

**ENVIRONMENTAL MONITORING AT  
ARGONNE NATIONAL LABORATORY  
ANNUAL REPORT FOR 1972**

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



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by

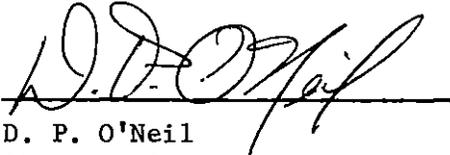
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ABSTRACT

The environmental monitoring program at Argonne National Laboratory for 1972 is described and the results are presented. To evaluate the effect of Argonne operations on the environment, measurements were made for a variety of radionuclides in air, surface water, soil, grass, benthos, and milk; for a variety of chemical constituents in surface and Argonne effluent water; and for 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 by AEC Manual Chapter 0513. 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 potentially toxic radioactive or chemical substance present in the environment. Of special interest is the detection of any such material released to the environment by Argonne. If releases by Argonne begin to approach significant levels, immediate steps will be taken to eliminate the release.

Argonne is a multi-disciplinary research and development laboratory with two principal objectives: It carries out a broad program of basic research activities, and it serves as an important center for the design and development of nuclear reactors. 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. Almost all of the work at the Laboratory is of an unclassified nature and deals with peacetime applications of nuclear energy.

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) is located on a 3,740-acre site in Du Page County, 27 miles southwest of downtown Chicago, and 24 miles due west of Lake Michigan. It lies south of Interstate Highway 55 and west of Illinois Highway 83. 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. Figures 1 and 2 are maps of the site and of the surrounding area.

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 0.5 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  $15^{\circ}$  to  $60^{\circ}$ , reaching an average elevation of

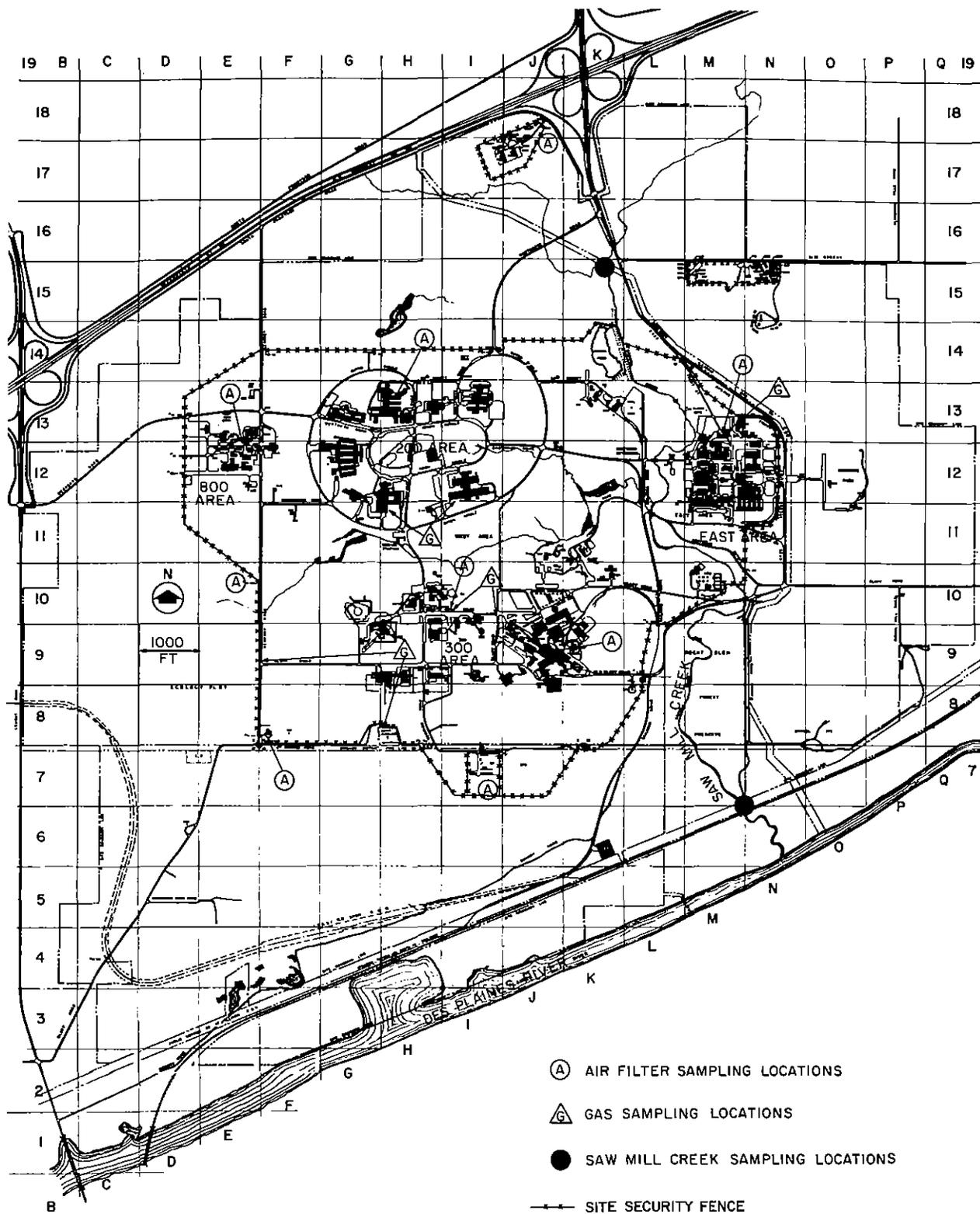


Fig. 1. Sampling Locations on the Site of Argonne National Laboratory

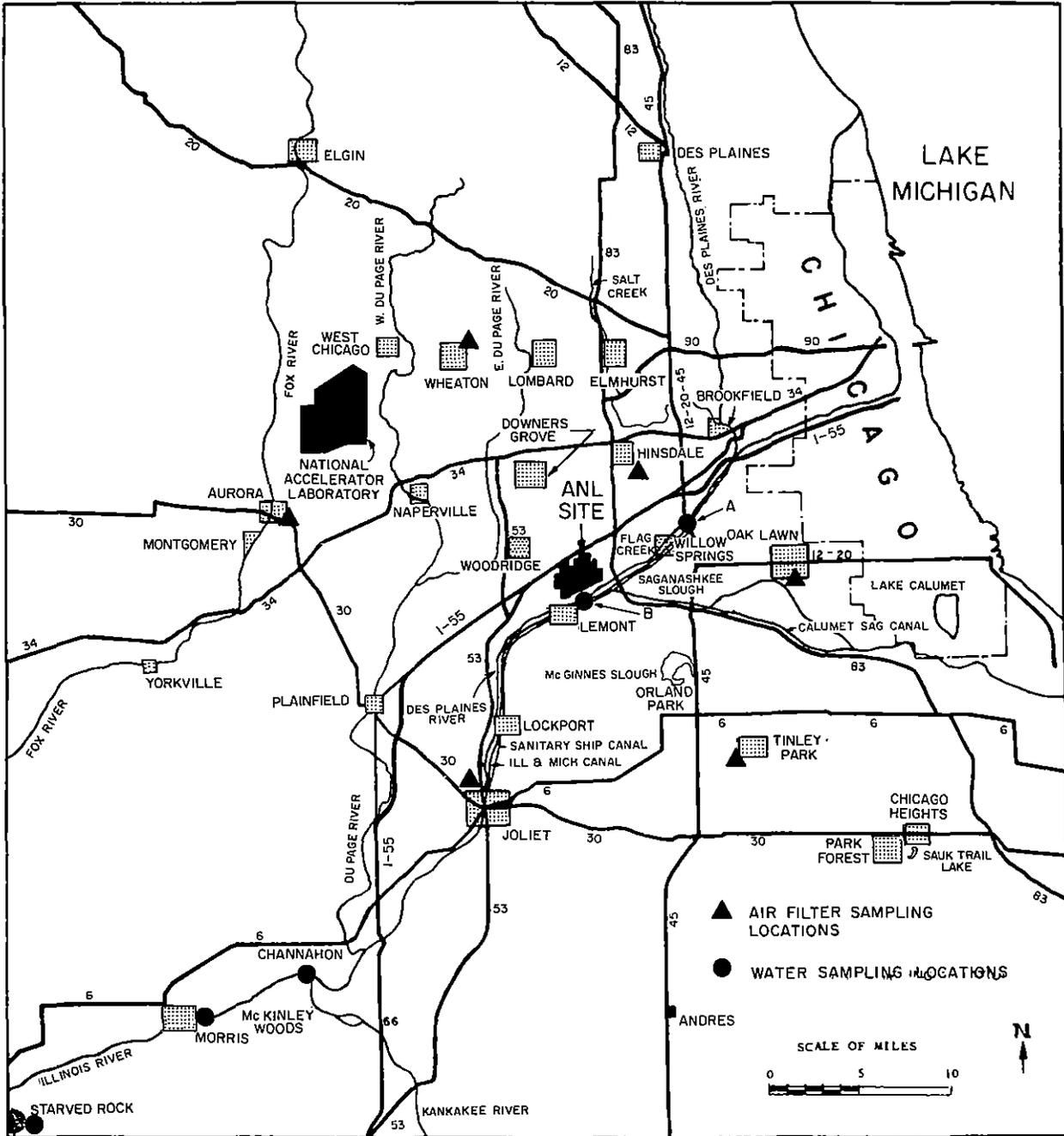


Fig. 2. Sampling Locations near Argonne National Laboratory

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 more than 25 feet in a distance of 500 horizontal feet.

The principal Laboratory facilities and buildings are located in the central 1,700 acres of the site. This area is surrounded by a security fence, and entrance is restricted to business purposes. The central area is surrounded by a 2,040-acre tract planted primarily with softwood trees. A pre-existing cemetery, to which visitors are allowed, is located in the southwest portion of the site (coordinate 8H, Figure 1). This is the closest permitted non-business approach to the security fence. In the southern portion of the site 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.<sup>(1)</sup> 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. The number 12.67 in the center of the rose represents the percent of observations of wind speed greater than 4 mph. The curves for 4 to 24 mph and less than 4 mph are almost identical.

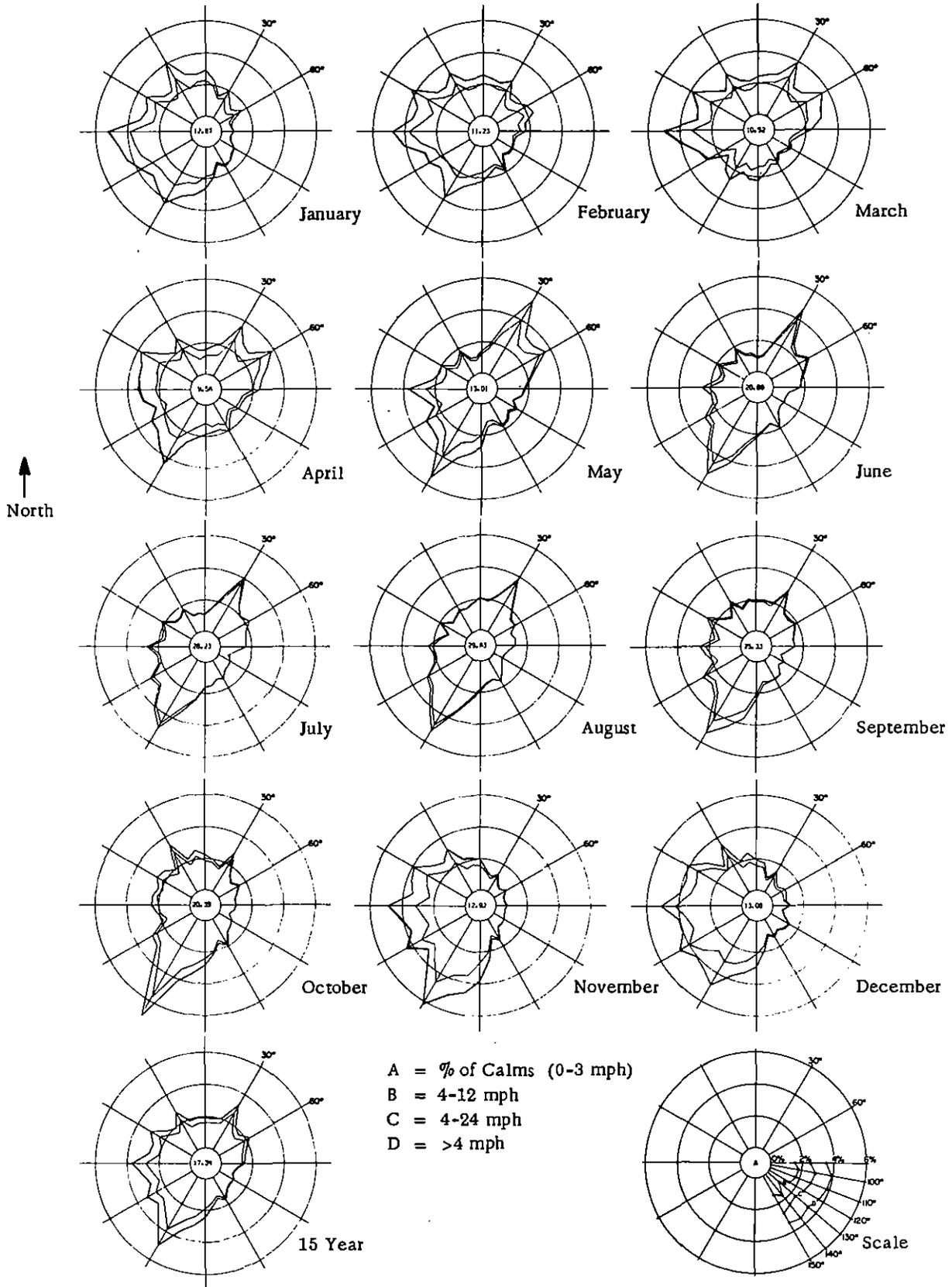


Fig. 3. Wind Roses Based on Hourly Observations, January 1950-December 1964, 19-Foot Level

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 varies from 5 mph in August to 10 mph in March. Gusts exceed 50 mph about once a year.

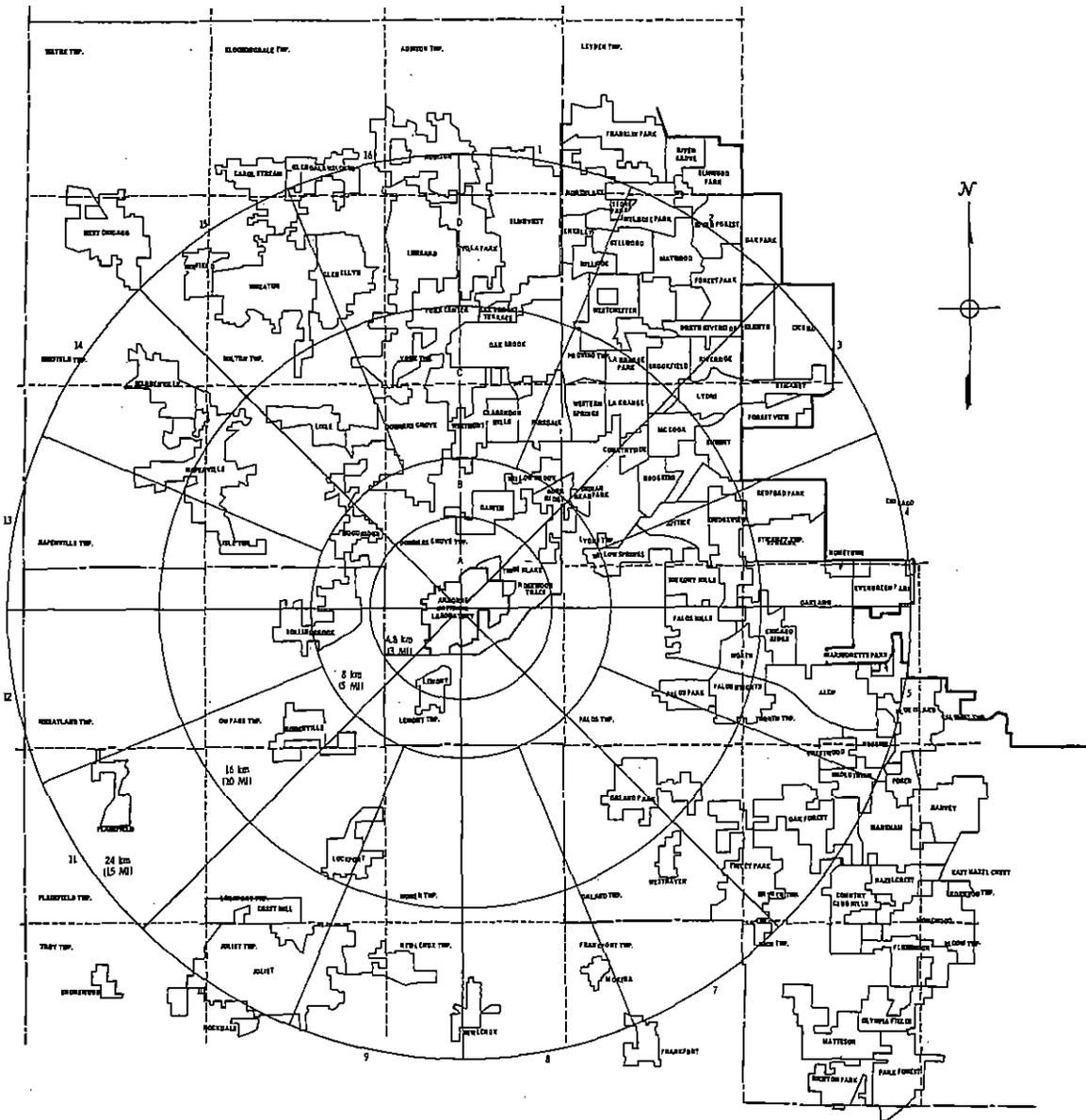
The air temperature data show a typical seasonal variation. Daily temperatures average from the low 20's in the winter to the 70's in the summer. The extreme temperatures recorded were  $-20^{\circ}\text{F}$  and  $100^{\circ}\text{F}$ .

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 on the northeast portion 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 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 the  $22.5^{\circ}$  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  $41.70^{\circ}$  No. lat. and  $87.98^{\circ}$  W. lon.



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

Fig. 4. Communities and Townships Within a 15 Mile Radius of ANL

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
Bloomingtondale	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	0	0	2592	310	33929	181886	257828	93833	185403
NNE	0	0	361	3420	4423	37082	336097	538710	112647	0
NE	0	0	3021	1175	1148	28825	803173	1071375	0	0
ENE	0	0	0	0	668	27658	737736	333866	0	0
E	0	0	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	1677	0	0	0	924	10503	6437	13373	18151
S	0	0	0	1542	0	2621	12007	1224	26042	38362
SSW	0	1848	3232	0	0	16232	104383	7940	14396	7063
SW	0	0	0	0	0	10260	24359	4470	13718	7223
WSW	0	0	0	462	0	5535	4201	1795	5560	9992
W	0	0	0	3387	4955	951	38397	16101	16475	7371
WNW	0	0	0	0	0	18162	82536	7700	3981	49352
NW	0	0	0	0	9311	11431	36959	63029	13843	11456
NNW	1515	0	1424	1188	3341	33270	92276	110962	85107	63205
Total	1515	3525	8038	13781	24156	271765	3194239	3047244	723432	470096
<u>Cumulative</u>										
Total	1515	5040	13078	26859	51015	322780	3517019	6564263	7287695	7757791

### E. Land and Water Use

Argonne waste water is discharged into Sawmill Creek at location 7M in Figure 1. 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 this area has resulted in the collection and channeling of additional runoff water into the Creek. The total water flow in the Creek for 1972 averaged about 7 million gallons per day. This unusually high volume was due partially to above-average precipitation (37.8 in.) and partially to the occurrence of heavy rains in the latter part of the year when the ground was saturated with water. The Laboratory effluent contribution to the Creek varied between 0.75 and 1 million gallons per day.

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. Communities closer to Lake Michigan use water from this lake.

The principal recreational area near Argonne is Rocky Glen Forest Preserve, bordering the site at the southeast corner (Figure 1), and part of the Du Page County Forest Preserve District. Sawmill Creek flows through Rocky Glen on its way to the Des Plaines River. The Preserve is used principally

for picnicking, hiking, and overnight camping by youth groups. During 1972, there were approximately 9,000 overnight stays by individuals. Based on the number of group permits issued, at least 15,000 people used the Forest Preserve during 1972 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 2,200 sq. mi. area around the Laboratory is shown by County in Table 4. The data are for 1971, the last year for which information is available. There is little commercial vegetable and fruit production near the Laboratory.

TABLE 4

Farm Production Near ANL, 1971

County	Corn	Acres Farmed			Hay	Milk Cows (number)
		Soy Beans	Oats	Wheat		
Du Page	22,800	17,900	2,100	1,300	2,400	700
Cook	24,500	22,100	3,800	1,600	6,800	1,400
Will	154,500	101,200	15,800	5,500	15,700	6,300

## 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. Included in this report are the results of measurements obtained in 1972 for a number of radionuclides in air, surface water, soil, grass, benthic materials, and milk; for a variety of chemical constituents in water; and the results of a continuing program to measure the environmental 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 by the action of cosmic rays on air), and fission and neutron activation products were measured in air filter samples. The concentrations were very similar on and off the site indicating that these radioactivities originated from widespread sources - global fallout from nuclear test detonations and naturally-occurring materials - and not from a localized source such as the Laboratory. The beta activity, due principally to fallout, averaged  $0.1 \times 10^{-12}$   $\mu\text{Ci/ml}$ , about a factor of three lower than in 1971. In terms of the concentration guides (CG)\*, the air activities ranged from  $6 \times 10^{-5}\%$  of the CG for cerium-141 to 0.97% of the CG for mixed beta activity. The detection of the short-lived fission products, iodine-131 and barium-140, at all sampling locations, during the first half of the year, along with an increase in the 40-day ruthenium-103 and 65-day zirconium-95 in the spring, indicates the presence of fallout from a recent series of atmospheric test detonations.

Air sampling for plutonium was conducted on the Argonne site beginning in March. The plutonium-239\*\* and plutonium-238 concentrations in monthly samples

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\* The hazard due to a given concentration of a radioactive nuclide is usually assessed in this report by comparison with the Concentration Guide (CG), 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.

\*\* The plutonium measurements were made by alpha particle spectrometry, which cannot distinguish between plutonium-239 and plutonium-240. Although only plutonium-239 is listed, it should be understood that the alpha activity due to the less abundant 240 isotope is also included.

varied from 1 to  $45 \times 10^{-18}$   $\mu\text{Ci/ml}$ , well within the range reported by the AEC Health and Safety Laboratory for fallout plutonium in air samples collected away from nuclear installations. The average concentration was  $27 \times 10^{-18}$   $\mu\text{Ci/ml}$  (0.003% of the CG for uncontrolled areas) for plutonium-239 and  $2.2 \times 10^{-18}$   $\mu\text{Ci/ml}$  (0.0002% of the CG) for plutonium-238.

Hydrogen-3 (as tritiated water vapor) and argon-41 concentrations in air were measured near the CP-5 reactor since the reactor is the primary source of these activities at Argonne. The argon-41 average and maximum concentrations ( $1.2 \times 10^{-7}$  and  $1.2 \times 10^{-6}$   $\mu\text{Ci/ml}$ , respectively) at this location were about 6% and 60% of the CG for controlled areas. The argon-41 concentration at the site security fence was less than  $8 \times 10^{-10}$   $\mu\text{Ci/ml}$ , equivalent to less than 2% of the CG for uncontrolled areas. The concentration of tritiated water vapor averaged  $1.4 \times 10^{-10}$   $\mu\text{Ci/ml}$ , which is 0.07% of the CG for uncontrolled areas and almost a factor of three lower than 1971. Samples collected at the site security fence closest to the CP-5 reactor had average and maximum concentrations of  $1.5 \times 10^{-11}$  and  $9.6 \times 10^{-11}$   $\mu\text{Ci/ml}$ , respectively, which are equivalent to 0.008 and 0.05% of the CG. The off-site concentration, which is the normal level for this area, averaged  $2.4 \times 10^{-12}$   $\mu\text{Ci/ml}$  (0.0012% of the CG) and is a factor of two lower than the 1971 average.

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 principal radionuclide added by Argonne waste-water was hydrogen-3. The average and maximum concentrations of  $5.6 \times 10^{-7}$  and  $1.9 \times 10^{-6}$   $\mu\text{Ci/ml}$ , respectively, are only 0.02 and 0.06% of the CG. The average tritiated water concentration in Sawmill Creek below the Laboratory waste-water outfall decreased by a factor of three compared to 1970 and by a factor of 20 compared to 1971. Traces of neptunium-237 and plutonium-239 were also added occasionally by the waste-water, but the maximum concentration of either of these nuclides in any one Creek sample amounted to only 0.005% of the CG.

Sawmill Creek flows into the Des Plaines River, which in turn becomes part of the Illinois River. The radioactivity levels in the latter two streams were normal and no radionuclides added to the Creek by Argonne waste-water could be found in the Rivers. The identification of the short-lived fission products, iodine-131 and barium-140, at all locations during May and plutonium-239 during the summer can be attributed to fallout from nuclear tests.

Plutonium concentrations in soil showed the same general range and average concentrations on and off the site. The average plutonium-239 concentration was  $(1.83 \pm 0.24) \times 10^{-3} \mu\text{Ci}/\text{m}^2$  on-site and  $(1.63 \pm 0.29) \times 10^{-3} \mu\text{Ci}/\text{m}^2$  off-site. These results fall within the range measured by other laboratories for fallout plutonium. The plutonium-239 content of grass was similar both on and off the site and ranged from 0.17 to  $0.67 \times 10^{-6} \mu\text{Ci}/\text{m}^2$ , a factor of two thousand to five thousand lower than in soil from the same location. An examination of the Sawmill Creek bed below the Laboratory waste-water outfall for plutonium showed the same concentrations as off-site benthic material.

Raw milk collected from a dairy farm near the Laboratory was analyzed monthly for several fission products. Strontium-89, iodine-131, and barium-140 were not present above the detection limit. The average cesium-137 concentration of  $< 11 \times 10^{-9} \mu\text{Ci}/\text{ml}$  is  $< 0.025\%$  of the CG for drinking water and the strontium-90 concentration,  $5.0 \times 10^{-9} \mu\text{Ci}/\text{ml}$ , is  $0.75\%$  of this CG. The average strontium-90 content decreased by 10% from last year while the cesium-137 concentration decreased about 30%.

Environmental penetrating radiation measurements were made with thermoluminescent dosimeters on and off the Argonne site, including 12 locations along the site security fence. Off-site values averaged  $105 \pm 5 \text{ mrem}/\text{year}$ , and this range is considered normal for the area. Two locations at the site security fence gave above-normal readings. These locations, both on the south fence, gave average readings about 20 mrem/year (8G in Figure 1) and 300 mrem/year (7I) above background. This dose was due to radioactive materials in a Waste Storage Area located at 7I. Based on extrapolations from the security fence measurements, the dose at the actual Argonne site south boundary would not be distinguishable from background. The dose rates at 8G and 7I are 4 and 60%, respectively, of the annual whole body exposure standard for critical individuals (500 mrem/year) and 12 and 176%, respectively, of the average exposure standard for a suitable sample of the exposed population. However, these locations are very rarely occupied and there are no individuals receiving this dose.

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 January and February, when the two-month average was about twice the state standard of 4 mg/l. However, samples obtained from Sawmill Creek both upstream and downstream of the ANL waste-water outfall indicated that the net effect of the effluent water was to lower ammonia nitrogen levels.

The levels of chemical constituents in the effluent were within acceptable limits with one exception. The average value for total mercury in the effluent was about 3.5 times the state standard. The exact source of this mercury has not as yet been determined, but the effect on the levels in Sawmill Creek was small. Only 3% of the individual samples contained mercury in excess of the detection limit of 0.0001 mg/l, and the average concentration was less than 20% of the state standard.

Levels of barium, iron, lead, nickel, silver, and zinc in Sawmill Creek below the outfall were comparable to previous years and below the state limit. Levels of copper are also comparable to past years, but the standard has been lowered to values approaching background levels. Beryllium concentrations were comparable to past results and appear to be at natural levels.

### 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 included in this report are those that are pertinent to the evaluation of Argonne's contribution to the environmental radioactivity and to the differentiation of Argonne activity from fallout and other sources.

The results of radioactivity measurements are expressed in this report in terms of microcuries per milliliter ( $\mu\text{Ci/ml}$ ) for water, air, and milk and microcuries per gram (g) and square meter ( $\text{m}^2$ ) for soil and vegetation. When a

nuclide was not detected, the result is given as less than (<) the minimum amount detectable (detection limit) by the analytical method used. Averages including individual results that were less than the detection limit were calculated by one of the following two methods. If 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 is 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, 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 given in AEC Manual Chapter 0524. 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.

#### A. Radioactivity in Air

The radioactivity of particulate matter in air was determined by drawing air through filter paper at a known rate and measuring the radioactivity of the particles collected by the paper. Samples were collected continuously, except for occasional equipment failures, at nine locations on the Argonne site and at six locations off the site. The sampling locations are shown in Figures 1 and 2. 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. Higher activities on the site are indicative of radioactivity released by Argonne if the differences are greater than the error in sampling and counting. This 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. The alpha and beta activities were measured 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 cm<sup>3</sup> lithium-drifted germanium diode as the detector. This detector was calibrated for each gamma-ray emitting nuclide given in Table 6.

The activities were very similar on and off the site and no significant differences between the locations were found. This indicates that Argonne did not add detectably to the average airborne particulate activity of the environment during the year, and that the activities originated in widespread sources - fallout from nuclear test detonations and naturally-occurring materials - and not in localized sources, such as the Laboratory.

The alpha activities averaged 25% lower than 1971 but were in their normal range. The alpha emitters in the air samples are principally naturally-occurring. About two-thirds of the gamma activity and a smaller fraction of the beta activity was due to beryllium-7, produced in the stratosphere by cosmic-ray interactions. Most of the remaining beta and gamma activity was due to fission and neutron activation products from nuclear test detonations.

TABLE 5

Total Alpha and Beta Activities in Air-Filter Samples, 1972\*

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	40	2.3	1.0	3.4	0.11	0.042	0.26
	off-site	22	2.6	0.8	6.1	0.13	0.023	0.22
February	on-site	32	2.4	1.3	4.0	0.087	0.062	0.13
	off-site	21	3.4	1.3	12.5	0.095	0.041	0.23
March	on-site	34	2.2	0.9	3.2	0.075	0.046	0.10
	off-site	26	2.8	1.3	8.2	0.078	0.035	0.11
April	on-site	34	2.7	1.3	4.5	0.094	0.045	0.32
	off-site	21	2.6	1.5	4.8	0.094	0.058	0.37
May	on-site	35	3.5	2.1	7.0	0.21	0.086	0.67
	off-site	23	3.3	1.7	6.1	0.23	0.065	0.60
June	on-site	34	3.1	1.0	8.0	0.21	0.11	0.44
	off-site	22	2.6	1.2	5.1	0.18	0.11	0.30
July	on-site	37	3.2	1.1	6.8	0.14	0.060	0.30
	off-site	23	2.5	1.1	3.8	0.14	0.069	0.27
August	on-site	32	2.1	0.6	4.1	0.072	0.042	0.12
	off-site	25	1.9	0.9	3.1	0.073	0.048	0.16
September	on-site	37	1.9	0.7	3.6	0.050	0.026	0.089
	off-site	24	1.8	0.8	3.0	0.048	0.022	0.082
October	on-site	37	1.9	0.7	4.1	0.036	0.017	0.074
	off-site	26	2.1	0.9	4.4	0.036	0.023	0.066
November	on-site	35	1.6	0.3	7.3	0.032	0.014	0.072
	off-site	22	1.8	0.9	5.6	0.028	0.017	0.054
December	on-site	34	1.9	0.3	4.3	0.046	0.023	0.28
	off-site	24	2.1	0.7	4.4	0.040	0.023	0.065
Annual Summary	on-site	421	2.4 $\pm$ 0.3	0.3	8.0	0.097 $\pm$ 0.035	0.014	0.67
	off-site	279	2.5 $\pm$ 0.3	0.7	12.5	0.098 $\pm$ 0.036	0.017	0.60

\* 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, 1972  
(concentrations in  $10^{-15}$   $\mu\text{Ci/ml}$ )

Nuclide	Location	January	February	March	April	May	June	July	August	September	October	November	December	Annual Average
Antimony-125	on-site	1	< 1	1	1	2	2	1	< 1	< 1	< 1	< 1	< 1	1 $\pm$ 1
	off-site	1	< 1	1	1	2	2	1	< 1	< 1	< 1	< 1	< 1	1 $\pm$ 1
Barium-140- Lanthanum-140	on-site	6	4	1	2	4	2	1	< 1	< 1	< 1	< 1	< 1	2 $\pm$ 4
	off-site	6	3	1	2	4	2	1	< 1	< 1	< 1	< 1	< 1	2 $\pm$ 4
Beryllium-7	on-site	70	81	116	103	150	140	105	85	95	91	67	85	99 $\pm$ 54
	off-site	70	74	108	105	158	120	97	80	88	87	66	81	95 $\pm$ 52
Cerium-141	on-site	2	2	1	2	10	10	6	2	1	< 1	< 1	< 1	3 $\pm$ 7
	off-site	3	2	1	1	10	9	6	2	1	< 1	< 1	< 1	3 $\pm$ 6
Cerium-144	on-site	10	10	13	11	23	23	14	7	6	3	2	2	10 $\pm$ 13
	off-site	9	9	14	12	22	20	14	6	5	3	2	2	10 $\pm$ 12
Cesium-137	on-site	1	2	2	2	3	3	2	1	1	1	< 1	< 1	2 $\pm$ 2
	off-site	1	1	2	2	3	3	2	1	1	1	< 1	< 1	2 $\pm$ 2
Cobalt-60	on-site	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
	off-site	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Iodine-131	on-site	50	10	< 10	10	10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	10 $\pm$ 35
	off-site	60	10	10	10	10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	11 $\pm$ 32
Manganese-54	on-site	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
	off-site	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Ruthenium-103	on-site	4	4	1	2	16	20	15	5	3	1	< 1	< 1	6 $\pm$ 12
	off-site	5	4	2	2	15	18	14	4	3	1	< 1	< 1	6 $\pm$ 11
Ruthenium-106- Rhodium-106	on-site	3	5	6	5	11	12	8	3	2	< 1	< 1	< 1	5 $\pm$ 7
	off-site	3	3	6	4	9	10	7	3	2	< 1	< 1	< 1	4 $\pm$ 6
Zirconium-95- Niobium-95	on-site	6	6	5	6	45	55	35	15	8	3	1	2	16 $\pm$ 35
	off-site	5	4	4	5	45	46	32	11	6	4	2	2	14 $\pm$ 31

Except for the shorter-lived fission products, the activities, including the naturally-occurring beryllium-7, increased in concentration from January to May, then generally decreased during the remainder of the year. This variation can be attributed to the usual "spring maximum" in the stratospheric fallout rate. The fact that the concentrations of the longer-lived fission products varied in approximately the same manner as beryllium-7 (except that they decreased at a greater rate during the latter part of the year through radioactive decay, since, unlike beryllium-7, they are not produced continuously) is evidence that the bulk of the fission products was also derived from the stratosphere.

The detection of short-lived fission products (iodine-131 and barium-140) at all sampling locations during the last quarter of 1971 and the first half of 1972, indicates the presence of fallout from a series of tests conducted during this period. The increase in 40-day ruthenium-103, 65-day zirconium-95, and 32-day cerium-141 in the spring implies a recent injection of fission products into the atmosphere at high altitudes. The variations with time in the concentrations of the short-lived fission products correlate with the dates of announced nuclear tests.

The average beta activity for the year,  $0.1 \times 10^{-12}$   $\mu\text{Ci/ml}$ , was about three times lower than in 1971. This was principally due to substantial decreases in the concentration of intermediate half-life fission products: cerium-144, ruthenium-106, and zirconium-95. The decreases are greater than can be accounted for on the basis of radioactive decay. Since the levels of beryllium-7 have been constant over the past years, these lower concentrations indicate a depletion of fission products in the stratospheric reservoir.

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.

Air was sampled continuously for radioiodine with activated charcoal in the 200 and 300 Areas because of the possibility of iodine releases in these areas. Very small amounts of iodine-131, up to  $0.15 \times 10^{-12}$   $\mu\text{Ci/ml}$  (0.15% of the uncontrolled CG), were detected in January and February and were probably

TABLE 7

Average Particulate Radioactivity in Air-Filter Samples, 1972  
(concentrations in  $10^{-12}$   $\mu\text{Ci/ml}$ )

Activity	Detection* Limit	Concentration* Guide (CG)	Concentration	Percent CG
Antimony-125	0.001	900	0.001	0.0001
Barium-140- Lanthanum-140	0.001	1000	0.002	0.0002
Beryllium-7	0.001	40000	0.099	(0.0002)
Cerium-141	0.001	5000	0.003	0.00006
Cerium-144	0.001	200	0.010	0.005
Cesium-137	0.001	500	0.002	0.0004
Cobalt-60	0.001	300	< 0.001	< 0.0003
Iodine-131	0.01	100	0.010	0.010
Manganese-54	0.001	1000	< 0.001	< 0.0001
Ruthenium-103	0.001	3000	0.006	0.0002
Ruthenium-106- Rhodium-106	0.001	200	0.005	0.003
Zirconium-95- Niobium-95	0.001	1000	0.016	0.002
Alpha	0.0002	10	0.0024	(0.024)
Beta	0.0005	10	0.097	0.97

\*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.

produced in nuclear tests conducted late in 1971, along with the other short-lived fission products found in the air filter samples. No iodine activity was detected at any other time during the year at either location.

Air sampling for plutonium was begun in March in the northeast area of the site (12N in Figure 1) and in October at 10H in the 300 Area close to the center of plutonium usage on the site. Off-site sampling for plutonium will be initiated early in 1973. Monthly samples were collected on a polystyrene filter medium at flow rates of 35 or 85 m<sup>3</sup>/hr, depending on the sampler. The total air volume filtered for each sample was about 30,000 and 65,000 m<sup>3</sup>, 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 has been ignited at 1000°C. The plutonium was separated from nitric acid solution on an anion-exchange column, electrodeposited, and its composition determined by alpha spectrometry. Alpha spectrometry cannot distinguish between plutonium-239 and -240, and although in the following discussion, and in 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 beginning with the June samples. To obtain the thorium recovery for these samples, thorium-234 was added prior to ignition. An aliquot of the sample solution was also analyzed for uranium by a standard type of 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 use of the specific activity of natural uranium,  $0.675 \times 10^{-6}$   $\mu\text{Ci}/\mu\text{g}$ . The results are given in Table 8.

The plutonium concentrations show the same general monthly variation discussed earlier for gamma-ray emitters (Table 6). The concentrations are also similar to those reported by other investigators<sup>(2,3,4)</sup> for samples collected at similar latitudes, but away from nuclear installations. For example, the AEC Health and Safety Laboratory obtained plutonium-239 results for New York City ranging from 30 in March to a maximum of 61 in June and decreasing to 46

TABLE 8

Plutonium, Thorium, and Uranium Concentrations in Air-Filter Samples on the ANL Site, 1972  
(concentrations in  $10^{-18}$   $\mu\text{Ci/ml}$ )

Month	Location	Plutonium-239	Plutonium-238	Thorium-232*	Thorium-228	Thorium-230	Uranium*
March	12N	31 ± 3	3.3 ± 1.0	-	-	-	-
April	12N	33 ± 3	4.2 ± 1.5	-	-	-	316 ± 41
May	12N	41 ± 3	-	-	-	-	146 ± 9
June	12N	45 ± 3	2.9 ± 0.9	20 ± 1	26 ± 1	33 ± 2	90 ± 8
July	12N	32 ± 3	1.5 ± 0.8	75 ± 2	75 ± 2	113 ± 3	77 ± 4
August	12N	28 ± 2	1.2 ± 0.8	23 ± 2	28 ± 3	36 ± 3	121 ± 28
September	12N	17 ± 2	1.5 ± 1.1	20 ± 2	16 ± 3	27 ± 3	63 ± 9
October	12N	7.6 ± 3.0	< 1	4.8 ± 1.8	4.1 ± 1.5	12 ± 3	40 ± 8
	10H	41 ± 3	2.5 ± 1.2	24 ± 3	20 ± 3	42 ± 2	79 ± 14
November	12N	6.6 ± 3.3	-	15 ± 2	20 ± 3	29 ± 3	89 ± 13
	10H	7.8 ± 2.0	-	20 ± 2	28 ± 2	36 ± 2	85 ± 18
December	12N	11.2 ± 1.4	2.4 ± 0.9	18 ± 2	23 ± 3	40 ± 3	125 ± 30
	10H	9.1 ± 0.8	1.2 ± 0.4	26 ± 2	21 ± 2	48 ± 2	83 ± 29
Monthly Average		27 ± 9	2.2 ± 0.7	27 ± 15	29 ± 15	45 ± 23	118 ± 52
Percent CG		0.003	0.0002	(0.003)	(0.015)	(0.015)	(0.003)
CG		$1 \times 10^6$	$1 \times 10^6$	$1 \times 10^6$	$2 \times 10^5$	$3 \times 10^5$	$4 \times 10^6$
Detection Limit		1	1	1	1	1	20

\*The concentrations in units of  $\mu\text{g/m}^3$  can be obtained by multiplying the value in  $\mu\text{Ci/ml}$  by  $1.48 \times 10^{12}$  for uranium and by  $9 \times 10^{12}$  for thorium-232. The amounts of the other two thorium isotopes in mass units are negligible in comparison to thorium-232. The average thorium and uranium concentrations are  $2.4 \times 10^{-4} \mu\text{g/ml}$  and  $1.7 \times 10^{-4} \mu\text{g/ml}$ , respectively.

in August, all in units of  $10^{-18}$   $\mu\text{Ci/ml}$ . The plutonium-238 concentrations were a factor of 5 to 10 lower. The monthly variations and the similarities between the samples collected by other laboratories indicate that the plutonium in the samples on the Argonne site is primarily from fallout. The October 10H concentration ( $41 \times 10^{-18}$   $\mu\text{Ci/ml}$ ), although within the yearly range, does not follow the monthly trend. The reason for this discrepancy is not clear, although at least two possibilities exist. The concentrations of the thorium isotopes are similar in most samples, about  $20 \times 10^{-18}$   $\mu\text{Ci/ml}$  for thorium-232 and thorium-228 and about  $30 \times 10^{-18}$   $\mu\text{Ci/ml}$  for thorium-230. The October 10H sample was in this range, while the October 12N sample is about a factor of four lower. This might imply that the 12N sample, for some reason, filtered relatively few particles and therefore the plutonium concentrations in the air were actually about four times greater than found at 12N. This implies, incidentally, that the plutonium fallout rate was unusually high during October. A second possibility is that the plutonium-239 fallout was actually closer to  $8 \times 10^{-18}$  than  $40 \times 10^{-18}$   $\mu\text{Ci/ml}$ , and that the additional plutonium, about  $30 \times 10^{-18}$   $\mu\text{Ci/ml}$  was due to plutonium from Argonne. An explosion involving plutonium occurred in Building 205 (location 12H), north of the 12N sampler on October 17, 1972, when the wind was from the north-northwest ( $330^\circ$ ), but was relatively calm (about 3-4 mph at the 19 foot level and about 6 mph at the 150 foot level). However, there is no unequivocal corroborating evidence that any plutonium escaped from Building 205 since, as will be seen, soil samples collected downwind from the building did not confirm a release. From whatever sources, the total plutonium-239 concentration,  $41 \times 10^{-18}$   $\mu\text{Ci/ml}$ , was very low compared to the exposure standards, 0.004% of the CG for uncontrolled areas and 0.0001% of the CG for controlled areas.

The thorium and uranium concentrations given in Table 8 are considered to be of natural origin. There is no indication that any of these elements in the air samples originated at Argonne. The percent of CG for these activities is included for purposes of completeness; the values are placed in parentheses since the concentrations are considered to be background levels.

Air sampling for argon-41, a beta emitter with a 1.8 hour 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 in Figure 1). Samples were collected by filling an evacuated "Marinelli type" container once a day from 2 to 5 times 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 yards 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. The average and maximum concentrations of  $1.2 \times 10^{-7}$  and  $1.2 \times 10^{-6}$   $\mu\text{Ci/ml}$  are, respectively, about 3 and 30 times the CG for uncontrolled areas and 0.060 and 0.6 times the CG for controlled areas. In this case, comparison with the CG for controlled areas is more appropriate since the sampling is conducted on the site and the argon-41 originated in the reactor. At the site security fence, the closest uncontrolled approach to the Laboratory, the concentrations were less than the detection limit,  $2 \times 10^{-8}$   $\mu\text{Ci/ml}$ . Based on penetrating radiation measurements (Section III.E.), the concentration at the fence averaged less than  $8 \times 10^{-10}$   $\mu\text{Ci/ml}$ , the concentration that would give a dose of 10 mrem/year.

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 absorption 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 yards east of the CP-5 reactor, and during the first half of the year, 200 yards from the reactor in various directions. 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 the off-site concentrations discussed below. 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 1972 ranged from  $0.2 \times 10^{-12}$  to  $8.6 \times 10^{-12}$   $\mu\text{Ci/ml}$  and averaged about  $2.4 \times 10^{-12}$   $\mu\text{Ci/ml}$ . Near CP-5 the average was  $1.4 \times 10^{-10}$   $\mu\text{Ci/ml}$ , and the difference was due to hydrogen-3 that left the reactor building. The average concentration near the reactor was almost three times lower than in 1971 because a leak that developed in the heat exchanger last year was repaired.

TABLE 9

Argon-41 Concentrations in Air, 300 Area, 1972

Month	No. of Samples	Conc. ( $10^{-9}$ $\mu$ Ci/ml)			Percent CG <sup>*</sup>		
		Av.	Min.	Max.	Av.	Min.	Max.
January	16	56	< 20	335	3	< 1	17
February	18	100	< 20	1160	5	< 1	58
March	12	180	< 20	1160	9	< 1	58
April	16	180	< 20	1000	9	< 1	50
May	14	125	< 20	500	6	< 1	25
June	16	82	< 20	420	4	< 1	21
July	16	140	< 20	950	7	< 1	48
August	8	81	< 20	220	4	< 1	11
September	16	120	< 20	1160	6	< 1	58
October	12	150	< 20	695	8	< 1	35
November	16	73	< 20	430	4	< 1	22
December	16	160	< 20	815	8	< 1	41
Annual Summary	176	121 $\pm$ 25	< 20	1160	6	< 1	58

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

TABLE 10

Hydrogen-3 Concentrations in Air, 300 Area, 1972

Month	No. of Samples	Conc. ( $10^{-12}$ $\mu\text{Ci/ml}$ )			Percent of CG*		
		Av.	Min.	Max.	Av.	Min.	Max.
January	8	210	1.5	880	0.10	0.00075	0.44
February	8	146	11.6	587	0.073	0.0058	0.29
March	16	117	0.94	781	0.058	0.00047	0.39
April	18	99	4.3	274	0.050	0.0022	0.14
May	18	145	3.4	1121	0.072	0.0017	0.56
June	14	194	6.1	718	0.097	0.0030	0.36
July	9	126	18.5	260	0.063	0.0092	0.13
August	9	166	86.0	367	0.083	0.043	0.18
September	9	78	24.3	178	0.039	0.012	0.089
October	9	117	8.7	497	0.058	0.0043	0.25
November	7	114	11.4	389	0.057	0.0057	0.19
December	9	151	30.7	357	0.076	0.015	0.18
Annual Summary	134	139 $\pm$ 22	0.94	1121	0.070	0.00047	0.56

\*This is the CG for uncontrolled areas.

As an extension of a study begun in 1971, air was also sampled for tritiated water vapor at two site security fence locations, 650 yards directly west (10E) and 530 yards directly south (8H) of the reactor. These two locations are the closest uncontrolled approaches to CP-5. Samples were also collected once a month at an on-site location in the East Area 2,000 yards northeast of the CP-5 reactor and off-site at a location approximately 6 miles northwest of the Laboratory. The results are given in Table 11. The fence line results show good correlation with wind direction and indicate that dilution to background levels occurs within 500-600 yards of the reactor in directions other than that from which the wind is blowing. The average and maximum concentrations were equivalent to 0.008 and 0.05% of the CG, respectively. The average fence line concentration corresponds to a dose of 0.038 mrem/year, about 85% from Argonne operations and the remainder from weapons testing.

The higher concentrations in the East Area compared to off-site concentrations may also be attributed to CP-5. Measurements from previous years 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 can be observed.

The off-site concentrations, which may be considered as normal for this area, decreased by a factor of two over the 1971 values. Most of this tritium was produced in nuclear test detonations, and a decrease is expected, since few atmospheric tests were conducted in 1971 and 1972. Much of the test-produced tritium has been transported from the atmosphere to the hydrosphere.

#### 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 results calculated in terms of activity assuming the isotopic composition 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

TABLE 11

Hydrogen-3 Concentrations On and Near the ANL Site, 1972

Month	Location	No. of Samples	Conc. ( $10^{-12}$ $\mu$ Ci/ml)			Percent CG*		
			Av.	Min.	Max.	Av.	Min.	Max.
January	East Area	1	-	-	2.6	-	-	0.0013
	Off-Site	1	-	-	0.68	-	-	0.00034
February	East Area	1	-	-	11.0	-	-	0.0055
	Off-Site	1	-	-	0.87	-	-	0.00043
March	East Area	1	-	-	0.43	-	-	0.00022
	Off-Site	1	-	-	0.18	-	-	0.00009
April	East Area	1	-	-	2.2	-	-	0.0011
	Off-Site	1	-	-	1.6	-	-	0.00080
May	East Area	1	-	-	2.7	-	-	0.0013
	Off-Site	1	-	-	2.9	-	-	0.0014
June	South Fence (8H)	2	35.2	13.7	56.7	0.018	0.0068	0.028
	East Area	1	-	-	6.1	-	-	0.0030
	Off-Site	1	-	-	8.6	-	-	0.0043
July	South Fence (8H)	9	8.4	2.5	13.2	0.0042	0.0012	0.0066
	East Area	1	-	-	14.7	-	-	0.0074
	Off-Site	1	-	-	6.5	-	-	0.0032
August	West Fence (10E)	9	5.8	2.8	8.1	0.0029	0.0014	0.0040
	East Area	1	-	-	3.9	-	-	0.0020
	Off-Site	1	-	-	2.5	-	-	0.0013
September	South Fence (8H)	9	13.5	0.77	55.6	0.0068	0.00038	0.028
	East Area	1	-	-	5.4	-	-	0.0027
	Off-Site	1	-	-	2.5	-	-	0.0013
October	South Fence (8H)	9	26.7	0.33	96.0	0.013	0.00016	0.048
	East Area	1	-	-	3.5	-	-	0.0017
	Off-Site	1	-	-	0.89	-	-	0.00044
November	South Fence (8H)	7	16.0	0.29	59.0	0.0080	0.00014	0.029
	East Area	1	-	-	2.3	-	-	0.0011
	Off-Site	1	-	-	0.84	-	-	0.00041
December	West Fence (10E)	9	1.8	0.82	3.7	0.00088	0.00041	0.0019
	East Area	1	-	-	2.4	-	-	0.0012
	Off-Site	1	-	-	0.38	-	-	0.00019
Annual Summary	Fence Line	54	15.3 $\pm$ 9.0.	0.29	96.0	0.0076	0.00014	0.048
	East Area	12	4.8 $\pm$ 2.4	0.43	14.7	0.0024	0.00022	0.0074
	Off-Site	12	2.4 $\pm$ 1.5	0.18	8.6	0.0012	0.00009	0.0043

\* This is the CG for uncontrolled areas.

hydrogen-3 and plutonium-neptunium. Most hydrogen-3 analyses were performed by counting 10 ml in a gel system. A few samples were analyzed by electrolytic enrichment of 250 ml aliquots prior to counting. Plutonium and neptunium analyses were performed on 10-liter samples by a plutonium chemical separation method,<sup>(5)</sup> modified to include neptunium, followed by alpha spectrometry. Plutonium-236 was used to determine the plutonium yield.

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 continuously, and the individual samples collected five times weekly. 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 collected twice a month and selected samples were analyzed for the same radionuclides as the below-outfall samples.

Annual summaries of the results obtained for Sawmill Creek are given in Table 12. Comparison of the results, and 95% confidence limits of the averages, for the two sampling locations show that the only nuclides whose presence in the Creek water can be attributed to Argonne operations were hydrogen-3, neptunium-237, plutonium-239, and possibly strontium-90. The fraction of individual samples containing activity attributable to Argonne was 90% for hydrogen-3 and 50% for plutonium and neptunium. The concentrations of all four nuclides were quite 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 of this nuclide to the Creek amounted to only 0.02% of the CG and the highest concentration in any single sample was equivalent to 0.06% of the CG. The average tritiated water concentration in Sawmill Creek during the past few years shows Argonne's hydrogen-3 contribution to have decreased by a factor of three over 1970 and by a factor of 20 over 1971, when a leak occurred in the CP-5 heat-exchanger. This is

TABLE 12

## Radioactivity in Sawmill Creek Water, 1972

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	24	$2.0 \pm 0.18$	1.1	2.8	(0.067)	(0.037)	(0.093)
	7M	253	$1.6 \pm 0.16$	0.64	3.8	(0.053)	(0.021)	(0.13)
Beta (nonvolatile)	15K	24	$17 \pm 2.3$	8.4	33	(0.57)	(0.28)	(1.1)
	7M	253	$16 \pm 1.5$	6.7	44	(0.53)	(0.22)	(1.5)
Hydrogen-3	15K	23	$270 \pm 45$	< 200	470	0.0090	< 0.0067	0.016
	7M	253	$565 \pm 70$	205	1870	0.019	0.0068	0.062
Strontium-89	15K	10	-	-	< 2	-	-	< 0.07
	7M	200	-	-	< 2	-	-	< 0.07
Strontium-90	15K	10	$0.76 \pm 0.25$	< 0.5	1.4	0.25	< 0.17	0.47
	7M	200	$1.1 \pm 0.13$	< 0.5	2.0	0.37	< 0.17	0.67
Iodine-131	15K	7	< 3.7	< 3	7.6	< 1.2	< 1	2.5
	7M	169	-	-	< 3	-	-	< 1
Barium-140	15K	9	-	-	< 2	-	-	< 0.007
	7M	169	-	-	< 2	-	-	< 0.007
Uranium (natural)**	15K	13	$1.6 \pm 0.21$	0.96	2.4	(0.0040)	(0.0024)	(0.0060)
	7M	253	$1.6 \pm 0.14$	0.95	2.7	(0.0040)	(0.0024)	(0.0068)
Neptunium-237	15K	11	-	-	< 0.002	-	-	< 0.00007
	7M	253	$0.011 \pm 0.0064$	< 0.002	0.15	0.00037	< 0.00007	0.005
Plutonium-239	15K	11	-	-	< 0.0005	-	-	< 0.00001
	7M	253	$0.0015 \pm 0.0013$	< 0.0005	0.035	0.000030	< 0.00001	0.00070

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

\*\*Uranium concentrations in units of  $\mu\text{g/l}$  can be obtained by multiplying the concentration given by  $1.48 \times 10^9$ . The average concentration in the Creek then becomes  $2.4 \mu\text{g/l}$ .

evidence of the Laboratory's continuing effort to reduce emissions to the lowest possible levels. The hydrogen-3 in the Creek above the site ( $270 \times 10^{-9}$   $\mu\text{Ci/ml}$ ) was similar in concentration to levels found away from the Laboratory site and is characteristic of the present normal levels of hydrogen-3 in surface waters. During 1972, the hydrogen-3 content of several Lake Michigan water samples was 275 to  $280 \times 10^{-9}$   $\mu\text{Ci/ml}$ ; other lakes and streams ranged from 220 to  $350 \times 10^{-9}$   $\mu\text{Ci/ml}$ . Over the past two years, the hydrogen-3 concentration in surface water samples has decreased by a factor of two as the hydrogen-3 from nuclear tests mixed with and was diluted by the hydrosphere.

The average total alpha and beta activities were higher above the site, indicating that at times Argonne waste-water contained less nonvolatile activity 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 nuclides, and increases the radioactivity in natural Creek water. The total alpha and beta activities were not appreciably different than in 1971.

In addition to the natural beta activity and that added by waste-water below the outfall, beta activity from nuclear detonations was detected at both sampling locations. The normal nonvolatile beta activity is approximately  $10 \times 10^{-9}$   $\mu\text{Ci/ml}$ . It is estimated that fallout activity added about  $5 \times 10^{-9}$   $\mu\text{Ci/ml}$  to the nonvolatile beta activity at both locations and that the Argonne contribution below the outfall averaged about  $1 \times 10^{-9}$   $\mu\text{Ci/ml}$ , equivalent to 0.03% of the CG. The Argonne contribution remained the same as 1971 levels, while the fallout contribution decreased by about a factor of two.

The same nuclides added to the Creek in Argonne waste-water are also produced in nuclear detonations and are constituents of fallout (although some are not present in amounts above the detection limits). 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 12 were very low.

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 13. The concentrations were in their normal ranges. The natural nonvolatile beta activity in the River is  $5-10 \times 10^{-9}$   $\mu\text{Ci/ml}$ , and the excess,  $4-5 \times 10^{-9}$   $\mu\text{Ci/ml}$ , was due to fallout.

Evidence of fission product fallout is apparent in some of the water samples. Positive iodine-131 concentrations in the Des Plaines River and at 15K in Sawmill Creek on the same day in early May correlate with air filter results that indicated a recent atmospheric nuclear test. Between June and October, plutonium-239 was detected at both locations in the River. Since the River consists mainly of surface runoff water, the presence of the 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 due to the fact that a substantial portion of this water is from deep municipal wells.

The activities in samples of Illinois River water (Table 14) were similar to those found in other bodies of water in the area and to the activities found previously at these same locations. The average alpha concentration of  $1.2 \times 10^{-9}$   $\mu\text{Ci/ml}$  and average beta concentration of  $8.5 \times 10^{-9}$   $\mu\text{Ci/ml}$  of 31 off-site surface water samples collected during the year is evidence that the Illinois River activity levels are normal. No radioactivity originating at Argonne could be detected in the Des Plaines or Illinois Rivers.

### C. Soil, Grass, and Benthic Materials

Plutonium deposition in 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. Soil samples consisted of a core  $173 \text{ cm}^2$  in area by 30 cm deep. The grass samples were obtained by collecting all the grass from a  $1 \text{ m}^2$  area. A grab sampling 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 40 g of the oven-dried plant.

TABLE 13

## Radioactivity in Des Plaines River Water, 1972

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	11	$1.8 \pm 0.39$	0.82	3.4	(0.060)	(0.027)	(0.11)
	B	24	$1.8 \pm 0.22$	0.85	3.1	(0.060)	(0.028)	(0.10)
Beta (nonvolatile)	A	11	$13 \pm 6.2$	5.1	43	(0.43)	(0.17)	(1.4)
	B	24	$11 \pm 2.0$	4.9	24	(0.37)	(0.16)	(0.80)
Hydrogen-3	A	11	$245 \pm 45$	< 200	352	0.0082	< 0.0067	0.012
	B	24	$230 \pm 35$	< 200	378	0.0077	< 0.0067	0.013
Strontium-89	A	9	-	-	< 2	-	-	< 0.07
	B	17	-	-	< 2	-	-	< 0.07
Strontium-90	A	9	$1.1 \pm 0.29$	< 0.5	1.6	0.37	< 0.17	0.53
	B	17	$1.2 \pm 0.22$	0.50	2.0	0.40	0.17	0.67
Iodine-131	A	7	-	-	< 3	-	-	< 1
	B	14	< 3	< 3	3.0	< 1	< 1	1.0
Barium-140	A	8	-	-	< 2	-	-	< 0.007
	B	15	-	-	< 2	-	-	< 0.007
Uranium (natural)**	A	10	$1.6 \pm 0.21$	0.95	2.1	(0.0040)	(0.0024)	(0.0052)
	B	23	$1.6 \pm 0.27$	0.40	3.1	(0.0040)	(0.0010)	(0.0078)
Neptunium-237	A	11	-	-	< 0.002	-	-	< 0.00007
	B	10	-	-	< 0.002	-	-	< 0.00007
Plutonium-239	A	11	$0.00072 \pm 0.00027$	< 0.0005	0.0014	0.000014	< 0.00001	0.000028
	B	10	$0.00055 \pm 0.00017$	< 0.0005	0.00092	0.000011	< 0.00001	0.000018

\*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  $1.48 \times 10^9$ . The average concentration is  $2.4 \mu\text{g/l}$ .

TABLE 14

Radioactivity in Illinois River Water, 1972  
(concentrations in  $10^{-9}$   $\mu$ Ci/ml)

Location	Date Collected	Alpha (nonvolatile)	Beta (nonvolatile)	Hydrogen-3	Uranium*	Neptunium-237	Plutonium-239
McKinley Woods State Park	June 27	1.2	9.7	390	0.61	< 0.001	< 0.0005
Below Dresden Power Station	June 27	1.2	7.4	285	1.1	< 0.001	< 0.0005
Morris	June 27	1.0	8.2	315	0.68	-	-
McKinley Woods State Park	October 5	1.1	8.4	290	1.5	< 0.0005	0.00053**
Below Dresden Power Station	October 5	0.57	5.3	290	1.1	< 0.0005	0.00022**
Morris	October 5	0.32	6.6	280	1.0	-	-
Starved Rock State Park	October 5	0.70	6.6	245	1.3	-	-

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

\*\* These analyses were made on a 45 l sample; all other plutonium analyses were made on 10 l samples.

The on-site and off-site soil results are given in Table 15 and Table 16, respectively. The limits given for the individual results are the statistical counting errors at the 95% confidence level, while the limits of the averages are the 95% confidence levels calculated from the standard deviation of the average. Comparison of the on-site and off-site samples shows that the same average and 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 (6-8) are in the same range as those reported here, namely about  $2 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ .

Several on-site locations are slightly higher than the highest off-site sample. These samples were collected near buildings in which plutonium had been used, so the possibility of plutonium in their vicinity exists. The levels of activity in these samples are not sufficiently above the fallout range to allow an unequivocal conclusion. Surveillance in these areas is continuing.

The samples collected downwind from Building 205 after the glove box explosion mentioned in Section III.A. averaged  $1.83 \times 10^{-3} \mu\text{Ci}/\text{m}^2$  and ranged from  $1.1$  to  $2.9 \times 10^{-3} \mu\text{Ci}/\text{m}^2$ . These values are also not sufficiently different from the off-site results to conclude with good probability that the samples contained plutonium from the explosion.

Composite monthly precipitation samples were analyzed for plutonium-239 by the same procedure as used on water samples. Concentrations, expressed in terms of ground deposition, ranged from  $7.5 \times 10^{-7}$  to  $1.2 \times 10^{-7} \mu\text{Ci}/\text{m}^2$  and averaged  $4.1 \times 10^{-7} \mu\text{Ci}/\text{m}^2$ . The total 1972 deposition by precipitation was only 0.3% of the amount previously deposited, and implies that in the absence of further testing the soil content will not increase greatly.

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. The thorium-228 concentrations averaged  $(0.40 \pm 0.04) \times 10^{-6} \mu\text{Ci}/\text{g}$  on site and  $(0.43 \pm 0.10) \times 10^{-6} \mu\text{Ci}/\text{g}$  off-site. The thorium-230 averaged  $(0.47 \pm 0.04) \times 10^{-6} \mu\text{Ci}/\text{g}$  on-site and  $(0.45 \pm 0.10) \times 10^{-6} \mu\text{Ci}/\text{g}$  off-site. The thorium-232 averaged  $(0.34 \pm 0.03) \times 10^{-6} \mu\text{Ci}/\text{g}$  on-site and  $(0.37 \pm 0.08) \times 10^{-6} \mu\text{Ci}/\text{g}$  off-site. No significant differences between on- and off-site concentrations were found. These are normal levels of the naturally-occurring thorium

TABLE 15

Plutonium Concentrations in On-Site Soil, 1972

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 7	12L	0.34 ± 0.24	0.13 ± 0.086	4.5 ± 0.5	1.74 ± 0.20	0.075
June 7	12L	0.61 ± 0.48	0.26 ± 0.20	6.7 ± 1.1	2.84 ± 0.47	0.092
June 7	12L	0.79 ± 0.30	0.24 ± 0.088	5.6 ± 0.6	1.70 ± 0.18	0.14
June 7	12L	0.51 ± 0.24	0.23 ± 0.10	3.9 ± 0.5	1.74 ± 0.21	0.13
June 7	12F	0.56 ± 0.25	0.25 ± 0.11	3.3 ± 0.5	1.45 ± 0.20	0.17
June 7	12-13F	0.54 ± 0.26	0.23 ± 0.10	3.7 ± 0.5	1.50 ± 0.20	0.15
June 7	9H	0.35 ± 0.24	0.13 ± 0.083	4.8 ± 0.5	1.77 ± 0.20	0.073
June 8	9I	0.42 ± 0.24	0.16 ± 0.085	8.0 ± 0.7	3.09 ± 0.27	0.052
June 8	11I	0.41 ± 0.24	0.16 ± 0.085	4.7 ± 0.5	1.77 ± 0.20	0.090
June 8	11I	0.71 ± 0.27	0.30 ± 0.12	3.8 ± 0.5	1.63 ± 0.22	0.18
June 13	11I	0.79 ± 0.26	0.26 ± 0.083	7.5 ± 0.6	2.47 ± 0.20	0.10
June 13	11I	0.71 ± 0.29	0.29 ± 0.12	5.5 ± 0.6	2.25 ± 0.25	0.13
October 27	11H	1.3 ± 0.29	0.34 ± 0.077	6.0 ± 0.5	1.66 ± 0.16	0.20
October 27	11H	1.3 ± 0.30	0.35 ± 0.083	9.4 ± 0.7	2.50 ± 0.19	0.14
October 27	11H	0.43 ± 0.23	0.15 ± 0.073	4.8 ± 0.5	1.66 ± 0.17	0.090
October 27	11H	0.54 ± 0.24	0.11 ± 0.046	6.1 ± 0.6	1.29 ± 0.12	0.085
October 27	11H	0.52 ± 0.24	0.17 ± 0.071	4.8 ± 0.6	1.53 ± 0.19	0.11
October 27	11H	0.97 ± 0.29	0.16 ± 0.048	8.7 ± 0.7	1.44 ± 0.11	0.11
October 30	11H	0.75 ± 0.29	0.21 ± 0.077	4.5 ± 0.5	1.27 ± 0.15	0.17
October 30	11H	0.63 ± 0.28	0.25 ± 0.10	7.3 ± 0.7	2.87 ± 0.26	0.087
October 30	11H	1.0 ± 0.30	0.28 ± 0.083	8.1 ± 0.7	2.24 ± 0.19	0.12
October 30	12H	0.55 ± 0.23	0.19 ± 0.071	3.2 ± 0.4	1.11 ± 0.13	0.17
October 30	12H	0.78 ± 0.29	0.26 ± 0.095	4.2 ± 0.5	1.40 ± 0.17	0.19
October 30	12G	0.59 ± 0.23	0.20 ± 0.071	2.7 ± 0.4	0.90 ± 0.13	0.22
Average			0.22 ± 0.027		1.83 ± 0.24	0.11

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

TABLE 16

## Plutonium Concentrations in Off-Site Soil, 1972

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 27	McKinley Woods State Park	$0.31 \pm 0.24$	$0.085 \pm 0.066$	$3.9 \pm 0.5$	$1.10 \pm 0.14$	0.077
June 27	Pioneer Park, Naperville	$0.68 \pm 0.25$	$0.26 \pm 0.10$	$4.0 \pm 0.6$	$1.50 \pm 0.21$	0.17
June 27	Saganashkee Slough	$0.59 \pm 0.26$	$0.20 \pm 0.09$	$6.3 \pm 0.6$	$2.09 \pm 0.21$	0.096
September 27	McGinnis Slough	$0.78 \pm 0.46$	$0.31 \pm 0.19$	$4.8 \pm 0.8$	$1.96 \pm 0.31$	0.16
September 27	McCormick Woods	$0.43 \pm 0.21$	$0.15 \pm 0.07$	$4.6 \pm 0.5$	$1.55 \pm 0.17$	0.097
October 3	Lake Delavan, Wisconsin	$0.48 \pm 0.23$	$0.14 \pm 0.06$	$5.8 \pm 0.5$	$1.58 \pm 0.16$	0.089
	Average		$0.19 \pm 0.07$		$1.63 \pm 0.29$	0.11

activities. Results of uranium analyses averaged  $(1.5 \pm 0.2) \times 10^{-6}$   $\mu\text{Ci/g}$  on-site and  $(1.3 \pm 0.3) \times 10^{-6}$   $\mu\text{Ci/g}$  off-site, and are quite similar to levels of uranium in soil found in previous years in this area.<sup>(9,10)</sup> No evidence of uranium from Argonne was found. In terms of mass the thorium concentrations were 3.1  $\mu\text{g/g}$  on-site and 3.3  $\mu\text{g/g}$  off-site and the uranium concentrations were 2.2  $\mu\text{g/g}$  on-site and 1.9  $\mu\text{g/g}$  off-site. Gamma-ray spectrometric analysis of these 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 17. The concentrations in terms of area were approximately the same on and off the site. Two on-site samples (June 7, 12L and 9I) are 1.5 to 2 times higher than the off-site samples, and these were collected near soil that also contained above average plutonium concentrations. However, it is not certain that any of the plutonium in these samples can be attributed to Argonne since grass collected in 1971 contained still higher concentrations both on and off the site<sup>(11)</sup> than were found this year. The differences observed are probably due to normal variations in fallout concentrations. In terms of deposition, the plutonium-239 concentration was a factor of two thousand to five thousand less than in soil from the same location. Uranium concentrations were similar to those reported last year.<sup>(12)</sup>

A study was made of the plutonium concentration in the Sawmill Creek bed below the Laboratory waste-water outfall. The results are collected in Table 18 along with the results from several control locations. Because the nature of the Creek bottom prevented collection from a known area, results are expressed in terms of concentration rather than deposition. The lower concentrations ( $< 10 \times 10^{-9}$   $\mu\text{Ci/g}$ ) were found in samples collected in areas where the flow rate is rapid and the bed was sandy and had little absorptive capacity. As the Creek slows 100 yards below the outfall the sedimentation increases, the bed becomes more silty and retentive, and higher plutonium concentrations occur. The highest Creek results are similar to those of the slower moving Des Plaines River. Thorium and uranium concentrations in the Creek bed followed the same pattern. Little, if any, plutonium released in the Laboratory waste-water is retained by the Creek bottom.

TABLE 17

Plutonium-239 Concentrations in Grass, 1972

Date Collected	Location*	Concentration	
		$10^{-9}$ $\mu\text{Ci/g}$	$10^{-6}$ $\mu\text{Ci/m}^2$
<u>On-Site</u>			
June 7	12L	$2.3 \pm 0.7$	$0.46 \pm 0.14$
June 7	12-13L	$1.5 \pm 0.4$	$0.22 \pm 0.07$
June 8	9I	$4.2 \pm 0.8$	$0.67 \pm 0.12$
June 8	12I	$1.6 \pm 0.5$	$0.35 \pm 0.12$
October 17	12H	$8.5 \pm 4.6$	$0.33 \pm 0.19$
October 17	12H	$4.5 \pm 3.7$	$0.21 \pm 0.18$
October 18	12H	$4.4 \pm 1.7$	$0.22 \pm 0.08$
October 30	12H	$2.3 \pm 1.6$	$0.32 \pm 0.21$
<u>Off-Site</u>			
June 27	McKinley Woods State Park	$0.87 \pm 0.3$	$0.17 \pm 0.07$
October 3	Lake Delavan, Wisconsin	$2.8 \pm 1.9$	$0.29 \pm 0.19$

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

TABLE 18

Plutonium Concentrations in Benthic Material, 1972

Location*	Plutonium-238 Conc. ( $10^{-9}$ $\mu$ Ci/g)	Plutonium-239 Conc. ( $10^{-9}$ $\mu$ Ci/g)	$^{238}\text{Pu}/^{239}\text{Pu}$
Des Plaines River, Willow Springs	1.7 $\pm$ 0.3	21.8 $\pm$ 1.1	0.078
Des Plaines River, Lemont	2.1 $\pm$ 0.4	33.3 $\pm$ 1.3	0.063
15K	1.1 $\pm$ 0.3	7.2 $\pm$ 0.6	0.15
50 Yds below 7M	1.1 $\pm$ 0.3	9.7 $\pm$ 0.8	0.11
100 Yds below 7M	1.3 $\pm$ 0.3	18.4 $\pm$ 1.0	0.071
150 Yds below 7M	2.7 $\pm$ 0.4	25.9 $\pm$ 1.3	0.10
200 Yds below 7M	2.2 $\pm$ 0.4	27.7 $\pm$ 1.3	0.079
250 Yds below 7M	2.1 $\pm$ 0.4	32.0 $\pm$ 1.4	0.066
300 Yds below 7M	2.0 $\pm$ 0.4	25.1 $\pm$ 1.2	0.080
400 Yds below 7M	2.5 $\pm$ 0.4	24.0 $\pm$ 1.2	0.10
Mouth of Sawmill at Des Plaines	3.0 $\pm$ 0.4	34.2 $\pm$ 1.4	0.088

\*Location 15K is above the ANL site. Location 7M is below the waste-water outfall. Both locations are in Sawmill Creek.

#### D. Radioactivity in Milk

Raw milk was collected monthly from a local dairy farm and analyzed for several fission products. Barium-140, strontium-89, and iodine-131 were not present in concentrations greater than the minimum detectable amounts of  $20 \times 10^{-9}$   $\mu\text{Ci/ml}$  for iodine-131 and  $2 \times 10^{-9}$   $\mu\text{Ci/ml}$  for the other two nuclides. The cesium-137 and strontium-90 concentrations are given in Table 19. These two nuclides are long-lived fission products from past nuclear tests and their presence in milk is not related to Argonne operations. The average strontium-90 content decreased by 10% from last year, while the cesium-137 concentrations decreased about 30%.

The concentrations given in Table 19 may be compared to the CGs for drinking water. These values are  $3 \times 10^{-7}$   $\mu\text{Ci/ml}$  for strontium-90 and  $2 \times 10^{-5}$   $\mu\text{Ci/ml}$  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 0.75% of the strontium-90 and  $< 0.025\%$  of the cesium-137 Concentration Guides. Using the standards specified by the Federal Radiation Council, one liter of milk is equivalent to  $< 0.3$  and 2.5% of the Daily Intake Guides for cesium-137 and strontium-90, respectively.

#### E. Penetrating Radiation

Measurements of gamma-ray dose were made with thermoluminescent dosimeters (extruded calcium fluoride chips, dysprosium-activated) calibrated with an NBS standard radium-226 source. Each measurement was the average of 3 to 6 chips exposed in the same package. Exposure times ranged from 44 to 74 days (except for one 133-day exposure). Individual results were calculated in terms of annual dose rate, and these were weighted according to their exposure times in calculating the average for a location.

The measurements were made throughout the year at a number of locations at the site security fence to determine the dose that might be due to Argonne operations at the closest uncontrolled approach to the Laboratory. The actual site boundary (the dashed line in Figure 1) varies from 150 to 1500 yards from the security fence, and over most of the perimeter is at least 500-1000 yards from the fence. The closest public access to potential radiation sources is at coordinate 8H. At this location, the public is allowed to visit a pre-existing cemetery (St. Patrick's Cemetery). Measurements were also made at several

TABLE 19

Cesium-137 and Strontium-90 Concentrations in Milk, 1972  
(concentrations in  $10^{-9}$   $\mu\text{Ci/ml}$ )

Date Collected	Cesium-137	Strontium-90
January 5	-	4.0
February 2	< 10	6.0
March 1	10	2.2
April 5	13	8.2
May 3	14	6.5
June 7	< 10	6.9
July 5	< 10	5.3
August 2	< 10	4.8
September 6	< 10	3.8
October 4	< 10	3.9
November 1	< 10	4.0
December 6	< 10	4.7
Average	< 11	5.0

locations on the site within the fence, and at several off-site locations for comparison purposes. The results are given in Tables 20-22 for off-site, security fence, and on-site locations, respectively.

The off-site measurements were quite uniform both by location and by period. Ninety-five percent of the results were in the range from 94 to 114 mrem/year. Only one anomalous result was obtained, 121 mrem/year for the second measurement at Lombard; all other results fell within 2 standard deviations (95% confidence limits) of the average for a single result,  $105 \pm 11$  mrem.

If it is assumed that the off-site readings accurately represent the average natural radiation background of the area, a result between 94 and 116 mrem/year may be considered normal with a 95% probability. Only 2.5% of the measurements of a natural background radiation field should be higher than 116 mrem/year and 2.5% lower than 94 mrem/year. The off-site average, 105 mrem/year, is almost identical with that obtained in 1971, 104 mrem/year.

Examination of the site security fence measurements (Table 21 and Figure 5) show that only three locations gave average readings outside the normal range and two locations gave occasional readings outside the normal range. The latter finding can simply be part of the normal spread indicated above. The dose rate at location 13D was well below the normal range, as it was in 1971, probably because of the large amount of subsurface gravel in the area containing below average concentrations of natural activities. Two locations were above normal, 8G and 7I, both on the south security fence. The abnormal dose rate at 7I, approximately 300 mrem/year, was due to radioactive materials in a Radioactive Waste Storage Area several hundred yards north of the fence. Radioactive waste is stored and packaged in this area for shipment to waste burial sites elsewhere. Readings in the center of the storage area (Table 22) were as high as 2.3 rem/year. The area between the site security fence and the site boundary in this vicinity is rugged and 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. Based on measurements made north and west of the storage area, the dose rate at the south boundary line, about 650 yards from the fence, would be indistinguishable from background levels. The Waste Storage Area may be considered as a stationary gamma-ray source in considering the variation of dose with distance. The strength of the source varies, however, as material is moved in and out of the area.

TABLE 20

Environmental Penetrating Radiation at Off-Site Locations, 1972

Location	Period of Measurement	Days	Dose Rate (mrem/yr)
Downers Grove	2/17 - 4/29 (2)	72	106
	5/12 - 6/29 (2)	48	96
	6/29 - 9/11	74	103
	7/6 - 9/11 (6)	67	111 ± 9
	9/11 - 11/21	74	112
	11/21 - 1/5	45	105
			Average 106 ± 11
Woodridge	2/17 - 5/1	74	109
	9/8 - 11/21	74	105
	11/21 - 1/4	44	103
			Average 106 ± 3
Lemont	2/17 - 5/1	74	103
	6/29 - 9/7	70	105
	9/11 - 11/21	71	101
			Average 103 ± 8
Lombard	6/29 - 9/8	71	107
	11/21 - 1/5	45	121
			Average 112
Naperville	6/29 - 9/8	71	94
Hinsdale	7/5 - 9/8	65	114
Plainfield	11/21 - 1/4	44	103
Oak Lawn	9/8 - 11/21	74	98
	11/21 - 1/4	44	102
			Average 99
			Average 105 ± 5

TABLE 21

Environmental Penetrating Radiation at ANL Site Security Fence, 1972

Location	Period of Measurement	Days	Dose Rate (mrem/yr)
13D	2/17 - 4/29	72	76
13D	7/5 - 9/11	74	85
			Average 81
14I	2/17 - 4/29	72	109
14I	9/8 - 11/21	74	111
14I	11/21 - 1/4	44	122
			Average 113 ± 14
18J	6/29 - 9/8	71	100
14L	2/17 - 5/1	74	104
14L	7/5 - 9/8	71	101
14L	9/8 - 11/21	74	100
			Average 102 ± 4
12-0	2/17 - 4/29	72	108
12-0	5/12 - 6/29	48	98
12-0	6/29 - 9/8	71	108
			Average 105 ± 11
12N	6/29 - 9/8	71	98
12N	9/8 - 11/21	74	107
			Average 103
10M	11/21 - 1/4	44	111
10L	2/17 - 4/29	72	111
7I	6/29 - 9/11	74	350
7I	9/11 - 11/21	71	509
			Average 428
8G	2/17 - 4/29	72	131
8G	5/12 - 6/29	48	111
8G	6/29 - 9/11	74	123
8G	9/11 - 11/21	71	134
8G	11/21 - 1/4	44	139
			Average 128 ± 21
8F	2/17 - 4/29	72	121
8F	5/12 - 6/29	48	108
8F	6/29 - 9/8	71	110
8F	9/8 - 11/21	74	116
8F	11/21 - 1/4	44	111
			Average 114 ± 10
9EF	2/17 - 4/29	72	114
9EF	5/12 - 6/29	48	112
9EF	6/29 - 9/8	71	112
			Average 113 ± 2
10E	2/17 - 6/29	133	98
10E	9/8 - 11/21	74	111
			Average 103

TABLE 22

Environmental Penetrating Radiation on the ANL Site, 1972

Location	Period of Measurement	Days	Dose Rate (mrem/yr)
9H - 75 m East of CP-5	2/17 - 4/29	72	1900
	6/29 - 9/8	71	1200
	9/8 - 11/21	74	435
	11/21 - 1/4	44	1640
			Average 1250
9G - 75 m West of CP-5	6/29 - 9/8	71	346
	9/8 - 11/21	74	452
8H - Heliport	2/17 - 4/29	72	149
	7/5 - 9/11	68	133
7I - No. Fence of Waste Storage Area	2/17 - 4/29	72	1270
	5/12 - 6/29	48	1170
	6/29 - 9/8	71	864
	11/21 - 1/4	44	2360
			Average 1330
7I - Waste Storage Area	5/12 - 6/29	48	860
	11/21 - 1/4	44	2360
7I - West Fence of Waste Storage Area	9/11 - 1/21	71	496
			Average 1105
10I - Bluff Road, NW of ZGS	5/12 - 6/29	48	106
	6/29 - 9/8	71	108
9H - No. of Bldg. 315 - 225 m So. of CP-5	6/29 - 9/8	71	248
	11/21 - 1/4	44	290
7/8J - NW of Storage Area	5/12 - 6/29	48	125
	6/29 - 9/11	71	135
11H - So. of 205, 500 m No. of CP-5	5/12 - 6/29	48	102
	6/29 - 9/8	71	115
9J - ZGS Area - South	7/5 - 9/11	68	109
	9/8 - 11/21	74	113
	9/11 - 11/21	71	120
			Average 114
9K - ZGS Area - East of Meson Bldg.	9/8 - 11/21	74	131
	11/21 - 1/4	44	188
13I - Bldg. 202 Parking Lot	2/17 - 4/29	72	78
14I - Bldg. 202 - South	6/29 - 9/8	71	118
13I - Bldg. 202 - North	6/29 - 9/8	71	89
			Average 95
13H - Bldg. 203 - North	9/8 - 11/21	74	122
	11/21 - 1/4	44	119
13G - Bldg. 206 - West	9/8 - 11/21	74	97

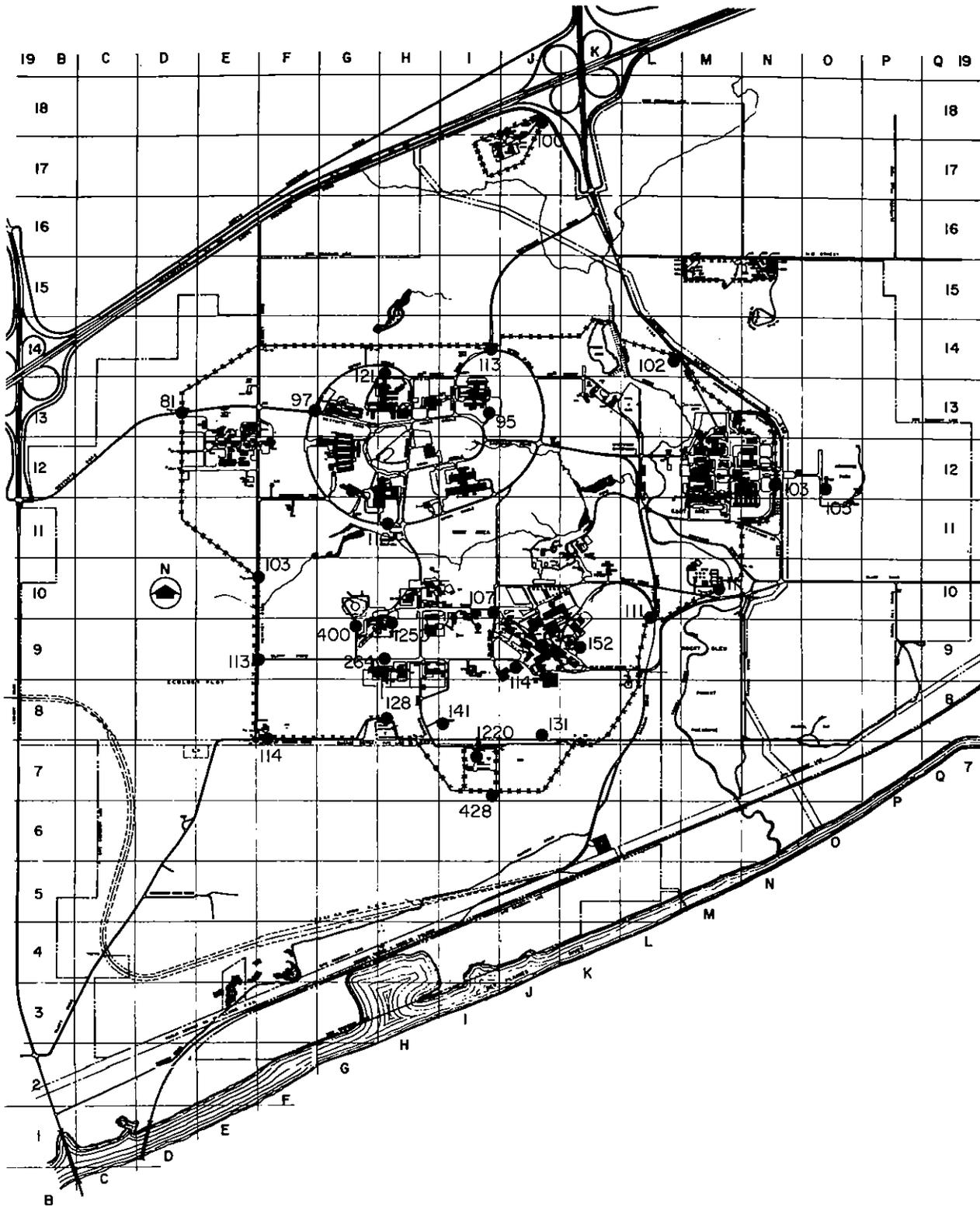


Fig. 5. Penetrating Radiation Measurements on the ANL Site

The dose rate at 8G, 128 mrem/year, is about 20 mrem/year above the normal average. At this location there are three possible sources for above-normal readings: direct radiation from the Waste Storage Area, direct radiation from the low power (10 W to 10 kW) reactors and from a tandem Van de Graaff generator in Building 315 (location 9H) and argon-41 from the CP-5 reactor at location 9H. The abnormal reading at 9H, north of Building 315 (264 mrem/year) makes this building a possible, but minor, contributor to the dose at 8H. The contribution of CP-5 is considered negligible since on-site readings at 11H (as far north of CP-5 as the 8H location is south) and at location 10I (west of CP-5 and slightly farther from CP-5 than 8H) were all normal. The wind roses (Figure 3) show a maximum from the west and southwest, so that higher doses from CP-5 should be observed to the north and east of the reactor. Elevated readings were obtained on-site at location 8H (141 mrem/year), between the Waste Storage Area and the 8G location and at location 7/8J (131 mrem/year), about the same distance from the Waste Storage Area as 8G, but to the east. These readings are consistent with the interpretation that the elevated 8G reading is due primarily to the radioactive materials in the Waste Storage Area. Although additional measurements in the area are indicated to determine the contribution of the various sources to the dose at 8G unequivocally, the Waste Storage Area is believed to be the major contributor.

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.<sup>(13)</sup> The dose from Argonne operations at location 8G (about 20 mrem/year) is well within these limits, particularly since the individuals visiting the cemetery are exposed for only a very small fraction of the year. The dose at the fence line south of the Waste Storage Area, about 300 mrem/year is 60% of the 500 mrem/year limit to individuals, although it is about twice the 170 mrem limit to the "suitable sample". However, as noted previously, there are no individuals being exposed at that location, and the dose at the south boundary of the site is normal. It is also appropriate here to consider the "as low as practicable" guideline limits being proposed for light water cooled power reactors.<sup>(14)</sup> Various limits are being considered, and at present, 10 mrem/year is the one most likely to emerge. The annual

dose at 8G exceeds this value, but as pointed out earlier, individuals receive exposures only when visiting the cemetery. There are no full-time residents living close enough to the site boundary to receive 10 mrem/year.

Other above-normal readings of interest on the site (Table 22) are those 75 yards east (1.3 rem/year) and west (400 mrem/year) of CP-5; just north of Building 315 at location 9H (264 mrem/year), and at 9K, just east of the ZGS meson building (152 mrem/year). The maximum occupational dose at these locations is about 20-25% of the annual dose considering the amount of time employees spend on the site. The applicable annual standard for occupational exposure is considerably higher, 5 rem.

#### 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.<sup>(15)</sup> The new regulations represent a comprehensive and well documented statement with regard to the quality of the aquatic sector. There are several changes and additions in the new regulations, the most notable being the lowering of the standard for copper from 0.04 mg/l to 0.02 mg/l and for silver from 0.05 mg/l to 0.005 mg/l. The copper level is essentially the value occurring naturally, while the silver level is about ten times the level found in treated drinking waters in the United States.<sup>(16)</sup>

The concentrations of barium, copper, iron, nickel, lead, 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.<sup>(18)</sup> Beryllium was determined fluorophotometrically as previously described.<sup>(19)</sup>

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 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 23). The detection limits for the atomic absorption methods are those used by Slavin.<sup>(17)</sup> Detection limits for ion selective methods are those listed by the manufacturer, since they depend entirely on solubility considerations.

#### A. Sawmill Creek

Sampling emphasis was placed on Sawmill Creek, which is tributary to the Des Plaines River, since this is the major route for waste-water leaving Argonne. Samples to be analyzed for dissolved oxygen, ammonia nitrogen, sulfate, and chloride were collected both upstream and downstream from the outfall once per week. Samples for all other analyses were collected three times per week downstream from the outfall.

The values obtained for the pertinent parameters are listed in Table 24. Other criteria not listed are dissolved oxygen and pH since these levels are expressed somewhat differently. The State law now provides that the oxygen levels of a stream shall not be less than 5 mg/l at any time. The oxygen levels varied from 3.9 to 13.5 mg/l above the outfall and from 4.3 to 14.1 mg/l below the outfall. On the basis of past data,<sup>(19)</sup> it has been shown that the water above the outfall is depleted in oxygen. Although the oxygen content of the Argonne effluent is high, it is not sufficient to raise the level substantially.

The pH criteria give a range of 6.5-9 units and these results are not subject to averaging. The pH levels measured above and below the outfall were essentially the same and all values were in the range from 6.7 to 8.1.

The ammonia nitrogen levels are well above the stream criteria both above and below the Argonne outfall. The source has been shown in past reports<sup>(20)</sup> to be well upstream of the Laboratory boundary. The range of ammonia nitrogen

TABLE 23

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	0.006
Fluoride	1.4	-	0.02
Fluoride (total)	-	2.5	0.02
Iron (total)	1.0	2.0	0.06
Lead (total)	0.1	0.1	0.002
Mercury	0.0005	-	0.0001
Mercury (total)	-	0.0005	0.0001
Nickel (total)	1.0	1.0	0.2
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	-	0.02
Zinc (total)	-	1.0	0.02
pH	6.5-9.0	5.0-10.0	

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

TABLE 24

Chemical Constituents in Sawmill Creek, 1972

Constituent	Location *	No. of Samples	Concentration (mg/l)			Percent of State Levels		
			Av.	Min.	Max.	Av.	Min.	Max.
Ammonia N	7M (up)	35	4.1 ± 1.0	0.3	12.0	270	20	800
	7M	47	4.2 ± 0.8	0.2	11.1	280	13	740
Barium (total)	7M	51	-	-	< 1.0	-	-	< 20
Beryllium	7M	12	0.00013 ± 0.00004	0.00005	0.00024	-	-	-
Chloride	7M (Up)	33	312 ± 42	84	669	62	17	134
	7M	33	266 ± 36	83	550	53	17	110
Chromium (VI)	7M	166	0.01 ± 0.002	< 0.006	0.08	20	< 12	160
Copper (total)	7M	51	0.018 ± 0.005	0.008	0.142	90	40	710
Fluoride	7M	45	0.38 ± 0.03	0.24	0.56	27	17	40
Iron (total)	7M	51	0.56 ± 0.18	0.08	3.66	56	8	366
Lead (total)	7M	21	0.011 ± 0.004	< 0.002	0.028	11	< 2	28
Mercury	7M	187	0.0001 ± 0.00001	< 0.0001	0.00055	20	< 20	110
Nickel (total)	7M	77	-	-	< 0.20	-	-	< 20
Silver (total)	7M	51	-	-	< 0.01	-	-	< 200
Sulfate	7M (up)	34	143 ± 10	55	183	29	11	37
	7M	34	148 ± 10	57	188	30	11	38
Zinc	7M	51	0.072 ± 0.011	0.03	0.232	7.2	3.0	23.2

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

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 for the first time because they are recent additions to the regulations. The levels for chloride are slightly lower below the outfall. Values for sulfate are essentially the same at both sampling locations.

The levels for barium, beryllium, hexavalent chromium, copper, lead, mercury, silver, and zinc during this report period remained essentially the same as for 1971. The level of iron increased substantially due to a change in sample treatment. The State law requires that total iron be determined, and substantial amounts of iron formerly filtered out before analysis are now measured. 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 major effluent at ANL is the waste treatment stream which enters Sawmill Creek at 7M. The stream is sampled at the waste treatment facility immediately after treatment by a proportional collector. 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. Additionally, the land between the site security fence and the site boundary may be released to the Du Page County Forest Preserve District. If this occurs, the cooling tower blowdown channels at 14J and 8J will leave the controlled area. 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 25) are comparable to the stream results with four exceptions, zinc, chromium, mercury, and beryllium. The levels of zinc are about five times higher in the plant effluent than in the stream. This has been previously observed,<sup>(21)</sup> and can be accounted for on the basis of stream dilution. The levels in the effluent averaged approximately 50% of the State limit of 1.0 mg/l but never exceeded it.

TABLE 25

Chemical Constituents in Effluent from ANL Treatment Plant, 1972  
(concentrations in mg/l)

Constituent	No. of Samples	Av.	Min.	Max.	Percent of Standard		
					Av.	Min.	Max.
Barium (total)	20	-	-	< 1.0	-	-	< 50
Beryllium	10	0.000027 ± 0.000037	< 0.000003	0.000170	-	-	-
Chromium (VI) (total)	54	0.036 ± 0.008	< 0.006	0.123	12	< 2	41
Fluoride (total)	11	0.46 ± 0.18	0.23	0.94	18	9.2	38
Iron (total)	20	0.29 ± 0.24	0.07	2.41	14	3.5	120
Mercury (total)	53	0.00178 ± 0.00068	0.00016	0.0194	356	< 32	3900
Mercury (dissolved)	32	0.00031 ± 0.00015	< 0.00010	0.0019	62	< 20	380
Nickel (total)	20	-	-	< 0.20	-	-	< 20
pH	53	-	6.80	7.80	-	-	-
Silver	20	-	-	< 0.01	-	-	< 10
Zinc (total)	20	0.45 ± 0.05	0.31	0.62	45	31	62

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 sampling program encompassed the latter third of 1972 and it is anticipated that sampling through the major cooling season will increase the value somewhat.

Sampling of the cooling tower effluents covered most of the year. The cooling tower blowdown channel at 14J averaged 0.20 mg/l and occasionally exceeded the effluent limit of 0.3 mg/l. 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 0.85 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) weekly. 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.

Mercury was determined in the plant effluent during the last quarter of 1972. The average value for total mercury exceeded the State limit by a factor of about 3.5. The average value for the dissolved mercury during this same period was about 20% of the total mercury concentration. The net effect on the average dissolved mercury concentration in Sawmill Creek was small. The exact source of the mercury in the effluent water is unknown. Studies are continuing to determine the source and to eliminate it.

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.<sup>(18)</sup> Results are as shown in Table 26.

Values for ammonia nitrogen in January and 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.

Coliform bacteria analyses were performed once per week using the fermentation tube technique. One result in December was elevated due to construction at

TABLE 26

Deoxygenating Waste and Coliform Measurements  
in ANL Effluent Water, 1972

Month	B.O.D. <sub>5</sub>	Concentration (mg/l)		Coliform Bacteria Coliform/100 ml
		Suspended Solids	Ammonia Nitrogen	
January	1.7	2.25	4.84	438
February	3.2	3.00	10.4	370
March	2.4	1.20	1.93	210
April	2.3	3.50	1.92	20
May	2.2	6.00	2.09	52
June	1.3	0.50	0.38	25
July	0.7	2.25	0.13	130
August	1.1	1.50	0.17	212
September	0.8	4.44	0.37	1508
October	1.7	1.00	0.66	426
November	2.0	1.75	0.98	648
December	3.6	3.14	1.96	6443
State Limit	30.0	37.0	2.5 (April-October) 4.0 (November-March)	5000

the plant. The remainder of the results were well within limits for total coliform set forth in SWB-11.<sup>(22)</sup> In the future, fecal coliform levels will be measured in conformity with the new State regulations.

## 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 27 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.

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%.

TABLE 27

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

Nuclide or Activity	Concentration Guide		Detection Limit	
	Water	Air	Water	Air
Antimony-125	-	$9 \times 10^{-10}$	-	$10^{-15}$
Argon-41	-	$4 \times 10^{-8}$ $2 \times 10^{-6}$	-	$2 \times 10^{-8}$
Barium-140	$3 \times 10^{-5}$	$1 \times 10^{-9}$	$2 \times 10^{-9}$	$10^{-15}$
Beryllium-7	-	$4 \times 10^{-8}$	-	$10^{-15}$
Cerium-141	-	$5 \times 10^{-9}$	-	$10^{-15}$
Cerium-144	-	$2 \times 10^{-10}$	-	$10^{-15}$
Cesium-137	-	$5 \times 10^{-10}$	-	$10^{-15}$
Cobalt-60	-	$3 \times 10^{-10}$	-	$10^{-15}$
Hydrogen-3	$3 \times 10^{-3}$	$2 \times 10^{-7}$ $5 \times 10^{-6}$	$2 \times 10^{-7(b)}$ $1.5 \times 10^{-8(c)}$	$10^{-13}$
Iodine-131	$3 \times 10^{-7}$	$1 \times 10^{-10}$	$3 \times 10^{-9}$	$10^{-14}$
Manganese-54	-	$1 \times 10^{-9}$	-	$10^{-15}$
Neptunium-237	$3 \times 10^{-6}$	-	$2 \times 10^{-12}$	-
Plutonium-238,239	$5 \times 10^{-6}$	$1 \times 10^{-12}$	$5 \times 10^{-13}$	$10^{-18}$
Ruthenium-103	-	$3 \times 10^{-9}$	-	$10^{-15}$
Ruthenium-106	-	$2 \times 10^{-10}$	-	$10^{-15}$
Strontium-89	$3 \times 10^{-6}$	-	$1 \times 10^{-9}$	-
Strontium-90	$3 \times 10^{-7}$	-	$2 \times 10^{-10}$	-
Thorium-228	-	$2 \times 10^{-13}$	-	$10^{-18}$
Thorium-232	-	$1 \times 10^{-12}$	-	$10^{-18}$
Uranium - natural	$4 \times 10^{-5}$	$4 \times 10^{-12}$	$2 \times 10^{-10}$	$2 \times 10^{-17}$
Zirconium-95	-	$1 \times 10^{-9}$	-	$10^{-15}$
Alpha <sup>(a)</sup>	$3 \times 10^{-6}$	$1 \times 10^{-10}$	$2 \times 10^{-10}$	$2 \times 10^{-16}$
Beta <sup>(a)</sup>	$1 \times 10^{-7}$	$1 \times 10^{-13}$	$10^{-9}$	$5 \times 10^{-16}$

(a) 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.

(b) By direct counting of 10 ml.

(c) By electrolytic enrichment of 250 ml.

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