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ENVIRONMENTAL RADIOACTIVITY AT ARGONNE NATIONAL LABORATORY
Report for 1962 and 1963

by

J. Sedlet and F. S. Iwami

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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
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ENVIRONMENTAL RADIOACTIVITY AT ARGONNE NATIONAL LABORATORY

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J. Sedlet and F. S. Iwami

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Industrial Hygiene and Safety Division

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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF TABLES

LIST OF TABLES

ENVIRONMENTAL RADIOACTIVITY AT ARGONNE NATIONAL LABORATORY

Report for the Years 1962 and 1963

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I. SUMMARY

This report presents the results of the environmental monitoring program at Argonne National Laboratory during 1962 and 1963. The purposes of the program are to measure the natural radioactive content of the ANL site and its surroundings, and to determine the origin, identity, and magnitude of any radioactivity not naturally present. Of primary interest is the detection of radioactive materials released to the environment as a result of Argonne operations.

The radioactive content of the environment was determined by radiochemical analyses, total activity measurements, and gamma-ray spectrometric measurements performed on several types of natural materials collected on the ANL site and from locations up to 100 miles from the site. The frequency of sampling decreased with increasing distance from the Laboratory. The sampling locations are described in Part II of this report. Since air and water are the most probable media for spreading radioactive contamination, the sampling program has concentrated on these materials. Argonne waste water is discharged into Sawmill Creek, which in turn flows into the Des Plaines River, and special emphasis was placed on these streams.

The average total activities in samples of water, material from lake and stream beds (bottom silt), soil, and plants collected during 1962 and 1963 are shown in Figures 1 through 6. For comparative purposes, the results obtained from 1952 through 1963 for these samples and for air-filter samples are shown in Figures 7 through 12.

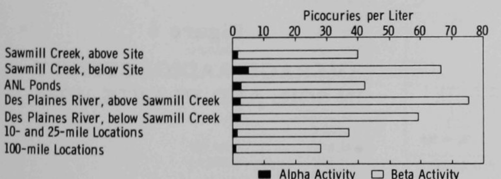


Figure 1

AVERAGE RADIOACTIVITY IN SURFACE WATER, 1962

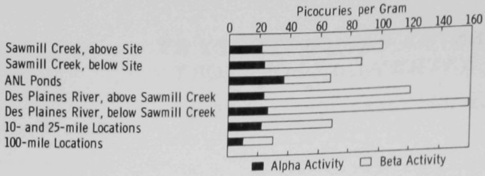


Figure 2

AVERAGE RADIOACTIVITY IN BOTTOM SILT, 1962

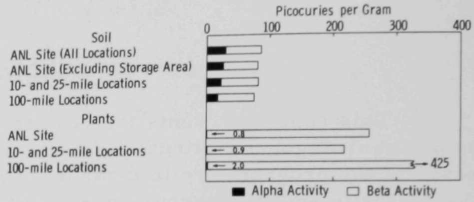


Figure 3

AVERAGE RADIOACTIVITY IN SOIL AND PLANTS, 1962

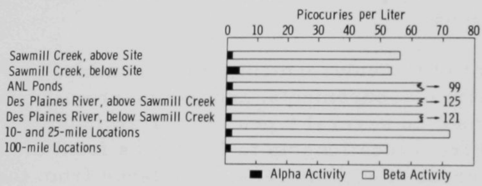
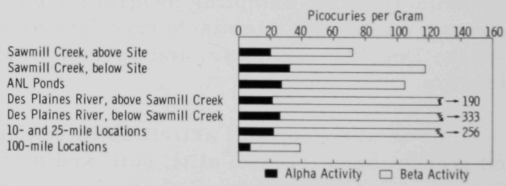


Figure 4

AVERAGE RADIOACTIVITY IN SURFACE WATER, 1963

Figure 5

AVERAGE RADIOACTIVITY IN BOTTOM SILT, 1963



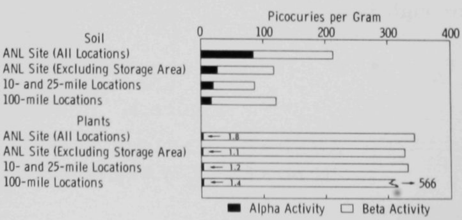


Figure 6

AVERAGE RADIOACTIVITY IN SOIL AND PLANTS, 1963

Figure 7

AVERAGE RADIOACTIVITY IN SURFACE WATER, 1952-1963

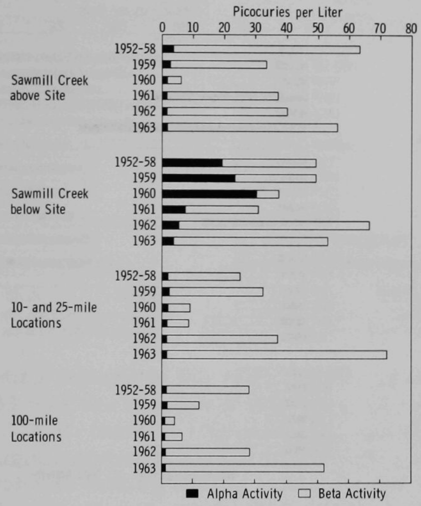


Figure 8

AVERAGE RADIOACTIVITY IN BOTTOM SILT, 1952-1963

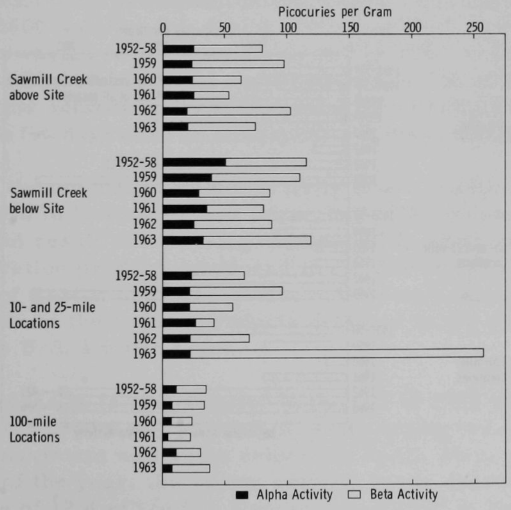


Figure 9

AVERAGE RADIOACTIVITY
IN SOIL, 1952-1963

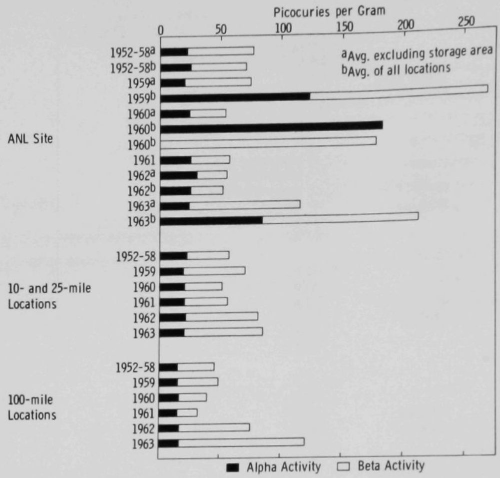


Figure 10

AVERAGE RADIOACTIVITY
IN PLANTS, 1953-1963

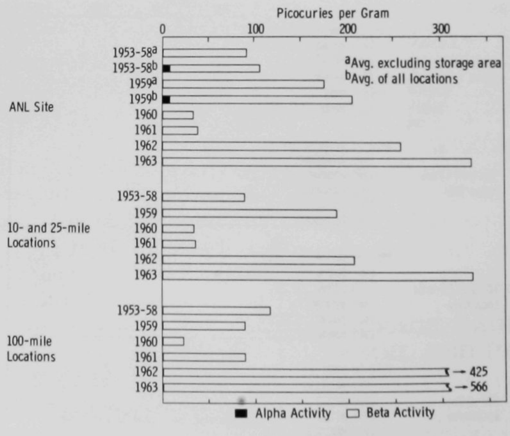


Figure 11

AVERAGE ALPHA ACTIVITY
IN WEEKLY AIR-FILTER
SAMPLES, 1953-1963

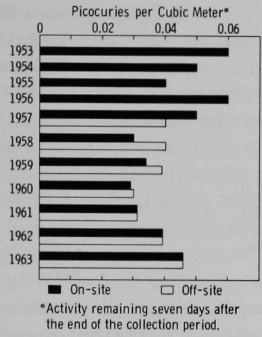
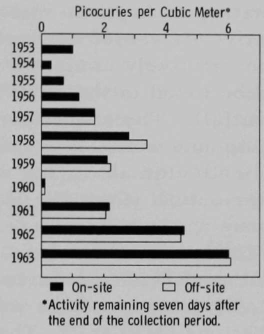


Figure 12

AVERAGE BETA ACTIVITY
IN WEEKLY AIR-FILTER
SAMPLES, 1953-1963



The outstanding feature in the radioactivity of the environment during 1962 and 1963 was the relatively high level of fallout activity resulting from the U. S. S. R. tests in 1961 and 1962 and the U. S. tests in 1962. Fallout averaged somewhat higher in 1963 than in 1962, although there was no atmospheric testing during 1963. By the end of 1963 the expected decrease in fallout was noted. The fallout activity, as measured by the beta activity in 24-hour air-filter samples (Part III-A), averaged 6.7 pCi/m^3 in 1962 and 7.2 pCi/m^3 in 1963. From 1953, when routine measurements of this type were begun, through 1961, the annual averages have ranged from 0.5 pCi/m^3 in 1954 to 4.1 pCi/m^3 in 1961. The amount of fallout activity deposited by precipitation (Part III-B) was also relatively high, approximately 2500 mCi/sq mi during 1962 and 1963. From 1953 to 1961, the annual totals varied from 640 mCi/sq mi in 1960 to about 950 mCi/sq mi in 1961. The highest total recorded was 11,000 mCi/sq mi in 1952. Most of this activity was carried down by a rainfall in June 1952, which occurred a few days after an atmospheric nuclear detonation in Nevada.

In 1962, the amount of fallout activity in air reached a peak in April and May and again in November and December. The increase in the spring was the combined result of a "spring maximum" in stratospheric fallout of fission and activation products produced in the fall of 1961, and of tropospheric fallout of fission products produced in tests conducted in the spring of 1962. The bulk of the fission products detected in the spring of 1962 originated in the U. S. Pacific tests.

The increase in fallout activity in the fall of 1962 was due primarily to fission products produced by U. S. S. R. tests conducted at that time. In 1963, a spring maximum was again evident in April, May, and June. During the second half of the year, the fallout activity in air decreased steadily, from an average of 12.4 pCi/m^3 in June to 1.0 pCi/m^3 in December.

Air-filter samples (Part III-A) were collected continuously from seven sampling stations on the site and from five sampling stations located from 6 to 20 miles from the Laboratory. The activities were essentially the same at the same times both on and off the site, and no indication of activity originating at Argonne was found in the samples. While the beta and gamma activities varied as described earlier due to fallout, the alpha activities were relatively constant. Radioactive materials originating at Argonne could be found in the environment only in Sawmill Creek below the waste-water outfall. The concentrations of radioactive materials added to the creek by Argonne waste water were determined by comparing the activity in water collected above the site with that in water obtained below the waste-water outfall (Part III-C). The principal activities added to the creek in Argonne waste water were Co^{58}, Co^{60}, uranium and its immediate descendants (Th^{234} and Pa^{234}), and Cs^{137}. Small amounts of plutonium, thorium, and other fission products (in addition to Cs^{137}) were also found occasionally. All concentrations were well below the maximum permissible concentrations (MPC's).* The average contribution of ANL waste water to the uranium content of the creek during 1962 and 1963 was about 2 pCi/liter, or 0.005% of the MPC. The natural uranium concentration in the creek during this period was 1.1 pCi/liter. The average plutonium and thorium concentrations in the creek due to ANL waste water were equivalent to approximately 0.003 and 0.004% of the MPC, respectively. The average Co^{58} content of the creek was 80 pCi/liter, or 0.08% of the MPC. The concentrations of the other contaminants for which analyses were made, and whose presence can be attributed to ANL waste water, ranged from 3 pCi/liter (0.006% of the MPC) for Co^{60} to 5 pCi/liter (0.03% of the MPC) for Cs^{137}. Fallout from nuclear detonations also contributed significantly to the radioactivity in the creek; the single largest contributor from this source in terms of MPC (0.5%, or 1.6 pCi/liter) was Sr^{90}. It is estimated that during 1962, ANL waste water and fallout each contributed approximately 30 pCi/liter to the total beta activity in the creek below the outfall, while approximately 6 pCi/liter was due to natural activities. During 1963, the average Argonne contribution decreased to approximately 5 pCi/liter, while the fallout contribution increased to about 40 pCi/liter.

The same activities found in the water were also detected in bottom silt (Part III-D) from the creek. Bottom silt collected below the outfall contained uranium, plutonium, Co^{60}, and the other nuclides present in ANL waste water, as well as fission-product fallout. Activity from ANL waste water was found in samples collected up to 40 yards downstream from the outfall.

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*The maximum permissible concentrations used in this report are those given in U. S. A. E. C. Manual Chapter 0524 (Standards for Radiation Protection). In applying these concentrations, the Chapter states that concentrations of radionuclides may be averaged over periods up to one year. This is usually done in this report in making comparisons with the MPC's, although averages over shorter periods and maximum concentrations are used at times when such comparisons are of interest.

Only on rare occasions could activity from ANL waste water be detected in the Des Plaines River (Part III-C). In October and November 1962, Co^{58} was found in the river below Sawmill Creek at a maximum concentration of 8 pCi/liter, or 0.008% of the MPC. During this period Sawmill Creek water samples contained from 500 to 3260 pCi of Co^{58} per liter. This corresponds to a dilution factor of the order of 1000. The volume of water carried by both streams is variable, and this dilution factor will not necessarily hold for other periods. At all other times during 1962 and 1963, the dilution factors and relative activities in the two streams were such that the activity in Sawmill Creek had no detectable effect on the activity in the river. The average activities were similar both above and below the mouth of Sawmill Creek. The average alpha activities and uranium concentrations, 2.1 and 1.3 pCi/liter, respectively, were essentially the same as those found in earlier years. Because of fallout, the total beta activities, approximately 65 pCi/liter in 1962 and 125 pCi/liter in 1963, were considerably higher than the normal range of 5 to 15 pCi/liter.

The activities in other bodies of water, both on and off the site, followed similar patterns (Part III-C). The alpha activities were within their normal range for these locations, approximately 0.1 to 6 pCi/liter. The actual value depends on a number of variables, including the level of water and the amount of solids, both dissolved and suspended. Lake Michigan generally contains the lowest activities, while small ponds and streams contain the highest. The predominant alpha emitter was uranium, although other natural alpha emitters could also be found. The beta activities averaged about 50 pCi/liter, compared to normal concentrations of 5-15 pCi/liter. The additional activity was due to fallout, and the beta (and gamma) activities in water followed the same variations as fallout activity in air during 1962 and 1963.

The radioactivity in bottom silt (Part III-D) followed the expected behavior at most locations. Fallout activity could be detected in all samples, but the amount varied greatly between locations and, with time, at the same location. Total beta activities in bottom silt in the area normally vary between about 10 and 90 pCi/g, depending on the composition of the silt. Fallout concentrations ranged from about 10 to 1900 pCi/g, and averaged about 50 pCi/g. The activity in bottom silt from Sawmill Creek below the waste-water outfall was mentioned earlier. Samples from this location that were analyzed in detail contained (per gram of sample) 5 to 10 pCi of uranium, 1.3 to 4 pCi of plutonium, and 13 to 23 pCi of Co^{60}, compared to normal values of 1 to 2 pCi of uranium, <0.1 pCi of plutonium, and <1 pCi of Co^{60}. The increased activity in the creek samples reflect the activity added in ANL waste water. Most bottom silt samples contain less than 35 pCi/g of total alpha activity and 1-2 pCi/g of Th^{232}. Some of the samples collected from the DuPage River, one pond on the ANL site, and the Fox River contained abnormally high concentrations of the Th^{232} and its decay products. The uranium content of these samples was normal.

The natural alpha and beta activity in surface soil (Part III-E) in the Chicago area is in the range of 10-40 and 40-90 pCi/g, respectively. The total alpha activities in soil from all locations, except near a radioactive-material storage area on the ANL site, were in the normal range. Near the storage area, the alpha activities varied from normal values up to 1600 pCi/g. The above-normal activities were due to uranium contamination from the storage area. Small amounts of Co^{60} and traces of plutonium and thorium were also found in soil near the storage area. The beta activities in soil from all locations were elevated due to fission products from fallout. The average fallout content of soil during 1962 and 1963 was approximately 20 pCi/g.

The alpha activities in grass samples (Part III-F) from all locations, except the radioactive-material storage area, were in the normal range. At the storage area, the alpha activities were up to a factor of ten greater than normal due to the presence of uranium. Co^{60} could not be found in the plant samples collected from the storage area although the soil from this area did contain this nuclide. The beta activities were unusually high at all locations due to fallout. The average beta activities, 300 pCi/g on the site, 270 pCi/g near ANL, and 500 pCi/g at the reference sites, were 10 to 20 times their natural values.

Three samples of milk were collected monthly (except February 1962) from the area and analyzed for several fission products (Part III-G). The activities found in these samples could be attributed entirely to fallout from nuclear detonations. There was no evidence that activity originating at Argonne was present in any of the samples. The Sr^{90} concentrations varied from approximately 3.5 pCi/liter during the first quarter of 1962 to approximately 15 pCi/liter during the last half of 1963. The Sr^{89} concentrations ranged from less than 5 pCi/liter during the first quarter of 1962 and during December 1963, to 78 pCi/liter in May 1962. Concentrations of Cs^{137}, Ba^{140}, and I^{131} are also given for some samples. The monthly variations in milk activities could be correlated with known periods of nuclear testing, with the spring maxima in stratospheric fallout, and with periods of pasture feeding.

II. PROGRAM AND PROCEDURES

Semiannual summaries of some of the results of the environmental monitoring program at Argonne National Laboratory for 1962 and 1963 have appeared in Radiological Health Data. This report presents the complete results of the program. The purposes of the program are to measure the natural radioactive content of the ANL site and its environment, and to determine the identity, magnitude, and origin of any radioactivity above the natural levels. Of primary interest is the detection of radioactive materials released to the environment by Argonne.

The radioactivity of the environment was determined by radiochemical analyses, total activity measurements, and gamma-ray spectrometric analyses of the types and numbers of samples listed in Table I. Since the most probable means of spreading radioactive contamination are by air and water, the sampling program has emphasized these types of materials. Samples were collected on the ANL site and from locations up to 100 miles from the site. The sampling locations are shown in Figures 13 and 14. Samples collected near ANL are intended to indicate the direction and extent of contamination if significant amounts of radioactive materials are accidentally released to the environment by Argonne. In general, it is expected that natural and fallout activity will be relatively uniform on the site and within 25 miles of the site, while activity released by Argonne should be present in higher concentrations on the site. Thus, higher activities on the site would indicate an activity release by Argonne, while similar activities on, and within 25 miles of, the site would indicate that no significant release has occurred. Interpretations based on such evidence have usually been valid, although localized differences in fallout and natural activity are sometimes observed, and conclusions regarding the origin of small amounts of nonnatural activity must be reached cautiously.

Table I

SAMPLES COLLECTED IN 1962 AND 1963

Type	Number	
	1962	1963
Water	349	364
Precipitation	78	74
Soil	92	95
Bottom silt	91	112
Plant	80	96
Air filters	911	899
Milk	33	39

The samples collected 100 miles from the Laboratory (referred to as "reference sites") were originally intended to serve primarily as continuous checks for contamination during collection, storage, and analysis, since their radioactive content was not expected to change with time. However, because of the widespread occurrence of fallout, the beta activities in these samples have served primarily to indicate the extent and magnitude of fallout activity. Since there is little nonnatural alpha activity in the environment, the primary purpose of the reference-site samples has been realized for total alpha-activity measurements. Because fallout activity was found in all environmental materials in varying amounts, beta activities have been more difficult to interpret, in terms of origin, than alpha activities. However, it was usually possible to distinguish between fallout, naturally occurring activity, and activity from Argonne operations, by making the proper choices of sampling locations and types of analyses.

Figure 13

SAMPLING LOCATIONS ON THE SITE OF ARGONNE NATIONAL LABORATORY

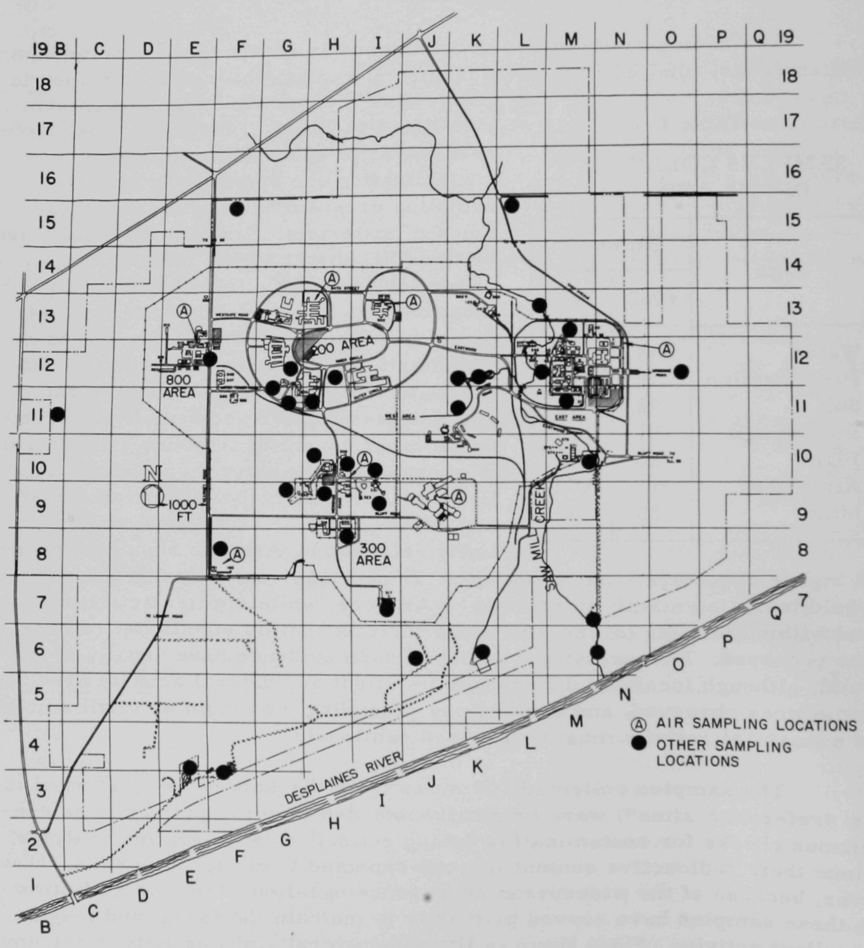
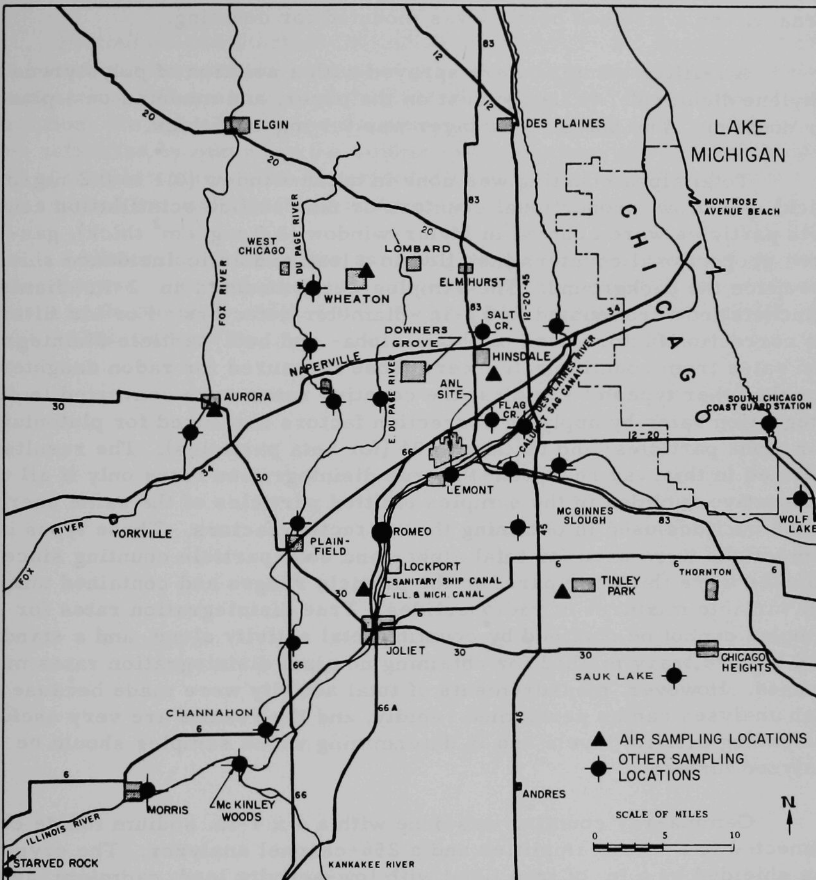


Figure 14

SAMPLING LOCATIONS NEAR ARGONNE NATIONAL LABORATORY



Total alpha and beta activity was measured by counting thick samples (5 to 75 mg/cm^2) after a minimum of sample preparation. Counting rates were converted to disintegration rates as described below, and expressed in terms of picocuries (pCi) per cubic meter (m^3), liter, or gram (g). Water and precipitation samples, acidified with nitric acid after collection to prevent hydrolysis, were evaporated to dryness. Several milligrams of silver iodide were precipitated from the rain and snow samples before evaporation to retain trace amounts of radioiodine. The residue was flamed, ground in a mortar, and spread uniformly on a weighed counting planchet with carbon tetrachloride. The residue was counted after it had been dried and the planchet reweighed.

Soil and bottom silt were dried at 110°C, ground in a mortar, and a weighed portion spread on a counting planchet. Plant samples were washed with water to remove dirt, dried at 110°C, and ashed in a muffle furnace, and a weighed portion was mounted for counting.

Air-filter samples were sprayed with a solution of polystyrene in ethylene dichloride, to fix the dust on the paper, and mounted on a planchet for counting. The polystyrene layer was 0.1 mg/cm^2 thick.

Total alpha counting was done in nylon-window (0.1 to 0.2 mg/cm^2 thick), gas-flow, proportional counters or zinc sulfide scintillation counters. Beta particles were counted in Mylar-window (0.9 mg/cm^2 thick), gas-flow, proportional counters installed in a lead and anticoincidence shield to reduce the background. The samples were mounted in 2-in.-diameter planchets and were counted in $2\frac{1}{4}$-in.-diameter detectors. For air filters, the correction factors used to obtain alpha- and beta-particle disintegration rates from counting rates were those measured for radon daughters. For the other types of samples, the counting rates were converted to disintegration rates by applying correction factors measured for plutonium-239 (for alpha particles) and thallium-204 (for beta particles). The results obtained in this way represent the true disintegration rates only if all the radioactive nuclides in the samples emitted particles of the same energies as the nuclides used in obtaining the correction factors. These types of corrections were used for total alpha- and beta-particle counting since the samples were thick compared to the particle ranges and contained unknown and variable mixtures of radionuclides. True disintegration rates for such samples cannot be obtained by counting total activity alone, and a standardized but arbitrary method for obtaining nominal disintegration rates must be used. However, measurements of total activity were made because many such analyses can be performed rapidly, and the results are very useful in comparing activity levels and in determining which samples should be analyzed further.

Gamma-ray counting was done with a 4 x 4-in. sodium iodide crystal connected to a linear amplifier and a 256-channel analyzer. The crystal was shielded by 6 in. of iron lined with low-activity lead, cadmium, and copper to reduce backscattering. Energy calibrations and counting-efficiency determinations were made with standardized solutions of the appropriate nuclides.

Fission-product and radiocobalt analyses were made by separating the desired element with carrier added, and counting the activity in anticoincidence-shielded and lead-shielded beta counters. Counting rates were converted to disintegration rates by applying correction factors experimentally determined for each nuclide.

Uranium analyses were made with a fluorometer, and the results converted to disintegration rates, by using the specific activity of natural uranium.

Plutonium and thorium (including the thorium-234 daughter of uranium) analyses were made by coprecipitation with cerium fluoride, followed by extraction with a solution of thenoyltrifluoroacetone in benzene. In the extraction, the two elements were separated from each other and from other activities by adjusting the acidity of the solution and the oxidation state of the plutonium. The separated plutonium and thorium fractions were counted for alpha activity in 2π proportional counters.

Additional details on the sampling program, instrumentation, counting techniques, and radiochemical analyses are contained in the previous reports in this series: ANL-5069, -5289, -5446, -5684, -5808, -5934, -6047, -6282, and -6736.

III. RADIOACTIVITY IN ENVIRONMENTAL SAMPLES

A. Air Filters

Airborne particulate matter was sampled at seven locations on the ANL site and at five locations off the site (see Figures 13 and 14). Samples were collected by drawing outside air through Hollingsworth and Vose No. 70 filter paper at a known rate. At one on-site location, the filter paper was changed and counted daily to record short-term changes in air activity. At all other locations, the papers were changed at weekly intervals. The weekly samples were used primarily to compare off-site and on-site activities. The alpha and beta activities in the on-site samples were measured during the first day after collection to obtain the radon and thoron concentrations (from the daughter activities), and again 4 and 7 days after collection to obtain the long-lived activity. The off-site air samples were counted only 4 and 7 days after collection since they could not be obtained sufficiently early for radon and thoron daughter measurement. The decrease in beta activity from the 4th to the 7th day was used to estimate the age of the fallout activity in the samples. The ratio of alpha to beta activity during the first day after collection was used to obtain a rapid fallout estimate, since this ratio is known for samples containing negligible amounts of fallout. Gamma spectra were obtained by combining and counting as one sample all the weekly samples collected each month from each location.

The long-lived alpha and beta activities are listed in Tables II through V. A comparison between the on- and off-site samples (Tables II and III) shows that the alpha and beta activities were similar at both locations. The average differences were less than the 10 to 20% error in collection and analysis, indicating that little, if any, of the airborne particulate activity in the air samples originated at Argonne. Higher activities would be expected on the site if a significant fraction of the activity were from Argonne.

Some individual samples differed from the average activity during a given period by factors up to about three. Such differences occurred at random at all locations, both on and off the site, and are to be expected since the amount of airborne activity depends on a number of variables, such as the amount and composition of the dust in the air and the meteorological conditions at the time of sampling. Because of their random nature, such differences tend to disappear when many samples are averaged.

The alpha activity remaining 4 days after collection was due to naturally-occurring alpha emitters present in dust, and to Po^{210} from the decay of Rn^{222} in the air. The alpha activity has remained fairly constant since sample collection was begun. As shown in Table VI, the average annual alpha activity has varied by a factor of two since 1953. There is a significant decrease in alpha activity from the 3rd to the 4th day, due to

Table II

LONG-LIVED ALPHA AND BETA ACTIVITIES IN WEEKLY AIR-FILTER SAMPLES, 1962

(picocuries per cubic meter)

Month	Location	No. of Samples	Alpha Activity				Beta Activity			
			After 4 Days		After 7 Days		After 4 Days		After 7 Days	
			Max.	Avg.	Max.	Avg.	Max.	Avg.	Max.	Avg.
January	On-site	23	0.0057	0.0035	0.0054	0.0036	10.5	5.6	9.8	5.5
	Off-site	12	0.0084	0.0042	0.0057	0.0039	7.8	5.2	7.6	5.0
February	On-site	23	0.0065	0.0037	0.0069	0.0039	7.9	4.8	7.8	4.6
	Off-site	12	0.0049	0.0038	0.0054	0.0040	6.3	4.6	6.1	4.4
March	On-site	25	0.0071	0.0036	0.0069	0.0036	9.3	5.0	8.9	4.9
	Off-site	16	0.0076	0.0038	0.0067	0.0036	8.5	4.9	8.1	4.8
April	On-site	24	0.0089	0.0051	0.0074	0.0043	11.6	5.9	11.2	5.7
	Off-site	17	0.0102	0.0055	0.0059	0.0042	8.1	5.6	7.7	5.4
May	On-site	25	0.0089	0.0058	0.0059	0.0048	10.6	5.6	10.0	5.4
	Off-site	18	0.0088	0.0054	0.0060	0.0042	7.8	5.2	7.3	5.1
June	On-site	27	0.0102	0.0053	0.0070	0.0042	8.0	4.9	7.9	4.8
	Off-site	19	0.0088	0.0048	0.0060	0.0036	7.8	4.4	7.9	4.4
July	On-site	27	0.0087	0.0045	0.0073	0.0035	4.9	3.2	4.9	3.1
	Off-site	19	0.0083	0.0047	0.0068	0.0038	5.3	3.4	5.2	3.3
August	On-site	27	0.0072	0.0046	0.0051	0.0033	3.8	2.4	3.7	2.3
	Off-site	16	0.0096	0.0057	0.0055	0.0037	3.5	2.4	3.4	2.3
September	On-site	25	0.0126	0.0055	0.0071	0.0032	13.9	4.6	9.1	3.7
	Off-site	19	0.0129	0.0048	0.0060	0.0033	8.4	4.1	6.9	3.7
October	On-site	27	0.0097	0.0052	0.0056	0.0034	8.1	4.2	7.2	3.8
	Off-site	19	0.0110	0.0056	0.0047	0.0034	5.9	4.1	5.4	3.7
November	On-site	27	0.0124	0.0055	0.0072	0.0038	19.5	7.6	17.3	6.8
	Off-site	18	0.0150	0.0060	0.0083	0.0040	13.9	7.3	13.4	6.6
December	On-site	19	0.0136	0.0056	0.0067	0.0046	10.0	5.0	9.6	4.5
	Off-site	13	0.0153	0.0064	0.0083	0.0050	9.4	5.6	8.6	5.1
Annual Summary	On-site	299	0.0136	0.0048	0.0074	0.0039	19.5	4.9	17.3	4.6
	Off-site	198	0.0153	0.0050	0.0083	0.0039	13.9	4.8	13.4	4.5

Table III

LONG-LIVED ALPHA AND BETA ACTIVITIES IN WEEKLY AIR-FILTER SAMPLES, 1963

(picocuries per cubic meter)

Month	Location	No. of Samples	Alpha Activity				Beta Activity			
			After 4 Days		After 7 Days		After 4 Days		After 7 Days	
			Max.	Avg.	Max.	Avg.	Max.	Avg.	Max.	Avg.
January	On-site	27	0.0062	0.0042	0.0069	0.0041	11.6	5.9	11.1	5.7
	Off-site	20	0.0085	0.0045	0.0070	0.0043	10.8	6.3	10.6	6.2
February	On-site	24	0.0083	0.0051	0.0070	0.0050	10.1	7.8	9.8	7.5
	Off-site	14	0.0084	0.0058	0.0068	0.0055	11.9	8.2	11.2	7.9
March	On-site	26	0.0077	0.0048	0.0075	0.0046	8.9	6.6	8.6	6.4
	Off-site	15	0.0113	0.0051	0.0066	0.0044	9.6	6.9	9.4	6.6
April	On-site	27	0.0089	0.0065	0.0076	0.0052	13.6	9.6	12.9	9.3
	Off-site	23	0.0132	0.0072	0.0087	0.0053	12.4	9.1	12.4	9.1
May	On-site	24	0.0101	0.0064	0.0086	0.0053	12.6	9.5	12.2	9.1
	Off-site	20	0.0096	0.0061	0.0069	0.0049	13.9	9.7	13.6	9.5
June	On-site	22	0.0089	0.0064	0.0078	0.0052	12.1	9.7	12.1	9.4
	Off-site	20	0.0101	0.0069	0.0076	0.0055	13.2	10.2	13.0	9.9
July	On-site	22	0.0096	0.0060	0.0063	0.0045	10.1	7.5	10.0	7.4
	Off-site	21	0.0133	0.0057	0.0088	0.0044	9.9	7.3	9.7	7.2
August	On-site	22	0.0098	0.0055	0.0051	0.0040	6.9	4.9	6.8	4.9
	Off-site	22	0.0090	0.0058	0.0084	0.0040	7.0	4.8	7.0	4.7
September	On-site	19	0.0087	0.0048	0.0053	0.0035	3.2	2.3	3.1	2.2
	Off-site	19	0.0094	0.0050	0.0050	0.0035	3.2	2.3	3.0	2.2
October	On-site	23	0.0180	0.0077	0.0134	0.0053	3.0	2.1	3.0	2.0
	Off-site	23	0.0112	0.0075	0.0097	0.0055	3.3	2.2	3.1	2.2
November	On-site	22	0.0070	0.0049	0.0055	0.0034	1.4	0.9	1.4	0.9
	Off-site	22	0.0075	0.0049	0.0052	0.0035	1.3	0.9	1.3	0.9
December	On-site	17	0.0063	0.0041	0.0056	0.0037	1.0	0.8	1.0	0.8
	Off-site	20	0.0076	0.0041	0.0056	0.0036	0.9	0.7	0.9	0.7
Annual Summary	On-site	275	0.0180	0.0055	0.0134	0.0045	13.6	5.6	12.9	5.5
	Off-site	237	0.0133	0.0057	0.0097	0.0045	13.9	5.7	13.6	5.6

Table IV

LONG-LIVED ALPHA AND BETA ACTIVITIES IN 24-hr AIR-FILTER SAMPLES ON ANL SITE, 1962

(picocuries per cubic meter)

Month	No. of Samples	Alpha Activity[1]		Beta Activity[1]		Beta Activity[2]	
		Max.	Avg.	Max.	Avg.	Max.	Avg.
January	37	0.0065	0.0036	16.2	6.8	15.1	6.8
February	44	0.0131	0.0041	12.8	6.2	12.1	6.2
March	31	0.0151	0.0063	15.5	6.8	14.9	6.6
April	30	0.0309	0.0109	13.4	7.3	12.9	7.0
May	29	0.0286	0.0121	9.6	6.4	9.3	5.4
June	30	0.0271	0.0118	10.2	5.2	10.1	5.1
July	31	0.0211	0.0097	8.6	3.9	8.2	3.8
August	31	0.0246	0.0131	12.2	3.2	11.7	3.1
September	30	0.0278	0.0160	56.5	7.8	52.2	7.2
October	31	0.0252	0.0119	21.4	5.8	18.7	5.5
November	27	0.0407	0.0148	40.5	12.1	34.6	9.9
December	30	0.0326	0.0136	22.9	9.2	20.6	7.5
Summary	381	0.0407	0.0107	56.5	6.7	52.2	6.3

[1]Activity remaining 4 days after end of sampling period.
[2]Activity remaining 7 days after end of sampling period.

Table V

LONG-LIVED ALPHA AND BETA ACTIVITIES IN IN 24-hr AIR-FILTER SAMPLES ON ANL SITE, 1963

(picocuries per cubic meter)

Month	No. of Samples	Alpha Activity[1]		Beta Activity[1]		Beta Activity[2]	
		Max.	Avg.	Max.	Avg.	Max.	Avg.
January	31	0.0217	0.0081	19.2	9.9	18.5	9.2
February	26	0.0197	0.0076	19.4	9.9	18.8	9.9
March	31	0.0252	0.0076	13.4	8.0	13.1	7.6
April	30	0.0255	0.0120	21.5	11.1	21.7	10.5
May	31	0.0229	0.0125	38.3	12.3	36.2	11.9
June	30	0.0334	0.0176	18.6	12.9	17.9	12.4
July	30	0.0227	0.0133	13.1	9.0	12.9	8.3
August	31	0.0253	0.0135	9.6	5.7	9.4	5.5
September	30	0.0297	0.0137	6.9	2.8	5.3	2.6
October	31	0.0415	0.0224	4.8	2.3	4.9	2.2
November	30	0.0293	0.0152	2.0	1.3	1.9	1.2
December	30	0.0357	0.0090	1.5	1.0	1.6	1.0
Summary	361	0.0415	0.0127	38.3	7.2	36.2	6.9

[1]Activity remaining 4 days after end of sampling period.
[2]Activity remaining 7 days after end of sampling period.

Table VI

AVERAGE ANNUAL RADIOACTIVITY IN AIR-FILTER SAMPLES, 1953-1963

(picocuries per cubic meter)

Year	Location	Weekly Samples				Daily Samples, 3-4 Days	
		Alpha Activity		Beta Activity		Alpha Activity	Beta Activity
		3-4 days	7 days	3-4 days	7 days		
1953	On-site	0.010	0.006	1.2	1.0	0.03	1.4
1954	On-site	0.010	0.005	0.4	0.3	0.03	0.5
1955	On-site	0.007	0.004	0.8	0.7	0.03	1.0
1956	On-site	0.010	0.006	1.3	1.2	0.03	1.6
1957	On-site	0.008	0.005	2.1	1.7	0.021	2.0
	Off-site	0.007	0.004	2.1	1.7	-	-
1958	On-site	0.007	0.003	2.8	2.5	0.024	3.0
	Off-site	0.007	0.004	3.4	2.7	-	-
1959	On-site	0.006	0.0034	2.2	2.1	0.021	2.4
	Off-site	0.006	0.0039	2.3	2.2	-	-
1960	On-site	0.007	0.0029	0.10	0.09	0.018	0.11
	Off-site	0.006	0.0030	0.09	0.09	-	-
1961	On-site	0.004	0.0031	2.7	2.2	0.006	4.1
	Off-site	0.004	0.0031	2.5	2.1	-	-
1962	On-site	0.0048	0.0039	4.9	4.6	0.011	6.7
	Off-site	0.0050	0.0039	4.8	4.5	-	-
1963	On-site	0.0055	0.0045	5.6	5.5	0.013	7.2
	Off-site	0.0057	0.0045	5.7	5.6	-	-

the decay of residual thoron daughters. From 1953 through 1960, the samples were measured 3 instead of 4 days after collection, so the difference between the 3- to 4-day activity and the 7-day activity was more pronounced before 1960. The alpha activity in an air-filter sample increases substantially over a period of months due to the growth of Po^{210} from Pb^{210}. The corresponding increase in beta activity from the growth of Bi^{210} is less noticeable because the initial beta activtiy is much greater than the initial alpha activity.

The beta activity remaining after the decay of the radon and thoron daughters was due almost entirely to fallout from nuclear detonations. The fallout activity increased sharply with the resumption of atmospheric nuclear testing by the U. S. S. R. in September 1961 and remained relatively high until the fall of 1963. The bulk of the airborne beta activity originated in the atmospheric nuclear tests conducted by the U. S. S. R. in 1961 and 1962, and by the U. S. in 1962. In addition, some of the fallout may have resulted from vented underground tests in Nevada, although the evidence is not conclusive. The increase in the fall of 1962 was accompanied by a decrease in the age of the fission products, and correlates well with the announced

U. S. S. R. atmospheric test series in September 1962. The rise in the spring of 1963 probably resulted from an increase in the stratospheric fallout ("spring maximum") of fission products produced primarily in 1962. This increase was not due to fission products produced in 1963. The steady decrease from June to December of 1963 was expected since atmospheric nuclear tests were not conducted in 1963.

Concentrations of the principal gamma-ray emitters in the air samples are given in Tables VII and VIII. These results were obtained from gamma-ray spectra determined by counting, as one combined sample, all the individual samples collected each month from each location. The results support the conclusions given earlier on the sources of the beta activity in the air samples. All the nuclides listed in Tables VII and VIII are fission products. Other nuclides were also detected in the air samples, including the neutron-activation products Mn^{54} and Sb^{124}, cosmic-ray-produced Be^{7}, and other fission products. However, quantitative determinations of these nuclides were not attempted.

Table VII

PRINCIPAL GAMMA-RAY ACTIVITY IN AIR-FILTER SAMPLES, 1962

(picocuries per cubic meter)

Month	Location	Barium-Lanthanum-140	Cerium-141	Cerium-144	Cesium-137	Ruthenium-103	Ruthenium-Rhodium-106	Zirconium-Niobium-95
January	On-site	-	0.67	0.69	0.019	0.45	0.21	2.0
	Off-site	-	0.70	0.76	0.013	0.51	0.19	2.1
February	On-site	-	1.0	0.76	0.018	0.38	0.25	1.8
	Off-site	-	0.93	0.79	0.024	0.42	0.37	1.8
March	On-site	<0.01	0.63	1.1	0.032	0.32	0.42	1.9
	Off-site	<0.01	0.96	1.5	0.033	0.36	0.35	2.4
April	On-site	<0.01	1.1	1.6	0.035	0.32	0.55	1.9
	Off-site	<0.01	1.0	1.6	0.040	0.31	0.54	2.0
May	On-site	0.068	1.2	1.6	0.051	0.21	0.50	1.7
	Off-site	0.064	1.2	1.5	0.054	0.25	0.54	1.6
June	On-site	0.073	1.5	1.5	0.13	0.15	0.46	1.2
	Off-site	0.067	1.2	1.3	0.10	0.17	0.46	1.5
July	On-site	0.11	1.8	1.3	0.056	0.22	0.54	1.0
	Off-site	0.13	1.4	1.5	0.060	0.29	0.55	1.0
August	On-site	0.29	0.50	0.93	0.037	0.32	0.31	0.59
	Off-site	0.36	0.60	1.0	0.040	0.36	0.34	0.68
September	On-site	1.4	1.1	0.73	0.028	0.30	0.23	0.99
	Off-site	1.5	1.1	0.62	0.027	0.33	0.20	0.76
October	On-site	1.2	1.4	0.49	0.025	0.29	0.22	0.81
	Off-site	1.3	1.3	0.61	0.023	0.30	0.23	0.92
November	On-site	2.7	1.7	1.0	0.024	0.63	0.30	1.1
	Off-site	3.2	1.0	1.4	0.026	0.69	0.35	1.4
December	On-site	0.68	1.5	2.0	0.035	0.41	0.37	1.9
	Off-site	0.77	1.5	2.0	0.040	0.43	0.32	1.8
Annual Summary	On-site	0.56	1.2	1.3	0.046	0.32	0.38	1.5
	Off-site	0.61	1.1	1.4	0.043	0.36	0.38	1.5

Table VIII
PRINCIPAL GAMMA-RAY ACTIVITY IN AIR-FILTER SAMPLES, 1963
(picocuries per cubic meter)

Month	Location	Antimony-125	Barium-Lanthanum-140	Cerium-141	Cerium-144	Cesium-137	Ruthenium-103	Ruthenium-Rhodium-106	Zirconium-Niobium-95
January	On-site	0.02	0.75	1.7	1.5	0.04	0.70	0.55	3.0
	Off-site	0.02	0.69	1.9	1.8	0.03	0.72	0.57	2.9
February	On-site	0.04	0.40	2.2	2.2	0.06	0.97	0.85	3.4
	Off-site	0.04	0.34	2.3	2.2	0.07	1.08	0.81	3.5
March	On-site	0.04	0.10	2.0	1.8	0.09	0.65	0.89	3.0
	Off-site	0.04	0.11	2.3	2.2	0.09	0.70	0.89	3.2
April	On-site	0.08	<0.01	1.7	3.1	0.14	0.69	1.7	4.4
	Off-site	0.09	<0.01	1.6	4.0	0.15	0.78	1.6	4.6
May	On-site	0.12	<0.01	1.9	3.6	0.17	0.73	1.9	4.5
	Off-site	0.13	<0.01	2.1	4.4	0.18	0.84	2.1	4.8
June	On-site	0.15	<0.01	2.1	4.4	0.24	0.56	2.1	4.4
	Off-site	0.16	<0.01	2.2	4.6	0.28	0.58	2.2	4.5
July	On-site	0.13	<0.01	0.8	3.3	0.16	0.29	1.2	2.6
	Off-site	0.11	<0.01	0.8	2.9	0.15	0.28	1.1	2.4
August	On-site	0.09	<0.01	<0.5	2.2	0.10	0.13	0.94	1.4
	Off-site	0.08	<0.01	<0.5	2.1	0.10	0.15	0.84	1.4
September	On-site	0.04	<0.01	<0.5	1.1	0.05	0.07	0.43	0.54
	Off-site	0.04	<0.01	<0.5	1.0	0.05	0.06	0.41	0.53
October	On-site	0.04	<0.01	<0.5	0.90	0.05	0.05	0.36	0.44
	Off-site	0.04	<0.01	<0.5	0.92	0.06	0.05	0.42	0.41
November	On-site	0.02	<0.01	<0.5	0.45	0.03	<0.05	0.20	0.14
	Off-site	0.02	<0.01	<0.5	0.49	0.03	<0.05	0.17	0.14
December	On-site	0.02	<0.01	<0.5	0.40	0.02	<0.05	0.19	0.08
	Off-site	0.02	<0.01	<0.5	0.43	0.02	<0.05	0.17	0.09
Annual Summary	On-site	0.06	0.11	1.1	2.1	0.10	0.40	0.94	2.3
	Off-site	0.05	0.10	1.2	2.3	0.10	0.45	0.94	2.4

The data in Tables VII and VIII are plotted, together with the total beta activities from Tables II and III, in Figure 15. The monthly variations

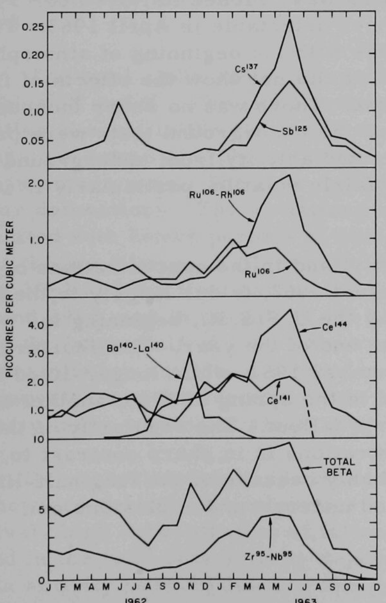


Figure 15

FALLOUT ACTIVITY IN AIR, 1962-1963

usually, but not always, parallel each other when the half-lives of the individual nuclides are considered. During the first 6 months of 1962, Zr^{95}-Nb^{95} and the cerium nuclides were the predominant gamma-emitters in fallout. The ruthenium nuclides also contributed significantly. The increase in total beta activity in the spring of 1962 must be attributed to an increase in stratospheric fallout of fission products produced in the fall of 1961 since the shorter-lived nuclides, such as Ba^{140}, Ru^{103}, and Zr^{95}-Nb^{95}, were either absent or decreasing in concentration during this period. In addition, the Ru^{103}/Ru^{106} activity ratio decreased with nearly its expected half-life of 45 days, indicating no injection of newly-generated fission products.

The origin of the fallout beginning in May 1962 is of interest. There is some evidence [E. A. Martell, Science, 143, 126 (1964)] that volatile fission products, particularly iodine-131, from some of the continental underground tests during 1962 contributed substantially to the atmospheric activity in some parts of the United States. Airborne iodine-131 concentrations were not determined during this period, and the measurements made do not permit a definite conclusion to be drawn regarding the presence of fission products from underground tests. However, the data do indicate that only a small fraction of the fallout activity measured could have originated in underground tests. Atmospheric testing in the Pacific was conducted by the U.S. from April 25, 1962, through October 1962. Most of the detonations occurred in May and June 1962. Underground tests in the U.S. were conducted during most of 1962. Barium-140 appeared for the first time in May 1962, increased until a peak was reached in November 1962, and then decreased until it was no longer detectable in April 1963. The appearance of Ba^{140} in May 1962 coincides with the beginning of atmospheric testing. In addition, the Ru^{103}/Ru^{106} ratio did not show the effects of freshly-produced activity beginning in May 1962. There was no sharp increase as would have occurred if ruthenium from the underground tests were present. The enrichment of ruthenium in the vented activity from underground tests might be expected since ruthenium is fairly volatile, particularly in its higher oxidation states.

The increase in the total activity, and in the concentrations of all nuclides except Cs^{137}, beginning in August 1962, is due largely to the large-scale atmospheric testing conducted by the U.S.S.R., beginning in August 1962 and continuing through the end of the year. The shorter-lived nuclides showed maxima in November 1962, while longer-lived activities and the total beta activity peaked in the spring of 1963, another example of a "spring maximum" in stratospheric fallout. The regularity of the monthly variations in the Cs^{137} concentrations is in sharp contrast to the variations in the other activities, probably because of the long half-life of Cs^{137}. There is no evidence that vented underground explosions contributed to the fallout in 1963.

During some months, significant differences in the concentrations of some nuclides were found between the off-site and on-site samples. However, these differences occurred at random in both directions and were due to experimental error in resolving the complex gamma-ray spectra, errors in sample collection (primarily flow-rate measurement), and probably to localized differences in the amount of fallout. The random differences in the monthly values and the small differences in the annual averages indicate that these gamma-ray emitters originated in a widespread source such as fallout, and not in a localized source such as Argonne. The data do not support any other interpretation. Because of the large area covered by the samples, large differences would be expected between the on-site and off-site results if Argonne activity were present.

B. Precipitation

Samples of all precipitation on the ANL site were collected and analyzed. The rain water collector was arranged to collect two successive one-gallon portions when the rainfall was sufficiently large. Each portion was equivalent to 0.48 in. of rain. A collector of this size is sufficient to collect more than 90% of the precipitation during a year and still provide samples sufficiently large for analysis from rainfalls totaling 0.01 in., or from rainfalls containing little activity. When two portions were collected, they were analyzed as separate samples. Snow was collected from a measured area of polyethylene-covered plywood sheet fixed to the ground.

The total alpha and beta activities during 1962 and 1963 are shown in Tables IX and X. The alpha activities were due primarily to natural activities present in the air and in dust blown into the rain collector. The average and range of alpha activities during the 2 years was similar to those in other years. The beta activity was primarily due to fallout from nuclear detonations. The variations in, and levels of, beta activity can be correlated with known periods of nuclear testing, and with the beta activities in the air filters. Fallout activity in precipitation was relatively high during 1962 and the first half of 1963. Maxima occurred in the spring of 1962 and 1963, and in the fall of 1962, and a general decrease was evident during the latter half of 1963. Exact correspondence between the air filter and precipitation activities cannot be expected since the air filters operate continuously near the surface, while precipitation is an occasional sampler of a very large volume of air of variable height.

The calculated ages, or detonation dates, also conform qualitatively to announced periods of nuclear tests in the spring and fall of 1962. The relatively high concentration of fallout in the air during the 2 years is reflected in the total beta activity deposited on the ground. The total amount of beta activity carried down by the precipitation was about 2500 mCi/sq mi

Table IX

TOTAL ALPHA AND BETA ACTIVITY IN PRECIPITATION ON ANL SITE, 1962

Month	Days after Precip.	No. of Samples	Alpha Activity (pCi/liter)		Beta Activity (nCi/liter)			Age Range* (days)
			Max.	Avg.	Max.	Avg.	Total (mCi/sq mi)	
January	1	3	1.9	1.5	2.28	1.88	-	-
	4	5	1.4	0.7	2.24	1.26	-	-
	7	5	1.2	0.7	2.11	1.20	304	57-95
February	1	4	6.8	2.9	1.89	1.28	-	-
	4	6	6.9	3.1	2.82	1.55	-	-
	7	6	5.7	2.0	2.73	1.48	143	57-120
March	1	4	19.6	8.3	6.41	3.52	-	-
	4	5	5.8	3.2	5.98	2.88	-	-
	7	5	4.6	2.8	5.89	2.83	147	47->300
April	1	7	69	21	9.60	2.70	-	-
	4	9	26.0	5.9	9.20	2.59	-	-
	7	9	6.0	3.1	8.70	2.47	259	45->300
May	1	7	5.9	2.8	7.70	2.44	-	-
	4	10	1.8	0.8	6.51	1.96	-	-
	7	10	1.8	0.8	5.14	1.66	432	17-250
June	1	5	15.0	7.4	1.31	0.72	-	-
	4	8	2.4	1.2	1.44	0.90	-	-
	7	8	1.5	0.8	1.36	0.84	147	47->300
July	1	10	49	12	10.30	2.48	-	-
	4	10	4.9	1.8	9.13	2.16	-	-
	7	10	4.3	1.8	8.29	1.96	554	25-55
August	1	3	4.1	2.2	0.74	0.45	-	-
	4	3	2.4	1.2	0.68	0.41	-	-
	7	3	2.1	1.0	0.64	0.40	38	55->300
September	1	5	47	13	2.95	1.81	-	-
	4	5	6.4	2.4	2.58	1.63	-	-
	7	5	5.1	2.1	2.40	1.48	121	26->300
October	1	8	38	14	5.20	2.66	-	-
	4	8	4.6	1.9	4.59	2.42	-	-
	7	8	3.9	1.9	4.20	2.20	178	22-160
November	1	4	7.4	5.2	4.36	2.87	-	-
	4	4	1.8	1.4	3.52	2.45	-	-
	7	4	1.6	1.0	3.03	2.11	136	20-30
December	1	4	58	31	11.1	5.5	-	-
	4	5	30.7	15.6	23.5	8.9	-	-
	7	5	29.0	12.1	23.1	8.2	86	28-45
Annual Summary	1	64	69	11	11.1	2.4	-	-
	4	78	30.7	3.1	23.5	2.2	-	-
	7	78	29.0	2.5	23.1	2.2	2437	-

*Calculated assuming beta-decay rate proportional to $T^{-1.2}$.

Table X

TOTAL ALPHA AND BETA ACTIVITY IN PRECIPITATION ON ANL SITE, 1963

Month	Days after Precip.	No. of Samples	Alpha Activity (pCi/liter)		Beta Activity (nCi/liter)		Beta Activity Total (mCi/sq mi)	Age Range* (days)
			Max.	Avg.	Max.	Avg.		
January	1	5	12.0	6.4	5.97	3.42	-	-
	4	5	9.4	5.3	5.43	3.14	-	-
	7	5	9.4	5.1	5.09	2.95	148	38-55
February	1	5	9.5	5.4	3.23	2.53	-	-
	4	4	6.4	3.2	3.09	2.42	-	-
	7	5	6.2	2.7	3.03	2.30	22	60-100
March	1	7	5.7	3.5	4.91	2.96	-	-
	4	7	5.7	2.8	4.76	2.79	-	-
	7	7	5.1	2.7	4.65	2.75	397	60-370
April	1	9	183	39	12.2	5.1	-	-
	4	9	24.2	6.5	11.6	4.9	-	-
	7	9	21.4	6.0	11.3	4.8	595	70-220
May	1	7	86	25	3.48	2.02	-	-
	4	7	6.2	3.7	3.41	1.93	-	-
	7	7	4.2	2.5	3.29	1.86	275	120-320
June	1	7	48	23	5.74	3.51	-	-
	4	7	10.6	6.2	5.63	3.38	-	-
	7	7	9.9	5.7	5.34	3.27	473	120-205
July	1	10	55	16	2.34	1.08	-	-
	4	10	3.4	1.6	2.15	1.03	-	-
	7	10	3.1	1.4	2.12	1.02	290	175-370
August	1	6	58	25	1.91	1.45	-	-
	4	6	14.2	5.7	1.88	1.39	-	-
	7	6	10.9	4.3	1.84	1.37	154	80->370
September	1	6	47	17	0.93	0.57	-	-
	4	6	2.9	1.7	0.84	0.54	-	-
	7	6	2.9	1.6	0.85	0.54	82	75->370
October	1	4	26	11	1.33	0.88	-	-
	4	5	7.7	4.0	1.29	0.85	-	-
	7	5	7.3	4.0	1.27	0.84	51	150->370
November	1	2	31	21	0.66	0.52	-	-
	4	4	7.6	3.5	0.62	0.32	-	-
	7	4	6.1	2.6	0.62	0.32	17	130->370
December	1	2	10.8	6.2	0.29	0.20	-	-
	4	3	10.3	4.2	0.28	0.18	-	-
	7	3	9.8	4.1	0.27	0.18	46	>370
Annual Summary	1	70	183	18	12.2	2.3	-	-
		73	24.2	4.0	11.6	2.1	-	-
		74	21.4	3.5	11.3	2.1	2550	-

*Calculated assuming beta-decay rate proportional to $T^{-1.2}$.

during each of the 2 years. This was the second highest annual total since precipitation collection was begun. The highest total recorded was 11,000 mCi/sq mi in 1952. From 1953 to 1961, the annual totals varied from 64 mCi/sq mi in 1960 to about 950 mCi/sq mi in 1961. The total amount of activity in the precipitation was the same during both 1962 and 1963, although there was very little atmospheric testing in 1963. The highest beta activities in the air-filter samples were also recorded during the spring of 1963 and resulted from fission products produced during 1962.

The results of analyses of 1962 precipitation samples for specific fission products (Table XI) indicate a gradually increasing age until May 10,

Table XI

FISSION-PRODUCT ACTIVITY IN SELECTED PRECIPITATION SAMPLES, 1962

(picocuries per liter)

Date	Amount (in.)	Total Beta Activity ($\times 10^3$)	I^{131}	Ba^{140}	Sr^{89}	Sr^{90}	Cs^{137}	Ce^{141} ($\times 10^2$)	Ce^{144} ($\times 10^2$)
January 4-5	0.25	2.11		151	772	14	5.6		1.1
January 5-8	0.5	1.08		36	175	1.2			
January 14-15	0.7	1.92		42	260	4.5			
January 26	0.42	0.30		10	82	1.8			
February 1	0.14	1.70		26	284	7.3			
February 15	0.05	2.73			1380	31			
February 17-19	0.34	0.60		13	237	6.8			
February 21-22	0.24	1.42		11	212	7.6			
February 25-26	0.32	1.48		27	767	24			
February 27-28	0.04	0.96		31	371	14	29		
March 4	0.03	5.89		26	830	32			6.7
March 8-9	0.20	0.94		5	149	6.2			
March 19	0.33	1.24			97	4.4			
March 21	0.20	3.38		20	634	31			
March 30		2.71		< 10	350	18		2.4	3.5
April 5		2.29		<10	311	18			
April 8	0.59	1.10			185	11		2.0	1.7
April 21-22	0.12	3.04			570	51			
May 10	0.99	5.14	1250	1220	572	26		7.2	2.7
May 12	0.71	1.67		284	300	21			4.7
June 4	1.04	0.82				7.6			0.45
June 8-10	1.44	0.54				9.2	12		
June 30									
July 2	1.58	8.29		23	55	8.8	15	1.5	2.0
July 11	0.97	2.82	250	217	346	14		0.6	1.3
July 22	0.53	1.65	64				23		
July 30		0.48	10				11		
August 5-6	1.53	0.25		<10				0.2	0.3
August 25-26	0.48	0.64	7						
September 8-10	1.24	0.46	21			3.4	8.6		1.6
September 8-10	1.24	0.48	14						
September 24	0.16	2.07	165			13			
September 30	0.03	2.4	96						
October 7	0.29	4.20	182	567	391	15			
October 16	0.22	0.74	32						
October 19-20	0.54	0.55	32				6.5		
November 2-3	0.20	1.87	135						
November 16	0.59	2.52	233			6.5			0.67
November 16	0.01	1.04	77						
December 13	0.03	9.50	58						
December 26	0.02	4.86	10	220	523	14	24		3.4

when the increase in the Sr^{89}/Sr^{90} ratio and the large Ba^{140} and I^{131} concentrations indicate the presence of recently produced fission products in the air. The Sr^{89}/Sr^{90} ratio in precipitation decreased from January through the April 21-22 sample with about a 54-day half-life, as would be expected if most of the activity were produced during the fall of 1961. The appearance of short-lived fission products, and the increase in the Sr^{89}/Sr^{90} ratio beginning with the May 10 sample, correlates with the U. S. atmospheric testing begun on April 25, 1962, at Christmas Island. The I^{131} concentration was not unusually large compared to the concentrations of other short-lived fission products. There is no indication from these results that fractionated debris from vented underground tests in the continental U. S. contributed to the fallout activity. The second increase in the I^{131}, Ba^{140}, and Sr^{89} concentrations during the fall and winter of 1962 correlates with the U. S. S. R. atmospheric testing during that period.

The results of fission-product analyses of the 1963 samples in Table XII show an increase in the age of the fallout during the year

Table XII

FISSION-PRODUCT ACTIVITY IN SELECTED PRECIPITATION SAMPLES, 1963

(picocuries per liter)

Date	Amount (in.)	Total Beta Activity (x 10^3)	I^{131}	Ba^{140}	Sr^{89}	Sr^{90}	Cs^{137}	Ce^{144}
January 14	0.3	5.09	61					
January 19	-	2.20	21					
January 23	0.01	1.38	28	100	22	4		
January 30	0.05	3.19	49	108	50	10		
February 10	0.15	2.92	9			17		300
February 20	0.20	1.73	5.1					
February 26	0.20	1.71	4.6					
March 13	0.39	4.65				70		
March 19	0.30	3.91			400	55		336
April 1	0.32	2.17	2.5		251	51		
April 17	0.19	10.0			680	113		
April 18	0.19	11.3			215	83		
April 29	0.50	2.04	<2					
June 9	1.21	2.88		<1	305	54	100	810
June 9	1.21	1.97		<1	210	37		
June 13	0.14	2.01		<1	356	71		
June 19	0.27	3.70		<1	356	71		
June 28	0.28	3.26		<1	210	45		
July 13	1.81	0.72		<1	93	26		
July 13	1.81	0.23		<1	41	9.9	27	
July 16	1.30	2.12		<1	172	44		
July 16	1.30	1.54		<1	129	33	95	
July 19	-	0.86		<1	63	16		
July 21	1.13	1.08		<1	91	26		
August 1	1.11	1.54		<1	114	28		
August 19	0.10	1.84		<1	205	82		
August 28	0.47	1.52		<1	46	22	75	
September 2	0.94	0.61		<1	25	15		
September 12	0.6	0.85		<1	31	18		
September 12	0.19	0.49		<1	15	12		
October 17	0.17	1.27			72	23		280
October 19	0.42	0.49				13	36	
November 24	0.15	0.37			<5	27		97
December 8	0.20	0.27				6.5	18	81

(the I^{131}, Ba^{140}, and Sr^{89} concentrations, and the Sr^{89}/Sr^{90} ratio decrease with time), although the absolute values for Sr^{89} and Sr^{90} are relatively high, especially in the spring, when it is considered that the fission products were produced during the previous fall and winter. A spring maximum in fallout, similar to that noted in the air-filter samples, is evident both in 1962 and 1963.

C. Water

1. Sawmill Creek and Des Plaines River

Argonne waste water is discharged into Sawmill Creek at location 7M in Figure 13, and this stream was sampled at location 13L, before it passed any of the ANL buildings, and below the waste-water outfall to determine if any radioactive materials were added to the stream in the waste water. Samples were collected below the outfall three times weekly and combined prior to analysis. In this way, the average activity in the samples collected each week was obtained. Since the activity below the outfall generally varied widely between samples, a representative average activity could be obtained only if all the samples were analyzed separately, or if the samples were combined prior to analysis. Since it was impractical to analyze all the samples for all nuclides of interest, the latter system was used.

Above-site samples were collected at weekly intervals, and at least one sample each month was analyzed for each nuclide of interest. When the stream contains little fallout activity, the radioactivity above the outfall consists primarily of natural activities and remains fairly constant with time. Under these conditions, the average activity during any period is close to the activity in a few samples collected during the same period, and the results obtained from the sampling and analytical schedule (composite below-outfall samples and weekly above-site samples) can be compared directly to determine the Argonne contribution to the below-outfall water.

When the fallout activity in the creek varied appreciably with time and contributed a significant portion of the total beta activity, the number of radiochemical fission-product and gamma-ray spectrometric analyses of above-site water was increased to obtain a better value for the average activity at that location. During such periods, direct comparisons of above- and below-outfall beta activities cannot solely be used to evaluate the Argonne contribution to the stream since fallout is added to the stream below the outfall directly from the air (primarily by precipitation) as well as by above-site water. Under these conditions it is difficult to evaluate the Argonne contribution of fission products directly, and additional information, such as detonation dates, air and precipitation activities, and fission-product ratios, is useful.

The alpha activities found in Sawmill Creek during 1962 and 1963 are presented in Tables XIII and XIV. The average alpha activity above the site during both years, 1.5 pCi/liter, was normal. At this location, the alpha activity was due almost entirely to naturally-occurring radioactive nuclides. The higher average alpha activity below the outfall indicates that the waste water contained slightly higher concentrations of alpha activity than natural creek water. Since Sawmill Creek water is diluted approximately in half by waste water, the average natural alpha activity below the outfall was approximately one-half of the above-site activity, and the contribution by ANL waste water was approximately 4.2 pCi/liter in 1962 and 3.2 pCi/liter in 1963. In other years, the average annual alpha activity added to the creek in waste water was greater, and varied from 6 to 30 pCi/liter. Concentrations of the alpha emitters most likely to be present in ANL waste water are also given in Tables XIII and XIV. As in the past, the largest contributor was uranium. During 1962 and 1963, the average uranium contribution was slightly greater than 2 pCi/liter, or only about 0.005% of the MPC*.

Table XIII

NONVOLATILE ALPHA ACTIVITY IN SAWMILL CREEK WATER, 1962

Month	Location[1]	Total Alpha Activity			Uranium			Plutonium			Thorium		
		No. of Samples	pCi/liter Max.	pCi/liter Avg.	No. of Samples	pCi/liter Max.	pCi/liter Avg.	No. of Samples	pCi/liter Max.	pCi/liter Avg.[2]	No. of Samples	pCi/liter Max.	pCi/liter Avg.[2]
January	13L	1	1.8	-	1	1.5	-	0	-	-	0	-	-
	7M	12	11.5	7.5	12	7.6	6.3	12	0.06	0.05	12	<0.05	-
February	13L	2	2.2	2.1	0	-	-	1	<0.05	-	1	<0.05	-
	7M	12	4.1	3.4	12	2.7	2.5	12	0.12	0.06	12	0.05	<0.05
March	13L	4	2.3	1.8	2	1.2	1.0	1	<0.05	-	1	<0.05	-
	7M	12	5.7	3.9	12	3.9	2.6	12	<0.05	-	12	0.10	0.06
April	13L	4	1.9	1.6	2	1.1	1.1	1	<0.05	-	1	<0.05	-
	7M	12	7.0	4.7	12	4.4	3.1	12	0.13	0.06	12	0.37	0.13
May	13L	5	2.5	1.9	2	1.3	1.2	1	<0.05	-	1	<0.05	-
	7M	12	7.6	5.5	12	5.5	3.8	12	0.18	0.06	12	0.05	<0.05
June	13L	5	2.3	1.8	2	1.0	0.9	1	<0.05	-	1	<0.05	-
	7M	12	4.4	3.9	12	3.9	2.6	12	<0.05	-	12	0.10	<0.05
July	13L	3	2.8	2.2	2	1.9	1.0	2	<0.05	-	2	0.10	0.10
	7M	15	7.7	3.6	15	2.0	1.5	15	0.11	0.05	15	0.48	0.11
August	13L	5	1.9	1.5	2	0.8	0.7	1	<0.05	-	1	<0.05	-
	7M	12	8.2	5.0	12	3.1	2.7	12	1.2	0.38	12	0.15	0.10
September	13L	4	2.0	1.2	2	0.5	0.5	1	<0.05	-	1	<0.05	-
	7M	12	3.5	2.7	12	3.7	2.2	12	<0.05	-	12	0.05	-
October	13L	5	1.0	0.5	2	1.1	0.9	1	<0.05	-	1	<0.05	-
	7M	15	11.2	6.4	15	5.1	2.7	15	0.11	0.05	12	0.13	0.09
November	13L	4	4.1	1.7	2	1.2	0.8	1	<0.05	-	1	<0.05	-
	7M	12	5.5	4.3	12	2.5	2.1	12	0.59	0.32	12	0.14	0.09
December	13L	4	2.8	1.4	2	0.8	0.7	1	<0.05	-	1	<0.05	-
	7M	15	16.4	8.4	15	4.4	2.4	15	0.91	0.19	15	0.18	0.08
Annual Summary	13L	46	4.1	1.6	21	1.9	0.9	12	<0.05	-	12	0.10	<0.05
	7M	153	16.4	5.0	153	7.6	2.9	153	0.91	0.11	153	0.48	0.07

[1]Location 13L is upstream from ANL site. Location 7M is downstream from waste-water outfall. (See Figure 13.)

[2]Averages were calculated assuming concentration of 0.025 pCi/liter, one-half of minimum detectable concentrations, for samples in which plutonium or thorium was not detected.

* The MPC for uranium and thorium is based upon a special definition of the Curie such that one "Curie" of recently extracted uranium or thorium is actually equivalent to (very nearly) 2 Ci of uranium (U^{238} + U^{234} + U^{235}) and thorium (Th^{232} + Th^{228}). Thus, the MPC must be multiplied by two for comparison with the concentrations given in this report, since these concentrations are based on the conventional definition of the Curie.

Table XIV

NONVOLATILE ALPHA ACTIVITY IN SAWMILL CREEK WATER, 1963

Month	Location[1]	Total Alpha Activity			Uranium			Plutonium			Thorium		
		No. of Samples	pCi/liter		No. of Samples	pCi/liter		No. of Samples	pCi/liter		No. of Samples	pCi/liter	
			Max.	Avg.		Max.	Avg.		Max.	Avg.[2]		Max.	Avg.[2]
January	13L	5	2.2	1.0	2	1.7	1.2	1	<0.05	-	1	<0.05	-
	7M	12	10.1	6.5	12	6.0	3.8	12	0.36	0.14	12	0.06	<0.05
February	13L	4	2.3	1.2	2	0.7	0.7	1	<0.05	-	1	<0.05	-
	7M	12	7.6	5.0	12	6.7	3.4	12	0.42	0.15	12	0.19	0.09
March	13L	4	2.7	1.5	2	1.3	0.8	1	<0.05	-	1	0.17	-
	7M	12	3.9	3.0	12	1.9	1.8	12	0.37	0.13	12	1.0	0.52
April	13L	4	3.3	2.3	2	1.3	1.2	1	<0.05	-	1	0.12	-
	7M	15	3.0	2.4	15	1.6	1.3	15	0.55	0.28	15	0.18	0.15
May	13L	5	2.3	1.6	2	1.7	1.5	1	<0.05	-	1	<0.05	-
	7M	12	6.6	3.3	12	5.0	2.7	12	0.26	0.10	12	0.38	0.22
June	13L	4	1.3	1.0	2	1.1	1.0	1	<0.05	-	1	0.07	-
	7M	12	3.4	2.3	12	2.2	1.7	12	0.10	0.06	12	0.18	0.13
July	13L	5	3.8	1.9	2	1.8	1.5	1	<0.05	-	1	0.15	-
	7M	15	7.0	3.4	15	3.5	2.7	15	0.18	0.09	15	0.27	0.12
August	13L	4	1.5	0.8	2	1.6	1.3	1	<0.05	-	1	0.14	-
	7M	14	10.9	4.7	14	4.3	2.3	14	0.21	0.09	14	0.30	0.12
September	13L	4	2.2	1.1	2	1.4	1.3	1	<0.05	-	1	0.09	-
	7M	12	8.6	5.8	12	2.9	2.2	12	1.2	0.70	12	0.40	0.22
October	13L	5	4.1	1.6	2	1.4	1.2	1	<0.05	-	1	0.08	-
	7M	12	11.8	6.6	12	8.0	3.9	12	0.62	0.34	12	0.17	0.12
November	13L	4	2.4	1.5	2	2.0	1.5	1	<0.05	-	1	0.13	-
	7M	12	4.9	3.1	12	4.8	2.8	12	0.30	0.23	12	0.07	0.06
December	13L	4	2.3	1.3	2	1.9	1.5	1	<0.05	-	1	<0.05	-
	7M	15	6.4	2.9	15	5.0	2.6	15	0.26	0.17	15	0.15	0.07
Annual Summary	13L	52	4.1	1.4	24	2.0	1.2	12	<0.05	-	12	0.17	0.09
	7M	155	11.8	4.0	155	8.0	2.6	155	1.2	0.20	155	1.0	0.16

[1]Location 13L is upstream from ANL site. Location 7M is downstream from waste-water outfall. (See Figure 13.)

[2]Averages were calculated assuming concentration of 0.025 pCi/liter, one-half of minimum detectable concentration, for samples in which plutonium or thorium were not detected.

Exact correspondence between the total alpha and uranium activities cannot be expected, even when uranium is the only alpha emitter present, because of the errors inherent in determining the total alpha activity by counting thick samples and because the isotopic composition of the uranium in the waste water is unknown. A fluorometric method was used for determining the uranium concentration because of its accuracy and sensitivity for small amounts of natural or depleted uranium and because of its speed. The concentration was converted to activity through the use of the specific activity of natural uranium. However, a variety of isotopic mixtures is used at Argonne, and the alpha activity due to uranium in a given sample can be (and has been) larger or smaller than the activity calculated from the fluorometric analysis when the sample contains enriched or depleted uranium.

Small amounts of plutonium and thorium were also detected in about one-half of the samples collected from Sawmill Creek below the waste-water outfall, evidently as a result of their presence in the waste water. The average plutonium concentrations in the creek were equivalent to 0.002 and 0.004% of the MPC during 1962 and 1963, respectively, while the highest sample encountered during both years was 1.2 pCi/liter, or 0.024% of the MPC. The thorium concentrations below the outfall, calculated by the same procedure as total activity and uranium concentration, amounted to 0.0025% of the MPC in 1962 and 0.0055% of the MPC in 1963. These values, as well as those given in Tables XIII and XIV, were calculated assuming a concentration of 0.025 pCi/liter, one-half of the minimum detectable concentration, for those samples in which plutonium or thorium were not detected. If such samples are assumed to have zero concentration, the average plutonium and thorium concentrations are about 20% lower. The isotopic composition of the thorium was not investigated completely, but the growth of alpha activity in the separated thorium fractions suggests that the activity was due primarily to thorium-232 and its descendant, thorium-228. The occurrence of plutonium and thorium in Sawmill Creek during 1962 and 1963 was similar to that of previous years. The concentrations have always been low, but on the average, higher below the outfall than above the site.

The beta activities found in Sawmill Creek water are presented in Tables XV and XVI. These results were supplemented by gamma-ray spectrometry on many of these same samples to assist in defining the origin of abnormally high beta activities. The natural beta activity in the creek usually varies between 5 and 10 pCi/liter. Activities above this range during 1962 and 1963 were due to fallout above the site and to fallout

Table XV

BETA ACTIVITY IN SAWMILL CREEK WATER, 1962

Month	Location[1]	Total Beta Activity			Colbalt-58			Colbalt-60			Strontium-89			Strontium-90			Cesium-137		
		No. of Samples	pCi/liter		No. of Samples	pCi/liter		No. of Samples	pCi/liter		No. of Samples	pCi/liter		No. of Samples	pCi/liter		No. of Samples	pCi/liter	
			Max.	Avg.		Max.	Avg.[2]		Max.	Avg.[2]		Max.	Avg.		Max.	Avg.[2]		Max.	Avg.[2]
January	13L	1	11	-	1	<5	-	1	<3	-	1	3.5	-	1	0.6	-	0	-	-
	7M	12	54	40	12	72	34	12	6.1	3.7	9	10	6	12	3.0	1.8	12	9.3	4.4
February	13L	2	65	48	1	<5	-	1	<3	-	2	41	31	1	2.4	1.6	0	-	-
	7M	12	75	49	12	260	71	12	7.5	5	12	22	17	12	1.8	1.5	12	7.5	4.0
March	13L	4	142	69	4	<5	-	4	<3	-	4	40	25	4	2.3	1.7	1	1.4	-
	7M	12	77	57	12	<5	-	12	<3	-	12	25	20	12	1.6	1.3	12	5.4	2.5
April	13L	4	25	19	4	<5	-	4	<3	-	4	11	10	4	1.6	1.3	1	<0.5	-
	7M	12	56	34	12	5.7	3.8	12	<3	-	12	11	9	12	1.6	1.2	12	3.1	1.7
May	13L	5	61	43	5	<5	-	5	<3	-	5	23	16	5	2.3	1.6	1	1.0	-
	7M	12	52	47	12	78	36	12	<3	-	12	16	13	12	2.6	1.4	12	4.7	4.1
June	13L	4	44	29	4	<5	-	4	<3	-	4	19	13	4	2.0	1.4	1	<0.5	-
	7M	12	76	45	12	283	81	12	3.0	1.7	12	11	7	12	3.1	2.3	12	10.3	4.2
July	13L	4	393	120	1	<5	-	1	<3	-	4	1.1	1.0	4	1.2	1.0	1	3.4	-
	7M	15	88	71	15	535	174	15	7.1	4.8	15	15	11	15	1.9	1.2	15	9.0	3.7
August	13L	5	29	22	1	<5	-	1	<3	-	3	3.4	2.6	3	2.0	1.5	1	3.6	-
	7M	12	47	36	12	147	96	12	10.4	4.6	12	6.8	3.5	12	<0.5	-	12	2.1	1.1
September	13L	4	28	23	1	<5	-	1	<3	-	2	3.5	3.0	2	1.5	1.3	1	1.2	-
	7M	12	61	43	12	228	86	12	5.2	2.5	12	15	8	12	1.6	0.9	12	7.6	4.4
October	13L	5	44	22	1	<5	-	1	<3	-	2	4.9	3.8	2	1.0	1.0	1	7.7	-
	7M	15	806	192	12	3260	669	12	46	10	12	10	6	12	2.6	1.0	15	3.7	2.1
November	13L	4	37	23	1	<5	-	1	<3	-	2	6.9	4.2	2	1.2	0.9	1	1.4	-
	7M	12	232	118	12	1005	362	12	19	14	12	10	6	12	2.2	1.4	12	3.0	2.7
December	13L	4	45	39	1	<5	-	1	<3	-	2	7.9	5.3	2	1.3	1.0	1	<0.5	-
	7M	15	104	56	15	91	33	15	1.7	1.2	15	5.0	3.8	15	1.5	0.9	15	254	55
Annual Summary	13L	46	393	40	25	<5	-	25	<3	-	35	41	11	34	2.4	1.3	10	7.7	2.0
	7M	153	806	66	150	3260	133	150	46	4	147	25	9	150	3.1	1.3	153	254	8

[1]Location 13L is upstream from ANL site. Location 7M is downstream from waste-water outfall. (See Figure 13.)

[2]Averages were calculated assuming concentration of one-half minimum detectable concentration for those samples in which given nuclide was not detected.

Table XVI

BETA ACTIVITY IN SAWMILL CREEK WATER, 1963

Month	Location[1]	Total Beta Activity			Cobalt-58			Cobalt-60			Strontium-89			Strontium-90			Cesium-137		
		No. of Samples	pCi/liter Max.	pCi/liter Avg.	No. of Samples	pCi/liter Max.	pCi/liter Avg.[2]	No. of Samples	pCi/liter Max.	pCi/liter Avg.[2]	No. of Samples	pCi/liter Max.	pCi/liter Avg.[2]	No. of Samples	pCi/liter Max.	pCi/liter Avg.	No. of Samples	pCi/liter Max.	pCi/liter Avg.[2]
January	13L	5	35	20	1	<5	-	1	<2	-	2	<2	-	2	0.6	0.5	1	<0.5	-
	7M	12	31	27	12	12.2	6.3	12	<2	-	12	3.2	1.1	12	3.2	1.1	12	3.4	2.2
February	13L	4	152	56	1	<5	-	1	<2	-	1	4.7	-	1	1.2	-	1	<0.5	-
	7M	12	114	74	12	27.4	19.5	12	<2	-	9	16.0	10.0	9	2.5	1.7	12	10.5	7.4
March	13L	4	489	239	1	<5	-	1	<2	-	1	23.5	-	1	5.3	-	1	1.2	-
	7M	12	169	137	12	6.5	3.7	12	<2	-	12	14.6	10.1	12	7.8	3.7	12	4.0	2.5
April	13L	4	178	90	1	<5	-	15	<2	-	1	1.1	-	1	5.1	-	1	2.1	-
	7M	15	116	72	15	5.7	3.5	15	<2	-	15	67.0	25.1	15	9.4	4.1	15	6.9	3.1
May	13L	5	78	48	1	<5	-	1	<2	-	2	30.3	20.3	2	4.5	3.1	1	0.6	-
	7M	12	70	55	12	10.0	5.4	12	4.0	2.5	12	21.4	12.2	12	3.2	2.7	12	2.9	2.0
June	13L	4	76	51	1	<5	-	1	<2	-	2	11.7	9.3	2	3.1	3.0	1	0.6	-
	7M	12	57	43	12	<5	-	12	3.0	2.4	12	7.4	4.7	12	2.8	2.2	12	2.6	2.0
July	13L	5	188	71	1	<5	-	1	<2	-	2	18.9	10.6	2	8.5	5.0	1	0.5	-
	7M	15	111	57	15	5.0	<5	15	<2	-	15	8.4	3.8	15	4.5	2.0	15	5.0	3.6
August	13L	4	66	37	1	<5	-	1	<2	-	2	9.5	5.6	2	5.1	4.4	1	<0.5	-
	7M	14	192	59	14	10.0	5.0	14	2.5	<2	14	10.1	3.1	14	7.0	2.3	14	2.9	2.0
September	13L	4	48	27	1	<5	-	1	<2	-	2	3.9	3.5	2	6.6	5.5	1	<0.5	-
	7M	12	176	58	12	20.5	10.2	12	4.9	2.6	12	<2	-	12	1.8	1.2	12	2.1	1.3
October	13L	5	22	17	1	<5	-	1	<2	-	2	<2	-	2	2.2	1.9	1	<0.5	-
	7M	12	28	18	12	25.0	7.4	12	5.9	2.9	12	<2	-	12	1.2	0.6	12	5.2	2.2
November	13L	4	22	17	1	<5	-	1	<2	-	2	<2	-	2	2.4	2.0	1	<0.5	-
	7M	12	19	17	12	5.0	3.0	12	2.0	1.1	12	<2	-	12	1.3	0.9	12	4.8	2.3
December	13L	4	32	18	1	<5	-	1	<2	-	2	2.6	1.8	2	1.4	1.4	1	<0.5	-
	7M	15	23	17	15	30.0	8.9	15	20.0	5.1	15	<2	-	15	1.2	0.5	15	2.1	1.7
Annual Summary	13L	52	489	56	12	<5	-	12	<2	-	21	30.3	6.5	21	8.5	3.1	12	2.1	0.6
	7M	155	192	53	155	27.4	6.4	155	20.0	1.9	152	67.0	6.3	152	9.4	1.9	155	10.5	2.7

[1]Location 13L is upstream from ANL site. Location 7M is downstream from waste-water outfall. (See Figure 13.)

[2]Averages were calculated assuming concentration of one-half of minimum detectable concentration for those samples in which given nuclide was not detected.

and/or activity in Argonne waste water below the outfall. When the fallout rate is high, and precipitation contributes much of the activity in the creek at both sampling locations, it is difficult to determine quantitatively the origin of the fission products in the stream below the outfall. However, reasonable estimates of the contributions made by fallout and Argonne waste water to the radioactivity in the creek can be made by comparing results of samples collected during the same week, by comparing fission-product ratios above and below the outfall, and by comparing creek activities with air and precipitation activities at the same time. From considerations of these types, it is estimated that during 1962, Argonne waste water and fallout each contributed approximately 30 pCi/liter to the total beta activity in the creek. During 1963, the average Argonne contribution decreased to approximately 5 pCi/liter, while the fallout contribution increased to about 40 pCi/liter.

The Argonne contribution was due primarily to Co^{58}, Co^{60}, and Cs^{137}. Occasional samples contained other fission products from Argonne waste water, but the bulk of the radiostrontium as well as other fission products (whose concentrations are not given in the tables) were added to the water from fallout. Iodine-131 and Ba^{140} from fallout were found in a few samples from both locations early in 1962, and during the spring of 1963, in amounts up to 20 pCi/liter. Iodine-131 from Argonne waste water was found below the site in July and October 1963 in concentrations ranging from 50 to 78 pCi/liter. Other common fission products, such as Zr^{95}-Nb^{95}, cerium, and ruthenium isotopes, were detected at both locations. On some days the total beta activity above the site was significantly greater than that below the outfall due to the addition of waste water containing less activity than natural creek water.

The Argonne contribution of the cobalt nuclides and uranium daughters (Th^{234} and Pa^{234}) can be obtained directly as before from the differences between the two sampling locations. In some samples, the Co^{58} concentration was greater than the total beta activity because only 15% of the Co^{58} disintegrations occur by positron emission and contribute to the total beta activity. The portion that decays by electron capture (85%) contributes little to the measured beta activity. The cobalt activities are believed to originate primarily in the Experimental Boiling Water Reactor and are produced by neutron irradiation of some of the materials of construction (iron, cobalt, and nickel). The uranium-daughter contribution to the total beta activity below the outfall is approximately 60% of the uranium concentration. This value is based on the measured self-absorption of the beta particles emitted by the uranium daughters and on the usual state of equilibrium between the uranium and its daughters in the water.

Regardless of source, the total concentration of a nuclide not naturally present in the environment must be considered when comparisons are made with the MPC's. Thus, in Table XVII, the total concentrations of the fission products are used rather than that fraction attributed to Argonne. As shown in the table, the most abundant beta emitter in the creek was Co^{58}, although the highest contribution in terms of fraction of MPC was made by Sr^{90}. Compared to the MPC's, the concentrations were relatively low and did not constitute a health hazard.

Table XVII

BETA EMITTING NUCLIDES IN BELOW-OUTFALL SAWMILL CREEK WATER, 1962-1963

Nuclide	Concentration (pCi/liter)		Percent MPC	
	Maximum	Average	Maximum	Average
Cobalt-58	3260	80	3.3	0.08
Cobalt-60	46	3	0.09	0.006
Thorium-234	5.3	1.6	0.03	0.008
Cesium-137	254	5	1.3	0.03
Strontium-89	67	8	2.2	0.27
Strontium-90	9.4	1.6	3.1	0.5
Iodine-131	78	<3	26	<1
Barium-140	20	<3	0.07	<0.01

On October 1, 1963, water and bottom silt from Sawmill Creek was sampled at several locations downstream from the waste-water outfall. The water results are listed in Table XVIII; the bottom-silt results are given in Section III-D. Since the radionuclides in Sawmill Creek water are also found in the bottom silt, this type of survey might give some information on the rate of removal of traces of ions by the bottom silt. Conclusive results can only be expected when the activity in the water is significantly above normal, that is, when the creek contains activity from the waste water that is not in equilibrium with the silt. In the past, the results from surveys of this kind have been inconclusive, but it appeared that the rate of removal from the water by the silt was slow, and that most of the activity traveled downstream for a considerable distance. In Table XVIII, the beta activities are nearly normal, but the alpha activities are two to three times their natural levels. It appears that the alpha activity is removed from the water only slowly, and that at any one time only a small fraction is lost from solution. There was a small decrease from 0 to 150 yards, but the 90% counting error at this level is about 1 pCi/liter, and the activity increased again at 200 yards.

Table XVIII

TOTAL ALPHA AND BETA ACTIVITY IN SAWMILL CREEK WATER AS FUNCTION OF DISTANCE DOWNSTREAM FROM WASTE-WATER OUTFALL, OCTOBER 1, 1963

Distance Downstream from Waste-water Outfall (yards)	Alpha Activity (pCi/liter)	Beta Activity (pCi/liter)
0	7.2	14.2
50	6.0	16.1
100	5.2	16.3
150	5.2	14.8
200	6.4	15.3
250	6.3	16.8

2. Des Plaines River

Since Sawmill Creek empties into the Des Plaines River about 500 yards downstream from the waste-water outfall, and since it is likely that much of the active material added to the creek in ANL waste water enters the river, the river was sampled at weekly intervals (except when the ice layer was very thick) to determine if the activity in the creek had any effect on the radioactivity in the Des Plaines River. All samples were analyzed for total alpha and beta activity. Analyses for specific elements and nuclides were performed less frequently. In addition to the analyses reported in Tables XIX and XX, one sample each month was analyzed for Co^{58}, Co^{60}, Ba^{140}, I^{131}, plutonium, and, in 1962, thorium. These results are not tabulated because positive values were rarely obtained.

Table XIX

RADIOACTIVITY IN DES PLAINES RIVER WATER, 1962

Month	Location[1]	Total Alpha Activity			Total Beta Activity			Uranium			Strontium-89			Strontium-90			Cesium-137		
		No. of Samples	pCi/liter		No. of Samples	pCi/liter		No. of Samples	pCi/liter		No. of Samples	pCi/liter		No. of Samples	pCi/liter		No. of Samples	pCi/liter	
			Max.	Avg.		Max.	Avg.		Max.	Avg.		Max.	Avg.		Max.	Avg.[2]		Max.	Avg.[2]
March	A	2	2.3	2.0	2	122	85	1	1.1	-	0	-	-	0	-	-	1	1.6	-
	B	2	2.4	2.2	2	98	73	2	1.0	1.0	0	-	-	0	-	-	1	1.6	-
April	A	4	3.7	2.7	4	47	35	2	1.9	1.5	1	13	-	1	1.8	-	1	<0.5	-
	B	4	2.8	2.3	4	39	33	4	1.6	1.4	1	16	-	1	1.4	-	1	<0.5	-
May	A	5	4.1	2.5	5	151	90	2	2.9	2.1	1	22	-	1	3.3	-	1	1.5	-
	B	5	2.9	2.5	5	80	66	5	1.8	1.4	1	18	-	1	2.4	-	1	2.7	-
June	A	4	4.3	1.6	4	61	48	2	1.4	1.3	1	20	-	1	2.1	-	1	<0.5	-
	B	4	4.5	2.6	4	51	43	3	1.2	1.0	1	16	-	1	2.6	-	1	<0.5	-
July	A	4	4.5	2.7	4	86	65	2	0.9	0.9	1	13	-	1	1.6	-	1	9.3	-
	B	4	4.8	2.7	4	237	108	4	1.7	1.1	1	21	-	1	1.9	-	1	2.9	-
August	A	5	4.4	2.7	5	67	48	2	1.8	1.4	1	19	-	1	6.8	-	1	1.1	-
	B	5	3.5	1.8	5	55	44	5	1.3	1.0	1	17	-	1	3.5	-	1	<0.5	-
September	A	3	3.3	2.7	3	96	59	2	1.2	1.2	1	13	-	1	4.3	-	1	<0.5	-
	B	3	3.1	2.2	3	55	48	4	1.3	1.1	1	18	-	1	3.9	-	1	<0.5	-
October	A	4	3.3	2.8	4	64	60	1	0.9	0.9	1	6.3	-	1	3.1	-	1	4.3	-
	B	4	2.7	2.2	4	55	50	4	1.0	0.8	1	8.2	-	1	<0.5	-	1	1.7	-
November	A	4	3.4	2.3	4	890	229	2	1.3	1.1	1	7.5	-	1	1.6	-	1	1.2	-
	B	4	1.6	1.4	4	72	60	4	1.7	1.0	1	9.6	-	1	<0.5	-	1	<0.5	-
December	A	2	1.9	1.9	2	69	57	1	1.2	-	1	11	-	1	0.6	-	0	-	-
	B	3	2.0	2.0	3	92	65	3	1.8	1.3	1	10	-	1	2.4	-	1	<0.5	-
Annual Summary	A	37	4.5	2.4	37	890	75	17	2.9	1.3	9	22	8	9	6.8	1.8	9	9.3	2.1
	B	38	4.8	2.3	38	237	59	38	1.8	1.1	9	21	9	9	3.9	1.3	10	2.9	1.1

[1]Location A is approximately 5 miles above mouth of Sawmill Creek. Location B is approximately 2.3 miles below mouth of Sawmill Creek. (See Figure 14.)

[2]Averages were calculated assuming concentration of one-half of minimum detectable concentration for samples in which given nuclide was not detected.

Table XX

RADIOACTIVITY IN DES PLAINES RIVER WATER, 1963

Month	Location[1]	Total Alpha Activity			Total Beta Activity			Uranium			Strontium-89			Strontium-90			Cesium-137			Thorium		
		No. of Samples	pCi / liter		No. of Samples	pCi / liter		No. of Samples	pCi / liter		No. of Samples	pCi / liter		No. of Samples	pCi / liter		No. of Samples	pCi / liter		No. of Samples	pCi / liter	
			Max.	Avg.		Max.	Avg.		Max.	Avg.		Max.	Avg.		Max.	Avg.		Max.	Avg.[2]		Max.	Avg.[2]
March	A	3	2.5	1.3	3	434	320	1	0.8	-	0	-	-	0	-	-	1	3.7	-	1	0.60	-
	B	3	1.9	1.5	3	427	286	3	0.9	0.7	0	-	-	0	-	-	1	6.1	-	1	0.40	-
April	A	4	2.2	1.7	4	280	206	2	1.0	1.0	1	13.7	-	1	6.0	-	1	1.9	-	1	0.10	-
	B	4	2.1	1.5	4	242	173	4	1.3	1.1	1	12.6	-	1	4.9	-	1	1.3	-	1	<0.05	-
May	A	5	3.4	2.5	5	125	107	2	1.8	1.4	1	47.0	-	1	6.0	-	1	1.3	-	1	<0.05	-
	B	5	4.5	2.4	5	215	142	5	1.7	1.2	1	45.5	-	1	6.9	-	1	1.0	-	1	<0.05	-
June	A	4	1.6	1.3	4	183	127	2	1.6	1.4	1	22.6	-	1	5.3	-	1	0.6	-	1	0.16	-
	B	4	2.6	1.8	4	164	140	4	1.5	1.3	1	27.8	-	1	7.8	-	1	1.1	-	1	0.09	-
July	A	5	3.9	2.9	5	215	148	2	1.4	1.1	1	24.4	-	1	7.8	-	1	1.3	-	1	0.10	-
	B	4	3.0	2.4	5	222	155	5	1.8	1.3	1	18.6	-	1	5.0	-	1	1.3	-	1	0.06	-
August	A	4	3.8	2.6	4	149	107	2	2.1	2.0	1	17.8	-	1	7.0	-	1	<0.5	-	1	<0.05	-
	B	4	4.8	3.0	4	108	94	4	2.0	1.7	1	16.0	-	1	9.2	-	1	<0.5	-	1	0.15	-
September	A	4	2.9	1.8	4	77	71	2	1.5	1.1	1	12.7	-	1	5.0	-	1	<0.5	-	1	<0.05	-
	B	4	2.4	1.5	4	66	63	4	1.7	1.3	1	11.9	-	1	4.9	-	1	<0.5	-	1	<0.05	-
October	A	5	1.6	0.9	5	71	59	2	1.8	1.3	1	7.2	-	1	4.8	-	1	<0.5	-	1	<0.05	-
	B	5	1.6	1.3	5	64	55	5	1.9	1.6	1	5.9	-	1	4.9	-	1	<0.5	-	1	0.13	-
November	A	4	3.2	2.1	4	73	56	2	1.5	1.5	1	2.1	-	1	2.8	-	1	<0.5	-	1	<0.05	-
	B	4	2.6	1.8	4	70	56	4	2.0	1.5	1	2.2	-	1	3.3	-	1	1.1	-	1	<0.05	-
December	A	2	1.7	1.6	2	56	49	1	1.5	-	1	3.6	-	1	3.2	-	0	-	-	1	<0.05	-
	B	2	1.9	1.7	2	60	50	2	1.6	1.6	1	2.2	-	1	3.3	-	0	-	-	1	<0.05	-
Annual Summary	A	40	3.9	1.9	40	434	125	18	1.8	1.3	9	47.0	16.8	9	7.8	5.3	9	3.7	1.1	10	0.60	0.11
	B	39	4.5	1.9	40	427	121	40	2.0	1.3	9	45.5	15.9	9	9.2	5.6	9	6.1	1.4	10	0.40	0.10

[1]Location A is approximately 5 miles above mouth of Sawmill Creek.
Location B is approximately 2.3 miles below mouth of Sawmill Creek. (See Figure 14.)

[2]Averages were calculated assuming concentration of one-half of minimum detectable concentration for those samples in which given nuclide was not detected.

The average concentrations shown in the tables were essentially the same at both locations, indicating that these activities were usually not present in Sawmill Creek in sufficiently high concentrations to be detected, after dilution, in the Des Plaines River below the mouth of the Creek. Significant differences in the total beta activities and in some fission-product concentrations at the two locations were found in some of the individual samples, but these differences occurred at random in both directions and were evidently the result of localized differences in the amount of fallout at the time of sampling. Either I^{131}, Ba^{140}, Co^{60}, and plutonium were not detected, or their concentrations were similar at both locations. Cobalt-58 was detected in the river below Sawmill Creek in October and November 1962 at a maximum concentration of 8 pCi/liter, or 0.008% of the MPC. The Co^{58} was found during a period when Sawmill Creek water samples contained from 500 to 3260 pCi of Co^{58} per liter, and must be attributed to Argonne waste water. During this period, the dilution factor for the creek in the Des Plaines River was of the order of 1000. The volume of water carried by both streams is quite variable, and this dilution factor cannot be applied at all times. This is the only instance during 1962 and 1963 when activity originating at Argonne could be positively identified off the site.

The average and range of uranium and total alpha activities during 1962 and 1963 were very similar to those found earlier, 1-2 pCi/liter for uranium and 1.5-3 pCi/liter for alpha activity, and were normal. The beta activities varied widely due to fallout. Natural beta activities range between 5 and 15 pCi/liter. Beta activities greater than this were due to fission-product fallout. Fallout levels during the last 2 years were at their highest levels since 1952, although they were still well below the MPC's. The variations in the fission-product concentrations during 1962 and 1963, and the ages of the fission-product mixtures were the same as those observed in air and precipitation. Maxima occurred during the spring and fall of 1962 and during the spring of 1963.

3. Other Waters

The total activities in other ponds and streams on the ANL site are shown in Table XXI. The alpha activities were in the range found at the same locations in other years. The alpha activities usually vary between 1.5 and 3 pCi/liter. Occasional samples contain as little as 0.5 pCi/liter and as much as approximately 6 pCi/liter. As noted in other years, the highest alpha activity among the natural ponds was found in a pond at location 11G in Figure 13. The activity is due primarily to uranium, and is probably of natural origin. The natural beta activities in these bodies of water are in the range of 5-10 pCi/liter. The additional beta activity during both years was due to fallout, and, as determined by fission-product analyses and gamma-ray spectrometry, consisted of the same fission products in the same ratios as those that were found in air and precipitation. As was the case for most other samples, fallout during the spring and summer of 1963 was at the highest level since 1952.

Table XXI

NONVOLATILE RADIOACTIVITY IN PONDS AND STREAMS ON ANL SITE, 1962-1963

Date Collected	No. of Samples	Alpha Activity (pCi/liter)		Beta Activity (pCi/liter)	
		Max.	Avg.	Max.	Avg.
April 11, 1962	4	3.4	2.6	55	46
May 24, 1962	2	2.5	1.6	24	22
May 31, 1962	2	3.0	2.9	85	74
July 30, 31, 1962	3	6.2	4.6	59	41
November 1, 1962	4	2.7	2.1	61	34
1962 Summary	15	6.2	2.8	85	42
March 27, 1963	4	4.9	2.1	247	141
April 26, 1963	1	2.7	-	248	-
May 15, 16, 1963	5	2.2	1.4	103	74
July 9, 1963	1	1.1	-	62	-
July 17, 1963	2	1.4	-	153	-
August 29, 1963	2	1.7	1.5	60	59
October 22, 1963	3	3.7	2.2	59	39
1963 Summary	18	4.9	1.9	248	99

Total activities in surface water collected within 25 miles of the Laboratory are listed in Tables XXII and XXIII and summarized in

Table XXII

NONVOLATILE RADIOACTIVITY IN SURFACE WATER NEAR ANL, 1962

(picocuries per liter)

Location	April 9, 10		May 23, 24		July 26-30		October 30, 31	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Des Plaines River, Brookfield	2.8	50	-	-	2.8	55	1.5	37
Des Plaines River, Romeoville	1.9	53	-	-	1.4	47	0.63	34
Illinois River, Morris	2.5	26	-	-	0.46	16	1.4	14
Confluence - Illinois, Kankakee, and Des Plaines Rivers	1.9	42	-	-	0.44	24	1.5	17
Du Page River, Naperville	2.9	52	-	-	3.8	24	-	-
Du Page River, Channahon	-	-	1.6	22	-	-	1.9	23
Flag Creek, German Church & Wolf Roads	-	-	1.2	92	-	-	0.62	24
Salt Creek, Wolf Road, Western Springs	-	-	4.1	77	-	-	3.5	56
McGinnis Slough, U.S. Rt. 45 and Ill. Rt. 7	1.5	81	-	-	-	-	5.8	119
Saganashkee Slough, 104th Ave. & Sag Canal	-	-	0.56	62	-	-	-	-
Sanitary & Ship Canal, Lemont	1.0	25	-	-	0.41	23	-	-
Fox River, Aurora	-	-	-	-	1.1	18	-	-
Cal-Sag Canal, 104th Ave.	1.5	53	-	-	-	-	1.1	17
Lake Michigan, 98th St., Chicago	-	-	0.10	8.0	0.11	16	0.53	9.7
Long Run Creek, 135th St. and Ill. Rt. 4A	-	-	1.7	23	-	-	3.2	20
Sauk Lake, 26th St., Park Forest	-	-	-	-	0.49	48	-	-
Average	1.8	40	1.5	47	1.2	30	2.0	34

Table XXIII

NONVOLATILE RADIOACTIVITY IN SURFACE WATER NEAR ANL, 1963

(picocuries per liter)

Location	March 26, 27		May 16		July 11		August 29		October 15	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Des Plaines River, Brookfield	-	-	-	-	-	-	0.94	96	-	-
Des Plaines River, Romeoville	-	-	1.8	100	-	-	1.9	85	-	-
Illinois River, Morris	1.9	67	-	-	-	-	-	-	2.2	17
Confluence - Illinois, Kankakee, and Des Plaines Rivers	1.7	91	-	-	1.5	19	1.7	27	1.7	15
Du Page River, Naperville	-	-	1.0	46	1.9	213	2.3	48	-	-
Du Page River, Channahon	1.5	88	-	-	-	-	1.7	39	-	-
Du Page River, Batavia Road, West Chicago	-	-	-	-	5.6	47	-	-	-	-
Flag Creek, German Church & Wolf Roads	1.2	129	-	-	-	-	-	-	-	-
Salt Creek, Wolf Road, Western Springs	-	-	-	-	-	-	2.6	131	-	-
McGinnis Slough, U.S. Rt. 45 and Ill. Rt. 7	-	-	-	-	1.8	105	-	-	-	-
Saganashkee Slough, 104th Ave. & Sag Canal	-	-	0.84	84	-	-	2.1	89	-	-
Sanitary & Ship Canal, Lemont	0.92	68	-	-	-	-	-	-	0.94	68
Fox River, Aurora	0.77	102	-	-	2.5	46	-	-	3.6	82
Lake Michigan, 98th St., Chicago	0.42	9.7	0.19	15	2.3	11	-	-	-	-
Sauk Lake, 26th St., Park Forest	2.2	221	1.2	90	-	-	-	-	5.6	64
Wolf Lake, 121st St. and U.S. Rt. 41	0.20	37	-	-	-	-	-	-	0.73	20
Average	1.2	90	1.0	67	2.6	73	1.9	74	2.5	44

Table XXIV. The alpha activities ranged from 0.1 to 5.8 pCi/liter and averaged 1.7 pCi/liter. In a given body of water, the alpha activity varies with the amount of solids, both soluble and insoluble, in the water, which in turn depends upon the water level. As usual, the lowest alpha (and beta) activities were found in Lake Michigan. The sample collected from Lake Michigan in July 1963 is abnormally high in alpha activity (2.3 pCi/liter). This sample contained considerable foreign material of unknown origin, but definitely not a normal lake constituent. The alpha activities in some of these water samples will be discussed further in connection with the results of bottom-silt samples (Part D below) from the same locations. Much of the beta activity in the water was due to fallout, and the variations with time corresponded to similar variations in air and precipitation. The natural beta activities at these locations vary from approximately 3 pCi/liter (in Lake Michigan) to 15 pCi/liter, depending on location and other variables. The average activities since 1952 have been above normal due to fallout, and have ranged from 9 pCi/liter in 1960 and 1961 to 43 pCi/liter in 1954. In 1962 and 1963, the averages were 37 and 72 pCi/liter, respectively.

The total activities in water from the reference sites are listed in Table XXV. The alpha activities were uniformly low and normal. The beta activities, naturally less than about 10 pCi/liter, showed the same

Table XXIV

NONVOLATILE RADIOACTIVITY IN SURFACE WATER NEAR ANL, 1962-1963

Date Collected	Distance from ANL (miles)	No. of Samples	Alpha Activity (pCi/liter)		Beta Activity (pCi/liter)	
			Max.	Avg.	Max.	Avg.
February 14, 1962	10	1	1.0	-	25	-
	25	1	0.33	-	4.3	-
April 9, 10, 1962	10	6	2.9	2.0	81	52
	25	3	2.5	2.2	42	28
May 23, 24, 1962	10	4	4.1	1.9	92	63
	25	2	1.6	0.84	22	15
July 26, 27, 1962	10	4	3.8	2.1	55	37
	25	5	1.1	0.53	48	24
October 30, 31, 1962	10	7	5.8	2.3	119	44
	25	4	1.9	1.3	23	16
1962 Summary	10	22	5.8	2.1	119	48
	25	15	2.5	1.1	48	20
	10 and 25	36	5.8	1.7	119	37
March 26, 27, 1963	10	2	1.2	1.1	129	99
	25	7	2.2	1.2	102	88
May 16, 1963	10	2	1.0	0.9	84	65
	25	3	1.8	1.1	100	68
July 11, 1963	10	2	1.9	1.9	213	159
	25	4	5.6	3.0	47	31
August 29, 1963	10	4	2.6	2.0	131	91
	25	3	1.9	1.8	85	50
October 15, 1963	10	1	0.9	-	68	-
	25	5	5.6	2.8	64	40
1963 Summary	10	11	2.6	1.5	213	98
	25	22	5.6	2.0	100	59
	10 and 25	33	5.6	1.8	213	72

Table XXV

NONVOLATILE RADIOACTIVITY IN SURFACE WATER FROM REFERENCE SITES, 1962-1963

(picocuries per liter)

Location	April 9-10, 1962		October 30-31, 1962		March 26-28, 1963		October 15-16, 1963	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Lake Delavan, Wisconsin	1.3	25	0.88	18	0.40	59	0.31	43
Fox River, Oak Point State Park, Wisconsin	0.85	44	1.5	17	0.68	109	0.33	15
Magician Lake, Michigan	0.26	12	0.54	53	0.14	150	1.0	20
Lake Michigan, St. Joseph, Michigan:								
Near shore	0.22	13	0.33	11	0.25	19	0.34	6.2
1500 ft from shore			0.19	16				
Illinois River, Starved Rock State Park, Illinois	2.3	32	2.5	12	3.0	82	1.9	16
Shafer Lake, Monticello, Indiana	2.0	21	1.2	11	2.6	85	1.4	5.9
Kankakee River, Kankakee River State Park, Indiana	0.69	27	1.4	102	1.3	97	1.8	16
Average	1.1	25	1.1	30	1.7	86	1.0	17

variations and increases due to fallout as did water collected on and near the ANL site. Fallout was present in all samples at all locations, but a decrease was noticeable in the fall of 1963.

D. Bottom Silt

Radioactivity in the beds of lakes and streams is of interest for several reasons. Where conditions are appropriate for the transfer of active materials from the water to the bed, plant and animal life may be exposed to higher radiation doses than expected from water activities alone. The bed, by concentrating activity, may show low-level stream contamination when water analyses do not. The bed can also retain activity for a considerable length of time and indicate water contamination that was undetected in the past, and thus give some information on previous contamination that might not otherwise be obtained for a moving stream of water.

The total activities in samples of bottom silt collected monthly from Sawmill Creek and the Des Plaines River are listed in Tables XXVI and XXVII. Although total activities in bottom silt vary considerably between locations, and between samples collected at different times at the same location, some generalizations can be made. Alpha activities in excess of 35 pCi/g and beta activities in excess of 90 pCi/g are abnormally high for this area. Samples composed largely of sand or organic matter contain less activity than samples composed largely of clay or loam. As a result, the concentrations of naturally-occurring activities vary by a factor of about five in this area. In some cases, specific analyses are required to determine the absence or presence of nonnatural activities.

In 1962, the alpha activities in Sawmill Creek and the Des Plaines River were normal at both sampling locations. All differences between locations were in the normal range and were due to small differences in the distribution of naturally-occurring radioactive elements. The beta activities fluctuated widely in the beds of both streams because of the widespread but variable occurrence of fallout. The total beta activities alone cannot be used to determine the origin of beta activity in the silt or to explain the differences in activity between samples collected at different locations from the same stream. However, from the rates of decay of the beta activities and from gamma-ray spectra it is evident that abnormally high values were due primarily to fallout. A few samples collected from Sawmill Creek below the outfall contained cobalt-58 and -60 originating at Argonne, but no evidence of activity originating at Argonne could be found in samples from the Des Plaines River below Sawmill Creek.

Similar results were obtained in 1963 (Table XXVII). The beta-decay rates and gamma-ray spectra indicated that fission products were present in all samples, that the relative amounts of fission products of different half-lives were generally the same at all locations, and that abnormally high beta activities, including the unusually high results from the

Des Plaines River, were due to fission products. The only exceptions were some of the below-outfall Sawmill Creek samples that contained cobalt-58 and -60. The concentrations of fission products from fallout were so large that the presence of fission products from Argonne waste water could not be established. However, if present, their concentrations were small compared to those from fallout.

Table XXVI

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM SAWMILL CREEK AND DES PLAINES RIVER, 1962

(picocuries per gram)

Date Collected	Location*	Sawmill Creek		Des Plaines River	
		Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
January 1	A	24	74	-	-
	B	25	56	-	-
April 4	A	20	71	24	183
	B	17	47	25	60
May 2	A	25	91	22	165
	B	16	57	26	81
June 6	A	29	275	24	131
	B	24	112	27	197
July 5	A	19	47	26	89
	B	20	97	26	208
August 1	A	23	226	29	134
	B	26	83	26	176
September 5	A	13	44	23	188
	B	23	115	26	165
October 3	A	18	45	21	60
	B	25	89	26	163
November 7	A	22	89	24	85
	B	29	85	25	206
December 5	A	20	52	15	43
	B	20	46	27	172
Average	A	21	101	23	120
	B	23	87	26	159

*Sawmill Creek locations were:
A - Above ANL Site (Location 13L in Figure 13).
B - Below waste-water outfall (Location 7M in Figure 13).

Des Plaines River locations were:
A - Willow Springs, approximately 5 miles above mouth of Sawmill Creek.
B - Lemont, approximately 2.3 miles below mouth of Sawmill Creek.
(See Figure 14.)

Table XXVII

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM SAWMILL CREEK AND DES PLAINES RIVER, 1963

(picocuries per gram)

Date Collected	Location*	Sawmill Creek		Des Plaines River	
		Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
January 2	A	18	44	-	-
	B	19	46	-	-
February 6	A	18	40	-	-
	B	33	92	-	-
March 6	A	26	53	-	-
	B	23	51	-	-
April 3	A	11	45	26	145
	B	24	96	30	138
May 1	A	25	91	20	199
	B	27	149	16	197
June 5	A	23	48	39	780
	B	31	119	27	435
July 3	A	21	98	22	149
	B	48	243	26	342
August 7	A	23	125	19	156
	B	29	173	28	556
September 4	A	23	150	14	86
	B	45	257	29	556
October 2	A	21	55	11	38
	B	45	77	29	275
November 6	A	14	42	19	112
	B	28	45	25	296
December 4	A	14	65	21	42
	B	33	53	23	203
Average	A	20	71	21	190
	B	32	117	26	333

*Sawmill Creek Locations were:
A - Above ANL site (Location 13L in Figure 13).
B - Below waste-water outfall (Location 7M in Figure 13).

Des Plaines River locations were:
A - Willow Springs, approximately 5 miles above mouth of Sawmill Creek.
B - Lemont, approximately 2.3 miles below mouth of Sawmill Creek.
(See Figure 14.)

The presence of alpha activity in Sawmill Creek bottom silt from ANL waste water is evident from the total activities in the July, September, and October, 1963 samples collected below the outfall. As shown in Table XXVIII, the same alpha emitters added to the creek in Argonne waste water were present in the bottom silt. The uranium content of bottom silt is normally in the range of 1-2 pCi/g, and plutonium concentrations are less than 0.1 pCi/g. The only location where larger values have been found, in 1963 and earlier, is in Sawmill Creek below the outfall.

Table XXVIII

RADIOACTIVITY IN SELECTED BOTTOM SILT SAMPLES, 1962-1963

(picocuries per gram)

Location	Date	Alpha	Beta	Uranium	Plutonium	Thorium	Cobalt-60	Strontium-90
Sawmill Creek, below outfall	November 7, 1962	29	85	-	-	-	13	<1
Sawmill Creek, 10 yards below outfall	July 3, 1963	48	243	5.3	4.0	<0.1	23	<1
Sawmill Creek, 40 yards below outfall	October 1, 1963	64	273	10.3	1.3	2.3	15	<1
Pond, ANL site, 4F	May 24, 1963	56	83	0.9	< 0.1	4.9	-	-
Pond, ANL site, 4F	July 30, 1962	100	167	1.1	<0.1	6.1	-	1.2
Pond, ANL site, 4F	March 27, 1963	35	74	-	<0.1	3.0	-	-
Pond, ANL site, 11K	November 1, 1962	34	50	1.6	-	-	-	<1
Fox River, Oak Point State Park, Wisconsin	April 9, 1962	57	67	2.0	<0.1	3.4	-	-
Flag Creek, Wolf Road	March 27, 1963	30	519	-	-	-	-	0.9
DuPage River, Naperville	July 27, 1962	60	196	1.1	<0.1	1.8	-	-
DuPage River, Naperville	July 11, 1963	55	1890	1.7	-	-	-	-
DuPage River, Batavia Road, West Chicago	July 11, 1963	49	156	1.6	<0.1	3.6	-	1.2
DuPage River, North Avenue and Fair Oaks Road	October 10, 1963	12	38	0.8	<0.1	0.55	-	-
DuPage River, Route 55 at Warrenville Road	October 10, 1963	129	209	2.1	<0.1	15.4	-	-

The monthly samples (Tables XXVI and XXVII) were collected about 10 yards below the outfall. A survey of the first 400 yards of the creek bed made in October 1963 gave results similar to those of the monthly samples. The total activities are presented in Table XXIX. It is seen that elevated

Table XXIX

NONVOLATILE RADIOACTIVITY IN BED OF SAWMILL CREEK, OCTOBER 1, 1963

Distance Downstream from Waste-water Outfall (yards)	Alpha Activity (pCi/g)	Beta Activity (pCi/g)
0	38	55
10	39	188
20	38	238
30	29	138
40	64	273
50	30	82
100	24	70
150	21	51
200	26	49
300	10	25
350	24	111
400	27	103

alpha and beta activities occurred during most of the first 50 yards, but that the maximum activity was at 40 yards. In the past, the location of the maximum, as well as the entire distribution pattern, has been quite variable. Maxima have occurred at locations from 0 to 150 yards from the outfall, and in some years several pronounced peaks, separated by distinct valleys, were found. In most cases, alpha and beta activities were both high at the same location since most abnormal activities in the creek bed resulted from contamination in ANL waste water. In a few samples during this reporting period and in the past, fallout produced abnormally high beta activities in samples that contained normal alpha activities. The excess alpha activity in the samples given in Table XXIX resulted from ANL waste water, while roughly 25-50% of the excess beta activity in the first 50 yards was due to the waste water and the remainder to fallout. The abnormal beta activities below 100 yards resulted from fallout. These distributions were determined from decay measurements and gamma-ray spectra. It is apparent that a quantitative measure of the extent, or absence, of contamination in a stream bed cannot be obtained by regular sampling at one location. It was impractical, however, to perform frequent surveys of the entire Creek bed below the outfall.

From 1952 to 1961, the annual average activity in the monthly samples below the outfall varied from 24 to 104 pCi/g for alpha activity and from 68 to 190 pCi/g for beta activity. In 1962 and 1963, the average alpha activities were in the lower portion of the previous ranges. Above the site, the alpha activities during 1962 and 1963 were very similar to those found earlier. At this location, the alpha activities have ranged from an average of 22 pCi/g in 1959 to 29 pCi/g in 1953. The beta activities above the site in 1962 and 1963 were elevated due to fallout, but were in the range of previous years, when the annual averages varied from 60 pCi/g in 1957 to 112 pCi/g in 1952.

The average alpha activities in the Des Plaines River at both locations were not significantly different from those found earlier, 16 to 26 pCi/g. The beta activities were generally higher than in other years, when the annual averages varied from 57 pCi/g in 1956 to 200 pCi/g in 1952. During 1963, the Des Plaines River bed contained the highest fallout level thus far detected. For example, the highest beta activity recorded in the river before 1963 was 369 pCi/g in June 1959. In 1963, five samples contained more than this value, and the average for the year, 333 pCi/g, was close to the previous maximum. The presence of fission-product fallout in these samples was confirmed by gamma-ray spectrometry and beta-decay measurements.

The total activities in the ponds and a few drainage ditches on the ANL site are shown in Table XXX. With the exception of samples from one pond at location 4F, the alpha activities were normal, ranging from 20 to 33 pCi/g. The beta activities varied considerably, from normal values of 50-90 pCi/g, to 400 pCi/g. With the exception of the pond at 4F, abnormal beta activities could be attributed to fallout. The total activities in the 4F pond varied from normal values of 25 and 75 pCi/g for alpha and beta activities, respectively, to 100 and 167 pCi/g for alpha and beta activities, respectively. As shown in Table XXVIII, the above-normal activities were due to thorium-232 and its decay products. The thorium content of water samples from the pond has been normal, indicating that thorium in the silt is very insoluble in this water. There is no indication that this thorium originated at ANL or is of other than natural occurrence.

Table XXX

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM PONDS AND STREAMS ON ANL SITE, 1962-1963

Date Collected	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
		Max.	Avg.	Max.	Avg.
April 11, 1962	3	33	25	64	59
May 24, 1962	4	56	35	83	68
July 30, 1962	3	100	51	167	96
November 1, 1962	4	47	35	55	50
1962 Summary	14	100	36	167	67
March 27, 1963	3	35	31	114	84
May 15 and 16, 1963	4	29	25	110	90
July 9 and 17, 1963	3	31	28	401	184
August 29, 1963	2	33	31	76	64
October 22, 1963	3	25	23	126	91
1963 Summary	15	35	27	401	104

The total activities in bottom silt collected within 25 miles of the ANL site (except for Des Plaines River samples in Tables XXVI and XXVII) are listed in Tables XXXI and XXXII and summarized in

Table XXXI

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT NEAR ANL, 1962

(picocuries per gram)

Location	April 10		May 23, 24		July 26, 27		October 31		November 1	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Des Plaines River, Brookfield	28	143	-	-	11	44	-	-	19	61
Des Plaines River, Romeoville	26	89	-	-	24	163	21	75	-	-
Illinois River, Morris	7	18	-	-	8	21	21	38	-	-
Confluence - Illinois, Kankakee, and Des Plaines Rivers	-	-	-	-	-	-	8	16	-	-
DuPage River, Naperville	18	41	-	-	60	196	-	-	-	-
DuPage River, Channahon	-	-	30	79	-	-	31	79	-	-
Flag Creek, German Church and Wolf Roads	-	-	31	168	-	-	-	-	25	90
Salt Creek, Wolf Road, Western Springs	-	-	28	57	-	-	-	-	12	30
McGinnis Slough, U.S. Rt. 45 and Ill. Rt. 7	29	49	-	-	-	-	10	31	-	-
Saganashkee Slough, 104th Ave. and Sag Canal	-	-	9	60	-	-	-	-	-	-
Fox River, Aurora	-	-	-	-	11	32	-	-	-	-
Lake Michigan, 98th St., Chicago	17	40	-	-	-	-	-	-	-	-
Long Run Creek, 135th St. and Ill. Rt. 4A	-	-	23	64	-	-	26	48	-	-
Sauk Lake, 26th St., Park Forest	-	-	-	-	22	60	-	-	-	-
Average	21	63	28	86	23	86	20	46	19	60

Table XXXII

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT NEAR ANL, 1963

(picocuries per gram)

Location	March		May		July		August		October	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Des Plaines River, Brookfield	-	-	-	-	-	-	17	44	-	-
Des Plaines River, Romeoville	-	-	23	606	-	-	23	200	-	-
Illinois River, Morris	12	30	-	-	-	-	-	-	5	28
DuPage River, Naperville	-	-	55	297	55	1890	31	416	14	30
DuPage River, Channahon	26	57	-	-	-	-	28	419	21	50
DuPage River, Batavia Road West Chicago	-	-	-	-	49	156	-	-	-	-
Flag Creek, German Church and Wolf Roads	30	519	-	-	-	-	-	-	-	-
McGinnis Slough, U.S. Rt. 45 and Ill. Rt. 7	-	-	-	-	21	85	20	63	-	-
Saganashkee Slough, 104th Ave. and Sag Canal	-	-	13	169	-	-	11	191	-	-
Fox River, Aurora	14	135	-	-	17	188	-	-	-	-
Sauk Lake, 26th St., Park Forest	19	310	19	79	-	-	-	-	22	140
Wolf Lake, 121st St. and U.S. Rt. 41	4	44	-	-	-	-	-	-	3	45
Average	17	182	28	288	36	580	22	222	13	59

Tables XXXIII and XXXIV. The alpha activities were normal at all locations except the DuPage River. The beta activities in the samples varied widely, and the sample collected from the DuPage River in July 1963 contained the highest beta activity (1890 pCi/g) found thus far, except for a lagoon on the ANL site formerly used for contaminated waste-water storage. The

Table XXXIII

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT NEAR ANL, 1962

Date Collected	Distance from ANL (miles)	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
			Max.	Avg.	Max.	Avg.
April 10	10	4	29	26	143	81
	25	2	17	12	40	29
May 23, 24	10	4	31	23	168	87
	25	1	30	-	79	-
July 26, 27	10	3	60	32	196	134
	25	3	22	14	60	38
October 31	10	3	26	19	75	51
	25	3	31	20	79	41
November 1	10	3	25	19	90	60
Annual Summary	10	17	60	24	196	83
	25	9	31	17	79	42
	10 and 25	26	60	22	196	69

Table XXXIV

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT NEAR ANL, 1963

Month	Distance from ANL (miles)	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
			Max.	Avg.	Max.	Avg.
March	10	1	30	-	519	-
	25	5	26	15	310	115
May	10	3	55	30	606	357
	25	1	19	-	79	-
July	10	2	55	38	1890	988
	25	2	49	33	188	172
August	10	4	31	21	419	218
	25	1	28	28	419	419
October	10	1	14	-	30	-
	25	4	22	13	140	66
Annual Summary	10	11	55	27	1890	406
	25	13	49	18	419	129
	10 and 25	24	55	22	1890	256

abnormally high beta activities were due to fallout, and during 1963, fallout at these locations was several times greater than during any previous year. Since these locations are sampled only at irregular intervals, comparisons between averages provide only a qualitative indication of relative fallout levels. However, the average beta activities at these locations have varied between 41 pCi/g in 1961 and 101 pCi/g in 1958.

Some, but not all, samples from the DuPage River contained up to four times the normal concentrations of alpha activity. Similar results have been obtained in previous years, and the excess activity was found to be due to thorium-232 and its decay products. Some of the above-normal beta activity in these samples was also due to the thorium, but in the presence of large amounts of fallout, as was the situation in 1962 and 1963, this effect on the total beta activity is not pronounced. To determine the extent to which this abnormal alpha activity occurs in the DuPage River, several locations between West Chicago and Channahon were sampled on the same day. The total activities are listed in Table XXXV in a downstream sequence. The total activities in water collected at the same time are also listed. As is usually the case, the activities varied considerably between locations. Abnormal alpha activities were found only at the second and third locations, although at other times high alpha activities have been found also at Naperville and Channahon. Thus, the occurrence of abnormal thorium concentrations in the bed of the DuPage River is widespread but variable. Additional sampling will be required to determine if abnormal alpha activities occur above West Chicago (Gary Mills Rd. and Route 30).

Table XXXV

NONVOLATILE RADIOACTIVITY IN DU PAGE RIVER, OCTOBER 10, 1963

(picocuries per gram)

Location	Bottom Silt		Water	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
North Ave. and Fair Oaks Rd.	13	38	1.0	27
Gary Mills Rd. and Route 30	66	310	1.2	51
Route 55 at Warrenville Rd.	129	321	-	-
Route 34, Naperville	14	30	3.3	22
Washington St. at 90th St.	16	46	-	-
135th St., Plainfield	21	75	1.7	31
Routes 52 and 59	25	250	-	-
Channahon, Illinois	21	50	0.8	27

The results of uranium, plutonium, and thorium analyses of some DuPage River bottom silt samples are given in Table XXVIII. The uranium and plutonium concentrations are normal in all samples, while elevated thorium concentrations are evident in some cases. The rate of growth of the alpha activity in the separated thorium fractions indicated the presence of thorium-232 and thorium-228 in approximately equal amounts. Some of the DuPage River water samples also contained elevated thorium concentrations, approximately 0.15 pCi/liter, several times the normal levels. The beta activities in both the bottom silt and water samples in Table XXXV were elevated due to fallout, and were well within the range found for samples collected from other bodies of water during October 1963. The presence of fission products in these samples was confirmed by gamma-ray spectrometry and radiochemical analyses.

The total activities in bottom silt collected from the reference sites are shown in Table XXXVI. The average activities as well as most of the

Table XXXVI

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT AT REFERENCE SITES, 1962-1963

(picocuries per gram)

Location	April 9, 10, 1962		October 30, 31, 1962		March 26, 28, 1963		October 15, 16, 1963	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Lake Delavan, Wisconsin	3.4	24	2.2	27	5.2	32	6.1	48
Fox River, Oak Point State Park, Wisconsin	57	67	11	30	7.9	43	15.3	48
Magician Lake, Michigan	2.6	22	5.1	41	4.2	46	4.2	32
Illinois River, Starved Rock State Park, Illinois	1.9	4.9	2.0	2.2	1.7	2.3	2.0	9.9
Lake Michigan, St. Joseph, Michigan	2.6	19	-	-	-	-	-	-
Shafer Lake, Monticello, Indiana	7.4	42	6.5	30	9.0	53	6.1	29
Kankakee River, Kankakee River State Park, Indiana	8.0	39	18	41	16.0	82	-	-
Average	12	31	7	29	7	43	7	33

individual results were in the ranges found earlier. These locations generally contain less natural activity than bottom silt from locations on and near the ANL site because of the sandy nature of the beds at the reference sites. Fallout was present in all samples, but in lower concentrations than in the samples collected near ANL. The alpha activity in the April 1962 sample from the Fox River was unusually high, 57 pCi/g. The alpha-to-beta activity ratio (neglecting about 30 pCi/g due to fallout) and the uranium and thorium concentrations (Table XXVIII) were typical of the high thorium-content samples found in the DuPage River. Such results have not been obtained at this location previously, and the Fox River bed in this area needs detailed examination.

E. Surface Soil

The natural activity in soil in the Chicago area is in the range of 10-40 and 40-85 pCi/g for alpha and beta activities, respectively. The natural alpha activity depends primarily on the natural uranium content. The natural beta activity depends, in addition, on the potassium content. The total natural activity in a particular soil sample is a function of its composition, with sandy soils containing less activity than soil consisting primarily of clay.

The total activities found in soil on the ANL site are listed in Table XXXVII. The alpha activities were normal at all locations except within 50 ft of a shed formerly used for the storage of active materials (location 12L in Figure 13). In this area, samples contained from 50 to 1625 pCi/g and from 200 to 2640 pCi/g for alpha and beta activities, respectively. The contamination was due primarily to uranium, but Co^{60}, in concentrations up to 550 pCi/g, and plutonium, in concentrations up to 0.3 pCi/g, were also found at a few locations. Fission products from fallout were also found in this area, as well as at all other locations sampled on the ANL site. Abnormal beta activities at all on-site locations except 12L are attributed to fallout, since they were due to fission products whose composition could not be distinguished from the fission-product mixtures found off the ANL site. Activity from ANL operations could not be detected in samples collected from the radioactive-waste storage area 7I, from the vicinity of the reactor buildings, and from the vicinity of Laboratory buildings in which large amounts of radioactive materials are used. Excluding the storage area at 12L, where traces of uranium contamination have been detected since 1956, the average annual alpha activities have varied in the past between 21 and 26 pCi/g. This is essentially the same as the average during 1962 and 1963, 25 pCi/g. Earlier average beta activities have ranged from 53 pCi/g in 1960 to 104 pCi/g in 1952. The former figure should be close to the natural beta activity since there was little fallout present in the environment during 1960. The averages during 1962 and 1963, 79 and 114 pCi/g, respectively, were elevated due to fallout, and except for 1952, are the highest thus far obtained.

Table XXXVII

NONVOLATILE RADIOACTIVITY IN SURFACE SOIL ON ANL SITE, 1962-1963

Date Collected	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
		Max.	Avg.	Max.	Avg.
January 5, 1962	4	27	23	59	56
April 11, 1962	3	32	27	111	94
May 24, 1962	2	24	22	129	92
May 31, 1962	2	25	23	167	112
June 1, 1962	6[a]	36	31	116	88
July 26, 1962	8[a]	34	21	94	66
July 31, 1962	7	32	23	100	79
	1[b]	176	-	228	-
November 1, 1962	5	33	25	111	81
	1[a]	28	-	72	-
1962 Summary	39	176[b]	28	228[b]	83
	38[c]	36	25	167	79
March 27, 1963	3	33	26	114	98
May 14, 1963	8[a]	1625	357	2640	666
July 12, 1963	6	31	27	354	212
	5[b]	51	31	168	102
August 29, 1963	6	27	24	143	99
November 29, 1963	17	31	25	164	88
1963 Summary	45	1625	85	2640	211
	32[c]	33	25	354	114

[a]Collected near radioactive-waste storage area 7I (Figure 13).

[b]Collected near former radioactive storage area 12L (Figure 13).

[c]Excluding samples collected near area 12L.

The total beta activities in soil collected near ANL are given in Table XXXVIII. The alpha activities varied between 11 and 39 pCi/g and averaged 21 pCi/g. These results are normal and similar to those found at these locations previously. As a result of fallout, the average beta activity, approximately 83 pCi/g in both 1962 and 1963, was significantly higher than in other years. The highest average beta activity previously found was 76 pCi/g in 1958. However, excluding the highest sample, 355 pCi/g, collected near Naperville in 1963, the range of beta activities was not significantly different in 1962 and 1963 than it was earlier. Although the beta activity was higher, on the average, on the site than off the site, the range of beta activities was the same both on and off the site, if the on-site storage area samples discussed earlier are excluded. The difference in the averages is merely a reflection of quantitative differences in the amount of fallout at each location at the time of sampling.

Table XXXVIII

NONVOLATILE RADIOACTIVITY IN SURFACE SOIL NEAR ANL, 1962-1963

Date Collected	Distance from ANL (miles)	No. of Samples	Alpha Activity (pCi/liter)		Beta Activity (pCi/liter)	
			Max.	Avg.	Max.	Avg.
April 9, 10, 1962	10	5	35	22	83	66
	25	3	25	21	135	73
May 23, 24, 1962	10	4	31	26	105	94
	25	2	20	17	185	117
July 26, 27, 30, 1962	10	3	23	16	118	68
	25	5	26	18	90	72
October 30, 31, November 1, 1962	10	4	29	22	77	63
	25	7	39	25	245	104
1962 Summary	10	16	35	22	118	73
	25	17	39	21	245	91
	10 and 25	33	39	22	245	82
March 27, 28, 1963	10	1	26	-	122	-
	25	7	21	18	102	78
May 16, 1963	10	2	25	23	102	87
	25	3	31	20	113	105
July 11, 1963	10	2	18	17	355	254
	25	4	25	20	61	48
August 29, 1963	10	5	20	20	63	59
	25	2	24	21	109	84
October 15, 1963	25	5	39	22	81	58
1963 Summary	10	10	26	21	355	110
	25	21	39	20	113	72
	10 and 25	31	39	20	355	84

The total activities in soil collected from the reference sites are given in Table XXXIX. The alpha activities varied from 9 to 24 pCi/g, and are normal for these locations. The average alpha activities are lower at

Table XXXIX

NONVOLATILE RADIOACTIVITY IN SURFACE SOIL AT REFERENCE SITES, 1962-1963

(picocuries per gram)

Location	April 9, 10, 1962		October 30, 31, 1962		March 26, 28, 1963		October 15, 16, 1963	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Delavan, Wisconsin	15	37	15	36	16	68	16	32
Oak Point State Park, Wisconsin	18	43	22	57	25	70	19	63
Magician Lake, Michigan	17	41	16	358	13	835	9	244
St. Joseph, Michigan	18	34	12	30	24	68	18	68
Starved Rock State Park, Illinois	16	33	21	89	14	51	24	34
Monticello, Indiana	12	69	16	50	13	52	7	27
Kankakee River State Park, Indiana	16	143	15	29	9	22	11	22
Average	16	57	17	93	16	167	15	70

the reference sites than on or near ANL because a larger fraction of the reference-site soils contain considerable sand. The same situation is true of the beta activities, but this effect is frequently obscured because of the presence of fallout. All reference-site samples contained some fallout, and some unusually high concentrations were noted at Magician Lake. These samples, because of the area from which they were collected, contained considerable decayed plant material, and the results apparently reflect the large amount of fallout activity that is frequently found in plants. The average beta activities at the reference sites has varied in the past from 32 pCi/g in 1961 to 76 pCi/g in 1952. The average during 1963 was considerably higher, primarily as a result of the large amount of fallout in the Magician Lake samples.

F. Plants

Plant collection was limited to grass because it was available at all locations and because comparison of results is more reliable if the same kind of plant is used. Total activities in grass are subject to wide fluctuations. Grass is a good collector of fallout and can accumulate large amounts of beta activity from this source, accentuating localized differences in fallout. The amount of natural alpha activity in grass also varies more widely than in soil, water, and other materials. An appreciable fraction of the natural activity is due to radium and Pb^{210}, and therefore, the alpha activity will increase with time after ashing due to the growth of radium decay products and Po^{210}.

The total activities in grass on the ANL site are listed in Table XL. The alpha activities were in the normal range except for some of the samples collected near the storage area at 12L. These samples contained from 4 to 15 pCi/g, compared to normal values of about 1 pCi/g. As for soil collected from this area, the abnormal activity was due to uranium. However, Co^{60} and abnormal amounts of plutonium and thorium were not found in the grass, although some amounts were detected in the soil. Excluding this storage area, the annual average alpha activity in the on-site grass samples in previous years has varied from 0.5 to 2.9 pCi/g. The averages during 1962 and 1963, 0.8 and 1.1 pCi/g, were similar. The beta activities were unusually high during 1962 and 1963, but were similar to those found off the site during the same period. The elevated beta activities were due to fission-product fallout, and no evidence of activity originating at Argonne could be found except near the storage area. The average beta activities on the site, 256 pCi/g in 1962 and 339 pCi/g in 1963, were appreciably higher than previous averages, which varied from 30 pCi/g in 1960 to 170 pCi/g in 1957.

Table XL

NONVOLATILE RADIOACTIVITY IN GRASS COLLECTED ON ANL SITE, 1962-1963

Date Collected	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
		Max.	Avg.	Max.	Avg.
January 5, 1962	4	1.4	1.0	489	390
April 11, 1962	3	1.4	0.8	599	464
May 24, 1962	2	0.32	0.24	174	119
May 31, 1962	2	0.81	0.67	356	303
July 31, 1962	8	2.4	1.0	312	197
November 14, 1962	6	0.95	0.72	327	260
1962 Summary	25	2.4	0.8	599	256
March 27, 1963	3	2.0	1.9	888	777
May 14, 1963	8[a]	14.7	5.3	785	400
July 12, 1963	11	2.4	1.1	1228	397
August 29, 1963	6	1.2	0.85	437	262
November 29, 1963	17	1.9	1.0	397	224
1963 Summary	37[b] 45	2.4 14.7	1.1 1.8	1228 1228	326 339

[a]Collected near former active storage area (12L in Figure 13).

[b]Average excluding samples collected from area 12L.

The total activities in grass obtained off the ANL site are given in Tables XLI and XLII. The alpha activities were all normal except for a sample collected near Lake Michigan in March 1963 (6.7 pCi/g) and one collected at Starved Rock State Park in April 1962 (8.3 pCi/g). The alpha activity in these samples is being investigated. The beta activities were unusually high due to the presence of fallout activity, as may be seen by comparing them with other years. Near the ANL site, the annual averages in other years have varied from 32 to 183 pCi/g, and at the reference sites the averages have varied from 22 to 253 pCi/g. Fallout activity was generally highest in the spring at most locations, presumably since fallout in air was also at a maximum at these times.

Table XLI

NONVOLATILE RADIOACTIVITY IN GRASS NEAR ANL, 1962-1963

Date Collected	Distance from ANL (miles)	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
			Max.	Avg.	Max.	Avg.
April 10, 1962	10	1	0.84	-	450	-
	25	2	3.7	2.5	404	358
May 23, 24, 1962	10	4	0.82	0.33	163	95
	25	2	0.34	0.32	144	120
July 26, 27, 1962	10	5	1.1	0.8	172	117
	25	3	0.64	0.45	197	157
October 30, 31, November 1, 1962	10	6	0.91	0.65	279	190
	25	5	3.4	1.5	256	189
1962 Summary	10	16	1.1	0.6	450	227
	25	12	3.7	1.2	404	198
	10 and 25	28	3.7	0.9	450	215
March 26, 28, 1963	10	1	1.9	1.9	662	662
	25	6	6.7	2.4	1223	845
May 16, 1963	10	3	0.65	0.38	174	142
	25	2	0.35	0.33	286	247
July 11, 1963	10	3	1.4	0.69	344	227
	25	3	0.90	0.63	228	182
August 29, 1963	10	5	0.97	0.37	221	169
	25	2	2.1	1.4	159	134
October 15, 16, 1963	25	4	0.73	0.56	182	145
1963 Summary	10	12	1.9	0.58	662	218
	25	17	6.7	1.6	1223	409
	10 and 25	29	6.7	1.2	1223	330

Table XLII

NONVOLATILE RADIOACTIVITY IN GRASS FROM REFERENCE SITES, 1962-1963

(picocuries per gram)

Location	April 9, 10, 1962		October 30, 31, 1962		March 26, 28, 1963		October 15, 16, 1963	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Delavan, Wisconsin	1.5	815	0.85	181	1.0	1200	0.38	170
Oak Point State Park, Wisconsin	2.5	500	0.79	134	1.5	684	1.5	160
Magician Lake, Michigan	-	-	0.91	339	2.8	1390	0.43	119
St. Joseph, Michigan	1.5	698	1.0	348	3.8	982	0.58	123
Starved Rock State Park, Illinois	8.3	356	0.88	221	0.72	382	0.64	170
Kankakee River State Park, Indiana	1.3	525	0.65	236	2.1	1585	0.28	120
Monticello, Indiana	-	-	-	-	2.0	736	0.91	90
Average	3.1	608	0.8	242	2.0	995	0.7	136

The results of the analyses of some samples for specific elements and nuclides are shown in Table XLIII. The abnormally high uranium concentration near the storage area at location 12L is evident.

Table XLIII

RADIOACTIVITY IN SELECTED GRASS SAMPLES, 1962-1963

(picocuries per gram)

Location	Date	Alpha	Beta	Uranium	Plutonium	Thorium	Strontium-90
Starved Rock State Park	April 10, 1962	8.3	356	0.26	0.15	0.20	-
ANL Site, 12L	July 31, 1962	2.4	126	4.0	0.06	0.14	-
Starved Rock State Park	October 31, 1962	0.88	221	-	-	-	1.5
98th St., Lake Michigan	April 16, 1963	6.7	1223	0.63	0.10	0.17	-
ANL Site, 12L	May 22, 1963	14	266	19	0.09	0.03	-
Naperville	September 6, 1963	1.4	344	0.10	0.08	-	3.9

G. Milk

Samples of raw milk were collected monthly (except February) during 1962 from three dairy farms within 15 miles of the Argonne site. Beginning in January 1963, the collections were made from three dairies that only process milk produced in the area. The collections were made by the DuPage County (Illinois) Health Department. All samples were analyzed for Sr^{89} and Sr^{90}. In addition, some of the samples were analyzed for other fission products. The results, except for I^{131} and Ba^{140}, are summarized in Table XLIV. Iodine-131 was detected in May, June,

Table XLIV

FISSION PRODUCT CONCENTRATIONS IN MILK, 1962-1963

(picocuries per liter)

Date Collected	Strontium-89		Strontium-90		Barium-140		Cesium-137	
	Max.	Avg.	Max.	Avg.	Max.	Avg.	Max.	Avg.
January 22, 1962	<5	-	4.0	3.6	<5	-	-	-
March 14, 1962	<5	-	4.1	3.8	<5	-	-	-
April 26, 1962	<5	-	3.4	3.3	<5	-	-	-
May 29, 1962	105	78	11.8	9.8	-	-	-	-
June 28, 1962	58	50	10.8	9.1	-	-	-	-
July 24, 1962	90	65	11.3	9.5	-	-	-	-
August 20, 1962	23	17	5.6	4.9	16	6.8	-	-
September 28, 1962	62	38	5.1	4.3	54	36	-	-
October 30, 1962	19	17	10.3	8.9	48	26	-	-
November 28, 1962	23	17	10.4	8.1	-	-	-	-
December 27, 1962	12	9	7.2	6.4	<5	-	-	-
January 31, 1963	13	6.4	7.1	6.8	<5	-	125	62
February 27, 1963	8.0	4.8	8.2	6.4	<5	-	118	59
March 27, 1963	7.8	4.5	7.2	6.3	<5	-	-	-
April 29, 1963	15	9.1	9.2	8.3	<5	-	206	159
May 27, 1963	71	60	19	16	<5	-	213	190
June 26, 1963	66	63	18	15	<5	-	180	172
July 29, 1963	50	43	21	16	<5	-	170	151
August 28, 1963	23	17	17	15	<5	-	168	109
September 30, 1963	10	8.5	13	11	<5	-	88	76
October 16, 1963	-	-	11*	-	<5	-	246	210
November 26, 1963	7.3	5.5	13	12	<5	-	172	150
December 30, 1963	<5	<5	17	16	<5	-	170	159

*One sample only.

September, and October of 1962. The concentrations ranged from about 10 pCi/liter in May and June, to a maximum of 260 pCi/liter in September. The January, March, and July 1962 samples were not analyzed for I^{131}, and this nuclide was not detected in the remaining months, including all of 1963. Barium-140 was detected in concentrations greater than 5 pCi/liter only in August, September, and October 1962. The average concentrations were 6.8, 26, and 26 pCi/liter, respectively. Analyses for this nuclide were not made in May, June, and July 1962, and the concentrations were less than 5 pCi/liter in the remaining months.

In January 1963, collections were made from the dairies as well as the three farms used in 1962 to determine if valid comparisons between the 1962 and 1963 results could be made in view of the change in method of milk collection. The radiostrontium concentrations were similar in both cases. The average Sr^{89} and Sr^{90} concentrations in the dairy milk were 6.4 and 6.8 pCi/liter, respectively, compared with 5.9 and 7.0 pCi/liter in the farm milk.

As shown in Table XLIV, the concentrations of the shorter-lived fission products, Sr^{89} and Ba^{140}, were low early in 1962. However, Sr^{90} was readily detectable. The appearance of Sr^{89} in May 1962 and the increase in the Sr^{90} content were apparently the result of the resumption of pasture feeding and an increase in the amount of fallout in the environment. Pasture feeding had not begun at the time of the April sampling. The possible sources of the increased activity in the May samples were fallout from (1) vented continental U. S. tests, (2) U. S. atmospheric tests in the Pacific during April and May 1962, and (3) U. S. S. R. atmospheric tests conducted in the fall and winter of 1961. The Sr^{89}/Sr^{90} ratio in the May samples suggests an age of several months for the activity. Even if it is assumed that 90% of the Sr^{90} in May was produced in 1961, the Sr^{89}/Sr^{90} ratio is about 80, which corresponds to an age of about 2 months. Thus, it appears that the fallout in the May samples was due principally to a mixture of fission products produced by the U. S. Pacific tests in the spring of 1962 and by the U. S. S. R. tests in the fall and winter of 1961. If a significant fraction of the activity originated in the U. S. continental tests, a much larger Sr^{89}/Sr^{90} ratio would have been expected since fission products only a few days old would have been present. The low concentration of I^{131} in May, 10 pCi/liter, also indicates that very small amounts of fresh fission products from vented underground explosions were present.

The activity in milk generally followed the variations in fallout during the 1962 period of pasture feeding in spring, summer, and early fall. The milk activity did not respond to the increased amount of shorter-lived fission products in the environment during November and December 1962, and produced primarily in the tests conducted by the U. S. S. R. in the fall of 1962, since the feed used at this time had been grown earlier in the year.

A sharp increase in the Sr^{89}/Sr^{90} ratio, as well as an increase in the Sr^{90} concentration was noted beginning in May 1963. The value of the strontium ratio and the absence of Ba^{140} and I^{131} indicates that the fission products in the milk were produced earlier, in 1962. The increase in activity was apparently due to the resumption of pasture feeding together with a spring increase in the rate of fallout. The concentrations of the longer-lived fission products during November and December 1963 did not decrease as much in milk as in air and other samples, probably because the feed in use during this period was grown earlier in the year when fallout activity in the air was higher.

All the fission-product activity in the milk samples is attributed to fallout from nuclear detonations. There was no evidence that any of the activity originated at Argonne.