RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

ANL-7104
Health and Safety
(TID-4500, 47th Ed.)
AEC Research and
Development Report

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

ENVIRONMENTAL RADIOACTIVITY AT
ARGONNE NATIONAL LABORATORY

Report for 1964

by

J. Sedlet and F. S. Iwami

Experimental Work Done by:

C. D. Eldred	F. S. Iwami
N. W. Golchert	K. K. Kamalick
C. M. Greene, Jr.	D. G. Kaplan
J. Sedlet	

Industrial Hygiene and Safety Division

October 1965

Operated by The University of Chicago
under
Contract W-31-109-eng-38
with the
U. S. Atomic Energy Commission

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TABLE OF CONTENTS

	<u>Page</u>
I. SUMMARY	7
II. PROGRAMS AND PROCEDURES	14
III. RADIOACTIVITY IN ENVIRONMENTAL SAMPLES.	19
A. Air	19
B. Precipitation	23
C. Water	24
D. Bottom Silt.	35
E. Surface Soil	39
F. Plants	42
G. Milk	44
H. Site A Samples	45

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Average Alpha Activity in Weekly Air-filter Samples, 1953-1964.	8
2.	Average Beta Activity in Weekly Air-filter Samples, 1953-1964.	8
3.	Average Radioactivity in Surface Water, 1964	9
4.	Average Radioactivity in Bottom Silt, 1964	9
5.	Average Radioactivity in Soil and Plants, 1964.	9
6.	Average Radioactivity in Surface Water, 1952-1964	9
7.	Average Radioactivity in Bottom Silt, 1952-1964	10
8.	Average Radioactivity in Surface Soil, 1952-1964	10
9.	Average Radioactivity in Plants, 1953-1964.	11
10.	Sampling Locations on the Site of Argonne National Laboratory	15
11.	Sampling Locations near Argonne National Laboratory	16
12.	Sampling Locations at Site A	45

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.	Samples Collected in 1964	14
II.	Long-lived Alpha and Beta Activity in Weekly Air-filter Samples, 1964	20
III.	Long-lived Alpha and Beta Activity in 24-hr Air-filter Samples on ANL Site, 1964.	20
IV.	Principal Gamma-ray Emitters in Air-filter Samples, 1964.	21
V.	Average Annual Radioactivity in Air-filter Samples, 1953-1964.	21
VI.	Total Alpha and Beta Activity in Precipitation on ANL Site, 1964.	23
VII.	Fission-product Activity in Selected Precipitation Samples, 1964.	24
VIII.	Nonvolatile Alpha Activity in Sawmill Creek Water, 1964. . .	26
IX.	Beta Activity in Sawmill Creek Water, 1964	28
X.	Beta-emitting Nuclides in Below-outfall Sawmill Creek Water, 1964.	30
XI.	Radioactivity in Des Plaines River Water, 1964.	32
XII.	Nonvolatile Alpha and Beta Activity in Ponds and Streams on ANL Site, 1964.	33
XIII.	Nonvolatile Alpha and Beta Activity in Water from Lakes and Streams near ANL, 1964	33
XIV.	Nonvolatile Alpha and Beta Activity in Surface Water near ANL, 1964.	34
XV.	Nonvolatile Alpha and Beta Activity in Water from Reference Sites, 1964	34
XVI.	Nonvolatile Alpha and Beta Activity in Bottom Silt from Sawmill Creek and Des Plaines River, 1964	35
XVII.	Radioactivity in Selected Bottom-silt Samples, 1964.	36
XVIII.	Nonvolatile Alpha and Beta Activity in Sawmill Creek Bottom Silt, 1964	37
XIX.	Nonvolatile Alpha and Beta Activity in Bottom Silt from Other Ponds and Streams on ANL Site, 1964	37
XX.	Nonvolatile Alpha and Beta Activity in Bottom Silt near ANL, 1964.	38

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
XXI.	Summary of Nonvolatile Alpha and Beta Activity in Bottom Silt near ANL, 1964	38
XXII.	Nonvolatile Alpha and Beta Activity in Bottom Silt from Reference Sites, 1964	39
XXIII.	Nonvolatile Alpha and Beta Activity in Surface Soil on ANL Site, 1964.	40
XXIV.	Nonvolatile Alpha and Beta Activity in Surface Soil near ANL, 1964.	41
XXV.	Nonvolatile Alpha and Beta Activity in Surface Soil from Reference Sites, 1964	41
XXVI.	Nonvolatile Alpha and Beta Activity in Grass on ANL Site, 1964	42
XXVII.	Nonvolatile Alpha and Beta Activity in Grass near ANL, 1964.	43
XXVIII.	Nonvolatile Alpha and Beta Activity in Grass from Reference Sites, 1964	43
XXIX.	Fission-product Concentrations in Milk, 1964	44

ENVIRONMENTAL RADIOACTIVITY AT ARGONNE NATIONAL LABORATORY

Report for 1964

by

J. Sedlet and F. S. Iwami

I. SUMMARY

This report presents the results of the environmental monitoring program at Argonne National Laboratory during 1964. 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 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.

Air-filter samples were collected continuously on the Argonne site and at locations from 7 to 20 miles from the site. The activities were very similar both on and off the ANL site, indicating that little, if any, of the radionuclides in these samples originated at Argonne. Higher activities would be expected on the site if a significant fraction of the activity were released at Argonne. The average alpha activities have varied by only a factor of two since 1953, from an average of 0.0031 pCi/m^3 in 1961 to an average of 0.006 pCi/m^3 in 1953. In 1964, the averages were 0.0037 pCi/m^3 on the ANL site and 0.0038 pCi/m^3 off the site. The alpha activity was due primarily to naturally-occurring radionuclides. The beta activity remaining after the decay of the radon and thoron daughters has been almost entirely due to fallout from nuclear-test detonations, and consequently has varied widely, from an average of 0.3 pCi/m^3 in 1954 to 5.6 pCi/m^3 in 1963. In 1964, the average was 1.0 pCi/m^3 . These results are summarized in Figs. 1 and 2. Concentrations of some of the fission products in the air-filter samples are discussed in Section III-A.

Fig. 1. Average Alpha Activity in Weekly Air-filter Samples, 1953-1964

Fig. 2. Average Beta Activity in Weekly Air-filter Samples, 1953-1964

In 1964, fallout activity increased from 1.3 pCi/m³ in January to a maximum of approximately 2.5 pCi/m³ in May and June. This peak was the usual "spring maximum" in stratospheric fallout, which this year consisted of long-lived fission products produced before 1964. The activity decreased to 0.32 pCi/m³ in September, and then increased to 0.5 pCi/m³ in October, and 0.7 pCi/m³ in November. The increase in October and November was due to fallout from the Communist Chinese test detonation of October 16, and was accompanied by the appearance, for the first time in 1964, of short-lived fission products (such as Ba-La¹⁴⁰, Ru¹⁰³, and Zr-Nb⁹⁵). Fallout from this detonation was first detected at Argonne on October 24. It is estimated that this test added an average of 0.2 pCi/m³ to the beta activity in the last quarter of the year. The activity in precipitation was generally similar to that in the air samples with respect to the nuclides present, their origin, and the variation of concentration with time.

Iodine-131 from Argonne was detected in only one air sample on the site, at a concentration of 2 pCi/m³ (0.67% of the MPC*). Iodine-131 from Argonne operations was not detected off the site.

The average total activities in samples of water, material from the beds of lakes and streams (bottom silt), soil, and plants collected in 1964 are shown in Figs. 3 through 5. For comparison purposes, the results obtained from 1952 through 1964 are shown in Figs. 6 through 9. Radioactive

*The maximum permissible concentrations (MPC's) 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.

Fig. 3. Average Radioactivity in Surface Water, 1964

Fig. 4

Average Radioactivity in Bottom Silt, 1964

Fig. 5
Average Radioactivity in Soil and Plants, 1964

Fig. 6

Average Radioactivity in Surface Water, 1952-1964

Fig. 7. Average Radioactivity in Bottom Silt, 1952-1964

Fig. 8. Average Radioactivity in Surface Soil, 1952-1964

Fig. 9. Average Radioactivity
in Plants, 1953-1964

materials originating at Argonne, and leaving the Argonne site, were found only in Sawmill Creek below the waste-water outfall and, to a much smaller extent, in the Des Plaines River below the mouth of the creek. The principal radionuclide added to the creek in Argonne waste water was H^3 (tritium) in the form of tritiated water. This activity was released when the heat exchanger of the CP-5 reactor ruptured. The primary heavy water, containing a large concentration of tritium and a smaller, but substantial, concentration of mixed fission products, contaminated the secondary cooling water. The secondary water, containing 1.0×10^{10} pCi/liter of tritium and approximately 1.8×10^6 pCi/liter of mixed fission products, was diluted with large volumes of uncontaminated waste water and released to the creek slowly during July and August.

The average tritium concentration in Sawmill Creek for the year was 2.8×10^6 pCi/liter, equivalent to 93% of the MPC. The average concentrations of other beta-emitting nuclides added to the stream in Argonne waste water ranged from 2.7 pCi/liter for Co^{60} (0.0054% of the MPC) to 7 pCi/liter for I^{131} (2.3% of the MPC).

Small amounts of uranium, thorium, and plutonium were also added to the creek in Argonne waste water. The uranium concentration due to the waste water was 1.9 pCi/liter (0.005% of the MPC). The corresponding average plutonium and thorium concentrations were 0.09 (0.0018% of the MPC) and 0.08 pCi/liter (0.0072% of the MPC), respectively.

The natural alpha activity in the creek above the outfall, 1.5 pCi/liter, was normal and similar to previous years. The total nonvolatile beta activity in the creek was distributed approximately as follows: 5-10 pCi/liter of naturally-occurring activity, 10-15 pCi/liter from fallout (a decrease from 1963 by about a factor of three), and 35-40 pCi/liter due to Argonne operations. Most of the latter activity was added as a result of the rupture of the CP-5 heat exchanger.

Tritium was the only nuclide from this release that could be detected in the Des Plaines River. The average tritium concentration in the river below the mouth of the creek was 2.8×10^4 pCi/liter (0.93% of the MPC). The maximum concentration in any one sample was 5.3×10^5 (18% of the MPC). The dilution factor for Sawmill Creek in the Des Plaines River, based on tritium concentrations, was about 1000 during the period when the contaminated cooling water was being released. All other activities in the Des Plaines River, including the fission products released with the tritium, were present in the river at essentially the same concentration above and below the mouth of the creek. This is reasonable with a dilution factor of 1000. The uranium and alpha activities in the river, 1.5 and 1.9 pCi/liter, respectively, were the same above and below the creek, and were normal compared to the previous years. The total nonvolatile beta activity, about 36 pCi/liter at both locations, consisted of about 10 pCi/liter of natural activity, with the remainder due to fallout. Fallout activity was about one-fourth of the 1963 value.

The alpha activities in other lakes and streams in the Chicago area were in their normal range, 0.3 to 3.9 pCi/liter. The beta activities varied from normal values of 5-10 to 110 pCi/liter. Above-normal beta activities were due to fallout. Fallout activity in surface water decreased from an average of about 65 pCi/liter in 1963 to about 20 pCi/liter in 1964. Similar behavior was found for the samples collected 100 miles from the ANL site (the "reference site" samples). At these locations, the alpha activities were uniformly low and normal, averaging 1.1 pCi/liter, while the beta activities averaged 22 pCi/liter in April and 9 pCi/liter in October. The decrease from April to October was due to the decrease in stratospheric fallout noted in the air samples. This trend was generally observed in most samples of all types. Fallout from the Chinese test in October was not significant except in air and precipitation.

Radioactivity in bottom silt followed the expected behavior at most locations. Abnormal concentrations of many of the nuclides added to Sawmill Creek in Argonne waste water were found in bottom silt below the waste-water outfall. These included uranium, plutonium, Co^{60} , Cs^{137} , and Sr^{90} . Below the outfall, the alpha and beta activities averaged 50 and 219 pCi/g, respectively, compared to above-site averages of 16 and 46 pCi/g. Activity from the waste water was readily apparent in the bed of the creek up to 150 yards from the outfall. Activity from Argonne operations was not detected in any off-site bottom-silt samples, including those from the

Des Plaines River. At all locations off the Argonne site, except the Du Page River, the alpha activities were in the normal range, 2 to 40 pCi/g, depending on the chemical and physical nature of the sample. Some of the Du Page River samples contained several times the normal concentrations of alpha activity due to the presence of abnormal concentrations of Th^{232} and its decay product, Th^{228} . Fallout was present in bottom silt from all locations and increased the average beta activity from a normal value of about 50 to about 100 pCi/g.

The natural alpha and beta activity in surface soil in the Chicago area is approximately 10-40 and 40-90 pCi/g, respectively. Only one location on the Argonne site, near a radioactive storage area, contained abnormally high concentrations of activity. The contamination at this location was primarily due to uranium and Co^{60} . The beta activities at locations within 25 miles of the site were unusually high, averaging 92 pCi/g, when it is considered that the fallout in the samples was due to fission products produced before 1964. In 1962 and 1963, years of high fallout, the soil activities at the same locations averaged 83 pCi/g. At the 100-mile locations, the average beta activity in soil was 51 pCi/g, compared to 75 pCi/g in 1962 and 119 pCi/g in 1963. Fallout activity in soil was generally greatest in the spring, and decreased greatly in the fall.

The alpha activities in grass from all locations were normal. The average at all locations was approximately 1 pCi/g. The beta activities, although three to four times smaller than those in 1963, remained fairly high due to fallout. The normal beta activity is about 20 pCi/g; the 1964 average was about 175 pCi/g. Fallout activity was greatest in the spring and decreased substantially later in the year. Iodine-131 from Argonne operations was detected in grass on the ANL site only. The concentrations ranged from 13 to 25 pCi/g.

Three samples of milk produced in the area were collected monthly and analyzed for the fission products normally found in milk. Barium-140, Sr^{89} , and I^{131} were not detected. The average monthly concentrations of Sr^{90} ranged from 11 to 18 pCi/liter; the corresponding range for Cs^{137} was 110 to 300 pCi/liter. The highest concentrations were found in the spring and early summer. All the fission products in the milk were derived from fallout. There was no evidence that any of the activity originated at Argonne.

II. PROGRAM AND PROCEDURES

This report presents the results of the environmental monitoring program at Argonne National Laboratory for 1964. The principal purposes of the program are to measure the natural radioactive content of the ANL site and its surroundings, and to determine the magnitude, identity, and

TABLE I. Samples Collected in 1964

Type	Number
Water	432
Precipitation	81
Soil	108
Bottom silt	112
Plant	81
Air (filter and charcoal)	981
Milk	36

origin of any radioactivity above the natural levels. Of primary interest is the detection of any radioactive materials released to the environment by Argonne.

The radioactive content of the environment was determined by radiochemical analyses, total activity measurements, and gamma-ray spectrometric analyses of the type and numbers of samples listed in Table I. Since the most probable means of spreading radioactive contamination is through air and water,

most of the effort was directed toward collecting and analyzing these two media. Samples were collected from the ANL site and from locations up to 100 miles from the site. The frequency of sampling decreased with increasing distance from ANL. The sampling locations on, and within 25 miles of the site are shown in Figs. 10 and 11, respectively. Samples collected within 25 miles of the site (referred to as "near the site" in this report) are intended to show the extent and direction of contamination if significant amounts of radioactive materials are accidentally released to the environment by Argonne. It is expected that natural and fallout activity will be relatively uniform on, and within 25 miles of, the site, while activity released by Argonne will be present in higher concentrations on the site. Thus, higher concentrations on the site indicate a release by Argonne, while similar activity on and near the site would indicate that no significant release has occurred. Interpretations based on such considerations 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 and be based on proper sampling and analyses.

The samples collected 100 miles from ANL (referred to as "reference sites" in this report) were originally intended to serve 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 total beta activities and fission-product concentrations in these samples have served primarily to indicate the extent and magnitude of fallout activity. Since there are only small amounts of nonnatural alpha activity in the environment, the primary

purpose of the reference-site samples has been achieved for total alpha-activity measurements, but not for environmental plutonium concentrations. 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.

149-1466B

Fig. 10. Sampling Locations on the Site of Argonne National Laboratory

149-1125B

Fig. 11. Sampling Locations near Argonne National Laboratory

Total alpha and beta activities were measured by counting thick samples (5 to 75 mg/cm²) after a minimum of sample preparation. All such samples were mounted in 2-in.-dia, stainless-steel planchets. Counting rates were converted to disintegration rates as described below, and expressed in terms of picocuries (pCi) per cubic meter (m³), 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 remaining after evaporation 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 and ground in a mortar, and a weighed portion was spread on a counting planchet. Plant samples were washed with water to remove dirt, dried at 110°C, and ashed in a muffle furnace. A weighed portion of the ash 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/cm² thick.

Alpha counting of thick samples was done in 2.5-in.-dia, zinc sulfide, scintillation counters with backgrounds of 0.03-0.06 count/min and a counting efficiency of 32% for a 2-in.-dia, weightless alpha source. Thick samples were counted for beta activity in gold-plated, Mylar-window (0.9 mg/cm² thick), gas-flow, proportional counters, 2.25 in. in diameter, and installed in a lead and anticoincidence shield. The backgrounds of these counters were 3.5-4.0 counts/min, and the counting geometry was 25% for a 2-in.-dia source. 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 Pu²³⁹ (for alpha particles) and Tl²⁰⁴ (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 usually thick compared to the particle ranges and contained unknown and variable mixtures of radio-nuclides. 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, total activity was measured 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., cylindrical, 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 either the beta counter described above or in a similarly shielded and windowed counter, except that it was 2 in. in diameter and had a background of 0.7-0.8 count/min. Counting rates were converted to disintegration rates by applying correction factors experimentally determined for each nuclide.

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

Plutonium and thorium (including the Th^{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 gas-flow proportional counters with backgrounds of 0.02-0.05 count/min.

Tritium determinations in water were made by counting the distilled sample, in a dioxane-based scintillation solution, with a Tri-Carb liquid scintillation counter (Packard Instrument Co.). The counting efficiency was 20% and the background was 40 counts/min.

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, -6736, and -6945.

III. RADIOACTIVITY IN ENVIRONMENTAL SAMPLES

A. Air

Airborne particulate matter was sampled at seven locations on the Argonne site and at five locations off the site (Figs. 10 and 11). The 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 papers were 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 twice 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 samples were counted only 4 and 7 days after collection since they could not be obtained sufficiently early for the radon and thoron daughter measurement. The ratio of the 4th- and 7th-day beta activity was used to estimate the age of the fallout. The fallout activity in the on-site samples was estimated rapidly from the alpha-to-beta activity ratio, since this ratio is known for samples containing negligible concentrations of fallout. Gamma-ray spectra were obtained by combining and counting as one sample all the weekly samples collected each month from each location. The spectra were resolved by the usual "stripping" technique using standards containing known amounts of the gamma-ray emitters that were detected in the samples. The standard sources duplicated the samples closely in size, shape, and average atomic number.

The long-lived alpha and beta activities in the weekly and daily samples are given in Tables II and III. The concentrations of the principal gamma-ray emitters in the weekly samples are listed in Table IV. The similarity between the average activities on and off the site indicates that little, if any, of the radionuclides detected in the samples originated at Argonne. Higher activities would be expected on the site if a significant fraction of the activity originated at Argonne. Some individual samples differed from the monthly averages by up to a factor of three. Such differences occurred at random at all locations, and are to be expected since the amount of airborne activity depends on many variables, such as the amount of 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 after the decay of the short-lived radon and thoron daughters was due primarily to naturally-occurring alpha emitters present in dust in the air, and to Po^{210} from the decay of Rn^{222} in air. As shown in Table V, the average long-lived alpha activity has remained fairly constant, varying by only a factor of two, since sample collection was begun in 1953. There is a significant decrease in alpha activity from the 3rd to 4th day after collection due to the decay of residual thoron daughters.

TABLE II. Long-lived Alpha and Beta Activity in Weekly Air-filter Samples, 1964

Month	Location	No. of Samples	Alpha Activity (pCi/m ³)				Beta Activity (pCi/m ³)			
			After 4 Days*		After 7 Days*		After 4 Days*		After 7 Days*	
			Max.	Avg.	Max.	Avg.	Max.	Avg.	Max.	Avg.
January	On-site	29	0.0055	0.0037	0.0068	0.0032	1.5	0.9	1.4	0.9
	Off-site	24	0.0048	0.0032	0.0042	0.0029	1.4	0.9	1.4	0.9
February	On-site	24	0.0059	0.0036	0.0050	0.0036	1.6	1.0	1.5	1.0
	Off-site	19	0.0054	0.0038	0.0047	0.0033	1.4	1.0	1.4	1.0
March	On-site	26	0.0085	0.0044	0.0071	0.0039	2.4	1.4	2.4	1.4
	Off-site	19	0.0081	0.0045	0.0080	0.0042	2.6	1.4	2.5	1.4
April	On-site	24	0.0085	0.0053	0.0084	0.0042	2.5	1.6	2.5	1.5
	Off-site	20	0.0080	0.0048	0.0074	0.0042	2.8	1.5	2.8	1.5
May	On-site	26	0.0144	0.0060	0.0127	0.0051	3.3	2.1	3.1	2.0
	Off-site	22	0.0099	0.0059	0.0070	0.0053	3.0	2.2	2.9	2.1
June	On-site	24	0.0070	0.0052	0.0050	0.0039	3.0	1.9	3.0	1.8
	Off-site	20	0.0128	0.0066	0.0085	0.0040	3.4	2.2	3.4	2.1
July	On-site	30	0.0092	0.0046	0.0058	0.0032	1.9	1.0	1.8	1.0
	Off-site	23	0.0087	0.0053	0.0056	0.0038	1.8	1.1	1.7	1.0
August	On-site	24	0.0090	0.0045	0.0043	0.0028	0.83	0.59	0.86	0.59
	Off-site	22	0.0094	0.0049	0.0048	0.0033	0.88	0.64	0.88	0.63
September	On-site	27	0.0065	0.0040	0.0050	0.0027	0.46	0.33	0.47	0.32
	Off-site	23	0.0057	0.0037	0.0049	0.0027	0.50	0.32	0.50	0.32
October	On-site	23	0.0098	0.0058	0.0060	0.0037	1.0	0.4	0.97	0.39
	Off-site	22	0.0122	0.0060	0.0077	0.0038	0.80	0.41	0.73	0.39
November	On-site	24	0.0110	0.0058	0.0075	0.0043	1.4	0.6	1.3	0.5
	Off-site	20	0.0086	0.0049	0.0086	0.0040	1.3	0.5	1.2	0.5
December	On-site	26	0.0052	0.0035	0.0053	0.0037	0.29	0.20	0.29	0.20
	Off-site	22	0.0060	0.0037	0.0057	0.0043	0.35	0.21	0.33	0.21
Annual Summary	On-site	307	0.0144	0.0047	0.0127	0.0037	3.3	1.0	3.1	1.0
	Off-site	256	0.0128	0.0048	0.0086	0.0038	3.4	1.0	3.0	1.0

*Time after the end of the collection period.

TABLE III. Long-lived Alpha and Beta Activity in 24-hr Air-filter Samples on ANL Site, 1964

Month	No. of Samples	Alpha Activity (pCi/m ³)				Beta Activity (pCi/m ³)			
		After 4 Days*		After 7 Days*		After 4 Days*		After 7 Days*	
		Max.	Avg.	Max.	Avg.	Max.	Avg.	Max.	Avg.
January	31	0.014	0.008	0.010	0.005	2.5	1.3	2.2	1.3
February	29	0.013	0.006	0.0067	0.0038	2.0	1.3	2.0	1.2
March	31	0.012	0.007	0.0084	0.0045	3.6	1.8	3.6	1.8
April	30	0.014	0.009	0.0097	0.0043	3.6	1.9	3.6	1.9
May	31	0.027	0.012	0.011	0.005	4.1	2.6	4.1	2.5
June	30	0.024	0.013	0.0098	0.0054	6.1	2.5	6.0	2.4
July	31	0.031	0.015	0.0089	0.0044	2.3	1.2	2.3	1.2
August	31	0.027	0.013	0.0089	0.0042	2.1	0.7	1.8	0.7
September	30	0.024	0.011	0.013	0.004	0.85	0.42	0.84	0.42
October	31	0.051	0.019	0.019	0.005	1.9	0.6	1.6	0.5
November	30	0.036	0.019	0.012	0.006	2.3	0.8	2.1	0.7
December	31	0.014	0.005	0.11	0.005	0.43	0.25	0.41	0.25
Summary	366	0.051	0.011	0.019	0.006	6.1	1.3	6.0	1.2

*Time after the end of the collection period.

TABLE IV. Principal Gamma-ray Emitters in Air-filter Samples, 1964
(picocuries per cubic meter)

Month	Location	Sb ¹²⁵	Ba-La ¹⁴⁰	Ce ¹⁴⁴	Cs ¹³⁷	Ru ¹⁰³	Ru-Rh ¹⁰⁶	Zr-Nb ⁹⁵
January	On-site	0.02	<0.01	0.54	0.04	<0.05	0.24	0.09
	Off-site	0.02	<0.01	0.53	0.04	<0.05	0.24	0.08
February	On-site	0.02	<0.01	0.52	0.04	<0.05	0.21	0.07
	Off-site	0.02	<0.01	0.49	0.04	<0.05	0.24	0.07
March	On-site	0.04	<0.01	0.63	0.07	0.06	0.27	0.09
	Off-site	0.04	<0.01	0.72	0.07	0.06	0.28	0.09
April	On-site	0.05	<0.01	0.80	0.07	0.05	0.28	0.08
	Off-site	0.04	<0.01	0.69	0.08	0.05	0.23	0.08
May	On-site	0.06	<0.01	1.10	0.11	0.06	0.39	0.09
	Off-site	0.07	<0.01	0.99	0.11	0.08	0.32	0.09
June	On-site	0.06	<0.01	0.85	0.11	0.04	0.38	0.07
	Off-site	0.06	<0.01	1.06	0.10	0.03	0.43	0.07
July	On-site	0.03	<0.01	0.49	0.06	0.02	0.17	0.02
	Off-site	0.03	<0.01	0.48	0.06	0.02	0.19	0.02
August	On-site	0.02	<0.01	0.28	0.04	0.01	0.13	0.01
	Off-site	0.02	<0.01	0.30	0.04	0.01	0.11	0.01
September	On-site	0.01	<0.01	0.14	0.02	0.01	0.07	<0.01
	Off-site	0.01	<0.01	0.16	0.02	0.01	0.07	<0.01
October	On-site	0.01	0.08	0.13	0.02	0.02	0.06	0.01
	Off-site	0.01	0.09	0.14	0.02	0.02	0.06	0.01
November	On-site	0.01	0.07	0.10	0.02	0.03	0.03	0.02
	Off-site	0.01	0.08	0.11	0.02	0.03	0.03	0.02
December	On-site	0.01	0.01	0.08	0.01	0.01	0.03	0.04
	Off-site	0.01	0.01	0.09	0.01	0.01	0.03	0.04
Average	On-site	0.03	0.02	0.47	0.05	0.03	0.19	0.05
	Off-site	0.03	0.02	0.48	0.05	0.03	0.19	0.05

TABLE V. Average Annual Radioactivity in Air-filter Samples, 1953-1964
(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	-	-
1964	On-site	0.0047	0.0037	1.0	1.0	0.011	1.3
	Off-site	0.0048	0.0038	1.0	1.0	-	-

*Time after the end of the collection period.

From 1953 to 1960, the samples were counted 3 days after collection; beginning in 1960, the count was made after 4 days. Consequently, the difference between the 3-4-day and 7-day activities in Table V was greater 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 activity is much greater than the initial alpha activity.

In contrast to the relatively constant alpha activity, the airborne beta activity has varied greatly with time, since most of the beta activity remaining after the decay of the radon and thoron decay products has been due to fallout from nuclear test detonations. In 1964 (Tables II and III), the total beta activity as well as the concentrations of individual gamma-ray emitters (Table IV) increased from January to May and June, and then decreased rapidly until October. The peak in May and June was due to an increase in the rate of stratospheric fallout of long-lived activities (fission and neutron-activation products) from earlier nuclear detonations. This is the so-called "spring maximum" observed in other years. The decrease from June to October was to be expected in the absence of the injection of additional nuclear test products into the atmosphere. The October increase in the total beta activity was due to fission products from the Communist Chinese test detonation of October 16. Activity from this detonation was first detected on October 24. The increase in the concentrations of the short-lived fission products (Ba-La^{140} , Ru^{103} , and Zr-Nb^{95}) at the same times confirms the presence of recently-produced fission products (Table IV). Their concentrations were very similar at all locations, indicating that they originated in a widespread source, such as fallout, and not in a localized source such as Argonne. Fallout from the Chinese test was present from October 24 through the end of the year, although the average concentration of beta activity from this source was only about 0.2 pCi/m^3 . The average beta activity during 1964, about 1 pCi/m^3 , was the lowest since 1960 (Table V) since there was no large-scale, atmospheric nuclear testing in 1963 or 1964.

Continuous sampling for gaseous radioiodine with activated charcoal was conducted in the 300 area on the ANL site (Fig. 10) because of the possibility of iodine releases in the vicinity. Iodine-131 was detected in only one sample, collected from August 7 to 10. The concentration was 2 pCi/m^3 (if it is assumed that the I^{131} was present in the air at a constant concentration during the collection period), or 0.67% of the MPC of 300 pCi/m^3 . Surface deposition, as determined by the I^{131} content of grass, was confined to within 100 yards of the point of release, Building 310. Bromine-82, a short-lived, shielded-nuclide, fission product, was present in one charcoal sample collected from April 6 to 10 at a concentration of about 10 pCi/m^3 , again assuming a constant concentration during collection. The MPC for this nuclide is $4 \times 10^4 \text{ pCi/m}^3$. The bromine probably did not originate in fission since I^{131} , with a much higher fission yield, although less volatile than bromine, was not present.

The Br^{82} and I^{131} in the air originated at Argonne. None of the other activities detected in the air samples, charcoal, or particulate, could be attributed to Argonne.

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

The total alpha and beta activities are given in Table VI. The results can be interpreted qualitatively in the same manner as the air-filter activities. That is, the alpha activity was due to the short-lived radon and thoron daughters and to longer-lived naturally-occurring nuclides, as uranium and its daughters in dust and to Po^{210} from the decay of Rn^{222} in air. The beta activity remaining after the decay of the radon and thoron descendants was due almost entirely to fission products from nuclear detonations. The activity in precipitation cannot be expected to follow the air-filter activity quantitatively because precipitation is an intermittent sampler of the atmospheric activity. However, precipitation is a sensitive sampling device since it collects and concentrates activity from large volumes of air.

The beta activity showed a broad maximum in the spring, not as sharply defined as in the air-filter samples, and then decreased rapidly to October, when a sharp increase occurred due to fission products from the Chinese test

TABLE VI. Total Alpha and Beta Activity in Precipitation on ANL Site, 1964

Month	Days after Precip.	No. of Samples	Alpha Activity		Beta Activity		
			(pCi/liter)		(nCi/liter)		Total (mCi/sq mi)
			Max.	Avg.	Max.	Avg.	
January	1	0	-	-	-	-	-
	4	1	2.0	-	0.31	-	-
	7	1	1.6	-	0.31	-	4
February	1	5	45.1	16.6	1.24	0.41	-
	4	5	34.5	9.2	1.17	0.39	-
	7	5	26.8	8.1	1.10	0.38	20
March	1	9	53.4	15.8	1.32	0.78	-
	4	9	10.0	3.8	1.24	0.76	-
	7	9	10.0	3.7	1.23	0.76	124
April	1	12	56.0	11.0	1.24	0.58	-
	4	12	7.5	2.3	1.13	0.55	-
	7	12	7.5	2.3	1.07	0.55	154
May	1	5	60.2	22.8	0.85	0.59	-
	4	6	10.5	5.8	1.10	0.66	-
	7	6	8.5	4.9	1.10	0.65	94
June	1	10	36.7	9.4	1.62	0.53	-
	4	10	7.4	2.8	1.62	0.52	-
	7	10	7.4	2.7	1.62	0.52	86
July	1	10	19.3	7.8	0.82	0.44	-
	4	10	4.0	2.0	0.81	0.43	-
	7	10	3.5	1.8	0.80	0.43	68
August	1	4	54.0	24.0	0.39	0.33	-
	4	7	5.4	2.5	0.40	0.26	-
	7	7	4.7	2.3	0.40	0.26	29
September	1	7	15.8	4.5	0.27	0.13	-
	4	7	4.6	4.2	0.26	0.11	-
	7	7	4.6	4.0	0.23	0.11	24
October	1	3	204.0	82.9	2.94	1.14	-
	4	3	10.5	4.2	2.44	0.95	-
	7	3	10.0	4.0	2.15	0.85	8
November	1	4	23.4	13.4	1.22	0.49	-
	4	4	14.7	6.2	1.15	0.48	-
	7	4	14.0	6.4	1.09	0.46	28
December	1	5	3.0	1.9	0.23	0.11	-
	4	7	2.5	1.3	0.21	0.10	-
	7	7	2.2	1.3	0.21	0.10	8
Annual Summary	1	74	204.0	14.6	2.94	0.50	-
	4	81	34.5	3.9	2.44	0.46	-
	7	81	26.8	3.3	2.15	0.45	647

detonation. The first rainfall after the detonation, on October 29, contained this fallout, as did many of the precipitation samples in the remainder of the year. As shown in Table VII, Ba^{140} and Sr^{89} were absent, as expected, until after the Chinese detonation. The Sr^{90} concentration was highest in the spring, and declined by winter, indicating that most of this nuclide came from the stratospheric reservoir of fission products produced before 1964.

TABLE VII. Fission-product Activity in
Selected Precipitation Samples, 1964
(picocuries per liter)

Date	Total Beta Activity ($\times 10^3$)	Sr^{90}	Sr^{89}	Ba^{140}
January 24	0.31	14	<2	<2
February 12, 13	0.29	8	<2	<2
February 23	1.10	11	<2	<2
March 5	0.79	37	<2	<2
March 7, 8	1.21	32	<2	<2
March 19, 20	0.49	24	<2	<2
April 2	0.98	41	<2	<2
April 17	0.80	35	<2	<2
April 28	0.55	45	<2	<2
May 12	0.77	42	<2	<2
November 2	1.09	9	79	122
November 15, 16	0.28	3	3	9
December 10	0.083	7	<2	<2
December 11	0.09	6	<2	<2
December 28	0.15	7	<2	<2

The total beta activity carried down by precipitation in 1964, 647 mCi/mi^2 , was considerably less than the 2500 mCi/mi^2 carried down in each of the two preceding years, but was 10 times greater than in 1960, which was, as in 1964, free from widespread atmospheric nuclear testing. The years 1962 and 1963 were periods of very high fallout because of the extensive thermonuclear testing in 1961 and 1962.

C. Water

1. Sawmill Creek and Des Plaines River

Argonne waste water is discharged into Sawmill Creek (location 7M in Fig. 10), 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 Argonne

waste water. Samples were collected below the outfall three times weekly and combined prior to analysis. This was done to obtain the average activity in the weekly samples, since the activity below the outfall can vary greatly between samples, and it was impractical to analyze all the samples individually for all the nuclides of interest. 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, or in the absence of nuclear testing, the radioactivity above the outfall consists of natural activities that do not vary greatly with time, and of the longer-lived debris from nuclear testing, whose activity and fallout rate do not change rapidly 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.

The Argonne contribution to the creek of nuclides not present in the air can be obtained directly from the difference in concentration above and below the outfall, with a correction for dilution of the creek by waste water. When the nuclide is added to the stream below the outfall directly from the air as well as by upstream water, the Argonne contribution is more difficult to determine. This is the situation for fallout nuclides; to establish their origin below the outfall, information such as detonation dates, air and precipitation activities, and fission-product ratios are useful.

The concentrations of alpha-emitting nuclides found in the creek during 1964 are listed in Table VIII. The average alpha activity above the site, 1.5 pCi/liter, was normal and similar to that found previously. The higher average alpha activity below the outfall indicates that the waste water contained somewhat higher concentrations of alpha-emitting nuclides than natural creek water. Since creek water is diluted approximately in half by waste water, the natural alpha activity below the outfall is about 0.8 pCi/liter; therefore the Argonne contribution to the alpha activity was about 2.2 pCi/liter. This is less than was found earlier; in previous years, the annual average alpha activity added in Argonne waste water has varied from 3.2 to 60 pCi/liter. As in the past, most of the alpha activity added by the waste water was due to uranium. The uranium contribution, calculated on the same basis as alpha activity, amounted to 1.9 pCi/liter (0.005% of the MPC* for natural uranium). Most of the uranium appeared to have very nearly the natural isotopic abundance, although this was not specifically investigated because the concentrations were quite low. Before 1964, the annual average uranium contribution varied between approximately 2 and 45 pCi/liter.

*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 VIII. Nonvolatile Alpha Activity in Sawmill Creek Water, 1964

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	3.3	1.3	2	1.3	1.3	1	<0.05	-	1	<0.05	-
	7M	15	5.9	4.5	15	6.8	4.7	15	0.22	0.10	15	0.10	<0.05
February	13L	4	3.3	1.4	2	2.1	1.9	1	<0.05	-	1	<0.05	-
	7M	12	6.9	5.0	12	8.3	5.6	12	0.20	0.14	12	0.50	0.16
March	13L	4	2.4	1.5	2	2.5	2.3	1	<0.05	-	1	0.19	-
	7M	12	6.6	3.8	12	2.6	1.7	12	0.35	0.19	12	0.60	0.38
April	13L	5	3.1	2.1	2	1.8	1.5	1	<0.05	-	1	<0.05	-
	7M	12	5.2	3.4	12	2.6	2.0	12	0.08	0.05	12	0.39	0.14
May	13L	4	3.8	1.7	2	1.1	1.0	1	<0.05	-	1	<0.05	-
	7M	15	3.6	2.7	15	2.5	1.7	15	0.17	0.10	15	0.13	0.06
June	13L	4	1.3	1.0	2	0.9	0.9	1	<0.05	-	1	<0.05	-
	7M	12	4.5	2.9	12	4.4	2.5	12	<0.05	-	12	<0.05	-
July	13L	5	1.5	1.1	2	0.8	0.8	1	<0.05	-	1	<0.05	-
	7M	15	2.2	1.4	15	2.4	1.7	15	0.10	<0.05	15	<0.05	-
August	13L	4	1.0	0.8	2	1.8	1.2	1	<0.05	-	1	<0.05	-
	7M	12	2.4	1.4	12	1.8	1.6	12	0.08	<0.05	12	0.05	<0.05
September	13L	5	2.1	1.0	2	1.1	0.9	1	<0.05	-	1	<0.05	-
	7M	12	3.7	2.0	12	3.2	1.8	12	0.06	<0.05	12	0.05	<0.05
October	13L	4	2.9	1.9	2	2.1	1.6	1	<0.05	-	1	0.07	-
	7M	15	8.3	3.6	15	6.3	2.9	15	0.12	0.09	15	0.11	<0.05
November	13L	4	2.2	1.8	2	2.6	2.1	1	<0.05	-	1	<0.05	-
	7M	12	4.7	3.2	12	6.4	3.6	12	0.36	0.20	12	0.16	0.10
December	13L	3	2.7	2.2	1	2.4	-	1	<0.05	-	1	<0.05	-
	7M	15	4.1	2.4	15	3.6	2.3	12	0.18	0.08	12	0.05	<0.05
Annual Summary	13L	51	3.8	1.5	23	2.6	1.5	12	<0.05	-	12	0.19	<0.05
	7M	159	8.3	3.0	159	8.3	2.7	159	0.36	0.09	159	0.60	0.08

¹Location 13L is upstream from the ANL site. Location 7M is downstream from the waste-water outfall. (See Fig. 10.)

²Averages were calculated assuming a concentration of 0.025 pCi/liter, one-half of the minimum detectable concentrations, for samples in which plutonium or thorium was 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. These concentrations were converted to activities 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.

Plutonium in excess of the minimum detectable concentration (0.05 pCi/liter) was detected only below the outfall; thorium was found with

higher frequency and at a higher average concentration below, than above, the outfall. Consequently, these elements must have been added to the creek primarily in the waste water. The average and maximum plutonium concentrations below the outfall were 0.0018 and 0.0072% of the MPC, respectively; the corresponding values for thorium were 0.0004 and 0.0030% of the MPC. Thus, the amounts were very small in terms of both concentration and fraction of MPC. The isotopic composition of the thorium was not specifically investigated, but the growth of alpha activity in the separated thorium fractions suggests that the activity was due primarily to Th^{232} and its descendant, Th^{228} . The occurrence of plutonium and thorium in the creek was similar to that of previous years. The concentrations have always been low, but on the average higher below than above the outfall.

The beta activities found in the creek are listed in Table IX. The results were supplemented by gamma-ray spectrometry on many of the samples.

The beta activity in the creek was due to naturally-occurring radionuclides, fallout activities, and, below the outfall, to beta emitters in Argonne waste water. The natural beta activity in the creek varies between about 5 and 10 pCi/liter. The additional beta activity above the outfall, approximately 15-25 pCi/liter averaged over the entire year, was due to fallout. As indicated by the relative concentrations of the short- and long-lived fission products in Table IX, the fallout activity was almost entirely due to long-lived fission products produced before 1964. This was true also in October and November, when fallout from the Chinese test was present in air and rain. Evidently the concentrations of short-lived fission products were not sufficiently large at the time of sampling to be detected in the creek water. This is reasonable when the amounts of rain and creek water and fission-product concentrations in rain during this period are considered.

During the first 6 months of the year, the beta activity below the outfall was less than above the site, indicating that Argonne waste water contained less beta activity than above-site creek water. This same effect is shown also by the Sr^{90} content during some of these months. However, the Cs^{137} , Co^{58} , and Co^{60} concentrations indicate that during this period these nuclides were added to the creek by the waste water.

The large increase in total beta activity below the outfall in July, August, and September was due to fission products produced in the CP-5 reactor on the ANL site. A rupture in the heat exchanger caused the primary heavy water in the reactor to contaminate the secondary cooling water. The contaminated cooling water was disposed of by diluting it, in batches, with large volumes of ordinary, uncontaminated waste water, and processing the mixture through the regular waste-disposal system. As shown in Table IX, the most abundant activity in this water was tritium (as tritium oxide) produced primarily by an ($\underline{n}$, $\underline{\gamma}$) reaction on the deuterium of

TABLE IX. Beta Activity in Sawmill Creek Water, 1964

Month	Location ¹	Total Beta Activity			Sr ⁹⁰			Sr ⁸⁹			Ba ¹⁴⁰			Cs ¹³⁷			I ¹³¹			Co ⁵⁸			Co ⁶⁰			H ³		
		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		No. of Samples	pCi/liter		No. of Samples	pCi/liter	
		Max.	Avg. ²			Max.	Avg. ²		Max.	Avg. ²		Max.	Avg. ²		Max.	Avg. ²		Max.	Avg. ²		Max.	Avg. ²		Max.	Avg. ²		Max.	Avg. ²
January	13L	5	56	21	2	<0.5	-	2	<2	-	2	<2	-	1	<0.5	-	2	<3	-	1	<5	-	1	<1	-	0	-	-
	7M	15	19	16	15	1.9	0.9	15	<2	-	15	<2	-	15	4.9	1.5	15	<3	-	15	5.8	<5	15	2.2	1.2	0	-	-
February	13L	4	49	32	2	4.3	4.0	2	<2	-	2	<2	-	1	<0.5	-	2	<3	-	1	<5	-	1	<1	-	0	-	-
	7M	12	21	16	12	0.9	0.7	12	<2	-	12	<2	-	12	1.0	0.8	12	<3	-	12	62	16	12	29	8	0	-	-
March	13L	4	66	18	2	4.0	3.7	2	<2	-	2	<2	-	1	<0.5	-	1	<3	-	1	<5	-	1	<1	-	0	-	-
	7M	12	25	19	12	3.7	1.8	12	<2	-	12	<2	-	12	1.1	0.6	12	<3	-	12	<5	-	12	1.0	<1	0	-	-
April	13L	5	40	28	2	4.9	4.5	2	<2	-	2	<2	-	1	<0.5	-	2	<3	-	1	<5	-	1	<1	-	0	-	-
	7M	12	42	28	12	5.6	3.6	12	<2	-	12	<2	-	12	1.5	0.7	12	<3	-	12	<5	-	12	<1	-	0	-	-
May	13L	4	73	32	2	4.0	3.7	2	<2	-	2	<2	-	1	<0.5	-	2	<3	-	1	<5	-	1	<1	-	0	-	-
	7M	15	23	21	15	3.8	2.5	15	<2	-	15	<2	-	15	0.8	0.7	15	<3	-	15	<5	-	15	<1	-	0	-	-
June	13L	4	26	19	2	4.8	4.0	2	<2	-	2	<2	-	1	0.6	-	2	<3	-	1	<5	-	1	<1	-	0	-	-
	7M	12	24	18	12	3.8	2.6	12	<2	-	12	<2	-	12	0.9	0.6	12	<3	-	12	14	5	12	38	8	0	-	-
July	13L	5	34	26	2	2.9	2.6	2	<2	-	2	<2	-	1	<0.5	-	2	<3	-	1	<5	-	1	<1	-	1	<4 x 10 ³	-
	7M	15	593	177	23	14.0	5.1	23	755	190	22	76	23	20	22	8	19	129	27	15	7.6	6.2	15	3.6	2.8	23	1.7 x 10 ⁸	1.9 x 10 ⁷
August	13L	4	12	11	2	2.8	1.9	2	<2	-	2	<2	-	1	0.9	-	2	<3	-	1	<5	-	1	<1	-	0	-	-
	7M	12	754	222	14	6.8	5.0	14	855	185	14	22	8	12	4.1	2.2	13	85	18	12	<5	-	12	2.3	1.3	18	1.0 x 10 ⁸	1.4 x 10 ⁷
September	13L	5	32	16	2	1.2	1.0	2	<2	-	2	<2	-	1	<0.5	-	2	<3	-	1	<5	-	1	<1	-	0	-	-
	7M	12	218	93	12	3.3	1.7	12	29	9	12	<2	-	12	4.5	2.0	12	<3	-	12	<5	-	12	5.6	1.8	12	<4 x 10 ³	-
October	13L	4	20	13	2	1.5	1.4	2	<2	-	2	<2	-	1	1.6	-	1	<3	-	1	<5	-	1	<1	-	0	-	-
	7M	15	66	43	15	3.0	2.3	15	<2	-	15	<2	-	15	2.8	2.0	9	<3	-	15	<5	-	15	20	7	12	<4 x 10 ³	-
November	13L	4	17	13	2	2.3	2.2	2	<2	-	2	<2	-	1	<0.5	-	1	<3	-	1	<5	-	1	<1	-	0	-	-
	7M	12	29	21	12	1.8	1.4	12	<2	-	12	<2	-	12	1.6	1.1	12	<3	-	12	<5	-	12	<1	-	0	-	-
December	13L	3	24	18	1	3.6	-	1	<2	-	1	<2	-	1	<0.5	-	0	-	-	1	<5	-	1	<1	-	0	-	-
	7M	15	27	22	15	3.1	2.2	15	<2	-	15	<2	-	12	1.4	1.1	12	<3	-	12	<5	-	12	1.4	<1	0	-	-
Annual Summary	13L	51	73	22	23	4.9	2.7	23	<2	-	23	<2	-	12	1.6	<0.5	19	<3	-	12	<5	-	12	<1	-	1	<4 x 10 ³	-
	7M	159	754	58	169	14.0	2.6	169	855	43	169	76	5	161	22	2.1	155	129	7	156	62	<5	156	38	2.7	65	1.7 x 10 ⁸	1.1 x 10 ⁷ 0.28 x 10 ⁷

¹Location 13L is upstream from ANL site. Location 7M is downstream from waste-water outfall. (See Fig. 10.)²Averages were calculated assuming a concentration of one-half of the minimum detectable concentration for those samples in which a given nuclide was not detected.

the heavy water. The tritium does not appear in the total (nonvolatile) beta activity, since the samples were evaporated to dryness prior to total beta counting. Separate portions were analyzed specifically for tritium.

The annual average tritium concentration was computed in two ways. The average based only on the samples analyzed is 1.1×10^7 pCi/liter. A more realistic annual average is obtained by assuming that the concentration during months when analyses were not made was one-half of the minimum detectable amount, 4×10^3 pCi/liter, and using 12 monthly values in calculating the average. This, in effect, spreads the tritium detected in July and August over the entire year. This method yields the lower figure, 0.28×10^7 pCi/liter.

The contaminated cooling water itself, before dilution in the waste-treatment system, contained, as of July 1, 1964, the following concentrations of those nuclides listed in Table IX:

Nuclide	Concentration (pCi/liter)
H ³	1.0×10^{10}
Sr ⁹⁰	2.6×10^3
Sr ⁸⁹	1.4×10^5
Ba ¹⁴⁰	2.0×10^5
Cs ¹³⁷	4.1×10^3
I ¹³¹	1.1×10^5
Co ⁶⁰ , Co ⁵⁸	$< 10^2$
Total (nonvolatile) beta activity	1.8×10^6

The radiochemical as well as gamma-ray spectrometric analyses of this water indicated that essentially all the activity was due to tritium and fission products. Concentrations of neutron activation products were small compared to those of the fission products. The dilution obtained in the disposal operation can be seen from the data. The concentrations of the longer-lived nuclides, H³, Sr⁸⁹, Sr⁹⁰, and Cs¹³⁷ were reduced by a factor of about 500; the total beta activity, Ba¹⁴⁰, and I¹³¹ concentrations decreased by about 10^4 , the combined effect of dilution and decay. The total beta activity in October indicates the presence of about 35 pCi/liter in the creek from the waste water. The absence of Sr⁸⁹ and tritium shows that this activity was not from the CP-5 heat exchanger but originated elsewhere at Argonne. It consisted of Sr⁹⁰, Cs¹³⁷, and Co⁶⁰, among other long-lived beta emitters.

The total beta activity above the site, 22 pCi/liter, was in the range found earlier. Since 1952, this average has varied from 6 to 248 pCi/liter, as the amount of fallout varied. The average activity below the outfall,

58 pCi/liter, was similar to earlier values. The annual averages at this location have not varied as greatly as those above the site, and have ranged from 31 pCi/liter in 1961 to 90 pCi/liter in 1952. At both locations other common fission products, such as Zr-Nb⁹⁵, Ce¹⁴¹, Ce¹⁴⁴, and Ru¹⁰⁶, were detected at various times. As was the case this year, both Argonne and fallout activities have been detected below the outfall in all years since sample collection was begun in 1952.

As mentioned previously, the contribution to the creek by the waste water of nuclides present in the atmosphere in significant amounts cannot be computed directly from the above- and below-outfall concentrations, but can be estimated by comparing concentrations at both locations on the same day. The results indicate that except for Sr⁹⁰, the nuclides listed in Table IX were present in the creek below the outfall, primarily as a result of their presence in Argonne waste water. However, regardless of source, the total concentration of a nuclide must be considered when comparisons are made with the MPC's. The values in Table X were calculated on this basis, without regard to origin. The activity of Th²³⁴, a decay product of U²³⁸, given in the table was approximated by assuming it to be 60% of the uranium concentration in Table VIII. This figure is based on the measured self-absorption of the uranium-daughter beta particles, and on the usual state of equilibrium between the uranium and its daughters in the water.

TABLE X. Beta-emitting Nuclides in
Below-outfall Sawmill Creek Water, 1964

Nuclide	Concentration (pCi/liter)		Percent MPC	
	Max.	Avg.	Max.	Avg.
Ba ¹⁴⁰	76.	5.	0.25	0.016
Cs ¹³⁷	22	2	0.11	0.01
Co ⁵⁸	62	<5	0.062	<0.005
Co ⁶⁰	38	2.7	0.076	0.0054
H ³	1.7 x 10 ⁸	2.8 x 10 ⁶	5.7 x 10 ³	93
I ¹³¹	129	7	43	2.3
Sr ⁸⁹	855	43	29	1.4
Sr ⁹⁰	14	2.6	4.7	0.87
Th ²³⁴	5.0	1.6	0.025	0.008

2. Des Plaines River

Since Sawmill Creek empties into the Des Plaines River about 500 yards downstream from the waste-water outfall, the river was sampled weekly (except when the ice was very thick) above and below the mouth of

the creek to determine if the activity in the creek had any effect on the radioactivity in the river. All samples were analyzed for total (nonvolatile) beta activity. At least one sample each month was analyzed for the same nuclides sought for in Sawmill Creek water. None of the samples contained detectable amounts of Ba^{140} , Co^{58} , Co^{60} , I^{131} , or Sr^{89} . The results of the other analyses are given in Table XI.

The alpha, uranium, and thorium activities were uniformly low and in the normal range for this location. The beta activities were elevated due to stratospheric fallout from nuclear detonations before 1964. As was the case for Sawmill Creek, fallout from the Chinese test was not detected. The natural beta activity in the river varies between 5 and 15 pCi/liter, depending on the amount of potassium and solid material in the sample. The decrease from the 1963 average, 125 pCi/liter, to 36 pCi/liter in 1964 was significant, and reasonable in the absence of widespread nuclear testing. The total (nonvolatile) beta activity, uranium, thorium, Sr^{90} , and Cs^{137} concentrations were quite similar at both locations, indicating that Sawmill Creek did not have a significant effect on the concentrations of these activities in the Des Plaines River. This could be due to a large dilution factor, or to very similar concentrations in the creek and the river. This was true also during July, August, and September when relatively large amounts of fission products were present in the creek from the failure of the CP-5 heat exchanger. The tritium concentrations from this accident were sufficiently large to be detected in some Des Plaines River samples collected below the creek in July and August, as shown in Table XI. A comparison of the tritium concentrations in Tables IX and XI indicates a dilution factor of about 100 for the creek into the river. However, this comparison is not valid because it is being made between samples collected at unrelated times. Samples were collected while the contaminated water was being discharged specifically to obtain dilution factors, and these samples show a dilution factor of about 1000. With this dilution factor, the detection of fission products from Sawmill Creek in the river would not be expected.

The average tritium concentration for the year was calculated by the same two methods described previously for Sawmill Creek. The lower value, 2.8×10^4 pCi/liter, is equivalent to 0.93% of the MPC. Strontium-90, attributed primarily to fallout, was also present at a concentration of about 1% of its MPC.

3. Other Waters

Total alpha and beta activities in water from other streams and ponds on the ANL site are given in Table XII. The alpha activities ranged from 0.4 to 2.4 pCi/liter, normal values for these locations. The beta activities varied with the potassium content, the amount of solids, and the fallout rate. Fallout, and therefore total beta activity, was greatest in the March samples and decreased later in the year. The natural beta

TABLE XI. Radioactivity in Des Plaines River Water, 1964

Month	Location ¹	Total Alpha Activity			Total Beta Activity			Uranium			Sr ⁹⁰			Cs ¹³⁷			H ³			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. ²		Max.	Avg. ²		Max.	Avg. ²
January	A	3	2.8	1.8	3	89	66	2	2.0	1.8	1	3.1	-	1	2.5	-	0	-	-	1	<0.05	-
	B	3	2.9	2.0	3	71	57	3	2.0	1.7	1	3.5	-	1	0.9	-	0	-	-	1	<0.05	-
February	A	3	2.0	1.9	3	79	58	2	1.6	1.5	1	3.1	-	1	2.0	-	0	-	-	1	<0.05	-
	B	4	2.8	1.7	4	48	33	4	2.1	2.0	1	4.2	-	1	1.4	-	0	-	-	1	0.09	-
March	A	4	2.1	1.6	4	64	42	2	2.1	1.4	1	1.9	-	1	<0.5	-	0	-	-	1	<0.05	-
	B	4	2.1	1.6	4	67	43	4	1.7	1.5	1	2.0	-	1	<0.5	-	0	-	-	1	<0.05	-
April	A	5	2.2	1.5	5	41	39	2	1.7	1.7	1	4.8	-	1	<0.5	-	0	-	-	1	0.07	-
	B	5	2.7	2.0	5	55	40	5	2.1	1.3	1	4.7	-	1	<0.5	-	0	-	-	1	0.22	-
May	A	4	3.0	2.3	4	44	30	2	1.9	1.7	1	3.8	-	1	<0.5	-	0	-	-	1	0.14	-
	B	4	2.5	2.0	4	36	30	4	1.4	1.3	1	4.7	-	1	0.5	-	0	-	-	1	<0.05	-
June	A	4	1.9	1.3	4	49	31	2	1.2	1.0	1	4.5	-	1	0.6	-	0	-	-	1	<0.05	-
	B	4	1.6	1.1	4	49	38	4	2.6	1.5	1	4.4	-	1	0.6	-	0	-	-	1	<0.05	-
July	A	5	3.7	1.9	5	46	37	2	1.4	1.3	1	6.1	-	1	0.7	-	1	<4 x 10 ³	-	1	<0.05	-
	B	5	2.5	1.8	5	37	34	5	1.5	1.2	5	6.1	5.6	3	0.8	0.7	10	5.3 x 10 ⁵	1.9 x 10 ⁵	1	<0.05	-
August	A	3	2.1	2.0	3	41	36	2	1.8	1.7	1	5.2	-	1	1.0	-	1	<4 x 10 ³	-	1	0.07	-
	B	4	3.9	2.1	4	34	29	4	2.0	1.5	2	5.2	5.1	1	0.8	-	4	3.0 x 10 ⁵	1.2 x 10 ⁵	1	<0.05	-
September	A	5	2.6	1.8	5	36	29	2	1.7	1.5	1	4.1	-	1	0.8	-	0	-	-	1	<0.05	-
	B	5	3.1	2.3	5	40	31	4	1.6	1.2	1	4.3	-	2	1.2	0.8	2	<4 x 10 ³	-	1	<0.05	-
October	A	4	3.4	3.2	4	34	31	2	2.0	1.9	1	3.6	-	1	<0.5	-	0	-	-	1	<0.05	-
	B	4	2.3	1.9	4	31	26	4	2.0	1.8	1	4.3	-	1	<0.5	-	2	<4 x 10 ³	-	1	<0.05	-
November	A	4	2.5	2.0	4	59	38	2	2.5	1.7	1	2.3	-	1	<0.5	-	0	-	-	1	0.05	-
	B	4	2.6	1.6	4	31	29	4	2.4	1.6	1	2.3	-	1	<0.5	-	2	<4 x 10 ³	-	0	-	-
December	A	2	2.3	2.3	2	22	23	1	2.1	-	0	-	-	1	0.5	-	0	-	-	0	-	-
	B	5	3.1	2.1	5	21	27	5	2.3	2.0	0	-	-	1	<0.5	-	2	<4 x 10 ³	-	1	<0.05	-
Annual Summary	A	46	3.7	1.9	46	89	38	23	2.5	1.5	11	6.7	3.9	12	2.5	0.8	2	<4 x 10 ³	-	11	0.14	<0.05
	B	51	3.9	1.9	51	71	34	50	2.6	1.5	16	6.1	4.5	15	1.4	0.6	22	5.3 x 10 ⁵	1.2 x 10 ⁵ 0.28 x 10 ⁵	11	0.22	0.05

¹Location A is approximately 5 miles above the mouth of Sawmill Creek. Location B is approximately 2.3 miles below the mouth of Sawmill Creek. (See Fig. 11.)

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

TABLE XII. Nonvolatile Alpha and Beta Activity in Ponds and Streams on ANL Site, 1964

Date Collected	No. of Samples	Alpha Activity (pCi/liter)		Beta Activity (pCi/liter)	
		Max.	Avg.	Max.	Avg.
March 11	3	2.4	2.1	79	60
June 9	3	1.2	0.8	11	9
June 30	1	1.0	-	24	-
November 4	1	1.2	-	25	-
Summary	8	2.4	1.4	79	32

activity in surface water in the area varies between 5 and 15 pCi/liter. Greater concentrations were due to fallout. In previous years, the average beta activity has varied from 7 pCi/liter in 1960 to 120 pCi/liter in 1952 (excluding a storage lagoon formerly used for contaminated waste water), depending on the amount of fallout present. The alpha and beta activities found in surface water within 25 miles of the site are listed in Table XIII and summarized in Table XIV. The alpha activities were all in the normal range for these locations, while the beta activities varied as the amount of fallout varied. As found in other samples, fallout was greatest in the spring but was present at all times during the year. The range of beta activities in these samples was similar to that observed in other years.

TABLE XIII. Nonvolatile Alpha and Beta Activity in Water from Lakes and Streams Near ANL, 1964 (picocuries per liter)

Location	March 11		April 16, 17		June 29, 30		October 28		November 12	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Des Plaines River, Brookfield	-	-	-	-	1.2	29	-	-	-	-
Des Plaines River, Willow Springs*	1.6	42	1.5	39	1.3	31	3.2	31	2.0	38
Des Plaines River, Lemont*	1.6	43	2.0	40	1.1	38	1.9	26	1.6	29
Des Plaines River, Romeoville	1.7	111	-	-	-	-	2.5	22	-	-
Illinois River, Morris	-	-	2.7	23	-	-	1.5	7.8	-	-
Confluence--Des Plaines, Kankakee, and Illinois Rivers	-	-	3.9	22	1.8	16	1.9	8.6	-	-
Du Page River, West Chicago	-	-	-	-	-	-	-	-	1.0	14
Du Page River, Naperville	-	-	2.6	19	2.9	62	0.3	17	-	-
Du Page River, Channahon	-	-	-	-	-	-	0.2	11	-	-
Sanitary and Ship Canal, Lemont	-	-	-	-	0.4	28	-	-	-	-
Flag Creek, German Church and Wolf Roads	-	-	1.8	25	-	-	0.6	18	-	-
Salt Creek, Wolf Road, Western Springs	-	-	-	-	-	-	-	-	2.5	25
McGinnis Slough, U. S. Rt. 45 and Ill. Rt. 7	1.3	48	-	-	-	-	-	-	1.9	58
Saganashkee Slough, 104th Ave. and Sag Canal	-	-	-	-	0.5	28	-	-	-	-
Fox River, Aurora	-	-	0.7	21	1.6	18	0.9	16	-	-
Lake Michigan, 98th St., Chicago	0.3	6.3	-	-	-	-	0.9	5.4	-	-
Long Run Creek, 135th St. and Ill. Rt. 4A	-	-	-	-	-	-	-	-	1.5	35
Wolf Lake, 121st St. and U. S. Rt. 41	-	-	0.5	23	-	-	0.4	6.1	-	-
Sauk Lake, Park Forest	0.5	62	-	-	-	-	-	-	-	-
Average	1.2	52	2.0	27	1.4	31	1.3	15	1.8	33

*The results for these locations were taken from the average monthly results given in Table XI.

TABLE XIV. Nonvolatile Alpha and Beta Activity
in Surface Water near ANL, 1964

Month	Distance from ANL (miles)	No. of Samples	Alpha Activity (pCi/liter)		Beta Activity (pCi/liter)	
			Max.	Avg.	Max.	Avg.
March	10	4	1.7	1.6	111	61
	25	2	0.5	0.4	62	34
April	10	4	2.6	2.0	40	31
	25	4	3.9	2.0	23	22
June	10	5	2.9	1.2	62	37
	25	3	1.8	1.5	29	21
October	10	6	3.2	1.6	31	20
	25	5	1.9	0.9	16	9
November	10	5	2.5	1.9	58	37
	25	1	1.0	-	14	-
Annual Summary	10	24	3.2	1.6	111	36
	25	15	3.9	1.3	62	19
	10 and 25	39	3.9	1.5	111	29

Table XV gives the results obtained for reference-site samples. Again, the alpha activities were all normal; the beta activities showed the presence of fallout, although the amount decreased appreciably in October. Fallout from the Chinese test was not evident from the total beta activities. Some of the samples from the reference sites and the 10- and 25-mile locations were analyzed for uranium. The results ranged from 2.2 pCi/liter in the Du Page River and Illinois River at Starved Rock to 2.8 pCi/liter in the Illinois River at Morris. Although these results are slightly higher than the average concentrations in the Des Plaines River and Sawmill Creek, above site, they are within the range encountered in the latter streams this year and are normal for their locations.

TABLE XV. Nonvolatile Alpha and Beta Activity
in Water from Reference Sites, 1964
(picocuries per liter)

Location	April 16, 17		October 28, 29	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Lake Delavan, Wisconsin	0.4	10	0.8	13
Fox River, Oak Point State Park, Wisconsin	1.6	22	0.3	4.6
Magician Lake, Michigan	0.3	15	0.3	3.9
Lake Michigan, St. Joseph, Michigan	0.3	5.1	0.6	4.3
Illinois River, Starved Rock State Park, Illinois	2.7	30	2.2	11
Shafer Lake, Monticello, Indiana	1.9	11	0.8	5.0
Kankakee River, Kankakee River State Park, Indiana	1.0	59	1.4	21
Average	1.2	22	0.9	9

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.

Although total activities in bottom silt vary considerably between locations, and between samples collected at different times from the same location, some generalizations can be made. Alpha activities in excess of 40 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.

The total activities in samples of bottom silt collected monthly from Sawmill Creek and the Des Plaines River are listed in Table XVI.

TABLE XVI. Nonvolatile Alpha and Beta Activity in Bottom Silt from Sawmill Creek and Des Plaines River, 1964
(picocuries per gram)

Date Collected	Location*	Sawmill Creek		Des Plaines River	
		Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
January 7	A	17	53	-	-
	B	31	49	-	-
February 5	A	20	67	-	-
	B	44	83	-	-
March 4	A	19	49	27	163
	B	75	278	28	202
April 1	A	22	80	28	65
	B	55	173	26	313
May 6	A	17	55	23	52
	B	50	175	29	344
June 3	A	17	51	26	90
	B	35	157	27	236
July 1	A	12	43	19	119
	B	34	80	27	227
August 19	A	14	40	14	55
	B	103	465	21	172
September 2	A	16	58	26	82
	B	41	535	24	119
October 7	A	8.4	41	14	43
	B	21	83	23	149
November 4	A	12	20	17	71
	B	24	90	22	157
December 2	A	14	40	-	-
	B	38	463	-	-
Average	A	16	50	22	82
	B	46	219	25	213

*Sawmill Creek locations were:

A--Above ANL site (Location 13L in Fig. 10).

B--Below waste-water outfall (Location 7M in Fig. 10).

Des Plaines River locations were:

A--Willow Springs, approximately 5 miles above the mouth of Sawmill Creek.

B--Lemont, approximately 2.3 miles below the mouth of Sawmill Creek (see Fig. 11).

In Sawmill Creek, the alpha activity was three times greater, and the beta activity four times greater, below the outfall than above the site. All of the excess alpha activity and much of the excess beta activity are attributed to contamination added to the creek in Argonne waste water. The same long-lived nuclides found in below-outfall water at abnormally high concentrations (uranium, plutonium, Co^{60} , Cs^{137} , and Sr^{90}) were also present in the bottom silt in above-normal amounts, as shown in Table XVII. It is difficult to determine if the thorium concentrations below the outfall, 4.4 and 2.8 pCi/g, are abnormal. Past experience has shown that the natural thorium content is variable, and these concentrations are in the normal range. It is noteworthy that below-outfall samples were either abnormally high or normal in both alpha and beta activity. If most of the high activities below the outfall were due to fallout, samples with normal alpha activity and abnormally high beta activity should have been detected.

TABLE XVII. Radioactivity in Selected Bottom-silt Samples, 1964
(picocuries per gram)

Location	Date Collected	Alpha	Beta	Uranium	Plutonium	Thorium	Cs ¹³⁷	Co ⁶⁰	Sr ⁹⁰
Sawmill Creek, below outfall	August 19	103	465	25	2.1	4.4	15	39	-
Sawmill Creek, below outfall	September 2	41	535	9.0	1.7	2.8	6.3	-	1.3
Sawmill Creek, below outfall	December 2	38	463	6.5	-	-	-	28	1.5
Des Plaines River, below Sawmill Creek	May 6	29	344	2.4	<0.05	0.5	-	<1	-
Des Plaines River, below Sawmill Creek	October 7	23	149	1.9	-	-	1.3	-	1.5
Du Page River, Naperville	June 29	63	362	2.3	<0.05	7.5	-	-	-
Long Run Creek, 135th St.	December 15	32	103	2.0	-	-	1.4	-	0.38

In the Des Plaines River, the alpha activities were normal at both locations, while the beta activity averaged about 2.5 times higher below Sawmill Creek. On the basis of beta decay rates, gamma-ray spectra, and the results in Table XVII, the excess beta activity below Sawmill Creek resulted from fallout. Apparently the nature of the bed at that location permits a high rate of adsorption of the activity from the stream water. If the abnormal beta activity were due to contamination from Argonne waste water, high alpha, uranium, and Co^{60} concentrations would be expected, but were not found.

The differences between sampling locations in Sawmill Creek and the Des Plaines River discussed above have been found in the past. Samples below the outfall have usually contained detectable amounts of activity from Argonne waste water. The activity distribution in the bed below the outfall, however, has varied with time, and this behavior is reasonable since the physical and chemical nature of the bed itself varies with time and location. The activity in the bed, as a function of distance at two different times in 1964, is shown in Table XVIII. Activity from the waste water is apparent down to at least 150 yards below the outfall. The variability shown by the data has been found in all such surveys conducted in other years. The results indicate that a quantitative measure of the natural activity or radioactive 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.

TABLE XVIII. Nonvolatile Alpha and Beta Activity
in Sawmill Creek Bottom Silt, 1964
(picocuries per gram)

Distance Downstream from Waste-water Outfall (yards)	July 22		October 3	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
0	21	51	22	71
10	49	238	42	261
20	58	377	32	170
30			54	1940
40			29	170
50			54	2250
60	11	37		
75	46	294		
100			68	2460
150			57	1950
200			16	53
250			31	265
300			23	63
350			22	86
400			13	50

Total activities in other ponds and streams on the site are given in Table XIX. The average alpha activity is in the normal range, although two or three of the samples (collected on March 11, June 9, and November 4) contain slightly elevated concentrations. Similar results have been found in the past, and such samples have contained above-average concentrations of Th^{232} , probably of natural origin. All abnormally high beta activities were due to fallout.

TABLE XIX. Nonvolatile Alpha and Beta Activity in Bottom Silt
from Other Ponds and Streams on ANL Site, 1964

Date Collected	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
		Max.	Avg.	Max.	Avg.
February 25	2	32	22	151	127
March 11	3	35	28	69	61
June 9	1	39	-	74	-
June 11	1	28	-	109	-
November 4	1	46	-	66	-
Annual Summary	8	46	30	151	86

Total activities in bottom silt from lakes and streams near ANL are listed in Table XX and summarized in Table XXI. The alpha activities were generally in the average range for this region except for some of the Du Page River samples. This behavior has been found in other years, and, as was the case for the Du Page River sample analyzed for thorium this year (Table XVII), the abnormal alpha activity has always been due to Th^{232} and its decay product Th^{228} . The isotopic composition of the separated thorium fraction was established from the alpha growth rate. All abnormally high beta activities, including those in the Du Page River samples, were due to fallout. In general, the amount of fallout was greater earlier in the year, and decreased in the October and November samples. This was the pattern shown by stratospheric fallout in 1964. The relatively small amount of fallout from the Chinese test did not affect the total beta activities significantly.

TABLE XX. Nonvolatile Alpha and Beta Activity in Bottom Silt near ANL, 1964
(picocuries per gram)

Location	March 11		April 16, 17		June 29, 30		October 28		November 12	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Des Plaines River, Brookfield	-	-	-	-	22	82	-	-	-	-
Des Plaines River, Willow Springs*	27	163	28	65	19	119	14	43	17	71
Des Plaines River, Lemont*	28	202	26	313	27	227	23	149	22	157
Des Plaines River, Romeoville	25	207	-	-	-	-	31	121	-	-
Illinois River, Morris	-	-	11	28	-	-	7.2	23	-	-
Du Page River, West Chicago	-	-	-	-	-	-	-	-	55	69
Du Page River, Naperville	-	-	-	-	63	362	65	60	-	-
Du Page River, Channahon	-	-	-	-	-	-	37	160	-	-
Flag Creek, German Church and Wolf Roads	-	-	32	42	-	-	28	94	-	-
Salt Creek, Wolf Road, Western Springs	-	-	-	-	-	-	-	-	13	33
McGinnis Slough, U. S. Rt. 45 and Ill. Rt. 7	37	73	-	-	-	-	-	-	33	63
Saganashkee Slough, 104th Ave. and Sag Canal	-	-	-	-	13	66	-	-	-	-
Fox River, Aurora	-	-	16	69	20	207	6.8	36	-	-
Long Run Creek, 135th St. and Ill. Rt. 4A	-	-	-	-	-	-	-	-	32	103
Wolf Lake, 121st St. and U. S. Rt. 41	-	-	15	31	-	-	2.6	31	-	-
Sauk Lake, Park Forest	22	97	-	-	-	-	-	-	-	-
Average	28	148	21	91	27	177	24	80	29	83

*Samples from these locations are also listed in Table XVI, and were collected on the dates given in that table.

TABLE XXI. Summary of Nonvolatile Alpha and Beta Activity in Bottom Silt near ANL, 1964

Month	Distance from ANL (miles)	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
			Max.	Avg.	Max.	Avg.
March	10	4	37	29	207	161
	25	1	22	-	97	-
April	10	3	32	29	313	140
	25	3	16	14	69	43
June	10	5	63	29	362	171
	25	1	20	-	207	-
October	10	5	65	32	149	93
	25	4	37	13	160	63
November	10	5	33	23	157	85
	25	1	55	-	69	-
Annual Summary	10	22	65	28	362	127
	25	10	55	19	207	74
	10 and 25	32	65	25	362	110

Table XXII lists the results obtained for bottom-silt samples from the reference sites. Because the activity in the Illinois River samples were abnormally low, the averages were calculated with and without these samples. The alpha activities were similar to those found in other years, when the annual averages have ranged from 6 to 16 pCi/g. Some of the April samples contained large amounts of fallout, and the decrease in the October samples is apparent. The average beta activity at the reference sites varied from 32 pCi/g in 1961 to 119 pCi/g in 1963.

TABLE XXII. Nonvolatile Alpha and Beta Activity in
Bottom Silt from Reference Sites, 1964
(picocuries per gram)

Location	April 16, 17		October 28, 29	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Lake Delavan, Wisconsin	13	29	5.0	32
Fox River, Oak Point State Park, Wisconsin	23	52	14	30
Magician Lake, Michigan	6.8	36	5.8	25
Illinois River, Starved Rock State Park, Illinois	2.1	3.7	2.7	3.5
Shafer Lake, Monticello, Indiana	3.1	23	11	39
Kankakee River, Kankakee River State Park, Indiana	16	464	15	40
Average	11 12*	101 121*	8.9 10.2*	28 33*

*Excluding samples from Illinois River.

E. Surface Soil

The natural activity in soil in the Chicago area is in the range of 10-40 pCi of alpha activity and 40-90 pCi of beta activity per gram. 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 results of total activity measurements on soil collected on the site are given in Table XXIII. The average activities are in the normal

range. Due to fallout, some of the samples were abnormally high in beta activity. One sample collected on January 23 contained 38 pCi of alpha activity per gram, a value close to the upper limit of the normal range. This sample contained an above-average concentration of uranium, 3.8 pCi/g, compared to average values of 1-2 pCi/g. Since seven alpha particles are emitted in the U^{238} decay series, the uranium content accounts for the above-average alpha activity. The activity in this sample appears to be of natural origin. The above-average activity in the sample collected on November 4, however, is attributed to contamination, primarily uranium and Co^{60} , from the radioactive storage area in the vicinity. Similar results have been found in the past. Activity from Argonne operations could not be detected at any other location.

TABLE XXIII. Nonvolatile Alpha and Beta Activity
in Surface Soil on ANL Site, 1964

Date Collected	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
		Max.	Avg.	Max.	Avg.
January 23	2	38	29	85	81
February 25	3	26	20	282	133
March 11	4	30	26	100	70
June 9	1	23	-	82	-
June 25	14	32	26	93	67
June 30	1	29	-	120	-
November 4	3	39*	29	121*	88
November 12	4	25	23	85	73
Annual Summary	32	39	26	282	79

*Collected near active storage area.

Total activities in soil collected near ANL and from the reference sites are listed in Tables XXIV and XXV. The alpha activities were all in the normal range. All abnormally high beta activities were due to fallout. For comparison purposes, previous annual averages near ANL were 20 to 25 pCi/g for alpha activity, and 46 to 84 pCi/g for beta activity. At the reference sites, the alpha activities have varied between 13 and 18 pCi/g, and the beta activities between 32 and 119 pCi/g. Thus, the beta activities near ANL were unusually high compared to previous years. The abnormal beta activities were due to fallout. Since samples are collected only at intervals of several months near the site, it is difficult to make quantitative comparisons of the fallout content of soil in different years. Qualitatively, however, fallout activity in soil was substantial in spite of the absence of widespread atmospheric testing in 1963 and 1964. Two samples that contained 30 and 32 pCi/g of alpha activity contained 3.3 and 4.1 pCi/g of uranium, respectively. The latter sample also contained 1.9 pCi/g of thorium.

TABLE XXIV. Nonvolatile Alpha and Beta Activity in
Surface Soil near ANL, 1964

Date Collected	Distance from ANL (miles)	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
			Max.	Avg.	Max.	Avg.
January 29	10	2	29	27	237	163
March 12	10	2	26	24	177	127
	25	2	19	17	234	160
April 16, 17	10	2	33	26	56	52
	25	5	32	19	76	59
June 29, 30	10	3	26	20	209	158
	25	2	18	18	63	65
October 28, 29	10	3	29	24	76	68
	25	6	36	20	150	72
November 12	10	3	27	22	103	83
	25	1	22	-	78	-
Annual Summary	10	15	33	23	237	107
	25	16	36	19	234	78
	10 and 25	31	36	21	237	92

TABLE XXV. Nonvolatile Alpha and Beta Activity in
Surface Soil from Reference Sites, 1964
(picocuries per gram)

Location	April 16, 17		October 28, 29	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Lake Delavan, Wisconsin	16	51	15	44
Oak Point State Park, Wisconsin	23	51	21	65
St. Joseph, Michigan	12	40	12	25
Magician Lake, Michigan	15	41	10	62
Starved Rock State Park, Illinois	21	56	17	57
Monticello, Indiana	15	56	15	41
Kankakee River State Park, Indiana	12	76	15	52
Average	16	53	15	49

F. Plants

Plant sampling was limited to grass because it was available at all locations, and intercomparison of results is more reliable if the same type of plants are collected at all locations. Total activities were determined by counting ashed plant samples, and calculated in terms of the oven-dried plant. The results are given in Tables XXVI, XXVII, and XXVIII. The alpha activities were in the normal range at all locations. Alpha activity from Argonne operations could not be found in any of the samples, including those collected near an active storage area where contaminated plant samples have been found previously. Small amounts of I^{131} were found near one of the buildings on the site following an iodine release in August. The concentrations varied from 13 to 35 pCi/g, and the contamination was confined to the first hundred yards around the building. These samples are not included in Table XXVI. Except for this case, all abnormal beta activities were due to fallout. In general, the fallout activity was greatest in the spring and decreased in the fall. This is the pattern exhibited by stratospheric fallout. Fallout activity in grass during 1964 was substantial, but considerably less than that in 1963, when the highest levels to date were found. In 1963, the average beta activities were 326 pCi/g on the site, 330 pCi/g near ANL, and 566 pCi/g at the reference sites. This year the values were one-third to one-half less than during 1963.

TABLE XXVI. Nonvolatile Alpha and Beta Activity
in Grass on ANL Site, 1964

Date Collected	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
		Max.	Avg.	Max.	Avg.
January 23	2	1.3	1.1	273	225
March 11	4	2.1	1.6	278	255
June 9	1	2.0	-	46	-
June 25	13	2.2	1.2	289	149
June 30	1	0.53	-	98	-
August 12	4	1.0	0.7	112	87
November 4	3	2.5	1.7	69	65
November 12	4	2.1	1.3	72	70
Annual Summary	32	2.5	1.2	289	137

TABLE XXVII. Nonvolatile Alpha and Beta Activity in Grass near ANL, 1964

Date Collected	Distance from ANL (miles)	No. of Samples	Alpha Activity (pCi/g)		Beta Activity (pCi/g)	
			Max.	Avg.	Max.	Avg.
January 29	10	2	1.6	1.1	416	304
March 12	10	2	1.4	1.3	244	223
	25	2	1.8	1.8	335	328
April 16, 17	10	2	2.1	1.9	508	484
	25	5	2.9	1.9	430	318
June 29, 30	10	2	0.64	0.42	76	74
	25	2	0.87	0.82	144	122
October 28	10	3	0.52	0.46	52	48
	25	6	1.7	0.8	70	63
November 12	10	3	1.2	0.9	60	49
	25	1	1.8	-	59	-
Annual Summary	10	14	2.1	1.0	508	176
	25	16	2.9	1.3	430	183
	10 and 25	30	2.9	1.2	508	180

TABLE XXVIII. Nonvolatile Alpha and Beta Activity in Grass from Reference Sites, 1964

(picocuries per gram)

Location	April 16, 17		October 28, 29	
	Alpha Activity	Beta Activity	Alpha Activity	Beta Activity
Lake Delavan, Wisconsin	2.3	426	0.36	54
Oak Point State Park, Wisconsin	1.0	192	0.80	53
St. Joseph, Michigan	2.1	374	0.55	70
Magician Lake, Michigan	-	-	0.44	56
Starved Rock State Park, Illinois	1.0	216	0.96	64
Monticello, Indiana	1.3	440	0.36	50
Kankakee River State Park, Indiana	1.2	327	0.27	40
Average	1.5	329	0.53	55

G. Milk

Samples of raw milk were collected monthly from three dairies that process milk produced only from the area near Argonne and analyzed for several fission products. This modest sampling and analysis program is intended to provide a base for a more complete program if an important activity release occurs at Argonne. Thus far, only activity from nuclear-test detonations has been detected in these samples. There is no evidence that the fission products in the milk originated at Argonne.

Cesium-137 and Sr^{90} concentrations in the milk samples are listed in Table XXIX. Analyses were also made for Ba^{140} , Sr^{89} , and I^{131} , but these nuclides were not detected. Thus, the Cs^{137} and Sr^{90} can be attributed to stratospheric fallout of debris from earlier tests. Fallout from the Chinese test in October was not sufficiently great to be detected in the milk.

TABLE XXIX. Fission-product Concentrations in Milk, 1964
(picocuries per liter)

Date Collected	Cs^{137}		Sr^{90}		Date Collected	Cs^{137}		Sr^{90}	
	Max.	Avg.	Max.	Avg.		Max.	Avg.	Max.	Avg.
January 27	260	240	14	11	July 3	325	300	17	15
February 24	160	155	16	14	August 7	180	175	16	15
March 30	180	170	13	11	September 4	185	140	14	13
April 27	185	165	14	12	October 2	140	110	13	11
May 8	240	215	17	16	November 17	125	115	16	13
June 5	135	130	19	18	December 9	195	180	16	14

As was the case for the long-lived fission products in air, fallout activity in milk was generally higher in the spring and early summer than at other times during the year. The decrease observed in the air activities later in the year did not occur in milk, probably because the fallout content of the feed did not decrease. In the winter months, feed produced during a time of higher fallout rate is being consumed.

The concentrations in Table XXIX can be compared to the environmental MPC's for drinking water of 300 pCi/liter for Sr^{90} and 2×10^4 pCi/liter for Cs^{137} , although these MPC's were developed for activity releases by atomic-energy installations and not for fallout. Since these MPC's are based on a daily liquid intake of 2.2 liters, the consumption of 1 liter of milk per day would result in an average intake of 0.4% of the Cs^{137} and 2% of the Sr^{90} maximum permissible daily intakes.

H. Site A Samples

A former portion of Argonne National Laboratory, known as Site A, is located about 4 miles east of the present site. An area of about 1600 sq ft at Site A, Plot M in Fig. 12, was used for the burial of contaminated waste

material. In 1954, as much as possible of this waste was removed, and the area covered with a concrete cap, 1 ft thick and 8 ft deep along the sides, to prevent disturbance of the remaining radioactive material. The ground north and east of the plot slopes downhill, and an intermittent stream flows in a northerly direction around the plot. Before the plot was covered with concrete in 1954, samples of water and bottom silt were collected from locations 1 through 4 and samples of soil, up to depths of 9 ft, were collected from locations 5 through 12. Samples of soil and bottom silt were also collected from some of these same locations in 1958, 1960, and 1961. The stream itself has been dry at these sampling times. All of these samples contained normal concentrations of alpha and beta activity. The water samples collected in 1954 ranged from 0.3 to 1.9 pCi/liter and from 5.0 to 9.6 pCi/liter in alpha and beta activity, respectively. All bottom-silt and soil samples contained less than 40 pCi/g of alpha activity and less than 90 pCi/g of beta activity. The samples collected in 1964 near locations 5, 7, 8, and 12, at depths up to 4.5 ft, were normal.

Fig. 12. Sampling Locations at Site A

However, at locations 15, 17, 18, and 19, sampled for the first time in 1964, the alpha activities ranged from 58 to 139 pCi/g, and the beta activities from 124 to 189 pCi/g. These activities are two to five times above normal values. Samples collected from locations 13, 14, and 16, close to the area of high activity, were normal. The abnormal activity appears to be due to normal uranium. For example, a sample whose alpha activity was 53 pCi/g contained 51 μ g of uranium per gram. This uranium concentration is equivalent to 34 pCi/g of natural uranium, and thus accounts for the above-normal alpha activity in the sample. Gamma-ray spectra of some of the abnormally high samples showed the presence of uranium and only the usual kinds and amounts of fallout nuclides.

Since samples collected in 1964 between the area of contamination and Plot M (locations 5, 7, 8, and 12) below the bottom of the concrete wall were normal, it is believed that the abnormal uranium concentrations are not due to the leaching of activity from Plot M. More likely, the activity was accidentally spread on the surface of the ground during work in the area before 1954, and remained undiscovered in the past because the levels are too low to be detected by ordinary survey methods, and sensitive measurements in this particular area were not made until 1964, when samples were collected and analyzed in the laboratory. Additional studies on the source and extent of the contamination, and on the isotopic composition of the uranium, are planned.