the reactor are obscured by direct radiation from a waste storage facility and a Van de Graaff generator 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, up to 123 mrem/year. Increases in excess of two standard deviations (22 mrem/year) of the off-site average (100 mrem/year) would have been readily recognized as abnormal, and on this basis the dose from argon-41 at the site boundary was less than about 20 mrem/year. Thus, the calculated doses and those measured by TLD agree within the ability of the TLD system to detect above-normal doses.

TABLE 12

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

Sector	Distance			
	1500 m	0-1 mi	1-2 mi	2-3 mi
N	3.2	7.9	1.5	0.6
NNE	3.7	9.2	1.7	0.7
NE	2.9	7.2	1.4	0.5
ENE	3.1	7.7	1.4	0.6
E	2.4	6.1	1.1	0.5
ESE	2.3	5.7	1.1	0.4
SE	2.2	5.5	1.0	0.4
SSE	1.9	4.6	0.9	0.4
S	2.0	5.0	0.9	0.4
SSW	2.3	5.7	1.1	0.4
SW	2.0	4.9	0.9	0.4
WSW	1.7	4.2	0.8	0.3
W	1.3	3.2	0.6	0.2
WNW	1.4	3.5	0.7	0.3
NW	1.7	4.1	0.8	0.3
NNW	2.0	4.9	0.9	0.4

The population data in Table 3 can be used to estimate the maximum individual dose and the population dose. The maximum dose to full-time residents occurs in the 0-1 mile annulus in the SW and NNW directions, where approximately 10 and 50 individuals, respectively, would be exposed to about 2 mrem/year, compared to the maximum dose to individuals of 500 mrem/year permitted by AECM 0524. The calculated population dose from argon-41 at several distances from the reactor is given in Table 13. The table also gives the estimated natural external radiation dose, based on the assumption that the average off-site, outdoor, radiation dose measured by TLD applies to the entire area within a 50 mile radius.

TABLE 13

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

Distance (miles)	Population	Dose			
		man-rem/year		Avg. mrem/year	
		Argon-41	Natural	Argon-41	Natural
0-1	180	0.75	18	4.2	100
1-2	1,720	1.81	172	1.3	100
2-3	8,131	3.82	813	0.64	100
0-50	7,757,789	93.8	7.76×10^5	0.012	100

At an argon-41 production rate of 72 Ci/day, the annual release from CP-5 is 2.6×10^4 Ci. Since the half-life of this nuclide is 110 minutes, approximately 5% will decay before reaching the site boundary, if the argon moves with 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. Based on the 72 Ci/day release rate, the highest concentration (in the NNE direction) for each incremental distance in Table 12 is 7.32×10^{-10} μ Ci/ml at 0.8 km, 2.94×10^{-10} μ Ci/ml at 1.5 km, 1.37×10^{-10} μ Ci/ml at 2.4 km, and 5.6×10^{-11} μ Ci/ml at 4 km.

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 0.011 mrem/year. The maximum individual exposure to full-time residents in the area occurs in the 0-1 mile annulus in the SW and NNW directions, where the annual dose is calculated to be 0.014 mrem/year. A brief summary of the results is given in Table 14. A comparison of the doses calculated from the meteorological model with the measured data from Table 11 is given in Table 15. In both cases concentrations are converted to dose as described in Part B of the Appendix. The doses were computed by the meteorological model for fixed distances of 0.8, 1.5, and 2.4 km, and the comparisons are made for the distances that correspond most closely with those at which the actual measurements were made. The agreement is considered reasonable in view of the large number of variables and parameters involved in obtaining both the calculated and measured values.

As mentioned earlier, other airborne effluents are quite low compared to those discussed above. A small amount of argon-41 is released from the JANUS reactor, about 3.5 Ci/year, and this activity would disperse and decay as the CP-5 argon-41. The total amount of other airborne effluents released from the Laboratory is estimated to be less than 0.5 Ci/year; the principal long-lived nuclide in this group is krypton-85. The concentration of iodine-131 (a nuclide of particular interest) in the CP-5 exhaust stack, was measured at about 80 pCi/m³, equivalent to a discharge of about 0.02 Ci/year. The maximum concentration at 1.5 km, assuming no ground deposition, would be about 1.6×10^{-16} μ Ci/ml, or 0.00016% of the CG.

B. 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

TABLE 14

Hydrogen-3 Population and Average Dose From the CP-5 Reactor

Distance (miles)	Population	man-rem/year	Avg. mrem/year
0-1	180	0.0022	0.012
1-2	1,720	0.0061	0.0043
2-3	8,131	0.0146	0.0023
0-50	7,757,789	1.12	0.00014

TABLE 15

Comparison of Measured and Calculated Hydrogen-3 Dose Rates

Direction	Calculated		Measured	
	Distance (km)	mrem/y	Distance (km)	mrem/y
W	0.8	0.0093	0.60	0.012
SW	0.8	0.014	0.85	0.024
NNE	1.5	0.011	1.6	0.012
NE	1.5	0.0089	1.9	0.020
	2.4	0.0045		

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, and neptunium. Hydrogen-3 analyses were performed by counting 10 ml in a gel system. Plutonium and neptunium analyses were performed on 10 or 45-liter samples by a plutonium chemical separation method,⁽¹²⁾ modified to include neptunium, followed by alpha spectrometry. Plutonium-236 was used to determine the plutonium yield. A very recent modification of the plutonium procedure allowed the group separation of a fraction containing the transplutonium elements. Americium-243 was added to determine chemical recovery, and individual nuclides were measured by alpha spectrometry. Some limited data is presented in the text for these elements.

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 yards 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 16. 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, neptunium-237, plutonium-239, and although not shown in the

TABLE 16

Radioactivity in Sawmill Creek Water, 1973

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)	15K	16	1.9 ± 0.3	1.2	3.2	(0.063)	(0.040)	(0.11)
	7M	252	1.5 ± 0.1	0.65	3.3	(0.050)	(0.022)	(0.11)
Beta (nonvolatile)	15K	16	13 ± 3	7.4	30	(0.43)	(0.25)	(1.0)
	7M	252	12 ± 1	7.0	24	(0.40)	(0.23)	(0.80)
Hydrogen-3	15K	16	< 242	< 200	360	< 0.0081	< 0.0067	0.012
	7M	252	1710 ± 1220	214	32,000	0.057	0.0071	1.1
Strontium-89	15K	12	-	-	< 2	-	-	< 0.07
	7M	252	-	-	< 2	-	-	< 0.07
Strontium-90	15K	12	0.90 ± 0.2	< 0.5	1.6	0.30	< 0.17	0.53
	7M	252	0.76 ± 0.08	< 0.5	1.5	0.25	< 0.17	0.50
Iodine-131	15K	12	< 5	< 3	24	< 1.7	< 1	8.0
	7M	252	< 4	< 3	9.8	< 1.3	< 1	3.3
Barium-140	15K	12	-	-	< 2	-	-	< 0.007
	7M	252	-	-	< 2	-	-	< 0.007
Uranium (natural)**	15K	12	1.6 ± 0.2	1.1	2.6	(0.0040)	(0.0028)	(0.0065)
	7M	252	1.5 ± 0.1	0.7	2.2	(0.0038)	(0.0018)	(0.0055)
Neptunium-237	15K	12	-	-	< 0.002	-	-	< 0.00007
	7M	252	0.17 ± 0.08	< 0.002	1.9	0.0057	< 0.00007	0.063
Plutonium-239	15K	12	-	-	< 0.0005	-	-	< 0.00001
	7M	252	0.0051 ± 0.0014	< 0.0005	0.0215	0.0001	< 0.00001	0.00043

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

** 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 in the Creek then becomes $2.4 \mu\text{g/l}$.

table, occasionally some transplutonium isotopes. The fraction of individual samples containing activity attributable to Argonne was 95% for hydrogen-3 and 90% for plutonium and neptunium. The concentrations of all these nuclides were low compared to the CGs. The principal radionuclide added to the Creek by Argonne waste water, in terms of both concentration and percent of CG, was hydrogen-3. The average Argonne contribution was three times greater than last year, but amounted to only 0.06% of the CG, and the highest concentration in any single sample was equivalent to 1.1% of the CG. If the highest result, 3.2×10^{-5} $\mu\text{Ci/ml}$, is excluded, the average decreases from 1.7×10^{-6} to 1.1×10^{-6} $\mu\text{Ci/ml}$ (0.036% of the CG).

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 waters. During 1973, the hydrogen-3 content of other lakes and streams ranged from $< 200 \times 10^{-9}$ $\mu\text{Ci/ml}$ to 345×10^{-9} $\mu\text{Ci/ml}$ and averaged $< 230 \times 10^{-9}$ $\mu\text{Ci/ml}$. The environmental hydrogen-3 levels have decreased to a point where a large number of results are less than the detection limit of the method used.

There was one positive iodine-131 result 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 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 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 natural Creek water. The total alpha and beta activities were not appreciably different than in 1972.

In addition to the natural beta activity and that added by ANL wastewater 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 $2-3 \times 10^{-9}$ $\mu\text{Ci/ml}$ 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 1972 levels, while the fallout contribution decreased 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 16 were low.

The total amount of radioactive substances released to the environment in Argonne waste water can be calculated from the measured water flow and from the activity concentrations measured either in the Creek water below the outfall or in the effluent water. These values are shown in Table 17, together with the calculated dose rates that would result if water at this concentration were used by an individual as the sole source of potable water. The nuclides found in the Creek and attributable to Argonne operations are hydrogen-3, plutonium-239, and neptunium-237. However, strontium-90 is also included since it is a major contributor to the non-natural dose from Creek water, although it was added to the Creek principally from nuclear test fallout. The dose rates are all well below the drinking water standards for individuals in uncontrolled areas. It should be noted, however, that this water is not used for drinking or recreational purposes, and there are very few fish in the stream and they do not constitute a significant source of food for any individual.

Analyses for transplutonium nuclides were performed on Sawmill Creek and Des Plaines River samples during the last four months of the year. The number of results is not considered adequate to give a meaningful annual average. Those nuclides identified were americium-241, curium-244 and/or californium-249, and californium-252. The alpha particles from curium-244

TABLE 17

ANL Radioactive Effluent and Dose
Values for Sawmill Creek Water, 1973

Nuclide	Annual Discharge (Ci)	Conc. (avg.) 10^{-9} μ Ci/ml	Dose Rate (mrem/year)	Percent of Standard*
Hydrogen-3	3.6	1.7×10^3	0.28	0.06
Strontium-90	-	0.8	8	0.3
Neptunium-237	0.0021	0.17	0.17	0.006
Plutonium-239	6.2×10^{-5}	0.0051	0.003	1×10^{-4}

*The standard is 500 mrem/year (whole body) for hydrogen-3 and 3 rem/year (bone-seekers) for the other nuclides.

and californium-249 have almost identical energies, and could not be distinguished by alpha spectrometry. The detection limit for these nuclides is 1×10^{-12} μ Ci/ml. The average concentration of americium-241 was similar to that of plutonium-239, while the average concentration of curium-244 and/or californium-249 was about twice that of the plutonium-239. The californium-252 concentration averaged less than the detection limit. Since the CGs for these nuclides are similar to those for plutonium-239, the dose resulting from these nuclides can be calculated from their concentrations relative to plutonium-239. The concentrations of the transplutonium nuclides in all the Des Plaines River and above-site Creek samples analyzed were less than the detection limit.

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 18. The average total alpha and beta activities,

TABLE 18

Radioactivity in Des Plaines River Water, 1973

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 ± 0.3	0.70	2.6	(0.050)	(0.023)	(0.087)
	B	24	1.6 ± 0.2	0.23	2.8	(0.053)	(0.008)	(0.093)
Beta (nonvolatile)	A	12	8.2 ± 1.9	5.2	18	(0.27)	(0.17)	(0.60)
	B	24	8.6 ± 1.0	5.2	15	(0.29)	(0.17)	(0.50)
Hydrogen-3	A	12	< 220	< 200	325	< 0.0073	< 0.0067	0.011
	B	24	< 210	< 200	265	< 0.0070	< 0.0067	0.0088
Strontium-89	A	12	-	-	< 2	-	-	< 0.07
	B	24	-	-	< 2	-	-	< 0.07
Strontium-90	A	12	0.76 ± 0.10	< 0.50	1.0	0.25	< 0.17	0.33
	B	24	0.76 ± 0.06	< 0.50	1.0	0.25	< 0.17	0.33
Iodine-131	A	12	-	-	< 3	-	-	< 1
	B	23	-	-	< 3	-	-	< 1
Barium-140	A	12	-	-	< 2	-	-	< 0.007
	B	24	-	-	< 2	-	-	< 0.007
Uranium (natural)**	A	11	1.3 ± 0.2	0.84	2.0	(0.0032)	(0.0021)	(0.0050)
	B	24	1.4 ± 0.2	0.67	1.9	(0.0035)	(0.0017)	(0.0048)
Neptunium-237	A	12	-	-	< 0.002	-	-	< 0.00007
	B	12	-	-	< 0.002	-	-	< 0.00007
Plutonium-239	A	12	< 0.0007	< 0.0005	0.0010	< 0.00001	< 0.00001	0.00002
	B	12	< 0.0013	< 0.0005	0.0093	< 0.00003	< 0.00001	0.00019

* 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 $2.1 \mu\text{g/l}$.

1.4×10^{-9} $\mu\text{Ci/ml}$ and 8.2×10^{-9} $\mu\text{Ci/ml}$, respectively, of 26 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 that it is not detectable as such in the Des Plaines River. The natural nonvolatile beta activity in the River is 5×10^{-9} $\mu\text{Ci/ml}$, and the excess, $2-3 \times 10^{-9}$ $\mu\text{Ci/ml}$, was due to fallout.

Between May and August, plutonium-239 was detected at both locations in the River. Since the River contains mainly surface runoff water, and slightly higher plutonium-239 concentrations were observed in the air during this period, the presence of plutonium-239 can be attributed to fallout, carried down by the rains and collected in the River. The fact that plutonium-239 could not be found at any time in Sawmill Creek above the site is probably due to the fact that a substantial portion of this water is from deep municipal wells.

The radioactivity in samples of Illinois River water, shown in Table 19, 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.

C. Soil, Grass, and Benthic Materials

The plutonium content of soil, grass, and benthic materials was measured on and off the site to study the plutonium fallout level in the area from nuclear testing and to determine if any plutonium is present in the environment that might be due to Argonne operations. Each soil sample consisted of two cores totalling 173 cm^2 in area by 30 cm deep. Two criteria were used in selecting sites for soil sampling, depending on the purposes of the sampling. The object of most off-site sampling was to determine the total deposition of plutonium from weapons testing for comparison with on-site samples, and with results obtained by other organizations for samples collected at large distances from nuclear installations. This latter comparison must be made to establish the state of normality of soil activity near Argonne. For this purpose, the criteria given by the AEC New York Health and Safety Laboratory^(8,13) were used. Sites were selected in several

TABLE 19

Radioactivity in Illinois River Water, 1973
(concentrations in 10^{-9} $\mu\text{Ci/ml}$)

Location	Date Collected	Alpha ¹	Beta ¹	Hydrogen-3	Uranium ²	Neptunium-237 ³	Plutonium-239 ³
McKinley Woods State Park	June 12	0.79	10	295	1.2	< 0.0005	0.00059
Below Dresden Power Station	June 12	1.1	5.2	215	1.0	< 0.0005	0.00034
Morris	June 12	0.20	6.7	300	1.1	-	-
Starved Rock State Park	June 12	0.25	4.4	315	1.0	-	-
McKinley Woods State Park	October 18	1.3	8.4	220	0.80	< 0.0005	0.00035
Below Dresden Power Station	October 18	1.2	8.8	< 200	0.80	< 0.0005	0.00024
Morris	October 18	1.9	8.3	255	1.0	-	-
Starved Rock State Park	October 18	1.4	10	< 200	1.0	-	-

¹Nonvolatile activity.

²Uranium concentrations in units of $\mu\text{g/l}$ can be obtained by multiplying the concentration by 1.48×10^9 .

³These analyses were made on 45 l samples.

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. For samples designed to detect recent deposition, particularly from localized sources, the site was selected on the basis of its location, such as downwind from a particular building or on-site activity source; for very recent deposition, the first two inches of soil was analyzed separately to increase the detection sensitivity.

The grass samples were obtained by collecting all 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. The samples were analyzed by the same method described in Section III.A. for air. After drying, grinding, and mixing, one hundred gram portions of soil and benthos were analyzed. The size of the grass samples was about 100 g of the oven-dried plant. Results are given in terms of oven-dried soil, benthos, or grass.

The on-site and off-site soil results are given in Tables 20 and 21, respectively. Comparison of the on-site and off-site samples 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 on-site samples resulted primarily from fallout of debris from nuclear detonations. Fallout deposition values found by other laboratories⁽¹⁴⁻¹⁶⁾ are in the same range as those reported here, about $2-3 \times 10^{-3} \mu\text{Ci}/\text{m}^2$.

The samples collected on-site on April 11, were each divided into two sections and analyzed separately, as shown in Table 20. For the purpose of obtaining the average, the plutonium deposition in these two sections was summed for each sample. These samples were taken in the area of the uranium fire mentioned in Section III.A. The plutonium content of two of these samples ($4.5 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ and $4.0 \times 10^{-3} \mu\text{Ci}/\text{m}^2$) was slightly higher than the usual fallout levels, as has been noted in this area previously,^(2,3) and this may be the result of the plutonium usage in this area in the past. Such concentrations are very localized, since normal amounts were present in other samples from the same vicinity. Surveillance in this area will be continued.

TABLE 20

Plutonium Concentrations in On-Site Soil, 1973

Date Collected	Location*	Plutonium-238		Plutonium-239		$^{238}\text{Pu}/^{239}\text{Pu}$
		10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	
April 11	12M (0-5 cm)	0.93 ± 0.33	0.051 ± 0.018	13.5 ± 0.9	0.73 ± 0.05	0.070
	12M (5-30 cm)	0.77 ± 0.31	0.22 ± 0.09	13.6 ± 0.9	3.75 ± 0.26	0.057
April 11	12M (0-5 cm)	1.3 ± 0.4	0.064 ± 0.019	25.4 ± 1.3	1.27 ± 0.07	0.051
	12M (5-30 cm)	0.28 ± 0.24	0.091 ± 0.076	3.8 ± 0.5	1.21 ± 0.16	0.075
April 11	12M (0-5 cm)	2.8 ± 0.5	0.14 ± 0.02	52.2 ± 1.9	2.55 ± 0.09	0.053
	12M (5-30 cm)	0.22 ± 0.21	0.072 ± 0.072	4.6 ± 0.5	1.49 ± 0.17	0.041
April 11	12M (0-5 cm)	1.1 ± 0.3	0.052 ± 0.016	23.4 ± 1.3	1.10 ± 0.06	0.048
	12M (5-30 cm)	0.37 ± 0.25	0.11 ± 0.07	4.8 ± 0.6	1.39 ± 0.16	0.046
June 28	10N	0.31 ± 0.27	0.10 ± 0.09	6.9 ± 0.7	2.28 ± 0.23	0.045
June 28	7E	0.52 ± 0.31	0.23 ± 0.13	5.0 ± 0.6	2.19 ± 0.27	0.10
June 28	14I	0.20 ± 0.27	0.08 ± 0.11	5.6 ± 0.6	2.18 ± 0.25	0.038
June 28	8H	0.40 ± 0.31	0.16 ± 0.13	5.6 ± 0.7	2.19 ± 0.26	0.072
June 28	8H	0.50 ± 0.28	0.20 ± 0.11	6.6 ± 0.7	2.50 ± 0.25	0.077
June 28	8H	0.34 ± 0.27	0.14 ± 0.12	6.2 ± 0.7	2.56 ± 0.27	0.055
June 28	8H	0.42 ± 0.26	0.15 ± 0.09	5.9 ± 0.6	2.13 ± 0.22	0.071
	Average		0.17 ± 0.03		2.68 ± 0.48	0.062

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*The locations are given in terms of the grid coordinates in Figure 1.
The depth is 30 cm, unless otherwise indicated.

TABLE 21

Plutonium Concentrations in Off-Site Soil, 1973

Date Collected	Location	Plutonium-238		Plutonium-239		$^{238}\text{Pu}/^{239}\text{Pu}$
		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 12	Channahon, Illinois	0.46 ± 0.25	0.16 ± 0.09	7.0 ± 0.7	2.43 ± 0.23	0.066
June 12	Dresden Lock and Dam	0.49 ± 0.28	0.16 ± 0.09	7.6 ± 0.7	2.40 ± 0.23	0.065
June 21	Lemont, Illinois	0.32 ± 0.25	0.11 ± 0.09	6.4 ± 0.6	2.19 ± 0.23	0.050
June 21	Western Springs, Illinois	1.8 ± 0.5	0.56 ± 0.16	7.8 ± 0.9	2.34 ± 0.26	0.24
October 16	Magician Lake, Michigan	0.12 ± 0.22	0.042 ± 0.078	6.4 ± 0.6	2.27 ± 0.23	0.019
October 18	Morris, Illinois	0.88 ± 0.30	0.29 ± 0.10	19.9 ± 1.1	6.58 ± 0.37	0.045
October 18	Romeoville, Illinois	< 0.1	-	10.1 ± 1.5	2.18 ± 0.32	-
October 25	Willow Springs, Illinois	< 0.1	-	4.2 ± 0.6	1.72 ± 0.25	-
October 25	West Chicago, Illinois	< 0.1	-	4.7 ± 0.5	1.65 ± 0.18	-
	Average		0.22 ± 0.15		2.64 ± 1.00	0.081

The sample collected at Morris, Illinois on October 18, 1973, gave unusual results. The plutonium-239 concentration is almost three times fallout level, the uranium level is slightly elevated, the thorium-230 concentration is five times normal, and gamma-ray spectrometry indicates an elevated radium-226 concentration. The thorium-232 and thorium-228 levels were normal. This area will be resampled to confirm these results.

Composite monthly precipitation samples were analyzed for plutonium-239. Concentrations, expressed in terms of ground deposition, ranged from 1.4×10^{-7} to 5.0×10^{-7} $\mu\text{Ci}/\text{m}^2$ and averaged 3.1×10^{-7} $\mu\text{Ci}/\text{m}^2$. The total 1973 deposition by precipitation was 25% less than 1972 and only 0.2% of the amount previously deposited. This implies that in the absence of further atmospheric nuclear testing, the soil content will not increase significantly.

The thorium and uranium content of the soil samples was also measured, the former by chemical separation and alpha spectrometry and the latter by a fluorophotometric method. Thorium-228 concentrations averaged $(0.43 \pm 0.05) \times 10^{-6}$ $\mu\text{Ci}/\text{g}$ on-site and $(0.43 \pm 0.12) \times 10^{-6}$ $\mu\text{Ci}/\text{g}$ off-site. Thorium-230 averaged $(0.52 \pm 0.06) \times 10^{-6}$ $\mu\text{Ci}/\text{g}$ on-site and $(0.70 \pm 0.45) \times 10^{-6}$ $\mu\text{Ci}/\text{g}$ off-site with the Morris result and $(0.49 \pm 0.12) \times 10^{-6}$ $\mu\text{Ci}/\text{g}$ off-site without the Morris result. Thorium-232 averaged $(0.39 \pm 0.03) \times 10^{-6}$ $\mu\text{Ci}/\text{g}$ on-site and $(0.37 \pm 0.08) \times 10^{-6}$ $\mu\text{Ci}/\text{g}$ off-site. With the exception of the Morris result, no significant differences between on-site and off-site concentrations were found. These are normal levels of the naturally-occurring thorium activities. Results of uranium analysis averaged $(1.6 \pm 0.4) \times 10^{-6}$ $\mu\text{Ci}/\text{g}$ on-site and $(1.5 \pm 0.6) \times 10^{-6}$ $\mu\text{Ci}/\text{g}$ off-site, quite similar to levels of uranium in soil found in previous years. In terms of mass, the thorium concentrations were 3.5 $\mu\text{g}/\text{g}$ and 3.3 $\mu\text{g}/\text{g}$ on and off the site, respectively; the uranium concentrations were 2.4 $\mu\text{g}/\text{g}$ on-site and 2.2 $\mu\text{g}/\text{g}$ off-site.

Examination of the results of samples collected in the vicinity of the uranium fire showed that the uranium content of the soil in this area was not increased measurably. Gamma-ray spectrometric analysis of the soil samples showed only traces of old fission products attributable to fallout, and no activity related to Laboratory operations.

The results of plutonium-239 measurements in grass are given in Table 22. The on-site concentrations in terms of deposition are similar to on- and off-site results of previous years. The off-site samples collected in October of this year contained somewhat less plutonium than the other samples perhaps because the air and precipitation concentrations were low also. All the results, on and off the site, were within the range expected and observed from fallout. In terms of deposition, the plutonium-239 concentration was a factor of about $1-2 \times 10^4$ less than in soil from the same location. Thorium results in plants appear to vary widely and may be due to residual soil analyzed with the plant. Elevated uranium concentrations in grass near the fire indicate that grass is a more sensitive detector of recent deposition than soil. Similar observations have been made in the past for fallout and local releases.⁽¹⁷⁾ The average uranium concentration in off-site grass samples was $(4.4 \pm 2.0) \times 10^{-8}$ $\mu\text{Ci/g}$, while two samples near the uranium fire contained 3.6×10^{-6} $\mu\text{Ci/g}$ and 0.52×10^{-6} $\mu\text{Ci/g}$.

A study was made of the plutonium concentration in the beds of the waterways that carry surface water to Sawmill Creek. The results are collected in Table 23 together with an off-site control sample. Results vary widely between locations, and plutonium-239 concentrations up to about 30×10^{-9} $\mu\text{Ci/g}$ may be considered normal based on previous data.⁽³⁾ Samples at 12K (lower lagoon) and 11K (upper lagoon) are 2 and 5 times this level, respectively. The lagoons are created by a small dam erected for water flow control. The effect of this dam is to create a type of natural settling pond which may allow fallout plutonium washed into it by runoff of surface water to accumulate in the bed at abnormal concentrations. No Laboratory radioactive waste is discharged into this water network. Thorium and uranium concentrations in these benthic samples were similar to soil. The average concentrations of these activities were: thorium-232, $(0.41 \pm 0.08) \times 10^{-6}$ $\mu\text{Ci/g}$; thorium-230, $(0.64 \pm 0.09) \times 10^{-6}$ $\mu\text{Ci/g}$; thorium-228, $(0.58 \pm 0.10) \times 10^{-6}$ $\mu\text{Ci/g}$; and uranium, $(1.4 \pm 0.2) \times 10^{-6}$ $\mu\text{Ci/g}$. In terms of mass, the thorium concentrations were 3.7 $\mu\text{g/g}$ and the uranium concentrations were 2.1 $\mu\text{g/g}$. Gamma-ray spectrometric analysis of these samples showed only traces of old fission products attributable to fallout.

TABLE 22

Plutonium-239 Concentrations in Grass, 1973

Date Collected	Location*	Concentration	
		10^{-9} $\mu\text{Ci/g}$	10^{-6} $\mu\text{Ci/m}^2$
<u>On-Site</u>			
April 11	12M	2.8 ± 0.3	0.27 ± 0.03
April 11	12M	2.4 ± 0.4	0.20 ± 0.03
June 28	8H	0.48 ± 0.19	0.19 ± 0.08
<u>Off-Site</u>			
June 12	Dresden Lock and Dam	0.45 ± 0.12	0.10 ± 0.03
June 21	Lemont, Illinois	0.63 ± 0.14	0.14 ± 0.03
October 16	Magician Lake, Michigan	0.77 ± 0.51	0.059 ± 0.040
October 18	Morris, Illinois	1.1 ± 0.2	0.098 ± 0.018
October 18	Romeoville, Illinois	0.74 ± 0.15	0.075 ± 0.015
October 25	Willow Springs, Illinois	0.68 ± 0.15	0.081 ± 0.018
October 25	West Chicago, Illinois	1.1 ± 0.2	0.082 ± 0.015

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

TABLE 23

Plutonium Concentrations in Benthic Material, 1973

Location*	Plutonium-238 Conc. (10^{-9} μ Ci/g)	Plutonium-239 Conc. (10^{-9} μ Ci/g)	$^{238}\text{Pu}/^{239}\text{Pu}$
Stream - 12L	< 0.1	3.7 ± 0.6	-
Lagoon - 12K	2.8 ± 0.5	62 ± 2	0.044
Lagoon - 11K	2.6 ± 0.5	147 ± 3	0.017
Stream - 11H	-	31.6 ± 2.0	-
Lagoon - 11G	0.39 ± 0.26	10.4 ± 0.8	0.038
Stream - 10E/F	< 0.1	3.2 ± 0.6	-
Stream - 8G/H	0.95 ± 0.33	30.8 ± 1.5	0.031
Stream - 8K	< 0.1	1.2 ± 0.3	-
Stream - 12I	< 0.1	3.2 ± 0.6	-
Des Plaines River, Brookfield	0.98 ± 0.33	15.7 ± 1.0	0.063

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

D. Radioactivity in Milk

Raw milk was collected monthly from a local dairy farm and analyzed for several radioactive **nuclides**. The tritiated water was removed from the milk by low temperature vacuum evaporation and the hydrogen-3 determined by liquid scintillation counting. Strontium-89, -90 was determined by chemical separation and beta counting. The other nuclides were measured by gamma-ray spectrometry. Barium-140, strontium-89, and iodine-131 were not present in concentrations greater than the detection limits of 20×10^{-9} μ Ci/ml for iodine-131 and 2×10^{-9} μ Ci/ml for the other two nuclides. The results for the other nuclides are given in Table 24. This is the first year we have reported hydrogen-3 in milk. 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 content was similar to last year, while the cesium-137 concentration appears to have decreased by a factor of two. These nuclides are long-lived products from past nuclear tests and their presence in milk is not related to Argonne operations.

The concentrations given in Table 24 may be compared to the CGs for drinking water. These values are 3×10^{-3} $\mu\text{Ci/ml}$ for hydrogen-3, 3×10^{-7} $\mu\text{Ci/ml}$ for strontium-90, and 2×10^{-5} $\mu\text{Ci/ml}$ for cesium-137, if the daily intake of water is 2.2 liters. The consumption of one liter of milk per day would then result in an average intake of $< 0.003\%$ of the hydrogen-3, 0.85% of the strontium-90, and $< 0.014\%$ of the cesium-137 Concentration Guides.

TABLE 24

Radioactivity in Milk, 1973
(concentrations in 10^{-9} $\mu\text{Ci/ml}$)

Date Collected	Hydrogen-3	Strontium-90	Cesium-137
January 3	214	1.9	5
February 7	268	4.2	5
March 7	205	3.5	6
April 4	245	8.7	< 5
May 2	220	11.9	5
June 6	< 200	7.8	< 5
July 5	< 200	3.3	< 5
August 1	232	6.7	< 5
September 5	< 200	5.9	< 5
October 3	< 200	4.2	< 5
November 7	206	3.5	< 5
December 5	< 200	5.3	< 5
Average	< 216	5.6	< 6

E. Penetrating Radiation

Gamma-ray dose measurements were made with dysprosium-activated calcium fluoride thermoluminescent chips. Each measurement was the average of 3 to 6 chips exposed in the same package. The response of the dosimeters was calibrated with a NBS standard radium-226 source, and the results calculated in terms of air dose. Measurements were made during six successive exposure periods ranging in length from 32 to 84 days, and averaging 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 on the site boundary to determine the dose due to Argonne operations at the closest uncontrolled approaches to the Laboratory and at several locations on the site. The latter locations were chosen for two principal 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 line. Measurements were also made at five off-site locations for comparison purposes. The results are summarized in Tables 25 and 26, and the site boundary and on-site readings are also shown in Figure 5.

The off-site results averaged 100 mrem/year with a standard deviation (for a single result) of 11 mrem/year. The standard error of the average is 2 mrem/year. During 1972, the corresponding average and standard deviation were 105 and 5 mrem/year, respectively, and a relatively few measurements made during the latter part of 1971 averaged 104 mrem/year. Thus, the off-site readings have remained constant, within **statistical fluctuations**, although the variability was greater in 1973 than 1972. When the 1973 off-site results are examined by measurement period, it appears that generally the readings at all locations varied in a similar manner from period to period. The same effect is apparent also at some on-site locations. The reasons for these results and the abnormally low values in Oak Lawn will be investigated.

The off-site measurements were sufficiently uniform and reproducible for comparison purposes. Twenty-six of the twenty-eight results lie within

TABLE 25

Environmental Penetrating Radiation at Off-Site Locations, 1973

Location	Dose Rate (mrem/year)						Average
	Period of Measurement						
	1/4-3/1	3/1-4/27	4/27-7/5	7/5-9/10	9/10-12/3	12/3-1/4	
Downers Grove	108	98	97	89	96	90	97 ± 17
Lockport	111	110	97	98	103	92	102 ± 20
Lombard	114	104	89	90	123	94	104 ± 34
Oak Lawn	97	88	75	75	107	90	89 ± 15
Oakbrook	-	-	113	107	114	99	110 ± 11
							100 ± 23

TABLE 26

Environmental Penetrating Radiation at ANL, 1973

Location	Dose Rate (mrem/year)						Average
	Period of Measurement						
	1/4-3/1	3/1-4/27	4/27-7/5	7/5-9/10	9/10-12/3	12/3-1/4	
14L - Boundary	103	97	88	80	76	85	87 ± 24
13D - Boundary	91	-	-	-	-	-	91
10E - Boundary	119	-	-	-	-	-	119
8H - Boundary	130	121	111	117	140	121	124 ± 24
7I - Boundary	552	499	511	275	390	271	423 ± 300
8F - Boundary	-	108	-	84	125	103	107 ± 43
9F - Boundary	-	124	101	97	110	101	107 ± 28
9L - Boundary	-	88	83	96	108	95	95 ± 26
7J - Boundary	-	183	164	-	-	-	173 ± 33
14I - Boundary	-	-	-	-	114	111	113
13H - 75 m N of 203	118	99	81	90	111	91	99 ± 34
9H - 75 m W of CP-5	358	-	-	-	-	-	358
9G - 75 m E of CP-5	1170	1100	-	865	2850	1920	1645 ± 1870
8H - 200 m NW (Heliport) of Waste Storage Area	162	136	124	124	179	134	145 ± 54
7I - Center - Waste Storage Area	2180	1980	2200	2600	3580	2790	2600 ± 1440
8J - 400 m N of Waste Storage Area	121	-	-	-	-	-	121
11F - 900 m NW of CP-5	-	97	85	79	114	95	95 ± 33
9H - 50 m S of 314/316	-	139	118	684	204	205	277 ± 600
9H - 25 m N of 314/316, 225 m S of CP-5	-	199	-	188	274	206	223 ± 103
10I - 500 m NE of CP-5	-	116	110	-	-	-	113
9J/K - ZGS Meson Area	-	142	-	-	-	-	142
11H - S of 205, 500 m N of CP-5	-	117	107	103	139	115	118 ± 36

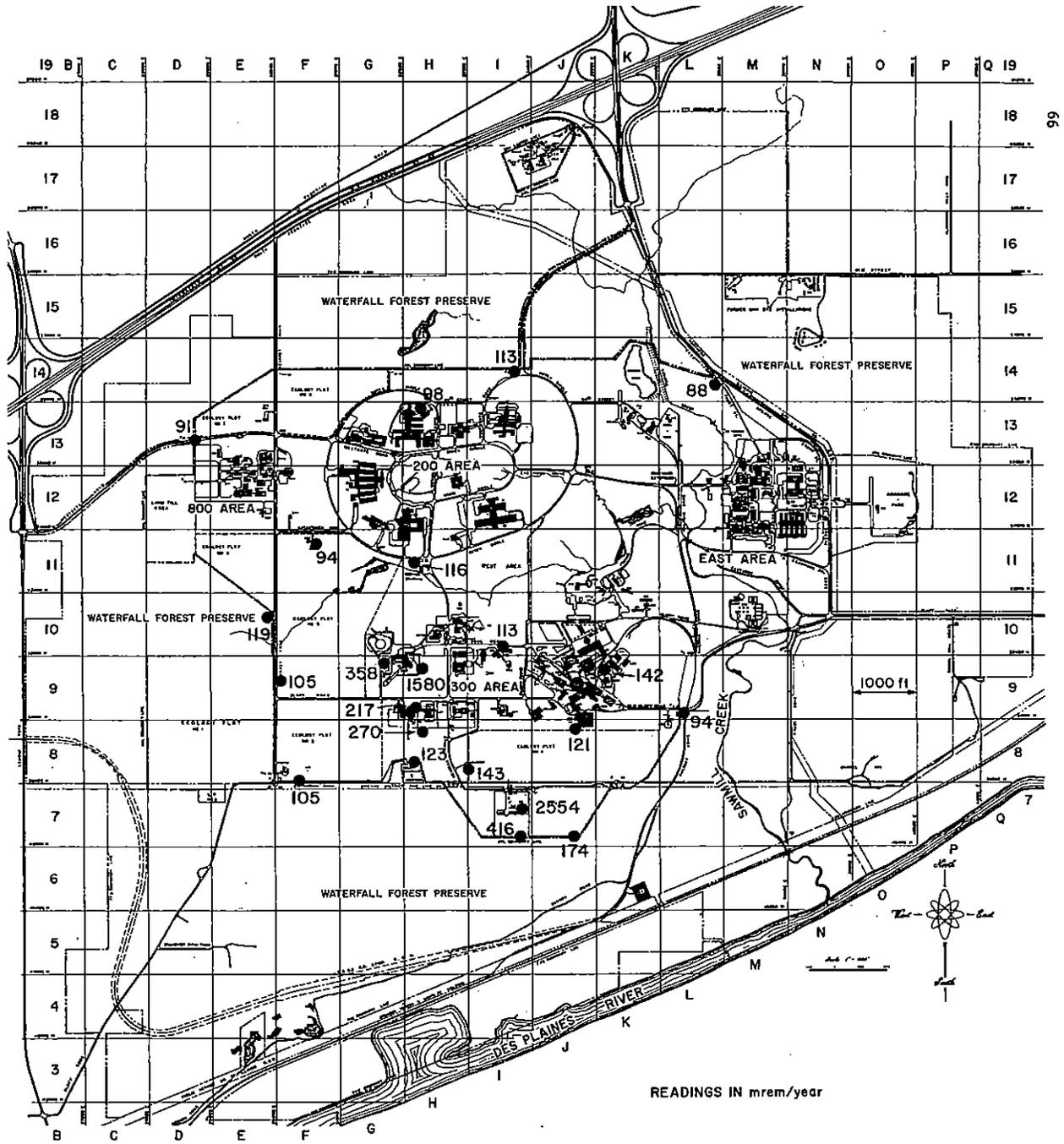


Fig. 5. Penetrating Radiation Measurements on the ANL Site

the 95% confidence limits of the average. If the off-site readings measure and sample the natural radiation background of the area, then a result in the range 100 ± 23 mrem/yr should be normal with a 95% probability. Only 2.5% of the measurements of a natural background radiation field should be higher than 123 mrem/year and 2.5% lower than 77 mrem/year.

Examination of the site boundary results shows that only at three locations were the results consistently above normal. The abnormal doses at location 7I, approximately 320 mrem/year, and at 7J, approximately 73 mrem/year, were 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 shipment for permanent storage elsewhere. The dose in the center of this storage area averaged 2.6 rem/year. The readings at 8H were taken at a cemetery in use before ANL was constructed (St. Patrick's Cemetery). While the average at this location, 124 mrem/year, is only slightly outside of the normal range, a better indication of the dose can be obtained by comparing readings for the same measurement period. The difference between the dose at 8H and the off-site average for the same sampling period ranged from 20-30 mrem/year. Similar results were obtained in 1972.

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 Van de Graaff generator and low-power reactors in Buildings 314 and 316 (location 9G/H), 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 314/316 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.

Location 8H is of particular interest because of the possibility of exposure to individuals visiting the cemetery. If the dose at this location due to ANL is 25 mrem/year, an individual spending 1 hour/week at the

cemetery would receive a dose of 0.15 mrem/year as a result of Argonne operations.

The applicable radiation protection standards for whole body external radiation dose to the general population is a maximum of 500 mrem/year to critical individuals, or, if individual doses are not known, 170 mrem/year 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.⁽¹⁸⁾ The dose at the cemetery is well below these limits. The doses at 7J and 7I are about 15% and 65%, respectively, of the 500 mrem/year limit to individuals, and the latter dose is about twice the 170 mrem 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 dose at any distance can be calculated assuming 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 0.008 mrem/year, if the energy of the radiation were 0.66 MeV, and 0.04 mrem/year, if the energy were 1.33 MeV. The energy spectrum of the radiation is unknown, 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.

Thus, 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/year.

IV. CHEMICAL CONSTITUENTS

The nonradioactive environmental data contained in this report have been collected in an effort to ascertain ANL 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.⁽¹⁹⁾ The new regulations represent a comprehensive and well-documented statement with regard to the quality of the aquatic sector.

The concentrations of barium, copper, iron, nickel, silver, and zinc were determined using conventional atomic absorption spectrophotometry. Mercury was determined using cold atomic absorption spectrophotometry. Fluoride and chloride levels were determined using ion selective electrodes, and the pH was determined using conventional pH electrode measurements. Dissolved oxygen, sulfate, ammonia nitrogen, and hexavalent chromium levels were determined using procedures described in Standard Methods.⁽²⁰⁾ Beryllium was determined fluorophotometrically as previously described.⁽²¹⁾

The results of the measurement of chemical constituents are expressed as milligrams per liter (mg/l) except as indicated. 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 and minimum detectable amounts are included for comparison (Table 27). 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.

TABLE 27

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
Chloride	500	-	0.5
Chromium (total hexavalent)	0.05	0.3	0.006
Copper (total)	0.02	1.0	0.009
Fluoride	1.4	-	0.02
Fluoride (total)	-	2.5	0.02
Iron (total)	1.0	2.0	0.08
Mercury	0.0005	-	0.0001
Mercury (total)	-	0.0005	0.0001
Nickel (total)	1.0	1.0	0.3
pH	6.5-9.0	5.0-10.0	-
Silver	-	0.1	0.01
Silver (total)	0.005	-	0.01
Sulfate	500	-	5.0
Total Dissolved Solids	1000	3500	2.0
Zinc	1.0	-	0.01
Zinc (total)	-	1.0	0.01

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

A. Sawmill Creek

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

During the early part of the year, grab samples were collected in Sawmill Creek three times per week in the morning (0900-1000). However, since the largest contribution of the chemical waste to the total effluent occurs in early afternoon, most of the samples reported in Sawmill Creek were collected in the period from 1300-1400. The reason for using non-random collection was to provide information needed to determine the source of contamination.

Since a different sampling technique is required, samples to be analyzed for dissolved oxygen, dissolved solids, 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 28. 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 varied from 4.1 to 13.2 mg/l above the outfall and from 6.0 to 13.0 mg/l below the outfall. The oxygen content of the stream is determined primarily by the quality of the water upstream of the Laboratory since the average dilution of Laboratory waste is about ten to one. These data represent little change in the ranges reported in the past, but the overall quality seems improved.

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 outfall were in the

TABLE 28

Chemical Constituents in Sawmill Creek, 1973

Constituent	Location *	No. of Samples	Concentration (mg/l)			Percent of State Levels		
			Av.	Min.	Max.	Av.	Min.	Max.
Ammonia N	7M (up)	42	2.19 ± 0.56	< 0.1	6.2	146	< 7	414
	7M (down)	43	1.82 ± 0.13	< 0.1	6.4	122	< 7	427
Barium (total)	7M	52	2.92 ± 0.31	0.63	4.93	58	13	99
Beryllium	7M**	12	0.088 ± 0.075	0.01	0.21	-	-	-
Chloride	7M (up)	9	294 ± 59	129	377	59	26	75
	7M (down)	9	302 ± 13	149	355	60	30	71
Chromium(VI)	7M (up)	20	0.027 ± 0.024	< 0.006	0.234	54	< 12	468
	7M (down)	151	0.037 ± 0.007	< 0.006	0.237	74	< 12	474
Copper (total)	7M	52	0.016 ± 0.002	0.009	0.036	80	45	180
Fluoride	7M	47	0.33 ± 0.02	0.22	0.51	24	16	36
Iron (total)	7M	52	0.65 ± 0.12	0.13	1.91	65	13	191
Mercury (total)	7M (up)**	43	-	-	< 0.1	-	-	< 20
	7M (down)**	108	0.5 ± 0.2	< 0.1	12.3	100	< 20	2460
Mercury (dissolved)	7M (down)**	92	0.23 ± 0.08	< 0.1	4.2	46	< 20	840
Nickel (total)	7M	52	-	-	< 0.3	-	-	< 30
Silver (total)	7M	52	-	-	< 0.01	-	-	< 200
Sulfate	7M (up)	16	152 ± 23	62	218	30	12	44
	7M (down)	16	154 ± 20	62	208	31	12	42
Zinc	7M	52	0.133 ± 0.015	0.048	0.299	13	5	30

* Location 7M 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 .

range from 7.1 to 9.1. Samples obtained above the outfall were in the range of 7.4 to 8.7.

The average ammonia nitrogen levels are slightly greater than the stream criteria both above and below the Argonne outfall. The source has been shown in past reports⁽²²⁾ to be well upstream of the Laboratory boundary. However, the values have been substantially reduced from past years, reflecting improved treatment upstream. The range of ammonia nitrogen found in the stream reflects the effect of temperature, the higher results being obtained in the colder weather.

Values for chloride and sulfate are reported on a limited basis, since past data indicated that no change from Argonne operations was occurring.

The average levels for beryllium, copper, fluoride, iron, and silver during this report period remained essentially the same as for 1972. The result for barium differs from the last reporting period due to increased sensitivity of the analytical technique. The average result is about 60% of the State limit. The source of the barium is not known, but it appears to be at least partially due to natural levels.

The levels of mercury are higher than the previous period primarily due to two reasons. The previous period reported only dissolved mercury as was then required. In anticipation of adoption of the new rule using total mercury, this value is reported herein. In general, the amount of mercury that is dissolved is a small percent of the total amount present. The second reason for higher results was that sampling was centered on the time period when maximum Laboratory waste was released. A twenty-four hour sample would produce much lower average results. However, it can be seen that mercury levels are substantially greater below the ANL outfall than above it. The average annual value was 0.5 $\mu\text{g}/\text{l}$, which is the Illinois limit.

The higher levels of hexavalent chromium both above and below the outfall as well as the higher values of zinc below the outfall result from a diversion of some of the blowdown effluents from the ZGS Area directly to the stream approximately one mile upstream from the ANL outfall. Levels

for hexavalent chromium averaged 54% of the State limit upstream of the Laboratory and 74% downstream of the outfall.

Since zinc chromate is used in the cooling towers, the average level of zinc also increased by about a factor of two from last year. These levels are substantially below the Illinois limit, and the average value is 13% of the limit.

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.

B. Effluent Water

The principal effluent from Argonne is the waste treatment stream which enters Sawmill Creek at 7M. The stream is sampled by a proportional collector at the waste treatment facility immediately after treatment. The water actually enters the Creek some 1,500 yards 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 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 enters Sawmill Creek and its contribution is assessed in the stream samples. Grab samples were obtained weekly at 8J and 14J, and they were analyzed for chromium immediately.

The results obtained in the waste treatment plant effluent samples (Table 29) are comparable to the stream results with four exceptions, zinc, chromium, mercury, and copper. The levels of zinc are about three times higher in the plant effluent than in the stream. The levels of the stream and effluent are more nearly the same than was reported last year due in part to releasing some of the effluent containing zinc chromate normally

TABLE 29

Chemical Constituents in Effluent from ANL Treatment Plant, 1973

Constituent	No. of Samples	Concentration (mg/l)			Percent of Standard		
		Av.	Min.	Max.	Av.	Min.	Max.
Barium (total)	54	2.35 ± 0.35	0.51	8.53	118	26	427
Beryllium*	48	0.051 ± 0.0004	< 0.007	0.16	-	-	-
Chromium(VI) (total)	254	0.054 ± 0.004	< 0.006	0.186	18	< 2	62
Copper (total)	54	0.0276 ± 0.00004	0.012	0.101	2.8	1.2	10.1
Fluoride (total)	50	0.3260 ± 0.0004	0.222	0.442	13.0	8.8	17.6
Iron (total)	54	0.42 ± 0.23	0.06	5.75	21.0	3.0	288
Mercury (total)*	256	1.96 ± 0.45	0.21	38.1	392	42	7620
Mercury (dissolved)*	256	0.33 ± 0.11	< 0.10	14.0	66	< 20.0	2800
Nickel (total)	54	-	-	< 0.30	-	-	< 30
pH	260	-	6.82	8.80	-	-	-
Silver	54	-	-	< 0.01	-	-	< 10
Zinc (total)	54	0.350 ± 0.031	0.12	0.73	35	12	73

*All concentration values multiplied by 10³.

found at 8J to the stream at 11L. The levels in the effluent averaged 35% of the State limit of 1.0 mg/l but never exceeded it.

The average value for hexavalent chromium was also higher than the stream values 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 50% greater 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 0.19 mg/l and exceeded the effluent limit of 0.3 mg/l about 25% of the time. This stream enters Sawmill Creek at a location above the ANL outfall and its contribution to the overall level of hexavalent chromium is assessed by the stream sample. The cooling tower blowdown at 8J averaged 0.93 mg/l, or almost three times the State effluent standard. As previously mentioned, this water does not usually reach the Des Plaines River, but soaks into the ground. To assess this source as well as any others, samples were obtained in the Des Plaines 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.

An extensive study was conducted throughout this year to determine the source of mercury in the effluent. The details of the study are given in a special report.⁽²³⁾ Basically all of the waste storage tanks and Laboratory waste lines were sampled for mercury content. The results indicated that one area of the Laboratory complex was the most likely site for contamination. Subsequently, the waste storage tanks in this area were sampled before and after a flocculation treatment designed to reduce mercury levels. Reductions in the content of mercury averaged approximately 80%. A recent study combining flocculation and evaporation reduced the mercury content of one tank by 99.9%. These removal studies were performed at the end of 1973 and the beginning of 1974. Lower average levels in the effluent were obtained during the treatment period and it is anticipated that future results will indicate substantially lower levels in both effluent and stream. This will continue

to be closely monitored. The average levels obtained in 1973 agreed with the levels reported in 1972. The average level in the effluent was 1.96 µg/l, which is three times the State limit. This value is heavily biased by high results in March and April. These months represented periods of mercury re-processing which we have identified as a major source. The treatment of the waste before release should greatly reduce subsequent effluent concentrations.

The copper levels in the effluent were about twice the levels found in the stream. These values are well below the State of Illinois limit and could be due to the use of copper tubing in Laboratory systems.

Fecal coliform studies were conducted in the stream from April through December. The samples were analyzed using the membrane filter technique. Values obtained above the ANL outfall ranged from 21 coliforms/100 ml to 3,400 coliforms/100 ml. The results are expressed as the monthly geometric mean. Results below the outfall ranged from < 1 coliform/100 ml to 17 coliforms/100 ml. The State limit is 200 coliforms/100 ml.

The ANL 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.⁽²⁰⁾ Results are as shown in Table 30.

The value for ammonia nitrogen obtained in February exceeded the State limit of 4.0 mg/l. The values for B.O.D. and suspended solids were well below State regulations during the entire period.

Total coliform bacteria analyses were performed once per week using the fermentation tube technique. Results obtained in May, July, September, and October were above the State limit. During these months, construction work was being conducted at the plant and a new automatic chlorinator was being installed. The remainder of the results were well within limits for total coliform set forth in SWB-11.⁽²⁴⁾

TABLE 30

Deoxygenating Waste and Coliform Measurements
in ANL Effluent Water, 1973

Month	Concentration (mg/l)			Coliform Concentration (MPN) Coliforms/100 ml
	B.O.D. ₅	Suspended Solids	Ammonia Nitrogen	
January	3.4	1.25	3.17	420
February	3.0	0.75	6.24	483
March	1.8	3.00	3.80	254
April	1.8	2.00	1.88	210
May	1.1	1.50	0.35	12,000
June	1.2	6.25	0.50	318
July	1.0	2.67	0.54	5,010
August	0.8	2.89	0.43	228
September	2.3	5.14	1.88	4,050
October	2.3	10.50	1.62	3,890
November	1.7	5.0	1.83	150
December	2.2	0.33	1.38	130
State Limit	30.0	37.0	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	5,000

V. APPENDIX

A. References

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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 31 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 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, non-occupationally, for 168 hours per week. Both CGs are given in the table for nuclides released by 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 (twice the standard deviation) is equal to the detection limit. The error 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%.

The factors used in this report to calculate internal dose from air and water concentrations are shown in Table 32. The source of these factors is discussed in the introduction to Part III.

TABLE 31

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

Nuclide or Activity	Concentration Guide		Detection Limit	
	Water	Air	Water	Air
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}
Cerium-141	-	5×10^{-9}	-	5×10^{-16}
Cerium-144	-	2×10^{-10}	-	10^{-15}
Cesium-137	2×10^{-5}	5×10^{-10}	-	5×10^{-16}
Cobalt-60	-	3×10^{-10}	-	10^{-16}
Hydrogen-3	3×10^{-3}	2×10^{-7} 5×10^{-6}	2×10^{-7}	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-238,239	5×10^{-6}	1×10^{-12}	5×10^{-13}	10^{-19}
Ruthenium-103	-	3×10^{-9}	-	5×10^{-16}
Ruthenium-106	-	2×10^{-10}	-	10^{-15}
Strontium-89	3×10^{-6}	3×10^{-10}	1×10^{-9}	10^{-16}
Strontium-90	3×10^{-7}	3×10^{-11}	2×10^{-10}	10^{-17}
Thorium-228	-	2×10^{-13}	-	10^{-18}
Thorium-230	-	3×10^{-13}	-	10^{-18}
Thorium-232	-	1×10^{-12}	-	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 *	$\left\{ \begin{array}{l} 3 \times 10^{-6} \\ \text{to} \\ 1 \times 10^{-7} \end{array} \right.$	$\left\{ \begin{array}{l} 1 \times 10^{-10} \\ \text{to} \\ 1 \times 10^{-13} \end{array} \right.$	2×10^{-10}	2×10^{-16}
Beta *				10^{-9}

*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.

TABLE 32

Concentration-to-Dose Conversion Factors

Nuclide	Medium	Concentration* ($\mu\text{Ci/ml}$)	Dose* (rem)	Critical Organ
^3H (H_2O)	Air	2×10^{-7}	0.5	Whole Body
	Water	3×10^{-3}	0.5	Whole Body
^{41}Ar	Air	4×10^{-8}	0.5	Whole Body
^{90}Sr	Water	3×10^{-7}	3	Bone
^{237}Np	Water	3×10^{-6}	3	Bone
^{239}Pu	Water	5×10^{-6}	3	Bone

*The concentrations and doses are the radiation standards specified in AECM 0524 for individuals in uncontrolled areas.(5)

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