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# Argonne National Laboratory

## ENVIRONMENTAL RADIOACTIVITY AT ARGONNE NATIONAL LABORATORY

Report for the Year 1959

by

J. Sedlet

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Development Report

ARGONNE NATIONAL LABORATORY
9700 S. Cass Avenue
Argonne, Illinois

# ENVIRONMENTAL RADIOACTIVITY AT ARGONNE NATIONAL LABORATORY

Report for the Year 1959

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# ENVIRONMENTAL RADIOACTIVITY AT ARGONNE NATIONAL LABORATORY

Report for the Year 1959

## I. SUMMARY

The results of the environmental monitoring program at Argonne National Laboratory for 1959 are given in this report. The 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 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 measured by performing radiochemical analyses and total activity measurements on several types of materials collected on the ANL site and from locations approximately 10, 25 and 100 miles from the Laboratory. The sampling locations are given in Part II. Since the most probable means of dispersal of radioactive contamination are by air and water, most of the samples analyzed were of these types. Argonne waste water is discharged into Sawmill Creek, which in turn flows into the Des Plaines River, and special emphasis was placed on sampling these streams.

The average total activities in samples of water, material from the beds of lakes and streams (bottom silt), soil, and plants during 1959 are given in Figures 1 to 3. For purposes of comparison, the results obtained from 1952 through 1959 are given in Figures 4 to 7. Air-filter results are given in Figure 8.

Fallout activity was present in most samples at all locations. The amount of fallout was relatively high during the first part of the year, but decreased markedly during the latter half. By the end of the year the shorter-lived fission products from fallout were at the lowest level since 1955. Airborne beta activity from fallout decreased from a maximum of about 5 micromicrocuries per cubic meter ($\mu\mu c/M^3$) in April to less than 0.1 $\mu\mu c/M^3$ in December. The average for the year, about 2.3 $\mu\mu c/M^3$, was 25% less than for 1958, but approximately twice as high as the average from 1953 to 1957. The long-lived airborne alpha activity has not changed appreciably since 1953.

Air-filter samples were collected both on the site and at four locations from six to twenty miles from the Laboratory. The activities were essentially the same both on and off the site, and no indication of activity originating at Argonne was found in the samples.

Figure 1

AVERAGE RADIOACTIVITY IN SURFACE WATER 1959

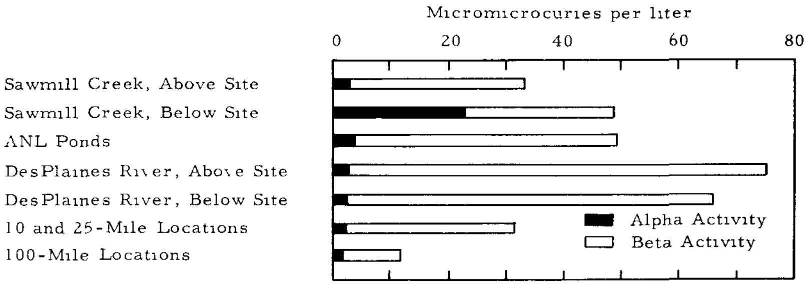



Figure 2

AVERAGE RADIOACTIVITY IN BOTTOM SILT, 1959

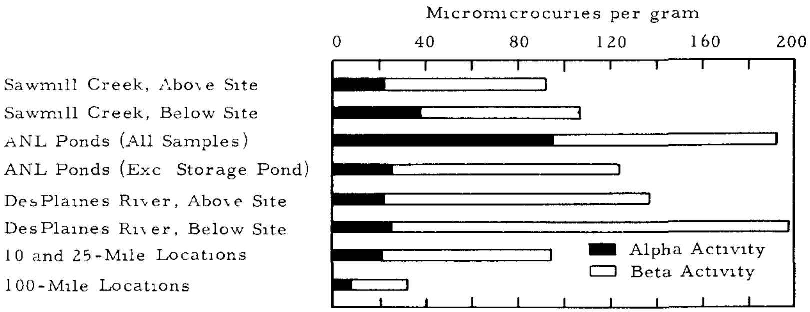



Figure 3

AVERAGE RADIOACTIVITY IN SOIL AND PLANTS, 1959

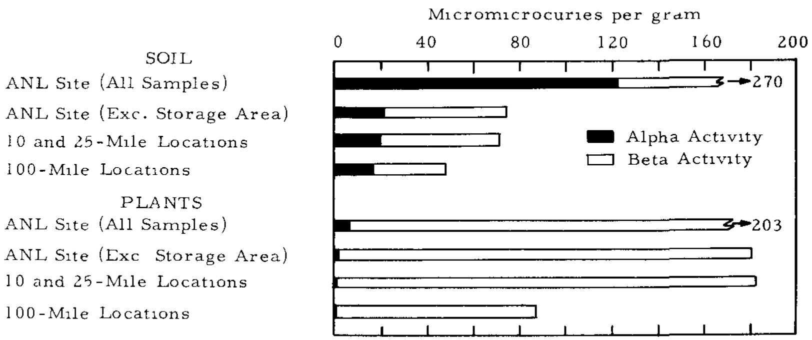



Figure 4

AVERAGE RADIOACTIVITY IN SURFACE WATER, 1952-59

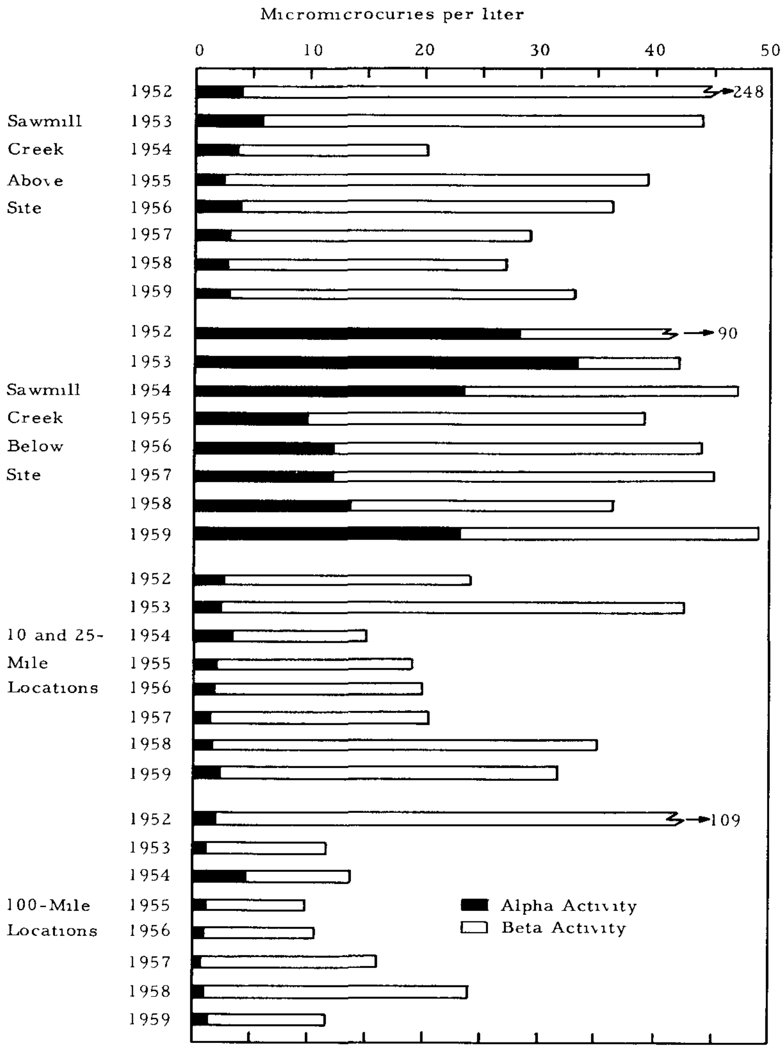


Figure 5

AVERAGE RADIOACTIVITY IN BOTTOM SILT, 1952-59

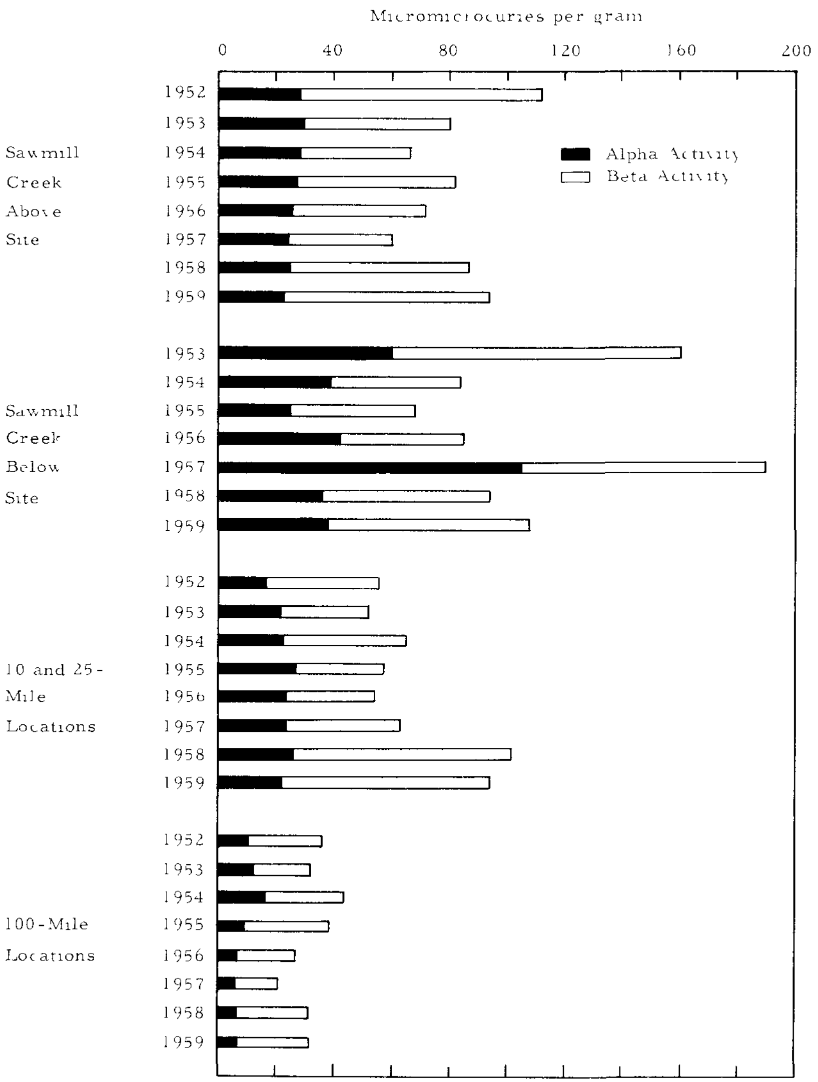



Figure 6

AVERAGE RADIOACTIVITY IN SURFACE SOIL, 1952-59

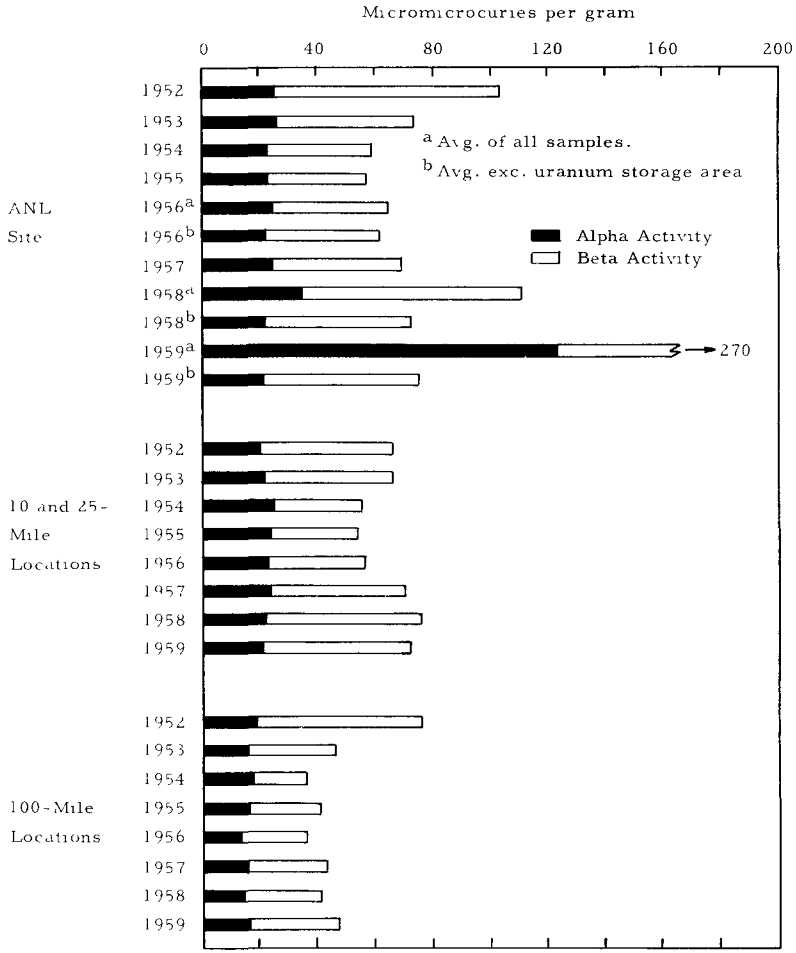


Figure 7

AVERAGE RADIOACTIVITY IN PLANTS, 1952-59

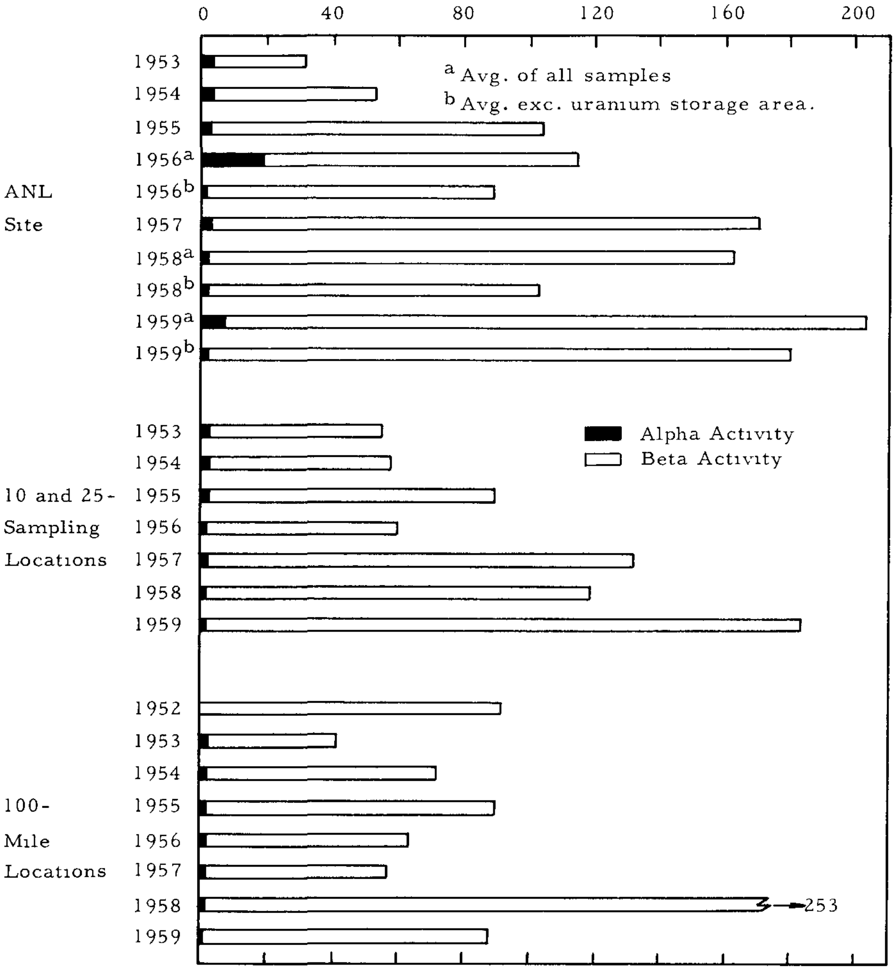
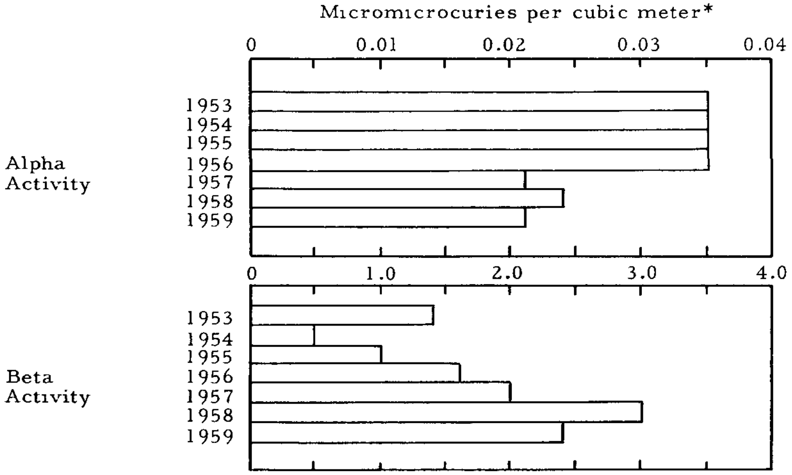



Figure 8

AVERAGE RADIOACTIVITY IN DAILY AIR FILTER SAMPLES ON ANL SITE, 1952-59




* Activity remaining three days after end of sample collection.

Radioactivity due to Argonne operations was found in Sawmill Creek below the waste water outfall, in a storage pond for contaminated waste water, and near a uranium storage shed. However, activity originating at Argonne and leaving the Argonne site was found only below the outfall in Sawmill Creek. The average alpha activity in water from this location was 23 micromicrocuries per liter ($\mu\mu c/l$), compared to an average of 2.8 $\mu\mu c/l$ in Sawmill Creek above the ANL site. The difference is attributed primarily to uranium added to the Creek in Argonne waste water. The average uranium concentration added to the Creek in waste water was approximately 21 $\mu\mu c/l$, or 0.1% of the maximum permissible concentration (MPC).* The maximum uranium concentration in a single sample was equivalent to 1.4% of the MPC. Normal uranium and uranium depleted in $U^{234}$ and $U^{235}$ were found below the outfall; enriched uranium was not detected. Most, but not all, samples collected below the outfall contained uranium from Argonne waste water. Small amounts of plutonium and thorium, also evidently from waste water, were found in below outfall water. Plutonium was detected in seventeen of thirty-six samples analyzed; the average concentration was 0.23 $\mu\mu c/l$ (0.005% of the MPC) and the maximum concentration was 1.2 $\mu\mu c/l$ (0.024% of the MPC). Plutonium was not found in the Creek above the site. Thorium was found in only four of the thirty-six below-outfall samples. The average thorium concentration in the Creek attributed to waste water was about 0.1 $\mu\mu c/l$, or 0.01% of the MPC. The relative amounts of the alpha-emitting elements in the Creek during 1959 were similar to those found earlier.

Beta activity due to contamination in Laboratory waste water was found in Sawmill Creek below the outfall. Beta activity from fallout was found both above and below the outfall. Above the ANL site the average beta activity was 33 $\mu\mu c/l$. Approximately 5 $\mu\mu c/l$ were naturally present in the stream, and 25 to 30 $\mu\mu c/l$ were due to fallout. Below the outfall the total beta activity averaged 49 $\mu\mu c/l$; the amount contributed by the waste water varied from 55% of the total in June to 95% in November and December. The remainder was due to fission product fallout and natural activity. Most of the beta activity added to the Creek in Argonne waste water was due to the $UX_1$ and $UX_2$ daughters of uranium and to $Co^{58}$. The average uranium daughter and $Co^{58}$ contributions were about 18 $\mu\mu c/l$ (0.1% of the MPC) and 10 $\mu\mu c/l$ (0.01% of the MPC), respectively. Smaller contributions were made by several of the fission products, including $Sr^{90}$ and $Cs^{137}$.

* The maximum permissible concentrations used in this report are those recommended in National Bureau of Standards Handbook 69 (June 5, 1959) for uncontrolled areas in the vicinity of a nuclear installation These values are one-tenth of those given in the Handbook for an occupational exposure of 168 hours per week. The MPC's given for water are for drinking water only, and although they are applied in this report to Sawmill Creek, it should be pointed out that Creek water is not used for human consumption The concentrations attributable to Argonne operations were low and only small fractions of the MPC

The average $Sr^{90}$ concentration below the outfall was 2.9 $\mu\mu c/l$ (2.9% of the MPC). Approximately one-third of the $Sr^{90}$ was due to fallout and the remainder to waste water. The average $Cs^{137}$ concentration, 2.0 $\mu\mu c/l$, amounted to 0.01% of the MPC. The average concentrations of the other fission products for which analyses were made were less than 0.05% of the MPC. During 1959 $Sr^{90}$ was detected with greater frequency than it had been earlier, particularly above the site, but the concentrations in individual samples were of the same order of magnitude. The $Sr^{90}$ concentrations ranged from 1 $\mu\mu c/l$ to 4.5 $\mu\mu c/l$ above the site and from less than one to 20 $\mu\mu c/l$ below the outfall.

Activity added to the Creek in Argonne waste water was also found in bottom silt samples obtained below the outfall. At this location the total activities ranged from 14 to 87 $\mu\mu c\alpha/g$ and from 35 to 208 $\mu\mu c\beta/g$. Normal activities in bottom silt are less than 30 $\mu\mu c\alpha/g$ and less than 75 $\mu\mu c\beta/g$, and about one-half of the samples contained activities in excess of these values. The concentrations of uranium, plutonium, and $Sr^{90}$ also were abnormally high in some of the samples at this location, whereas the thorium concentrations were normal.

The concentrations of alpha activity, uranium, and thorium in the Des Plaines River were normal both above and below Sawmill Creek, and plutonium was not detected at either location. Beta activity from Sawmill Creek could not be detected in the River in the presence of the much larger amounts present from fallout and naturally occurring radionuclides. Dilution of Sawmill Creek water was apparently sufficiently great so that the radioactivity in the Creek had no detectable effect on the radioactive content of the Des Plaines River.

Some of the samples of water and bottom silt from the storage lagoon for contaminated waste water contained approximately ten times the normal concentrations of total activity, as well as of uranium, plutonium, $Sr^{90}$, and $Cs^{137}$. Soil and grass collected up to 40 feet from a uranium storage shed on the site contained an average of about 200 times more uranium than is normally found in such materials. Normal concentrations are 1 to 2 $\mu\mu c/g$ in soil and about 0.2 $\mu\mu c/g$ in grass. In both cases similar results were obtained in previous years, and the contamination is, of course, the result of the use made of the two areas.

Two of four samples collected from one of the natural ponds on the ANL site contained about twice the usual surface water concentrations of 1 to 3 $\mu\mu c\alpha/l$ and 1 to 2 $\mu\mu cU/l$. This increase was probably of natural origin. Except for these samples, those from Sawmill Creek below the outfall and from the storage lagoon, samples from all locations both on and off the site contained normal concentrations of alpha activity and up to ten times the normal beta activities of 5 to 10 $\mu\mu c/l$. The increased beta activities were due to fallout. The amount of fallout activity in water was at a maximum in the spring, and decreased regularly during the remainder of the year.

Bottom silt samples from all locations except those mentioned previously contained only natural and fallout activity. Average values for the natural activity were about 20 $\mu\mu c\alpha/g$ and 70 $\mu\mu c\beta/g$, although some individual samples differed greatly from these averages. Normal concentrations of uranium and thorium varied between 1.2 and 2.6 $\mu\mu cU/g$ and between 1 and 2 $\mu\mu cTh/g$. Some of the samples from the DuPage River at Naperville and from a pond on the ANL site contained two to four times the usual thorium concentrations, but contained normal amounts of uranium. The alpha activities in these samples were two to three times the normal amounts, but the beta activities were only slightly elevated. Fallout activity in bottom silt produced an average increase of about 25 $\mu\mu c/g$ in the total beta activity. Fallout activities of the 100-mile samples were not pronounced at the times of sampling.

Excluding the area near the uranium storage shed, surface soil samples collected on and near the ANL site contained comparable amounts of activity. Average values on the site (for all samples other than those near the storage area) were 21 $\mu\mu c\alpha/g$ and 75 $\mu\mu c\beta/g$. Near the site the averages were 20 $\mu\mu c\alpha/g$ and 71 $\mu\mu c\beta/g$. Average activities at the 100-mile locations were lower, namely, 16 $\mu\mu c\alpha/g$ and 48 $\mu\mu c\beta/g$, but usual for these locations. Due to fallout, increases in beta activity up to twice the normal concentrations were found in some samples at all locations.

The alpha activities in plant samples collected near the uranium storage shed ranged from 2.8 to 96 $\mu\mu c/g$. At all other locations the alpha activities varied from 0.25 to 3.2 $\mu\mu c/g$. The beta activities decreased from a maximum of several hundred micromicrocuries per gram early in the year to about 30 $\mu\mu c/g$ in December. This decrease followed the decline in fallout activity in air. The beta activities during the first part of the year were the highest found since plant collection was begun. Plutonium concentrations in excess of the detection limit of 0.005 $\mu\mu c/g$ of the oven-dried plant were found in three of the five samples analyzed. In these samples the plutonium concentrations ranged from 0.015 to 0.072 $\mu\mu c/g$. The plutonium presumably was due to fallout.

## II. PROGRAM AND PROCEDURES

The results of the environmental monitoring program at Argonne National Laboratory for the year 1959 are given in this report. The 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 origin of any radioactivity above the natural levels. Of primary interest is the detection of any radioactive materials released to the environment by Argonne National Laboratory.

The radioactive content of the environment was measured by performing radiochemical analyses and total activity measurements on the types and numbers of samples given in Table I. These samples were collected on the Argonne site and from locations approximately 10, 25, and 100 miles from the site. Sampling locations on the site are shown in Figure 9. The sampling locations within 25 miles of the Laboratory (referred to as "near" ANL in this report) are given in Figure 10. Samples collected near ANL should indicate the extent and direction of contamination in the event that significant amounts of activity are released at Argonne. The samples collected 100 miles from the Laboratory (referred to as "reference sites" in this report) were originally intended to serve as continuous checks for contamination during collection, analysis, and storage, since their radioactive content was not expected to change with time. This purpose has been realized for alpha activity, but because of the widespread occurrence of fallout from nuclear detonations the beta activities in these samples have served primarily to indicate the extent and magnitude of fallout activity. Since the amount of fallout decreased sharply during the latter half of 1959 and very little bomb debris has been added to the atmosphere since late in 1958, the original purpose of the 100-mile samples may be realized in the future. Since fallout activity could be found in varying amounts in all environmental materials collected in the area being sampled, beta activities at the levels found have been more difficult to interpret in terms of origin than has been true for alpha activities. However, by making the proper choices of sampling location and type of analysis, it was usually possible to distinguish between fallout, naturally occurring activity, and activity from Argonne operations.

TABLE I

SAMPLES COLLECTED IN 1959

| Type | Number |
|---|---|
| Water | 327 |
| Precipitation | 84 |
| Soil | 91 |
| Bottom Silt | 167 |
| Plant | 81 |
| Air Filter | 712 |
| Animal | 7 |

Total alpha and beta activity measurements were made by counting thick samples (5 to 75 $mg/cm^2$) after a minimum of sample preparation. Water and precipitation samples, acidified with nitric acid after collection to prevent hydrolysis, were evaporated to dryness. The residue was flamed, ground in a mortar, and spread uniformly on a weighed counting planchet with carbon tetrachloride. The residue was counted after it had been dried and the planchet reweighed. Soil and bottom silt were dried at 110°C, ground in a mortar, and a weighed portion spread on a counting planchet. Plant samples were washed with water to remove dirt, dried at 110°C, ashed, and a weighed portion mounted for counting. Animal samples were air dried at room temperature, ashed, and a weighed portion counted. Air-filter samples were sprayed with a solution of polystyrene in ethylene dichloride to fix the dust on the paper prior to mounting on a planchet.

Figure 9

SAMPLING LOCATIONS ON THE SITE OF ARGONNE NATIONAL LABORATORY

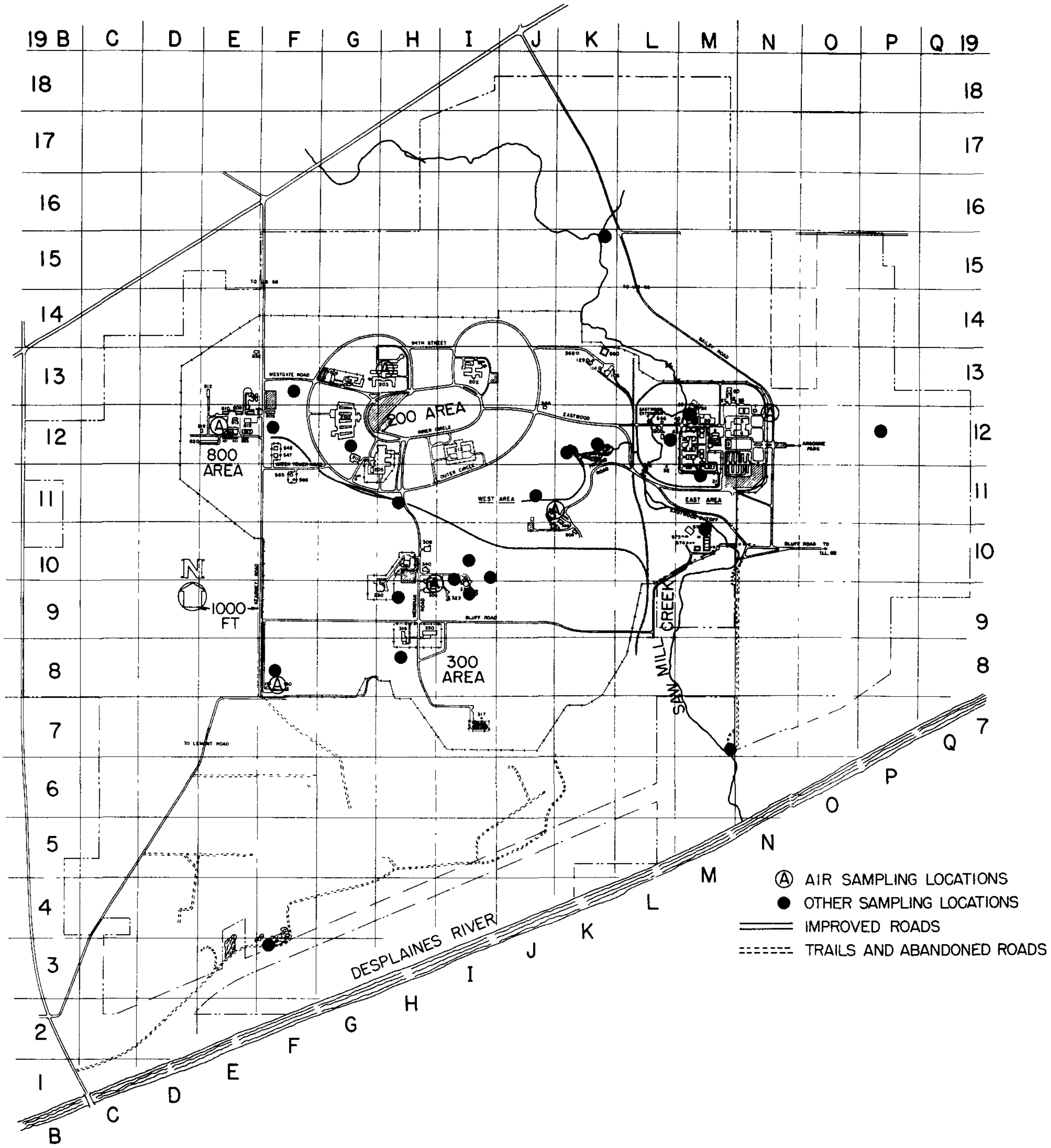
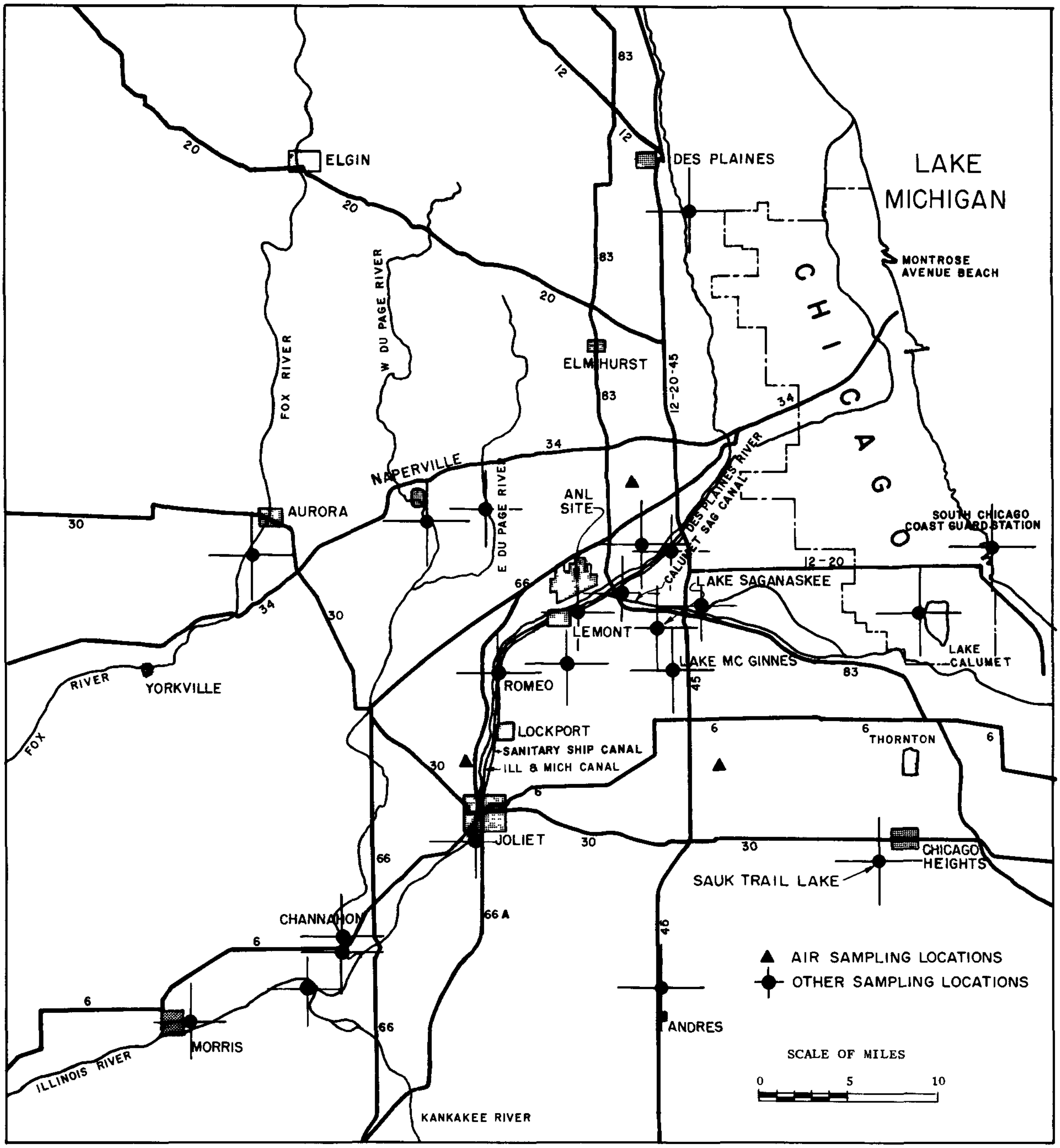


Figure 10

SAMPLING LOCATIONS NEAR ARGONNE NATIONAL LABORATORY



The polystyrene layer was 0.1 $mg/cm^2$ thick. Total alpha counting was done in a nylon-window gas-flow proportional counter; beta particles were counted in the same type of chamber installed in a lead and anticoincidence shield to reduce the background. The thicknesses of the nylon windows ranged from 0.1 to 0.2 $mg/cm^2$. For air filters, the correction factors used to obtain 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 $Pu^{239}$ (for alpha particles) and $Tl^{204}$ (for beta particles). The results obtained in this way represent the true disintegrations rates 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 are used for total alpha and beta counting since the samples were thick and contained unknown and variable mixtures of radionuclides. True disintegration rates in such samples cannot be obtained by counting total activity alone, and a standardized but arbitrary method for obtaining nominal disintegration rates must be used. However, measurements of total activity are made because large numbers of such analyses can be made rapidly, and the results are very useful in comparing activity levels and in determining which samples should be analyzed further.

Gamma-ray counting was done with a 4 x 4-in. sodium iodide (thallium) crystal connected to a 200-channel analyzer. The crystal was shielded by six inches of iron lined with $\frac{1}{8}$ inch of low-activity lead to reduce backscattering. Energy and counting efficiency calibrations were made with standardized solutions of the appropriate nuclides. Specific fission product analyses were made by separating the desired element with carrier added and counting the activity in the anticoincidence shielded beta counters. Counting rates were converted to disintegration rates by applying correction factors experimentally determined for each nuclide being counted. Uranium analyses were made with a fluorophotometer, and the results converted to disintegration rates, assuming the natural isotopic composition of uranium. Plutonium and thorium (including the $UX_1$ 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 $2\pi$ proportional counters.

Additional details on the sampling program, instrumentation, counting techniques, and radiochemical analyses are given in the previous reports in this series. A list of these reports is given in the report for 1958, ANL-6047.

## III. RADIOACTIVITY IN ENVIRONMENTAL SAMPLES

### A. Precipitation

Samples of precipitation were collected on the Argonne site at location 8F in Figure 9. The rain collector was arranged to collect two successive one-gallon samples when the rainfall was sufficiently large. Each one-gallon sample was equivalent to 0.48 inch of rain. The capacity of the collector was sufficient to collect over 90% of all the rain during the year. When two portions were collected from one rainfall, they were analyzed and tabulated as separate samples. In general, the first portion contained the larger amount of activity and solids, although occasionally the reverse situation was found. Snow also was collected from the rain tray whenever possible in order to obtain a sample from a known area. Analyses were made as indicated in Part II.

The total alpha and beta activities in precipitation collected during 1959 are given in Table II. The bulk of the alpha activity was due to naturally occurring radionuclides present in air. These included uranium, thorium, and their daughters present in dust, as well as radon and thoron daughters. Preliminary results also indicated that plutonium contributed a small fraction of the alpha activity in some samples. Most of the beta activity resulted from fission product fallout from nuclear detonations. The presence of fission products was confirmed by gamma-ray spectroscopy and chemical separations on selected samples. Fallout activity, expressed in millicuries per square mile, reached a maximum in March and April. In terms of $\mu\mu c$ per liter, the maximum fallout occurred in January. By either method of expressing the results, fallout activity was relatively high during the first $4\frac{1}{2}$ months of the year. During May the beta activity in rainfall dropped sharply by about a factor of five and, except for occasional fluctuations, remained low for the remainder of the year. Only one rainfall after May 20 contained more than 1000 $\mu\mu c$ of beta activity per liter. The beta activity decreased from May until September, when both the average and total beta activity increased as a result of increased activity in two of the seven September rains. These two rainfalls contained about 500 and 200 $\mu\mu c$ per liter, respectively. The alpha activity in these rains also increased, so the same factors may have caused increases in both types of activity. Since the age of the fission products in these rains was essentially the same as in the previous few months, the increase was not due to recently produced fission products. After a decrease in October, there was little change in beta activity during the remainder of the year. The beta activities during the last three months of the year were the lowest since October 1955. The decrease in beta activity during the last half of 1959 and the gradual increase in the age of the fission products during the year were to be expected since nuclear test detonations were not conducted during 1959.

# TABLE II

## NONVOLATILE RADIOACTIVITY IN PRECIPITATION AT ANL SITE, 1959

| Month | Time[1] (days) | Alpha Activity | | | Beta Activity | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | No of Samples | μμc/liter Max | μμc/liter Avg | No of Samples | μμc/liter Max | μμc/liter Avg | mc/sq mi | Age Range[2] (days) |
| | | | | | | x $10^2$ | x $10^2$ | | |
| January | 3 | 4 | 37 9 | 13 8 | 4 | 59 0 | 39 5 | - | 60-90 |
| | 7 | 4 | 37 8 | 13 8 | 4 | 53 9 | 31 8 | 269 | |
| February | 1 | 5 | 3 1 | 1 7 | 3 | 17 1 | 12 4 | - | |
| | 3 | 5 | 2 7 | 1 1 | 5 | 15 7 | 10 1 | - | 80-120 |
| | 7 | 5 | 1 6 | 0 7 | 5 | 15 1 | 10 0 | 142 | |
| March | 1 | 10 | 14 1 | 4 1 | 7 | 29 9 | 19 2 | - | |
| | 3 | 10 | 4 2 | 1 8 | 10 | 35 8 | 20 7 | - | 90->300 |
| | 7 | 10 | 3 9 | 1 8 | 10 | 30 0 | 19 5 | 374 | |
| April | 1 | 7 | 18 5 | 7 6 | 7 | 34 3 | 20 4 | - | |
| | 3 | 6 | 11 9 | 4 1 | 7 | 33 0 | 19 5 | - | 70-300 |
| | 7 | 7 | 9 9 | 3 1 | 7 | 32 2 | 18 7 | 309 | |
| May | 1 | 12 | 16 3 | 4 2 | 12 | 27 9 | 9 3 | - | |
| | 3 | 8 | 5 7 | 2 6 | 12 | 27 6 | 9 1 | - | 70->300 |
| | 7 | 12 | 3 3 | 0 9 | 12 | 27 3 | 8 8 | 258 | |
| June | 1 | 7 | 18 8 | 12 6 | 7 | 25 3 | 5 5 | - | |
| | 3 | 5 | 2 2 | 1 0 | 6 | 4 8 | 1 9 | - | 70->300 |
| | 7 | 7 | 7 2 | 1 4 | 7 | 24 5 | 5 0 | 58 | |
| July | 1 | 11 | 5 4 | 1 7 | 4 | 1 21 | 1 08 | - | |
| | 3 | 11 | 3 8 | 1 0 | 11 | 2 79 | 1 40 | - | 90->300 |
| | 7 | 11 | 1 1 | 0 4 | 11 | 2 78 | 1 36 | 22 | |
| August | 1 | 3 | 1 4 | 1 0 | 3 | 1 05 | 0 52 | - | |
| | 3 | 4 | 0 7 | 0 4 | 4 | 1 80 | 0 79 | - | >300 |
| | 7 | 4 | 0 7 | 0 5 | 4 | 1 66 | 0 74 | 7 9 | |
| September | 1 | 5 | 29 2 | 8 1 | 5 | 6 55 | 2 16 | - | |
| | 3 | 7 | 4 3 | 1 5 | 7 | 5 97 | 1 56 | - | >300 |
| | 7 | 7 | 4 7 | 1 6 | 7 | 4 86 | 1 41 | 26 | |
| October | 1 | 7 | 4 3 | 1 8 | 6 | 0 54 | 0 35 | - | |
| | 3 | 7 | 0 9 | 0 4 | 7 | 0 94 | 0 39 | - | >300 |
| | 7 | 7 | 0 9 | 0 3 | 7 | 0 89 | 0 39 | 7 4 | |
| November | 1 | 2 | 3 4 | 2 3 | 2 | 1 11 | 0 63 | - | |
| | 3 | 5 | 1 7 | 0 7 | 5 | 1 04 | 0 49 | - | >300 |
| | 7 | 5 | 1 6 | 0 7 | 5 | 0 97 | 0 46 | 5 7 | |
| December | 1 | 2 | 1 6 | 0 9 | 2 | 0 30 | 0 23 | - | |
| | 3 | 5 | 1 2 | 0 6 | 5 | 0 73 | 0 42 | - | >300 |
| | 7 | 5 | 0 7 | 0 5 | 5 | 0 70 | 0 41 | 4 4 | |

[1] Days after end of precipitation (approximately)

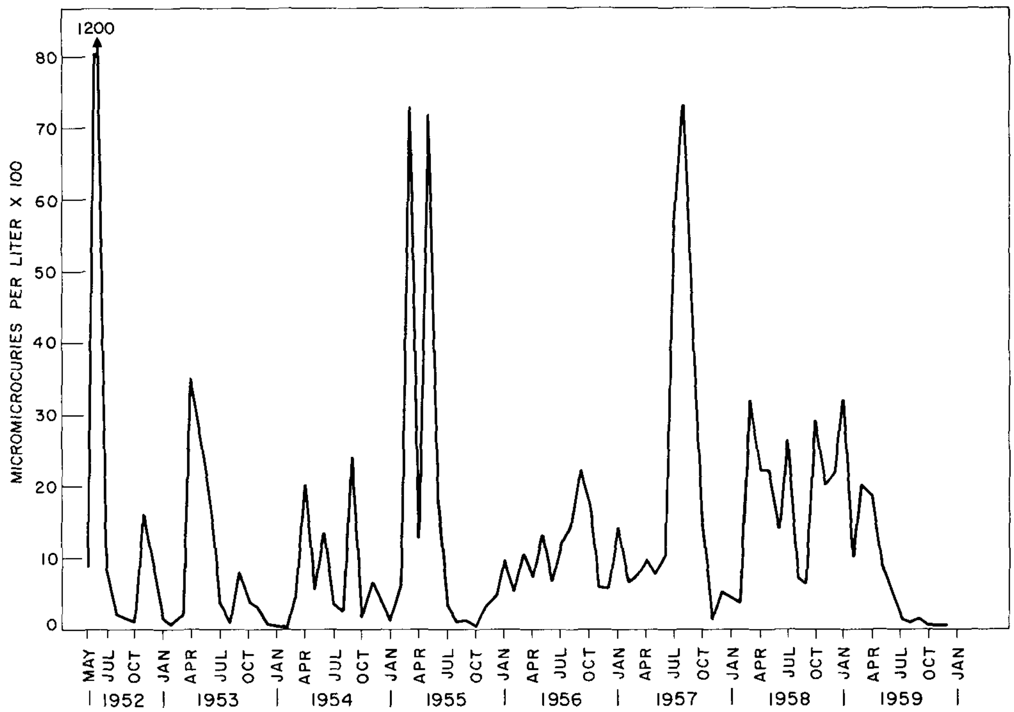
[2] Calculated for each sample assuming a $T^{-1\ 2}$ decay rate

It should be mentioned that the age calculation based on a $T^{-1.2}$ decay rate can give only an approximate value, since each rain contained fission products from a large number of detonations occurring at many different times.

The average monthly "seventh day" beta activity since 1952, when precipitation collection was begun, is given in Figure 11.

Figure 11

AVERAGE MONTHLY BETA ACTIVITY IN PRECIPITATION AT ANL SITE, 1952-58




The fluctuations may be correlated with announced periods of nuclear detonations. Increases were noted during and shortly after each test series; in general, two pronounced increases were observed each year, corresponding to the spring and fall test series. These increases also were associated with increased concentrations of the shorter-lived fission products (and therefore with a decrease in apparent age), indicating that the increase was primarily due to fission products produced during the tests being conducted at the time. As was mentioned previously, this was not the situation for the small increase in September 1959. In this case there was apparently an increase in the amount of fallout from the reservoir of activity already present in the upper atmosphere. From 1952 through 1955, when the test periods were relatively short, the activity dropped to 200 $\mu\mu c$ per liter or less between peak activities ranging from about 600 to $1.2 \times 10^5$ $\mu\mu c$ per liter. From 1956 through 1958 the valleys were usually higher, being about 500 to 700 $\mu\mu c$ per liter, probably because the periods between testing were shorter. The behavior during 1959 has already been discussed. The total amount of beta activity carried down by precipitation during 1959 was approximately 1500 millicuries per square mile, somewhat higher than the values from 1953 through 1958 (600 to 1100 mc per sq. mi.), but considerably less than the 11,000 mc per sq. mi. carried down in 1952.

Some of the samples collected during 1959 were analyzed for $Sr^{89}$, $Sr^{90}$, $Cs^{137}$, and $Ce^{144}$. These results are given in Table III.

TABLE III

FISSION PRODUCT ACTIVITY IN SELECTED PRECIPITATION SAMPLES, 1959

| Date of Rainfall | Amount (inches) | Micromicrocuries per liter* | | | |
|---|---|---|---|---|---|
| | | $Sr^{89}$ | $Sr^{90}$ | $Cs^{137}$ | $Ce^{144}$ |
| January 19 | 0.30 | 438 ± 54 | 14 ± 2 | 46 ± 5 | 267 ± 8 |
| January 21 | 0.41 | 518 ± 60 | 24 ± 1 | | |
| January 25 | 0.20 | 440 ± 80 | 24 ± 2 | | 270 ± 8 |
| February 10 | 0.91 | 265 ± 53 | 18 ± 2 | | |
| February 12 | 0.17 | 150 ± 10 | 6.7 ± 0.3 | | 70 ± 6 |
| February 23 | 0.48 | 288 ± 39 | 16 ± 2 | | 161 ± 20 |
| February 23 | 0.08 | 149 ± 21 | 8.3 ± 0.8 | | |
| March 5 | 0.58 | 118 ± 25 | 9.1 ± 2.7 | | 124 ± 10 |
| March 8 | 0.06 | 244 ± 44 | 19 ± 2 | | |
| March 15 | 0.66 | 675 ± 85 | 31 ± 2 | | |
| March 26 | 1.0 | 407 ± 73 | 23 ± 2 | | |
| May 3 | 0.29 | 262 ± 25 | 18 ± 4 | | |
| May 11 | 0.88 | 251 ± 25 | 27 ± 3 | 37 ± 2 | |
| July 19 | 0.56 | 21 ± 2 | 5.3 ± 0.5 | | |
| July 30 | 0.49 | 19 ± 2 | 4.8 ± 1.0 | | |
| October 8 | 0.55 | $<2$ | 1.8 ± 0.5 | 2.8 ± 1.0 | |
| November 4 | 1.81 | $<2$ | 0.76 ± 0.30 | | |

*Corrected for decay to the midtime of the precipitation period.

Variations in the concentrations of these nuclides generally followed the trend expected from the total beta activities and the relative half-lives of the nuclides. From the middle of March through July, the $Sr^{89}/Sr^{90}$ ratio decreased with about the half-life of $Sr^{89}$, indicating that the age of the fission products remained constant during this period. The ratios could not, of course, be determined for the samples analyzed after July, since $Sr^{89}$ was not detected. Prior to March, the $Sr^{89}/Sr^{90}$ ratio fluctuated and was lower than expected relative to the ratios found later. The reasons for this are not apparent.

B. Air Filters

Airborne particulate matter was sampled at six locations on the Laboratory site and at four locations off the site. The sampling locations are shown in Figures 9 and 10. The off-site samplers were located six to twenty miles from the Laboratory. Samples were collected by drawing outside air through Hollingsworth-Vose No. 70 filter paper at an air flow rate of approximately 30 cubic meters per hour. At the off-site locations and at five of the on-site locations the filter papers were changed and counted at weekly intervals. One on-site sample was changed daily to obtain short-term changes in air activity. The on-site weekly samples were counted during the first day after the end of the collection period to obtain the radon and thoron concentrations, and again three and seven days after collection to obtain the longer-lived activities. The off-site samples could be counted only three and seven days after collection; the daily samples were counted once after three days. Sampling was continuous except for occasional equipment failures and the few minutes required to change filter papers. Additional details on the sampling and counting techniques are given in reports ANL-5808 and ANL-5934.

The long-lived alpha and beta activities found in samples collected during 1959 are summarized in Tables IV to VI. The alpha activity remaining three days after the end of the filtering period was due primarily to natural activities present in dust in the air. The average "third day" alpha activities (about $0.006 \mu\mu c/M^3$ in the weekly samples) were essentially the same both off and on the ANL site, although some individual samples differed from the average by about a factor of three. Such differences are normal and depend on a variety of variables, particularly the amount of dust in the air. The maximum difference in the monthly averages between off-site and on-site samples was $0.0014 \ \mu\mu c\alpha/M^3$. Differences of this magnitude were observed when comparisons were made between either off-site or on-site samples only. The difference between the annual off-site and on-site averages was only $0.0005 \ \mu\mu c\alpha/M^3$. The alpha activities in the daily samples were higher than in the weekly samples, probably because the same self-absorption corrections were used for both types of samples, and the daily samples, containing considerably less dust, required less correction. The results, however, are self-consistent and comparisons between the same type of sample are valid. The beta activities are not as sensitive to variations in the amount of dust on the sample.

TABLE IV

LONG-LIVED RADIOACTIVITY IN WEEKLY AIR FILTER SAMPLES ON ANL SITE, 1959

| Month | No. of Samples | $\mu\mu c/M^3$ after 3 Days of Decay | | | | $\mu\mu c/M^3$ after 7 Days of Decay | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | Alpha Activity | | Beta Activity | | Alpha Activity | | Beta Activity | |
| | | Max | Avg | Max | Avg | Max | Avg | Max | Avg |
| January | 15 | 0.0066 | 0.0041 | 6.00 | 4.20 | 0.0062 | 0.0040 | 5.57 | 4.02 |
| February | 15 | 0.0048 | 0.0031 | 5.28 | 4.43 | 0.0055 | 0.0036 | 5.09 | 4.29 |
| March | 17 | 0.0157 | 0.0064 | 7.23 | 4.56 | 0.0092 | 0.0048 | 7.00 | 4.41 |
| April | 17 | 0.0126 | 0.0083 | 8.08 | 5.19 | 0.0094 | 0.0040 | 7.90 | 5.09 |
| May | 15 | 0.0115 | 0.0055 | 5.65 | 3.53 | 0.0056 | 0.0031 | 5.66 | 3.44 |
| June | 14 | 0.0204 | 0.0092 | 4.83 | 2.45 | 0.0056 | 0.0035 | 4.79 | 2.18 |
| July | 15 | 0.0236 | 0.0076 | 3.88 | 1.25 | 0.0072 | 0.0028 | 3.60 | 1.22 |
| August | 14 | 0.0143 | 0.0069 | 0.93 | 0.36 | 0.0046 | 0.0027 | 0.91 | 0.35 |
| September | 11 | 0.0181 | 0.0070 | 0.32 | 0.23 | 0.0056 | 0.0032 | 0.27 | 0.21 |
| October | 17 | 0.0156 | 0.0054 | 0.23 | 0.12 | 0.0045 | 0.0022 | 0.22 | 0.11 |
| November | 16 | 0.0143 | 0.0053 | 0.17 | 0.11 | 0.0051 | 0.0034 | 0.16 | 0.11 |
| December | 18 | 0.0097 | 0.0051 | 0.21 | 0.086 | 0.0048 | 0.0029 | 0.21 | 0.084 |
| Average | | 0.0062 | | 2.21 | | 0.0034 | | 2.13 | |

TABLE V

LONG-LIVED RADIOACTIVITY IN WEEKLY AIR FILTER SAMPLES NEAR ANL, 1959

| Month | No. of Samples | $\mu\mu c/M^3$ after 3 Days of Decay | | | | $\mu\mu c/M^3$ after 7 Days of Decay | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | Alpha Activity | | Beta Activity | | Alpha Activity | | Beta Activity | |
| | | Max | Avg | Max | Avg | Max | Avg | Max | Avg |
| January | 18 | 0.0073 | 0.0046 | 7.10 | 4.14 | 0.0076 | 0.0049 | 5.47 | 4.04 |
| February | 14 | 0.0054 | 0.0037 | 5.27 | 4.08 | 0.0056 | 0.0037 | 5.10 | 3.98 |
| March | 16 | 0.0121 | 0.0064 | 7.08 | 4.49 | 0.0109 | 0.0057 | 6.94 | 4.29 |
| April | 14 | 0.0147 | 0.0078 | 8.08 | 5.81 | 0.0086 | 0.0053 | 7.82 | 5.59 |
| May | 16 | 0.0096 | 0.0056 | 6.82 | 4.04 | 0.0067 | 0.0043 | 6.61 | 3.91 |
| June | 13 | 0.0154 | 0.0090 | 4.50 | 2.92 | 0.0065 | 0.0044 | 4.37 | 2.90 |
| July | 17 | 0.0153 | 0.0077 | 2.01 | 1.28 | 0.0057 | 0.0036 | 1.98 | 1.22 |
| August | 18 | 0.0079 | 0.0054 | 0.91 | 0.40 | 0.0040 | 0.0026 | 0.87 | 0.34 |
| September | 16 | 0.026 | 0.0073 | 0.36 | 0.20 | 0.0060 | 0.0028 | 0.28 | 0.19 |
| October | 11 | 0.0067 | 0.0040 | 0.19 | 0.10 | 0.0041 | 0.0025 | 0.22 | 0.10 |
| November | 12 | 0.0128 | 0.0040 | 0.20 | 0.093 | 0.0069 | 0.0037 | 0.19 | 0.092 |
| December | 13 | 0.0133 | 0.0042 | 0.16 | 0.070 | 0.0049 | 0.0028 | 0.16 | 0.070 |
| Average | | 0.0058 | | 2.30 | | 0.0039 | | 2.23 | |

TABLE VI

LONG-LIVED RADIOACTIVITY IN 24-HOUR AIR-FILTER SAMPLES ON ANL SITE, 1959

| Month | No. of Samples | Alpha Activity ($\mu\mu c/M^3$)* | | Beta Activity ($\mu\mu c/M^3$)* | |
|---|---|---|---|---|---|
| | | Max | Avg | Max | Avg |
| January | 26 | 0.0097 | 0.0068 | 5.95 | 4.28 |
| February | 26 | 0.0056 | 0.0042 | 6.55 | 3.94 |
| March | 27 | 0.028 | 0.012 | 6.95 | 4.06 |
| April | 23 | 0.052 | 0.024 | 6.93 | 5.76 |
| May | 22 | 0.053 | 0.023 | 6.06 | 4.18 |
| June | 23 | 0.073 | 0.040 | 6.92 | 3.58 |
| July | 19 | 0.10 | 0.041 | 2.79 | 1.74 |
| August | 20 | 0.026 | 0.018 | 0.63 | 0.37 |
| September | 25 | 0.073 | 0.035 | 0.45 | 0.29 |
| October | 31 | 0.071 | 0.017 | 0.30 | 0.13 |
| November | 25 | 0.038 | 0.014 | 0.19 | 0.093 |
| December | 27 | 0.037 | 0.014 | 0.11 | 0.085 |
| Average | | 0.021 | | 2.38 | |

*Activity remaining three days after end of collection period.

The beta activity remaining after three days was primarily due to fallout from nuclear detonations. The presence of fission products in the air samples was confirmed by gamma-ray spectroscopy. The similarity in beta activity between the off-site and on-site samples suggests a widespread and reasonably uniform source, such as fission product fallout, for the bulk of the beta activity. This similarity was noted in comparing individual samples as well as in the monthly averages given in the tables. The results for individual samples are not given in this report because of space requirements. The variations in beta activity during the year followed the pattern expected for fallout. As was the case for precipitation, the beta activities were relatively high during the first part of the year, reaching a maximum in March and April, and declined rapidly in the latter part of the year. Also, there was little change during the last quarter of the year. Two differences from the precipitation results were noted. The beta activity in air did not increase in September as did the beta activity in rain, and the air activities did not fluctuate as greatly. These differences are probably due to meteorological conditions. The precipitation samples indicate the amount of activity present at the time of precipitation in the large volume of air through which the rain or snow falls. The air samples, since they are continuous, give an average air concentration at the point of sampling. As a result, some differences between the two types of samples might be expected.

There was no indication from the air-filter results that activity originating at Argonne added significantly to the airborne activity off the site. If radioactive materials used at Argonne were leaving the site, higher activities would be expected on the site than off the site. However, both the alpha and beta activities were essentially the same at both locations. On-site air samplers were located at the periphery of the site as well as near buildings containing large amounts of radioactive materials. No significant differences in activity among such locations were found. The average "third day" activity both on and off the ANL site was a few percent of the natural radon daughter activity and about 2.2% of the MPC for a mixture of unidentified radionuclides in air.

TABLE VII

AVERAGE ANNUAL "THIRD DAY" RADIOACTIVITY IN AIR FILTER SAMPLES, 1953-1959

| Year | Weekly Samples ($\mu\mu c/M^3$) | | Daily Samples ($\mu\mu c/M^3$) | |
|---|---|---|---|---|
| | Alpha | Beta | Alpha | Beta |
| 1953 | 0.010 | 1 2 | 0 03 | 1 4 |
| 1954 | 0 010 | 0 4 | 0 03 | 0 5 |
| 1955 | 0 007 | 0 8 | 0 03 | 1 0 |
| 1956 | 0 010 | 1 3 | 0 03 | 1 6 |
| 1957 | 0 008<br>0 007* | 2 1<br>2 1* | 0 021 | 2 0 |
| 1958 | 0 007<br>0 007* | 2 8<br>3 4* | 0.024 | 3 0 |
| 1959 | 0 006<br>0 006* | 2 2<br>2 3* | 0 021 | 2 4 |

*Results obtained at off-site sampling locations All other results are for on-site locations

The annual average activities since sample collection was begun are given in Table VII. The steady increase from 1954 through 1958 is apparent. The decrease from about 3.0 $\mu\mu c/M^3$ in 1958 to about 2.4 $\mu\mu c/M^3$ in 1959 was to be expected since nuclear testing was not conducted during 1959. While the average beta activity varied by about a factor of six from 1953 to 1959, the alpha activity varied by only about 40% during the same period. This also indicates that the bulk of the alpha and beta activities had different origins.

## C. Water

### 1. Sawmill Creek and Des Plaines River

Argonne waste water is discharged into Sawmill Creek at location 7M in Figure 9, and this stream was sampled before it passed through the Laboratory site and below the waste water discharge. During January, February, part of August, September, and October, there was insufficient water (or ice) in the stream to permit sample collection. At such times the water collected below the outfall consisted entirely of waste water. At other times during the year the below-outfall samples were collected after the waste water had mixed with the natural Creek water.

The alpha activities found in Sawmill Creek water during 1959 are given in Table VIII. The increased alpha activity below the outfall was evidently due to radioactivity added to the Creek in Argonne waste water. Above the site water was collected weekly when available; below the outfall water was sampled three times weekly. Since above the site the Creek contained only natural water, and essentially only naturally occurring alpha activity, the average of the weekly samples at this location, 2.8 $\mu\mu c/liter$, is very similar to the average that would have been obtained if above-site water had been collected and analyzed at the same times as below-outfall water. Therefore, a comparison of the averages of all the samples collected from the Creek will give the activity added to the Creek in the waste water at the times of below-outfall sampling. Since Creek water is diluted on the average approximately in half by Argonne waste water, the average natural activity below the outfall was 1.4 $\mu\mu c/l$, one-half of the total activity above the site. The total alpha activity below the outfall was 23 $\mu\mu c/l$. Consequently, Argonne's contribution to the alpha activity of the Creek below the outfall was about 22 $\mu\mu c/l$. This value is not greatly affected if the average below-outfall activity is taken from only those samples collected on the same days as the Creek was sampled above-site. In this case the below-outfall average was 20 $\mu\mu c/l$ and Argonne's contribution was 19 $\mu\mu c/l$, or 3 $\mu\mu c/l$ less.

A comparison of the total alpha activities with the uranium concentrations indicates that the alpha activity added to the Creek in Argonne waste water was almost entirely due to uranium. Since above the site the Creek water contained an average of 1.8 $\mu\mu c$ of uranium per liter, the natural Creek water contributed approximately 0.9 $\mu\mu c/l$ to the uranium concentration below the outfall. The average uranium concentration in all below-outfall samples analyzed for uranium was 35 $\mu\mu c/l$. Using this average, Argonne waste water added approximately 34 $\mu\mu c/l$ to the Creek below the outfall during 1959. This uranium concentration amounts to 0.17% of the maximum permissible concentration. About 30% of the below-outfall samples were analyzed for uranium. Since many of the below-outfall samples analyzed for uranium contained above-average concentrations of total alpha activity (and, therefore, of uranium), the average below-outfall uranium concentration given previously is higher than the true average. A closer approximation to the true average is obtained from those below-outfall samples collected only on days when above-site water was also sampled. This represents a more random sampling for uranium analysis, since these samples were analyzed for uranium regardless of the alpha activity.

TABLE VIII

NONVOLATILE ALPHA ACTIVITY IN SAWMILL CREEK WATER, 1959

| Month | Location* | Total Alpha Activity | | | Uranium | | | Plutonium | | | | Thorium | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | No of Samples | μμc/l | | No of Samples | μμc/l | | No of Samples | Samples >0 05 μμc/l | | | No of Samples | Samples >0 05 μμc/l | | |
| | | | Max | Avg | | Max | Avg | | No | Max | Avg | | No | Max | Avg |
| January | B | 13 | 40 6 | 18 2 | 6 | 37 8 | 25 0 | 2 | 1 | 0 2 | - | 2 | 0 | - | - |
| February | B | 12 | 80 6 | 37 7 | 6 | 79 5 | 56 7 | 2 | 0 | - | - | 2 | 0 | - | - |
| March | A | 4 | 2 8 | 2 5 | 4 | 1 9 | 1 6 | 2 | 0 | - | - | 2 | 0 | - | - |
| | B | 13 | 80 7 | 25 7 | 8 | 87 2 | 38 8 | 2 | 0 | - | - | 2 | 0 | - | - |
| April | A | 5 | 3 1 | 2 3 | 5 | 2 3 | 1 6 | 3 | 0 | - | - | 3 | 0 | - | - |
| | B | 13 | 51 1 | 23 0 | 7 | 42 4 | 24 9 | 3 | 3 | 1 2 | 0 9 | 3 | 1 | 0 3 | - |
| May | A | 4 | 2 8 | 2 4 | 4 | 1 4 | 1 2 | 2 | 0 | - | - | 2 | 0 | - | - |
| | B | 14 | 95 0 | 18 3 | 6 | 123 | 39 4 | 2 | 0 | - | - | 2 | 0 | - | - |
| June | A | 3 | 4 6 | 4 0 | 4 | 3 8 | 2 8 | 2 | 0 | - | - | 2 | 1 | 0 07 | - |
| | B | 12 | 9 3 | 5 7 | 6 | 10 6 | 6 7 | 2 | 0 | - | - | 2 | 0 | - | - |
| July | A | 4 | 4 5 | 2 7 | 4 | 1 4 | 1 2 | 2 | 0 | - | - | 2 | 1 | 0 1 | - |
| | B | 13 | 16 4 | 7 2 | 7 | 14 7 | 7 1 | 3 | 1 | 0 2 | - | 3 | 1 | 0 2 | - |
| August | A | 1 | 2 7 | - | 1 | 1 4 | - | 2 | 0 | - | - | 2 | 1 | 0 2 | - |
| | B | 12 | 12 4 | 6 0 | 5 | 13 0 | 6 3 | 4 | 1 | 0 06 | - | 4 | 1 | 2 0 | - |
| September | B | 12 | 20 2 | 11 1 | 7 | 33 3 | 13 2 | 4 | 2 | 1 0 | 0 7 | 4 | 0 | - | - |
| October | B | 13 | 93 4 | 20 4 | 6 | 87 6 | 30 1 | 4 | 3 | 0 5 | 0 4 | 4 | 0 | - | - |
| November | A | 4 | 3 0 | 2 7 | 4 | 2 3 | 1 8 | 2 | 0 | - | - | 2 | 0 | - | - |
| | B | 11 | 265 | 43 8 | 6 | 285 | 70 3 | 5 | 3 | 0 9 | 0 5 | 5 | 1 | 0 3 | - |
| December | A | 4 | 3 9 | 3 7 | 4 | 2 7 | 2 3 | 2 | 0 | - | - | 2 | 1 | 0 07 | - |
| | B | 11 | 218 | 63 5 | 7 | 229 | 89 1 | 3 | 3 | 0 4 | 0 3 | 3 | 0 | - | - |
| Annual | A | 29 | 4 6 | 2 8 | 30 | 3 8 | 1 8 | 17 | 0 | - | - | 17 | 4 | 0 2 | 0 05 |
| Summary | B | 149 | 265 | 22 8 | 77 | 285 | 34 5 | 36 | 17 | 1 2 | 0 23 | 36 | 4 | 2 0 | 0 09 |

*Location A is above the ANL site
Location B is below the ANL waste water outfall

In these samples the average uranium concentration was 21 $\mu\mu c/l$, and on this basis Argonne added approximately 20 $\mu\mu c$ of uranium per liter to the Creek (0.1% of the MPC).

In several of the below-outfall samples the total alpha activity was less than the uranium activity. The differences were greater than the 5 to 10% error in the analyses. This may be seen from Table VIII in the maximum columns for some of the months. This behavior, however, was not limited to the more active samples, but was noted at all levels of activity. The uranium was separated from some of these samples and the isotopic composition determined mass spectroscopically. This uranium was depleted in $U^{235}$ and $U^{234}$, and therefore had a lower specific activity than normal uranium. The $U^{235}$ content varied from 0.4 to 0.6% as compared with about 0.7% for $U^{235}$ in normal uranium. The amount of $U^{234}$ was about 1% of the $U^{235}$ content. Since the uranium concentration was determined fluorometrically and converted to activity units using the specific activity of normal uranium, the presence of depleted uranium will result in a value for uranium activity that is too large. For example, the below-site sample collected on November 20 contained 493 $\mu g$ of uranium per liter as determined fluorometrically. Since the specific activity of this uranium was 1.28 dpm/$\mu g$, the uranium activity was 285 $\mu\mu c/l$. The total alpha activity in this sample was 265 $\mu\mu c/l$. This is very good agreement in view of the 5% error in each of the measurements, including the $U^{234}$ determination.* If the specific activity of normal uranium were used, the uranium activity in this sample would be 333 $\mu\mu c/l$, about 25% higher than the value based on the measured isotopic ratio.

This type of error is, of course, inherent in the conversion of weight of uranium to activity in cases for which the specific activity or isotopic composition is variable. The samples were analyzed fluorometrically because the method is rapid and very sensitive for uranium of low specific activity. It was impractical, with the time and personnel available, to analyze each below-site sample for uranium by a counting method or to determine the isotopic composition of each sample. Since most samples contained normal uranium, the error in the average uranium activity calculated from the fluorometric results was affected only slightly by the presence of depleted uranium in some samples. This is particularly true when the correct activity or isotopic composition is known for the few samples of high activity. Another advantage in performing a fluorometric analysis in conjunction with a total activity measurement is the qualitative information obtained on the isotopic composition of the uranium. This information would not have been obtained if a counting method were used for uranium.

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*The specific activity of a mixture of uranium isotopes is very sensitive to $U^{234}$ content, and in depleted uranium this quantity is difficult to measure accurately since $U^{234}$ is present only in small amounts.

The uranium content of below-outfall water varied widely during the year, from 1.7 to 285 $\mu\mu c/l$ (1.4% of the MPC). This large variation also indicates that the uranium in below-outfall water was not of natural origin. Above the site the uranium concentration ranged only from 1.0 to 3.8 $\mu\mu c/l$.

In addition to uranium, small amounts of plutonium and thorium were also found in some of the below-outfall samples as a result of their presence in Argonne waste water. Seventeen of the thirty-six samples analyzed contained plutonium in excess of the minimum amount detectable by the analytical method used, namely, 0.05 $\mu\mu c/l$. The maximum plutonium concentration was 1.2 $\mu\mu c/l$, or 0.024% of the MPC; the average concentration was 0.23 $\mu\mu c/l$, or 0.005% of the MPC. Since it was not detected above the site, the plutonium found below the outfall was added by the waste water. Thorium was found in four of thirty-six below-outfall samples and in four of the seventeen above-site samples analyzed. The average below-outfall concentration was 0.09 $\mu\mu c/l$ (about 0.01% of the MPC); above-site the average concentration was 0.05 $\mu\mu c/l$. The results given in the table show that most of the thorium detected below the outfall originated in waste water. The identity of the thorium isotopes was not investigated. The average concentrations of plutonium and thorium were calculated assuming a concentration of 0.025 $\mu\mu c/l$, one-half of the minimum detectable amount, in those samples in which they were not detected. If it is assumed that the plutonium and thorium concentrations were zero in those samples in which the concentrations were less than the minimum detectable amounts, then the average thorium concentrations become 0.03 $\mu\mu c/l$ above the site and 0.08 $\mu\mu c/l$ below the site, but the plutonium average is not significantly affected.

The distribution of the alpha activity in Sawmill Creek during 1959 was similar to that found earlier. The alpha activity has usually been higher below the outfall, and this increase has been due primarily to uranium in Argonne waste water. Contributions by plutonium, thorium, and other alpha emitters have been smaller and less frequent. The average increase in alpha activity below the outfall has ranged from 7 $\mu\mu c/l$ in 1955 to 27 $\mu\mu c/l$ in 1953.

Concentrations of beta activity in Sawmill Creek during 1959 are given in Table IX. In addition to the results given in the table, approximately four analyses per month were made for $Ba^{140}$ and $I^{131}$ on below-outfall samples and on above-site samples when available. All $Ba^{140}$ concentrations were less than 3 $\mu\mu c/l$. One sample, collected on February 25, below the outfall, contained 17 $\mu\mu c$ of $I^{131}$ per liter; all other samples analyzed for radioiodine contained less than 3 $\mu\mu c/l$. These analyses, as well as the fission product analyses given in the table, were usually performed on samples selected at random.

TABLE IX

NONVOLATILE BETA ACTIVITY IN SAWMILL CREEK WATER, 1959

| Month | Loc.[a] | Total Beta Activity | | | $Sr^{90}$ | | | | $Sr^{89}$ | | | | $Cs^{137}$ | | | | $Ce^{144}$ | | | | $Ce^{141}$ | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | No. of Samples | $\mu\mu c/l$ | | No. of Samples | Samples >1 $\mu\mu c/l$ | | | No. of Samples | Samples >1 $\mu\mu c/l$ | | | No. of Samples | Samples >0.5 $\mu\mu c/l$ | | | No. of Samples | Samples >1 $\mu\mu c/l$ | | | No. of Samples | Samples >2 $\mu\mu c/l$ | | |
| | | | Max | Avg | | No. | Max | Avg | | No. | Max | Avg | | No. | Max | Avg | | No. | Max | Avg | | No. | Max | Avg |
| January | B | 13 | 342 | 63 | 4 | 3 | 2.4 | 1.7 | 4 | 3 | 10.3 | 5.1 | 1 | 1 | 3.1 | - | 2 | 1 | 3.0 | - | 2 | 1 | 3.1 | - |
| February | B | 12 | 859 | 145 | 5 | 5 | 14.2 | 4.4 | 4 | 3 | 13.2 | 6.7 | 3 | 2 | 6.0 | 3.5 | 2 | 2 | 2.5 | 1.1 | 2 | 0 | - | - |
| March | A | 4 | 64.6 | 47.8 | 4 | 4 | 2.2 | 1.7 | 4 | 4 | 17.6 | 14.7 | 1 | 0 | - | - | 4 | 3 | 2.7 | 2.2 | 4 | 1 | 3.0 | - |
| | B | 13 | 111 | 38 | 4 | 4 | 5.0 | 2.5 | 4 | 4 | 6.9 | 5.3 | 1 | 1 | 2.5 | - | 4 | 4 | 4.5 | 2.2 | 4 | 0 | - | - |
| April | A | 5 | 150 | 74 | 5 | 5 | 3.2 | 2.5 | 5 | 5 | 37.9 | 21.1 | 2 | 2 | 0.8 | - | 5 | 5 | 6.9 | 2.5 | 5 | 0 | - | - |
| | B | 13 | 90.0 | 39.7 | 6 | 6 | 5.7 | 2.8 | 6 | 5 | 11.5 | 6.7 | 2 | 1 | 4.1 | - | 5 | 5 | 2.7 | 2.1 | 5 | 2 | 31.0 | 18.8 |
| May | A | 4 | 51.7 | 41.7 | 2 | 2 | 3.8 | 2.9 | 2 | 2 | 19.0 | 18.3 | 2 | 0 | - | - | 2 | 2 | 1.9 | 1.6 | 2 | 1 | 5.3 | - |
| | B | 14 | 81.7 | 46.1 | 3 | 2 | 3.4 | 2.6 | 3 | 2 | 17.4 | 15.4 | 2 | 0 | - | - | 2 | 2 | 1.9 | 1.8 | 2 | 0 | - | - |
| June | A | 3 | 36.0 | 28.7 | 2 | 2 | 1.9 | 1.5 | 2 | 2 | 10.3 | 7.0 | 1 | 0 | - | - | 2 | 1 | 1.5 | - | 2 | 0 | - | - |
| | B | 12 | 146 | 32 | 3 | 3 | 2.2 | 1.9 | 3 | 2 | 6.9 | 6.0 | 2 | 1 | 3.2 | - | 4 | 4 | 34.0 | 9.4 | 4 | 0 | - | - |
| July | A | 4 | 33.5 | 18.2 | 2 | 2 | 4.5 | 3.8 | 2 | 2 | 12.2 | 7.9 | 1 | 1 | 0.5 | - | 2 | 1 | 11.0 | - | 2 | 0 | - | - |
| | B | 13 | 67.6 | 23.0 | 3 | 2 | 20.0 | 11.2 | 3 | 1 | 8.6 | - | 2 | 1 | 1.4 | - | 2 | 1 | 3.7 | - | 2 | 0 | - | - |
| August | A | 1 | 13.2 | - | 2 | 2 | 1.9 | 1.6 | 2 | 2 | 4.0 | 2.7 | 1 | 0 | - | - | 2 | 2 | 1.9 | 1.9 | 2 | 0 | - | - |
| | B | 12 | 34.4 | 15.4 | 2 | 2 | 1.3 | 1.2 | 2 | 2 | 3.8 | 2.7 | 3 | 2 | 1.8 | 1.8 | 2 | 1 | 2.6 | - | 2 | 0 | - | - |
| September | B | 12 | 161 | 36 | 4 | 4 | 5.2 | 2.3 | 2 | 1 | 1.2 | - | 1 | 1 | 3.6 | - | 2 | 1 | 1.0 | - | 2 | 0 | - | - |
| October | B | 13 | 52.6 | 18.2 | 2 | 1 | 1.2 | - | 2 | 0 | - | - | 1 | 1 | 4.0 | - | 2 | 1 | 1.2 | - | 2 | 0 | - | - |
| November | A | 4 | 15.7 | 8.4 | 2 | 2 | 1.0 | 1.0 | 2 | 2 | 1.2 | 1.2 | 1 | 0 | - | - | 1 | 0 | - | - | 1 | 0 | - | - |
| | B | 11 | 441 | 72 | 3 | 3 | 4.5 | 2.2 | 3 | 2 | 1.7 | 1.6 | 2 | 1 | 3.6 | - | 1 | 0 | - | - | 1 | 0 | - | - |
| December | A | 4 | 8.5 | 6.6 | 2 | 2 | 2.2 | 1.6 | 2 | 0 | - | - | 2 | 2 | 0.8 | 0.8 | 2 | 0 | - | - | 2 | 0 | - | - |
| | B | 11 | 246 | 66 | 2 | 1 | 11.8 | - | 2 | 0 | - | - | 1 | 1 | 3.0 | - | 3 | 1 | 21.7 | - | 3 | 1 | 6.8 | - |
| Annual Summary | A | 29 | 150 | 33 | 21 | 21 | 4.5 | 2.2 | 21 | 19 | 37.9 | 11.4 | 11 | 5 | 0.8 | 0.5 | 20 | 14 | 11 | 2.1 | 20 | 2 | 5.3 | 1.3 |
| | B | 149 | 859 | 49 | 41 | 36 | 20 | 2.9 | 38 | 25 | 17.4 | 4.2 | 21 | 13 | 6.0 | 2.0 | 31 | 23 | 34 | 3.2 | 31 | 4 | 31.0 | 2.4 |

[a]Location A is above the ANL site.
Location B is below the waste water outfall.

Analyses for other beta-emitting nuclides were made on some of the more active samples. These analyses showed that $Co^{58}$, $Co^{60}$, and the $UX_1$ and $UX_2$ daughters of uranium were present below the outfall and that fission products other than those previously mentioned were present at both locations. The results will be discussed in more detail below.

Activity from fallout was found at both sampling locations in Sawmill Creek. Beta activity added in Argonne waste water was found only below the outfall. Fallout activity was relatively high during the first part of the year and began to decrease in May. This may be seen from the total beta activity and $Sr^{89}$ concentrations above the site, where the Creek contained only naturally occurring activity (about 5 $\mu\mu c\beta/l$) and fallout activity. Although above-site water was not available during January and February, the results obtained for the Des Plaines River during these two months (see below) indicated that fallout in surface water was about the same in January and February as in May and March, respectively. The relative amounts of activity from fallout and from waste water in below-outfall water may be approximated as described below from the uranium and $Co^{58}$ concentrations below the outfall and from the fission product concentrations at both sampling locations. The measured beta-disintegration rate due to $UX_1$ and $UX_2$ in equilibrium with the parent uranium is equivalent to about 90% of the uranium concentration with the type of sample preparation, counting, and self-absorption and other corrections used in obtaining the total beta activities in water. Analyses for $UX_1$ in some of the below-outfall samples indicated that about 80% of the equilibrium amount was present on the average in below-site water. Thus, the contributions of $UX_1$ and $UX_2$ in waste water to the total beta activity below the outfall may be obtained from the amount of uranium added to the Creek in the waste water.

In most of the samples containing unusually high beta activities (>100 $\mu\mu c/l$) and relatively low alpha activities, the activity was due primarily to $Co^{58}$. This was the case for the maximum values listed in Table IX for January, February, March, June, September, and November, as well as for several other samples of lower activity. Since $Co^{58}$ was not detected above the site, it was added to the Creek below the outfall by Argonne waste water. Small amounts of $Co^{60}$ and probably $Co^{57}$ were also present, but these nuclides did not contribute more than 5% of the cobalt activity. The $Co^{58}$ had a large effect on the average beta activity for the year. Two samples, containing beta activities (due primarily to $Co^{58}$) of 859 and 441 $\mu\mu c/l$, respectively, contributed a total of about 17% of the beta activity found below the outfall during the year.

The concentrations of $Sr^{89}$ and $Sr^{90}$ as well as their ratios at both locations also aided in assigning sources to the beta activity below the outfall. For example, in March and April, the $Sr^{89}/Sr^{90}$ ratio was about 8.5 above-site and about 2.3 below the outfall. The $Sr^{89}$ concentrations were about three times lower below the outfall while the $Sr^{90}$ concentrations were 10 to 45% higher. It is evident that below the outfall the natural Creek water was diluted by Argonne waste water containing a lower $Sr^{89}/Sr^{90}$ ratio than the natural Creek water. To fit the data, the $Sr^{89}/Sr^{90}$ ratio in the waste water must have been less than four and the fraction of waste water in the Creek must have been 0.7 or greater. Therefore, in March and April one-half to two-thirds of the beta activity in those below-outfall samples analyzed for strontium resulted from Argonne waste water.

In January and February natural Creek water was not present in samples obtained below the outfall and the only fallout in the samples was that collected between the outfall and the sampling location, about 20 yards. In this case fallout should have made an even smaller contribution to the total beta activity than it did during March and April. This is confirmed by the Des Plaines River samples. In February the $Sr^{89}/Sr^{90}$ ratio in Sawmill Creek was 3.5, compared to about 20 for the Des Plaines River samples. It is, of course, possible for the strontium ratios in fallout and in the waste water, and for the concentrations of each strontium nuclide above and below the outfall, to be similar. In this situation, which occurred in May and June, the strontium results were not useful in distinguishing between fallout and Argonne activity except that since dilution of Creek water did occur at the outfall it may be concluded that some strontium was present in waste water. If this were not the case, the strontium activities would have been lower below the outfall, and this was observed in other years. When the differences above and below outfall are sufficiently great, the differences alone are sufficient to approximate the contribution of waste water to the total activity in the Creek. This could be done from the cerium results in June, the strontium results in July, and the total activities late in the year when fallout activity was relatively low. Differences in the ages of the fission products at the two sampling locations, which indicated a difference in origin, could also be seen from the cerium results in the spring.

From these considerations it is estimated that the waste water contribution to the total beta activity below the outfall ranged from about 55% in June to 95% in November and December, when the amount of fallout was at a minimum for the year. The average contribution for the year was about 33 $\mu\mu c/l$, about 70% of the average total beta activity of 49 $\mu\mu c/l$. Of the remaining activity, 2 to 3 $\mu\mu c/l$ were due to natural activity in the Creek, and approximately 13 $\mu\mu c/l$ were due to fallout. Above the site, the average beta activity was 33 $\mu\mu c/l$. Of this activity, 25 to 30 $\mu\mu c/l$ resulted from fallout and approximately 5 $\mu\mu c/l$ were due to activity naturally present in the stream.

The average uranium daughter concentration added to the Creek in waste water was 15 to 20 $\mu\mu c/l$, or about 0.1% of the MPC. The average $Co^{58}$ activity, also added by waste water, was about 10 $\mu\mu c/l$ (0.01% of the MPC). The nuclide $Co^{60}$ was present in concentrations equivalent to less than 0.01% of the MPC. The average $Sr^{90}$ concentration below the outfall, 2.9 $\mu\mu c/l$, amounted to 2.9% of the MPC. Approximately one-third of the $Sr^{90}$ was due to fallout, but all sources of non-natural activity must be considered when comparisons are made with the MPC. The average concentrations of the other fission products for which analyses were made were less than 0.05% of the MPC.

The total beta activities in Sawmill Creek in 1959 were in the range found earlier. From 1953 to 1958 the average yearly beta activity above the site varied from 27 to 44 $\mu\mu c/l$, compared with 33 $\mu\mu c/l$ in 1959. During the same period the average beta activity below the outfall varied from 36 to 47 $\mu\mu c/l$, compared with 49 $\mu\mu c/l$ during 1959. The origin and identity of the radioactive nuclides were also the same during 1959 as in previous years, except for $Co^{58}$ and $Co^{60}$. Radioactive cobalt was not detected in Sawmill Creek prior to 1959. Also, $Sr^{90}$ was detected with greater frequency during 1959 than earlier, particularly above the site, but the concentrations in individual samples were of the same order of magnitude.

2. <u>Des Plaines River</u>

Since Sawmill Creek empties into the Des Plaines River, the river was sampled above and below the mouth of Sawmill Creek to determine if the Creek had any effect on the radioactive content of the Des Plaines River. The total activities and uranium concentrations in Des Plaines River water are given in Table X. The alpha activities and uranium concentrations were very similar at both locations. The counting error in the alpha activity measurement was 20 to 30%, and the error in the uranium analysis was 10 to 20%. Most individual samples collected from both locations on the same day, and the annual averages, agreed within this error. Small differences greater than the analytical errors were found occasionally, but they occurred in both directions and appeared to depend on the amount of suspended solids in the samples. Samples containing only small amounts of insoluble material were not filtered prior to analysis, and the presence of dirt in the sample tends to increase the results. However, no important or consistent difference in alpha or uranium activity between the two locations was found. The range of values and the averages were essentially the same as found during previous years, and are normal for the Des Plaines River. Three pairs of samples were also analyzed for plutonium and thorium. At both locations the plutonium concentrations were less than 0.05 $\mu\mu c/l$, and the thorium concentrations ranged from 0.1 to 0.17 $\mu\mu c/l$. No differences between the locations were found.

The beta activities varied greatly during the year due to fallout. By December the beta activity had decreased to about 7 $\mu\mu c/l$, essentially normal for the River. Beta activities greater than this were due to fallout. This was confirmed by chemical separation of some fission products and by gamma-ray spectroscopy.

TABLE X

NONVOLATILE RADIOACTIVITY IN DES PLAINES RIVER WATER, 1959

| Date | Location* | Micromicrocuries per liter | | |
|---|---|---|---|---|
| | | Alpha Activity | Uranium | Beta Activity |
| January 28 | A | 0.7 | 0.43 | 41.1 |
| January 28 | B | 1.0 | 0.62 | 80.2 |
| February 19 | A | 1.5 | 0.95 | 164 |
| February 19 | B | 1.2 | 1.3 | 154 |
| February 27 | B | 1.3 | 0.62 | 158 |
| March 6 | A | 2.0 | - | 168 |
| March 6 | B | 2.1 | - | 125 |
| March 13 | A | 2.6 | - | 118 |
| March 13 | B | 1.2 | - | 79.0 |
| March 18 | A | 3.0 | 1.4 | 108 |
| March 18 | B | 1.5 | 1.2 | 74.4 |
| April 15 | A | 3.6 | 1.9 | 69.9 |
| April 15 | B | 4.2 | 2.7 | 69.5 |
| May 20 | A | 1.8 | 1.6 | 82.4 |
| May 20 | B | 2.2 | 1.8 | 45.6 |
| June 17 | A | 2.6 | - | 34.6 |
| June 17 | B | 3.4 | - | 42.1 |
| July 15 | A | - | 1.9 | - |
| July 15 | B | 2.0 | 1.3 | 27.5 |
| August 20 | A | - | 1.1 | - |
| August 20 | B | 1.3 | 1.4 | 16.6 |
| September 16 | A | 3.8 | 1.7 | 25.0 |
| September 16 | B | 2.0 | 1.6 | 22.7 |
| October 21 | A | 3.6 | 1.8 | 17.5 |
| October 21 | B | 4.5 | 1.8 | 19.0 |
| December 16 | A | 3.0 | 1.9 | 7.4 |
| December 16 | B | 3.3 | 1.8 | 6.7 |
| Average | A | 2.6 | 1.5 | 76 |
| | B | 2.2 | 1.5 | 66 |

*Location A is approximately 5 miles above the mouth of Sawmill Creek. Location B is approximately 2.3 miles below the mouth of Sawmill Creek.

Differences between samples collected on the same day from both locations, such as were found on January 28 and March 13, were due to differences in the amount of fallout activity. Usually the samples collected upstream from the mouth of Sawmill Creek were higher, and this is reflected in the averages. The average values, about 70 $\mu\mu c/l$, were about two times greater than in the past several years as a result of the relatively large amount of fallout during the first part of 1959. The $Sr^{90}$ content of the River at both locations decreased from 3.6 $\mu\mu c/l$ in February to about 1 $\mu\mu c/l$ in December; during the same period the $Sr^{89}$ content decreased from 68 $\mu\mu c/l$ to less than 1 $\mu\mu c/l$. The $Cs^{137}$ concentrations were about the same in the Des Plaines River as in Sawmill Creek above the site.

The results indicate that the activity in Sawmill Creek did not increase the activity in the Des Plaines River to any significant degree. This is reasonable in view of the large dilution of Sawmill Creek water by the much greater volume of water in the River.

3. Other Waters

The total activities in water samples collected from ponds on the ANL site are given in Table XI. The average alpha activity in the natural ponds, 2.4 $\mu\mu c/l$, was normal and similar to previous yearly averages. The two highest alpha activities in this group of samples, 5.9 and 3.5 $\mu\mu c/l$, were obtained in September and December from the same pond (location 11G in Figure 9). These results are unusual in that more than 75% of the surface water samples collected in the Chicago area contained less than 3 $\mu\mu c/l$. About 90% of the alpha activity in the two samples from the pond at 11G was due to uranium. This is also abnormal since uranium accounts for 50 to 75% of the total activity in ordinary surface water. Plutonium and thorium were not detected in these samples; the total beta activities were normal and several times greater than the alpha activities. The reason for this small increase in uranium content is not known at present, but is being investigated. Contamination by ANL operations is possible but unlikely. The samples collected from this pond in April and May contained normal amounts of both alpha activity and uranium, approximately 2 $\mu\mu c\alpha/l$ and 1.3 $\mu\mu cU/l$.

The highest alpha activities in the ANL ponds were found in water from a pond used occasionally to store contaminated waste water. As shown in the table, some of the samples from this pond contained alpha activities three to five times higher than in normal surface water. About three-quarters of this activity was due to uranium and plutonium. For example, the sample collected on July 31 contained 14 $\mu\mu c$ of alpha activity/l, 5.6 $\mu\mu c$ of uranium/l, and 3.5 $\mu\mu c$ of plutonium/l. The thorium concentration was only 0.2 $\mu\mu c/l$. This pond is also used to store excess uncontaminated waste water, and at times contains only rain water. However, the bottom of the pond has adsorbed considerable activity from contaminated water in the past, so some of the activity found in the 1959 samples may have been leached from the bottom by uncontaminated waste water or rain water present in the pond at the time of sampling.

TABLE XI

NONVOLATILE RADIOACTIVITY IN PONDS ON ANL SITE, 1959

| Date Collected | No. of Samples | Alpha Activity ($\mu\mu c/l$) | | Beta Activity ($\mu\mu c/l$) | |
|---|---|---|---|---|---|
| | | Max | Avg | Max | Avg |
| March 13 | 3 | 3.2 | 2.0 | 134 | 78 |
| | 1[a] | 3.8 | - | 59.6 | - |
| April 30 | 2 | 2.6 | 2.4 | 90.2 | 85.5 |
| | 1[a] | 6.7 | - | 72.1 | - |
| May 28 | 2 | 1.8 | 1.8 | 40.0 | 39.5 |
| | 1[a] | 6.3 | - | 75.0 | - |
| July 31 | 1 | 1.5 | - | 20.7 | - |
| | 1[a] | 14.0 | - | 78.0 | - |
| September 30 | 2 | 5.9 | 3.3 | 15.2 | 11.8 |
| December 8 | 3 | 3.5 | 3.0 | 15.6 | 14.0 |
| | 1[a] | 3.0 | - | 18.9 | - |
| Annual Summary | 18 | 14.0[a] | 3.6 | 134 | 49 |
| | 13[b] | 5.9 | 2.4 | 134 | 44 |

[a]Storage pond for contaminated waste water.

[b]Excluding samples from storage pond.

The beta activities in the ANL ponds were up to ten times higher than normal during the first part of the year as a result of fallout. Beta activities in the past have varied between 7 and 15 $\mu\mu c/l$ during periods of little fallout. By December the beta activities had decreased to nearly normal values. Similar beta activities were found throughout the year at sampling locations off the Argonne site. The decrease during the year followed the decrease in fallout in air and rain. Because of the large amount of fallout early in the year, the average beta activity, about 45 $\mu\mu c/l$, was about 20 $\mu\mu c/l$ higher than the averages for the past several years.

The total activities in samples collected within 25 miles of the Laboratory are listed in Table XII and summarized in Table XIII. Both the average and range of alpha activities were normal. The average for 1959 was 2.2 $\mu\mu c/l$; previous yearly averages ranged from 1.6 to 3.6 $\mu\mu c/l$. The beta activities were high during the first part of the year, but decreased to nearly normal values by the end of the year as the amount of fallout diminished. Except for Lake Michigan, which normally contains less activity than the other bodies of water, the range of activities during periods of high fallout, 12 to 118 $\mu\mu c\beta/l$, was essentially the same as found in the ANL ponds. The beta activity in the ponds and in the off-site samples decayed at the rate expected for mixed fission products.

The total activities in samples collected 100 miles from the Laboratory are given in Table XIV. The alpha activities were low and in the range normally found at these locations. The beta activities in the May samples averaged about three times higher than normal due to the presence of fission products from fallout. In the November samples the beta activities had decreased to normal values. The beta activities in the Illinois River at Starved Rock and Henry agreed very well with those in the samples collected from the River at Morris.

The samples collected at the confluence of the Des Plaines, Kankakee, and Illinois Rivers and those collected from the Illinois River farther downstream are of interest since Argonne waste water from Sawmill Creek follows this waterway and since the Dresden Nuclear Power Plant is situated at the confluence. No indication of activity other than fallout or natural activity was found in any of these samples.

D. Bottom Silt

The activities in the beds of lakes and streams are of interest for several reasons. Where bed conditions are appropriate for removing and concentrating activity from water, plant and animal life may be exposed to higher concentrations of activity than is 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 may indicate water contamination that was undetected in the past.

The total activities in bottom silt samples from Sawmill Creek and the Des Plaines River during 1959 are given in Table XV. In Sawmill Creek above the ANL site, the alpha activities varied from 20 to 26 $\mu\mu c/g$ and averaged 22 $\mu\mu c/g$. These are normal values for the Creek, and agree well with earlier results. Below the outfall the alpha activities ranged from 14 to 87 $\mu\mu c/g$, averaging 38 $\mu\mu c/g$. The increase in the maximum and average values below the outfall is reasonable in view of the increased activity found in water below the outfall, and is the result of the activity added to the stream in Argonne waste water.

TABLE XII

NONVOLATILE RADIOACTIVITY IN SURFACE WATER NEAR ANL, 1959

| Location | March | | April | | May | | July | | September | | December | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | $\mu\mu c\alpha/l$ | $\mu\mu c\beta/l$ | $\mu\mu c\alpha/l$ | $\mu\mu c\beta/l$ | $\mu\mu c\alpha/l$ | $\mu\mu c\beta/l$ | $\mu\mu c\alpha/l$ | $\mu\mu c\beta/l$ | $\mu\mu c\alpha/l$ | $\mu\mu c\beta/l$ | $\mu\mu c\alpha/l$ | $\mu\mu c\beta/l$ |
| Des Plaines River, 31st St., Brookfield[a] | 1.7 | 107 | - | - | 2.2 | 37.2 | 2.7 | 20.4 | 3.0 | 32.2 | 3.5 | 7.5 |
| Des Plaines River, Willow Springs[a] | 2.6 | 118 | 3.6 | 69.9 | 1.8 | 82.4 | - | - | 3.8 | 25.0 | 3.0 | 7.4 |
| Des Plaines River, Lemont[b] | 1.2 | 79.0 | 4.2 | 69.5 | 2.2 | 45.6 | 2.0 | 27.5 | 2.0 | 22.7 | 3.3 | 6.7 |
| Des Plaines River, Romeoville[b] | 1.9 | 82.4 | - | - | 1.6 | 35.0 | 1.7 | 19.9 | 0.92 | 12.0 | 3.2 | 9.2 |
| Illinois River, Morris | 2.5 | 26.2 | - | - | 2.1 | 18.0 | 3.9 | 13.3 | - | - | 2.8[c] | 7.9[c] |
| Confluence, Illinois, Kankakee and Des Plaines Rivers | - | - | - | - | 1.7 | 32.7 | 1.6 | 14.2 | - | - | - | - |
| DuPage River, Naperville | 3.2 | 41.4 | - | - | - | - | 2.6 | 21.5 | - | - | 3.3 | 7.6 |
| DuPage River, Channahon | 2.9 | 29.2 | - | - | - | - | - | - | - | - | 2.4 | 3.8 |
| Flag Creek, German Church and Wolf Roads | 2.7 | 68.7 | - | - | - | - | - | - | 1.2 | 10.1 | 0.8 | 13.0 |
| Cal-Sag Canal, 109th and Willow Springs Road | 1.3 | 53.6 | - | - | - | - | 1.3 | 12.7 | - | - | - | - |
| Salt Creek, Wolf Road., Western Springs | - | - | 3.5 | 62.1 | - | - | 3.7 | 15.8 | - | - | - | - |
| McGinnis Slough, U.S. Route 45 and Ill. Route 7 | - | - | 1.3 | 97.6 | - | - | - | - | 2.1 | 16.0 | - | - |
| Saganaskee Slough, 104th Ave and Sag Canal | - | - | 0.9 | 80.2 | - | - | - | - | 3.0 | 13.2 | 2.5 | 11.8 |
| Long Run Creek, 135th St. and Archer Ave | - | - | - | - | 1.9 | 15.8 | - | - | - | - | 1.7 | 5.5 |
| Lake Calumet, 98th St., Chicago | - | - | - | - | 1.3 | 60.2 | 1.6 | 26.4 | - | - | 2.6 | 10.7 |
| Lake Michigan, 98th St., Chicago | - | - | - | - | 0.22 | 7.8 | 0.39 | 5.2 | - | - | 0.36 | 1.9 |
| Lake Michigan Tap Water, South Side, Chicago | - | - | - | - | - | - | 0.22 | 5.2 | - | - | - | - |
| Sanitary and Ship Canal, Lemont | - | - | - | - | 0.54 | 31.9 | - | - | - | - | 0.66 | 5.8 |
| Fox River, Aurora | - | - | - | - | - | - | 2.9 | 28.9 | - | - | 1.5 | 7.1 |
| Sauk Trail Lake, Park Forest | - | - | - | - | - | - | 2.8 | 30.2 | - | - | 2.7 | 7.7 |
| Average | 2.2 | 67 | 2.7 | 75.9 | 1.6 | 36.7 | 2.1 | 18.6 | 2.3 | 18.7 | 2.3 | 7.6 |

[a]Above Sawmill Creek.

[b]Below Sawmill Creek.

[c]This sample collected November 24.

TABLE XIII

NONVOLATILE RADIOACTIVITY IN SURFACE WATER
NEAR ANL, 1959

| Month | Distance from ANL (miles) | No. of Samples | Alpha Activity ($\mu\mu c/l$) | | Beta Activity ($\mu\mu c/l$) | |
|---|---|---|---|---|---|---|
| | | | Max | Avg | Max | Avg |
| March | 10 | 7 | 3.2 | 2.1 | 118 | 79 |
| | 25 | 2 | 2.9 | 2.7 | 29.2 | 27.7 |
| April | 10 | 5 | 4.2 | 2.7 | 97.6 | 75.9 |
| May | 10 | 6 | 2.2 | 1.7 | 82.4 | 41.3 |
| | 25 | 4 | 2.1 | 1.3 | 60.2 | 29.7 |
| July | 10 | 6 | 3.7 | 2.3 | 27.5 | 19.6 |
| | 25 | 6 | 3.9 | 1.9 | 30.2 | 17.6 |
| September | 10 | 7 | 3.8 | 2.3 | 32.2 | 18.7 |
| December | 10 | 10 | 3.5 | 2.5 | 13.0 | 8.2 |
| | 25 | 5 | 2.8 | 1.9 | 10.7 | 6.3 |
| Annual Summary | 10 | 41 | 4.2 | 2.3 | 118 | 37 |
| | 25 | 17 | 3.9 | 1.9 | 60.2 | 18.3 |
| | 10 and 25 | 58 | 4.2 | 2.2 | 118 | 32 |

TABLE XIV

NONVOLATILE RADIOACTIVITY IN WATER
FROM REFERENCE SITES, 1959

| Location | May 26, 27 | | November 23, 24 | |
|---|---|---|---|---|
| | $\mu\mu c\alpha/l$ | $\mu\mu c\beta/l$ | $\mu\mu c\alpha/l$ | $\mu\mu c\beta/l$ |
| Fox River, Oak Point State Park, Wis. | 1.2 | 19.4 | 0.9 | 4.5 |
| Lake Delavan, Wis. | 0.2 | 17.1 | 1.6 | 7.2 |
| Illinois River, Henry, Ill. | 1.6 | 21.1 | - | - |
| Illinois River, Starved Rock State Park, Ill. | 1.7 | 19.0 | 3.4 | 7.8 |
| Lake Shafer, Monticello, Ind. | 1.6 | 13.9 | 2.8 | 4.7 |
| Magician Lake, Mich. | 0.2 | 20.9 | 1.3 | 7.4 |
| Lake Michigan, St. Joseph, Mich. | 0.2 | 11.3 | 0.3 | 2.0 |
| Kankakee River, Kankakee River State Park, Ind. | - | - | 1.3 | 4.2 |
| Average | 1.0 | 17.5 | 1.7 | 5.4 |

TABLE XV

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM SAWMILL CREEK AND DES PLAINES RIVER, 1959

| Date | Location[a] | Sawmill Creek | | Des Plaines River | |
|---|---|---|---|---|---|
| | | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ |
| January 28 | B | 87 | 147 | - | - |
| February 19 | B | 27 | 70 | - | - |
| March 13 | A | - | - | 20 | 95 |
| | B | - | - | 25 | 100 |
| March 18 | A | 24 | 69 | 18 | 77 |
| | B | 19 | 84 | 28 | 207 |
| April 15 | A | 21 | 132 | 21 | 68 |
| | B | 18 | 43 | 20 | 107 |
| May 20 | A | 26 | 122 | 23 | 75 |
| | B | 24[b] | 74[b] | 23 | 327 |
| June 17 | A | 22 | 95 | 23 | 369 |
| | B | 30[c] | 100[c] | 26 | 288 |
| July 15 | A | 21 | 88 | 24 | 308 |
| | B | 20 | 49 | 20 | 228 |
| August 20 | A | 21 | 91 | 21 | 57 |
| | B | 27 | 68 | 25 | 198 |
| September 16 | A | - | - | 27 | 210 |
| | B | 23 | 94 | 26 | 166 |
| October 21 | A | - | - | 16 | 49 |
| | B | 51 | 144 | 23 | 161 |
| November 18 | B | 75 | 208 | - | - |
| December 16 | A | 20 | 54 | 23 | 59 |
| | B | 56 | 203 | - | - |
| Average | A | 22 | 93 | 22 | 137 |
| | B | 38 | 107 | 24 | 198 |

[a]Sawmill Creek locations were:
- A - Above the ANL site.
- B - Below the waste water outfall.

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.

[b]Average of five samples. The individual results ranged from 14 to 35 $\mu\mu c\alpha/g$ and 42 to 108 $\mu\mu c\beta/g$.

[c]Average of four results. The individual results ranged from 15 to 47 $\mu\mu c\alpha/g$ and 45 to 226 $\mu\mu c\beta/g$.

Similar increases were noted in other years. The magnitudes of the increases have been variable, as may be seen from the range of alpha activities in the 1959 samples and from the average activities previously found. Earlier averages below the outfall varied from 68 $\mu\mu c/g$ in 1955 to 190 $\mu\mu c/g$ in 1957.

The presence of activity from waste water below the outfall is confirmed by the results given in Tables XVI and XVII. Uranium concentrations in bottom silt from Sawmill Creek above the outfall and from other locations (except Lake Michigan) ranged from 1.2 to 2.6 $\mu\mu c/g$. In the first 20 yards below the outfall in Sawmill Creek the uranium concentrations were significantly larger, from 3.3 to 31.7 $\mu\mu c/g$, and decreased to normal values after 20 yards. Plutonium was found below the outfall, but was not detected in concentrations greater than 0.1 $\mu\mu c/g$ at any other location. The maximum plutonium concentration (2.9 $\mu\mu c/g$) in the samples collected on May 20 occurred 20 yards below the outfall, whereas the uranium concentrations decreased regularly with distance from the outfall. Approximately the same behavior was observed in a series of samples collected in 1958. In the latter case the maximum plutonium concentration (26 $\mu\mu c/g$) occurred at the outfall, but the concentration at 20 yards (11 $\mu\mu c/g$) was higher than at either 10 or 30 yards. This effect is probably due to differences in the nature and properties of the bottom silt at different locations. Activity due to $Sr^{90}$ was found in the two samples below the outfall that contained the largest amount of beta activity. The $Sr^{90}$ concentrations in these samples, about 2 $\mu\mu c/g$, were appreciably higher than in samples from other locations. Consequently it appears that most of the uranium, plutonium, and $Sr^{90}$ in below-outfall bottom silt originated in Argonne waste water. There are insufficient data for $Cs^{137}$ to determine the principal source of the below-outfall concentrations of 4 and 8 $\mu\mu c/g$. Because of the relatively large amount of $Cs^{137}$ from fallout at other locations, additional analyses will be required. Thorium concentrations below the outfall were in the normal range, as would be expected from the low thorium content of Creek water at this location.

The average beta activity in Sawmill Creek was 25 to 35 $\mu\mu c/g$ above the normal values. Beta activity from fallout was found at various times at both sampling locations in the Creek, and activity from Argonne waste water was found below the outfall. Fallout activity was sufficiently high above the site so that the average beta activity at that location was only about 15% lower than below the outfall. Gamma-ray spectroscopy and beta-decay rates aided in distinguishing fallout activity from waste-water activity. The alpha-to-beta activity ratios also indicated a difference in origin between the abnormally high activities at the two sampling locations. Increases in the beta activity below the outfall were usually accompanied by increases in alpha activity, while the alpha activity remained normal in above-site samples containing abnormal beta activities. This is consistent with the relative alpha and beta activities found in water at both locations.

TABLE XVI

RADIOACTIVITY IN BED OF SAWMILL CREEK, 1959

| Location | Date | Micromicrocuries per gram | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | Alpha | Beta | U | Th | Pu | $Sr^{90}$ | $Cs^{137}$ |
| At waste water outfall | May 20 | 35 | 108 | 10 8 | 0 64 | 1 2 | <0 4 | - |
| 10 yards below outfall | May 20 | 30 | 93 | 5 9 | 2 3 | 1 3 | <0 5 | - |
| 20 yards below outfall | May 20 | 26 | 76 | 3 3 | 1 2 | 2 9 | <0 4 | - |
| 30 yards below outfall | May 20 | 14 | 72 | 1 3 | 0 75 | 0 30 | <0 2 | - |
| 40 yards below outfall | May 20 | 14 | 51 | 1 0 | 0 55 | 0 13 | <0 4 | - |
| 20 yards below outfall | Jan 28 | 87 | 147 | 23 0 | - | 18 2 | 2 4 | 8 1 |
| 20 yards below outfall | Nov 18 | 75 | 208 | 31 7 | - | 9 4 | 1 6 | 4 0 |
| Below outfall, East Bank | June 17 | 47 | 61 | - | - | - | - | - |
| Below outfall, 5 ft from East Bank | June 17 | 15 | 45 | - | - | - | - | - |
| Below outfall, 5 ft from West Bank | June 17 | 37 | 226 | - | - | - | - | - |
| Below outfall, West Bank | June 17 | 22 | 67 | - | - | - | - | - |
| Above ANL Site | May 20 | 26 | 122 | 2 2 | 3 1 | <0 1 | <0 2 | - |

TABLE XVII

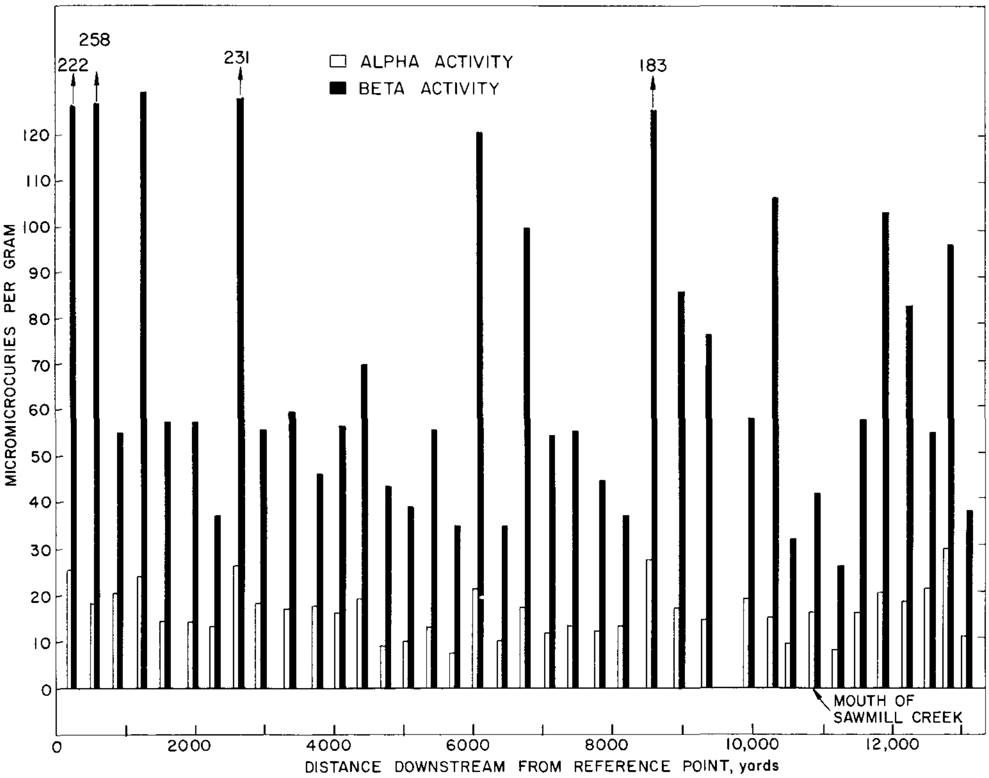
RADIOACTIVITY IN SELECTED BOTTOM SILT SAMPLES, 1959

| Location | Date | Micromicrocuries per gram | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | Alpha | Beta | U | Th | Pu | $Sr^{90}$ | $Cs^{137}$ |
| Des Plaines River, Lemont (below Sawmill Creek) | May 20 | 23 | 327 | 1 9 | 1 6 | <0 1 | 0 45 | - |
| Des Plaines River, Lemont (below Sawmill Creek) | Aug 20 | 25 | 198 | 1 3 | - | - | <0 4 | 2 7 |
| Des Plaines River, Willow Springs (above Sawmill Creek) | May 20 | 23 | 75 | 1 4 | 1 3 | <0 1 | 0 23 | - |
| Des Plaines River, Willow Springs (above Sawmill Creek) | Sept 29 | 26 | 231 | 2 6 | - | - | <0 3 | 3 7 |
| DuPage River, Naperville | Mar 13 | 94 | 94 | 2 3 | 8 2 | <0 1 | <3 | - |
| DuPage River, Channahon | Mar 13 | 29 | 195 | 1 4 | - | - | <2 | - |
| McGinnis Slough, U S Rt 45 and Ill Rt 7 | Sept 30 | 25 | 39 | 2 5 | 2 0 | <0 05 | - | - |
| Lake Michigan, 98th St , Chicago | May 28 | 4 | 28 | 0 53 | - | - | <0 04 | - |
| Storage Pond, ANL Site (10M) | Mar 13 | 485 | 546 | 23 4 | - | 272 | 8 5 | 49 6 |
| Pond, ANL Site (3F) | Mar 13 | 60 | 81 | 1 2 | 3 8 | <0 05 | - | - |
| Pond, ANL Site (3F) | Sept 30 | 30 | 54 | 2 0 | <0 1 | <0 1 | - | - |
| Pond, ANL Site (11G) | May 17 | 23 | 69 | 2 1 | 1 8 | <0 1 | <1 2 | - |
| Creek, ANL Site (13G) | June 11 | 27 | 197 | 1 5 | - | - | 0 45 | 7 1 |

In Des Plaines River bottom silt (Table XV), the alpha activities were normal at both locations, but the average beta activity below Sawmill Creek, 198 $\mu\mu c/g$, was about 60 $\mu\mu c/g$ higher than above the Creek. Among individual samples collected on the same day, differences in both directions were found, and the average difference in beta activity was due to fission product fallout, and not to activity added to the River by Sawmill Creek water. The amounts of activity in river-bottom sediment can vary considerably between locations depending on the nature of the bottom material. Samples consisting primarily of clay generally contain larger activities than those composed largely of sand or organic matter, and both types of bottom material can be found in the same stream. The samples collected from the Des Plaines River in March are examples of the variability of fallout activity in bottom silt. On March 13, the beta activities were essentially the same at Willow Springs (above Sawmill Creek) and at Lemont (below the Creek), while on March 18 the activity was about three times greater below the Creek. The highest activity in the March samples, 227 $\mu\mu c/g$, was found above Sawmill Creek at Brookfield (see Table XIX on p. 42). Similar variations have been found in the past, and to determine if there was any consistent pattern in the activity of the Des Plaines River bed in the vicinity of Sawmill Creek, the River bed was sampled in a two-day period in September. The results, plotted in Figure 12, show that the activity varied with location in a random manner, and no significant change occurred below the mouth of Sawmill Creek.

Figure 12

RADIOACTIVITY IN BED OF DES PLAINES RIVER, SEPT., 1959



None of the alpha activities was above normal. About one-third of the samples contained above-average concentrations of beta activity due to fallout, and the high samples were about equally distributed, on a percentage basis, above and below Sawmill Creek. In those samples containing normal amounts of beta activity, the alpha-to-beta ratio was fairly constant. However, unusually high beta activities were not accompanied by high alpha activities. If Argonne waste water had added significantly to the activity of the Des Plaines River bed below the Creek, high alpha as well as high beta activities would be expected. All the results indicate that Sawmill Creek had little, if any, effect on the activity in the bed of the Des Plaines River.

The total activities in ponds and in a few drainage ditches on the ANL site are given in Table XVIII.

TABLE XVIII

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM PONDS ON ANL SITE, 1959

| Date Collected | No. of Samples | Alpha Activity ($\mu\mu c/g$) | | Beta Activity ($\mu\mu c/g$) | |
|---|---|---|---|---|---|
| | | Max | Avg | Max | Avg |
| March 13 | 3 | 60 | 35 | 151 | 95 |
| | 1[a] | 485 | - | 546 | - |
| April 30 | 2 | 24 | 24 | 215 | 142 |
| | 1[a] | 486 | - | 644 | - |
| May 28 | 2 | 24 | 22 | 66 | 61 |
| | 1[a] | 473 | - | 648 | - |
| June 11 | 3 | 28 | 26 | 197 | 187 |
| July 31 | 7 | 28 | 25 | 439 | 167 |
| | 1[a] | 109 | - | 145 | - |
| September 30 | 2 | 30 | 26 | 65 | 60 |
| December 8 | 4 | 23 | 21 | 117 | 84 |
| | 1[a] | 505 | - | 506 | - |
| Annual Summary | 28 | 505[a] | 94 | 648[a] | 192 |
| | 23[b] | 60 | 25 | 439 | 125 |

[a]Storage pond for contaminated waste water.

[b]Excluding samples from storage pond.

Samples from the storage pond for contaminated waste water contained from 4 to 20 times the normal concentrations of alpha activity and 2 to 8 times the normal beta activity. The sample collected on March 13 was analyzed for uranium, plutonium, $Cs^{137}$, and $Sr^{90}$, and abnormally large amounts of these activities were found. The large amount of activity in the bed of this pond had accumulated over a period of years from the contaminated water stored at intervals in the pond.

Samples were collected on June 11, July 31, and December 8, from a drainage ditch at location 13G that carries scrubbing water from a burner used for disposal of metallic sodium. Both inactive and neutron-irradiated sodium is burned in this equipment, but contaminated scrub water is not knowingly discharged to the ditch. None of the samples contained detectable amounts of $Na^{22}$ or $Na^{24}$. The alpha activities were all normal, and some samples contained appreciable concentrations of fission products, presumably from fallout.

One sample collected on March 13, from a pond near the southern edge of the ANL site (location 3F) contained 60 $\mu\mu c$ of alpha activity per gram, about twice the normal concentration. The beta activity, 81 $\mu\mu c/g$, was only slightly above normal. Additional analyses performed on this sample (see Table XVII on page 38) showed that it contained a normal uranium concentration but about twice the normal thorium concentration. Similar results were obtained for a sample collected on March 13 from the DuPage River at Naperville. In this sample the alpha activity (94 $\mu\mu c/g$) was about three times the normal value and the thorium concentration (8.2 $\mu\mu c/g$) about four times normal. Growth curves of the thorium separated from these samples indicated that the thorium consisted primarily of $Th^{232}$ and its daughter $Th^{228}$. In both cases the thorium probably occurs naturally. The increased thorium concentrations apparently are localized, since other samples from the same beds contained normal amounts of thorium.

Except for the storage lagoon and the pond at location 3F, the alpha activities in samples collected on the ANL site were in the normal range. The average value, 25 $\mu\mu c\alpha/g$, was similar to the alpha activities found off the site. The average beta activity, 125 $\mu\mu c/g$, was about 30 $\mu\mu c/g$ higher than the average activity found near ANL in 1959 and 25 to 60 $\mu\mu c/g$ higher than the yearly averages found on the site prior to 1959. This was due to increased amounts of fallout activity; except for the storage lagoon there was no indication that the on-site samples contained significant amounts of activity from Argonne operations. The relative amounts of fission products of different half-lives were the same in samples collected on and off the site at the same time, indicating that the ages of the fission products were the same.

The total activities in bottom silt collected within 25 miles of ANL are listed in Table XIX and summarized in Table XX.

TABLE XIX

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT NEAR ANL, 1959

| Location | March | | April | | May | | July | | October | | December | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ |
| Des Plaines River, Brookfield[a] | 25 | 227 | - | - | 25 | 120 | 14 | 36 | 21 | 80 | 24 | 63 |
| Des Plaines River, Willow Springs[a] | 20 | 95 | 21 | 68 | 23 | 75 | 24 | 308 | 16 | 49 | 23 | 59 |
| Des Plaines River, Lemont[b] | 25 | 100 | 20 | 107 | 23 | 327 | 20 | 228 | 23 | 161 | - | - |
| Des Plaines River, Romeoville[b] | 23 | 154 | - | - | 24 | 151 | 26 | 185 | 25 | 130 | 23 | 90 |
| Illinois River, Morris | 15 | 31 | - | - | 3.2 | 23 | 7.4 | 18 | - | - | 3.1[c] | 18[c] |
| DuPage River, Naperville | 94 | 94 | - | - | - | - | 45 | 128 | - | - | - | - |
| DuPage River, Channahon | 29 | 195 | - | - | - | - | - | - | - | - | 25 | 38 |
| Flag Creek, German Church and Wolf Roads | 18 | 60 | - | - | - | - | - | - | 8 9 | 23 | 8 4 | 27 |
| Salt Creek, Wolf Road, Western Springs | - | - | 30 | 138 | - | - | 27 | 59 | - | - | - | - |
| McGinnis Slough, U S Route 45 and Ill Route 7 | - | - | 21 | 108 | - | - | - | - | 25 | 39 | - | - |
| Saganaskee Slough, 104th Ave. and Sag Canal | - | - | 19 | 50 | - | - | - | - | 14 | 65 | 19 | 57 |
| Fox River, Aurora | - | - | - | - | 15 | 149 | 15 | 75 | - | - | 12 | 32 |
| Long Run Creek, 135th St and Archer Ave | - | - | - | - | 23 | 78 | - | - | - | - | 22 | 66 |
| Lake Calumet, 111th St., Chicago | - | - | - | - | 16 | 55 | 14 | 34 | - | - | 23 | 62 |
| Lake Michigan, 98th St., Chicago | - | - | - | - | 4.1 | 28 | - | - | - | - | - | - |
| Sauk Trail Lake, Park Forest | - | - | - | - | - | - | 13 | 54 | - | - | 27 | 71 |
| Average | 31 | 120 | 22 | 94 | 17 | 112 | 21 | 113 | 19 | 78 | 19 | 53 |

[a]Above Sawmill Creek.

[b]Below Sawmill Creek.

[c]This sample collected November 24.

TABLE XX

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT NEAR ANL SITE, 1959

| Month | Distance from ANL (miles) | No. of Samples | Alpha Activity ($\mu\mu c/g$) | | Beta Activity ($\mu\mu c/g$) | |
|---|---|---|---|---|---|---|
| | | | Max | Avg | Max | Avg |
| March | 10 | 6 | 94 | 34 | 227 | 122 |
| | 25 | 2 | 29 | 22 | 195 | 113 |
| April | 10 | 5 | 30 | 22 | 138 | 94 |
| May | 10 | 5 | 25 | 24 | 327 | 150 |
| | 25 | 4 | 16 | 10 | 149 | 64 |
| July | 10 | 6 | 45 | 26 | 308 | 157 |
| | 25 | 4 | 15 | 12 | 75 | 45 |
| October | 10 | 7 | 25 | 19 | 161 | 78 |
| December | 10 | 6 | 24 | 20 | 90 | 60 |
| | 25 | 5 | 25 | 18 | 71 | 44 |
| Summary | 10 | 35 | 94 | 24 | 327 | 109 |
| | 25 | 15 | 29 | 15 | 195 | 59 |
| | 10 and 25 | 50 | 94 | 21 | 327 | 94 |

Results of additional analyses on some of the samples have been given in Table XVII. The alpha activities showed so significant change from previous years and, except for samples from the DuPage River at Naperville and Lake Michigan, were all in the normal range. The DuPage River samples have been discussed above. As usual, the sand bottom of Lake Michigan contained little alpha and uranium activity.

The beta activities in many of the samples were above-normal due to fission product fallout. The average beta activity, 94 $\mu\mu c/g$, was 7 $\mu\mu c/g$ lower than in 1958, but 30 to 40 $\mu\mu c/g$ higher than in samples collected from 1952-1957. In most of the monthly groups of samples collected in 1959, samples collected 10 miles from the Laboratory contained higher beta activities than the 25-mile samples. This was due to normal variations in fallout activity between locations and was not related to distance from the Laboratory. The 25-mile locations include bodies of water, such as Lake Michigan and the Illinois River, that usually contain below average amounts of fallout, and this decreases the average beta activity as well as the probability of obtaining a sample at 25 miles that contains very high fallout activities.

The alpha and beta activities in bottom silt from the reference sites (Table XXI) were very similar to those found earlier. No unusual amounts of fallout activity were found in these samples although water samples collected in May from these locations contained readily detectable amounts of fallout.

TABLE XXI

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM REFERENCE SITES, 1959

| Location | May 26, 27 | | November 23, 24 | |
|---|---|---|---|---|
| | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ |
| Fox River, Oak Point State Park, Wis. | 5.0 | 36 | 9.8 | 37 |
| Lake Delavan, Delavan, Wis. | 11 | 51 | 5.6 | 25 |
| Illinois River, Henry, Ill. | 9.7 | 39 | - | - |
| Illinois River, Starved Rock State Park, Ill. | 1.4 | 5.8 | - | - |
| Lake Shafer, Monticello, Ind. | 5.8 | 31 | 4.1 | 34 |
| Magician Lake, Mich. | 3.5 | 20 | 10 | 34 |
| Kankakee River, Kankakee River State Park, Ind. | - | - | 6.7 | 35 |
| Average | 6 | 31 | 7 | 33 |

## E. Surface Soil

The total activities in surface soil collected on the ANL site are given in Table XXII. The samples collected near a uranium storage shed at location 12L are listed separately since these were the only soil samples that contained activity from ANL operations. These samples contained from 2 to 70 times the normal alpha and beta activities of about 25 $\mu\mu c\alpha/g$ and 60-75 $\mu\mu c\beta/g$. As indicated in Table XXIII, the contamination was primarily due to uranium in the sample analyzed chemically for uranium and plutonium. The presence of abnormally high amounts of uranium in other samples from the same area was confirmed by gamma-ray spectroscopy. During 1959 this contamination was confined to within 30 to 40 feet of the shed.

TABLE XXII

NONVOLATILE RADIOACTIVITY IN SOIL ON ANL SITE, 1959

| Date Collected | No. of Samples | Alpha Activity ($\mu\mu c/g$) | | Beta Activity ($\mu\mu c/g$) | |
|---|---|---|---|---|---|
| | | Max | Avg | Max | Avg |
| March 13 | 3 | 22 | 19 | 73 | 53 |
| | 1[a] | 54 | - | 123 | - |
| April 30 | 3 | 23 | 20 | 75 | 71 |
| May 28 | 4 | 29 | 21 | 150 | 99 |
| July 31 | 3 | 22 | 20 | 93 | 77 |
| | 3[a] | 123 | 95 | 254 | 175 |
| September 30 | 2 | 32 | 29 | 56 | 56 |
| December 8 | 4 | 23 | 18 | 130 | 78 |
| | 5[a] | 1870 | 542 | 4140 | 1095 |
| Annual Summary | 28 | 1870 | 123 | 4140 | 270 |
| | 19[b] | 29 | 21 | 150 | 75 |

[a]Samples collected near a uranium storage shed.

[b]Excluding uranium storage shed area.

TABLE XXIII

RADIOACTIVITY IN SELECTED SOIL SAMPLES, 1959

| Location | Date | Micromicrocuries per Gram | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | Alpha | Beta | U | Th | Pu | $Sr^{90}$ | $Cs^{137}$ |
| Willow Springs | Mar. 13 | 31 | 70 | 1.8 | - | - | <0.6 | - |
| Morris | Mar. 13 | 27 | 107 | 2.3 | - | - | 0.7 | - |
| Naperville | July 29 | 22 | 93 | 1.0 | 0.04 | <0.05 | 0.6 | 1.7 |
| Brookfield | Sept. 30 | 21 | 147 | 2.1 | 0.57 | <0.05 | 1.0 | 2.8 |
| Brookfield | Dec. 7 | 21 | 191 | 1.6 | - | - | 1.2 | 5.6 |
| ANL Site (12-0) | Dec. 8 | 18 | 130 | 1.9 | - | - | 0.39 | 1.0 |
| ANL Site (12L)* | Dec. 8 | 369 | 417 | 326 | - | <0.03 | - | - |

*Collected near uranium storage shed.

The alpha and beta activities in all other samples collected on the ANL site were similar to those found near the site in 1959 and to those found both on and near the site in previous years. The alpha activities varied from 14 to 29 $\mu\mu c/g$ and averaged 21 $\mu\mu c/g$. These values are all in the normal range. The beta activities varied from normal values of about 60 $\mu\mu c/g$ up to 130 $\mu\mu c/g$. Gamma-ray spectra of many of the samples and the $Cs^{137}$ and $Sr^{90}$ concentrations given in Table XXIII indicated that above normal beta activities were due to fission product fallout.

The total activities in samples collected off the ANL site are given in Tables XXIV and XXV. At these locations the alpha activities were very similar in range and average to those found earlier. Beta activity from fallout was present in varying amounts in all samples. Near the ANL site the average fallout activity decreased only slightly in the last half of the year relative to that in the spring, although pronounced decreases were noted in air and rain. The sample collected in Brookfield on December 7 contained the highest concentrations of beta activity, $Sr^{90}$, and $Cs^{137}$ found during the year.

TABLE XXIV

NONVOLATILE RADIOACTIVITY IN SOIL NEAR ANL, 1959

| Date Collected | Distance from ANL (miles) | No. of Samples | Alpha Activity ($\mu\mu c/g$) | | Beta Activity ($\mu\mu c/g$) | |
|---|---|---|---|---|---|---|
| | | | Max | Avg | Max | Avg |
| March 13 | 10 | 5 | 31 | 22 | 98 | 66 |
| | 25 | 3 | 27 | 23 | 107 | 79 |
| April 30 | 10 | 3 | 22 | 19 | 118 | 103 |
| May 26,27 | 10 | 3 | 24 | 20 | 119 | 92 |
| | 25 | 4 | 24 | 19 | 66 | 49 |
| July 29,30,31 | 10 | 5 | 25 | 20 | 93 | 72 |
| | 25 | 6 | 26 | 18 | 69 | 53 |
| September 30 | 10 | 5 | 27 | 22 | 147 | 85 |
| December 7,8 | 10 | 6 | 28 | 20 | 191 | 78 |
| | 25 | 7 | 25 | 17 | 94 | 59 |
| Annual Summary | 10 | 27 | 31 | 21 | 191 | 80 |
| | 25 | 20 | 27 | 19 | 107 | 58 |
| | 10 and 25 | 47 | 31 | 20 | 191 | 71 |

TABLE XXV

NONVOLATILE RADIOACTIVITY IN SOIL
AT REFERENCE SITES, 1959

| Location | May 26,27 | | November 23,24 | |
|---|---|---|---|---|
| | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ |
| Oak Point State Park, Wis. | 17 | 43 | 13 | 28 |
| Delavan, Wis. | 19 | 37 | 22 | 44 |
| Henry, Ill. | 21 | 42 | - | - |
| Starved Rock State Park, Ill. | 27 | 119 | 16 | 56 |
| Monticello, Ind. | 21 | 72 | 14 | 53 |
| St. Joseph, Mich. | 10 | 42 | 7 | 28 |
| Magician Lake, Mich. | 11 | 45 | 13 | 34 |
| Kankakee River State Park, Ind. | - | - | 10 | 26 |
| Average | 18 | 57 | 14 | 38 |

At the reference sites the beta activities decreased from an average of 57 $\mu\mu c/g$ in May to 38 $\mu\mu c/g$ in November, due to a decrease in fallout activity in most of the November samples. The average beta activity during 1959, 48 $\mu\mu c/g$, was 20 to 25% higher than in the past several years.

F. Plants

Plant sampling was limited to grass because it was available at all locations, and intercomparison of results would be more reliable if only one type of plant were collected. The total activities in grass collected during 1959 are given in Tables XXVI, XXVII, and XXVIII in terms of the oven-dried sample. Additional results for some of the samples are given in Table XXIX. Excluding the samples collected near the uranium storage shed on the ANL site, the alpha activities at all locations were in the range found previously, and no unusual values were found in the individual samples. The annual averages varied slightly between locations; on the site the average was 1.2 $\mu\mu c/g$, whereas off the site the averages were somewhat lower, 0.9 $\mu\mu c/g$ near ANL and 0.6 $\mu\mu c/g$ at the reference sites.

Three of the four samples collected near the uranium storage shed contained alpha activities above the normal range of 0.4 to 3 $\mu\mu c/g$. In these samples the alpha activities ranged from 4 to 96 $\mu\mu c/g$. As was the case for soil samples from this area, the additional alpha activity was due to uranium. When the uranium concentration was sufficiently large, as in one sample collected on December 8, the $UX_1$ and $UX_2$ daughters also produced a marked increase in the total beta activity.

TABLE XXVI

NONVOLATILE RADIOACTIVITY IN GRASS ON ANL SITE, 1959

| Date Collected | No. of Samples | Alpha Activity ($\mu\mu c/g$) | | Beta Activity ($\mu\mu c/g$) | |
|---|---|---|---|---|---|
| | | Max | Avg | Max | Avg |
| March 13 | 3 | 1.6 | 1.4 | 490 | 464 |
| April 30 | 2 | 3.1 | 2.7 | 838 | 741 |
| May 28 | 4 | 2.0 | 1.0 | 164 | 137 |
| June 11 | 1 | 0.6 | - | 85 | - |
| July 31 | 2 | 0.8 | 0.6 | 69 | 52 |
| | 2[a] | 4.0 | 3.4 | 80 | 74 |
| September 30 | 2 | 2.2 | 1.4 | 78 | 59 |
| December 8 | 5 | 1.8 | 1.1 | 35 | 25 |
| | 2[a] | 96 | 52 | 114 | 73 |
| Annual Summary | 23 | 96 | 5.8 | 838 | 180 |
| | 19[b] | 3.1 | 1.2 | 838 | 203 |

[a]Samples collected near a uranium storage shed.
[b]Excluding uranium storage shed area.

TABLE XXVII

NONVOLATILE RADIOACTIVITY IN GRASS NEAR ANL, 1959

| Date Collected | Distance from ANL | No. of Samples | Alpha Activity ($\mu\mu c/g$) | | Beta Activity ($\mu\mu c/g$) | |
|---|---|---|---|---|---|---|
| | | | Max | Avg | Max | Avg |
| March 13 | 10 | 6 | 1.7 | 1.4 | 645 | 481 |
| | 25 | 2 | 1.2 | 1.1 | 661 | 615 |
| April 30 | 10 | 3 | 1.5 | 0.9 | 813 | 486 |
| May 26 | 10 | 3 | 0.45 | 0.32 | 136 | 109 |
| | 25 | 3 | 1.7 | 0.9 | 431 | 213 |
| June 29,30 | 10 | 5 | 3.2 | 1.3 | 97 | 74 |
| | 25 | 6 | 1.1 | 0.7 | 94 | 71 |
| September 30 | 10 | 5 | 0.91 | 0.62 | 58 | 47 |
| December 7,8 | 10 | 6 | 1.0 | 0.7 | 36 | 30 |
| | 25 | 4 | 2.8 | 1.2 | 47 | 34 |
| Annual Summary | 10 | 28 | 3.2 | 0.9 | 813 | 195 |
| | 25 | 15 | 2.8 | 0.9 | 661 | 162 |
| | 10 and 25 | 43 | 3.2 | 0.9 | 813 | 183 |

TABLE XXVIII

NONVOLATILE RADIOACTIVITY IN GRASS
FROM REFERENCE SITES, 1959

| Location | May 26,27 | | November 23,24 | |
|---|---|---|---|---|
| | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ | $\mu\mu c\alpha/g$ | $\mu\mu c\beta/g$ |
| Oak Point State Park, Wis | 0 65 | 115 | 0 63 | 33 |
| Delavan, Wis | 0 25 | 132 | 0 62 | 54 |
| Starved Rock State Park, Ill | 0 29 | 110 | 0 55 | 26 |
| Monticello, Ind | 0 57 | 57 | 0 90 | 46 |
| St Joseph, Mich | 0 44 | 258 | 0 96 | 46 |
| Magician Lake, Mich | 0 30 | 153 | 0 95 | 75 |
| Kankakee River State Park, Ind | - | - | 0 68 | 32 |
| Average | 0 42 | 138 | 0 76 | 45 |

TABLE XXIX

RADIOACTIVITY IN SELECTED GRASS SAMPLES, 1959

| Location | Date | Micromicrocuries per Gram | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | Alpha | Beta | U | Th | Pu | $Sr^{90}$ | $Cs^{137}$ |
| Morris | Mar 13 | 1 0 | 569 | 0 074 | 0 046 | 0 072 | 3 7 | 5 0 |
| Willow Springs | Mar 13 | 0 7 | 217 | 0 027 | - | - | 1 1 | 1 0 |
| 104th Ave and Sag Canal | Apr 30 | 1 5 | 813 | 0 12 | 0 020 | 0 015 | 5 8 | 6 8 |
| ANL Site (10M) | Apr 30 | 3 1 | 838 | 0 33 | 0 36 | 0 14 | 6 3 | 10 7 |
| ANL Site (12L) | May 28 | 2 0 | 99 | 1 1 | 0 014 | <0 005 | 0 83 | 0 63 |
| Bemis Woods (near Western Springs) | July 29 | 3 2 | 97 | 0 22 | <0 005 | <0 005 | - | - |
| ANL Site (11G) | Sept 30 | 2 2 | 78 | 0 21 | - | - | 0 79 | 0 18 |

Samples from all locations contained fission products from fallout. The fallout activity was high early in the year and decreased sharply during the latter half of 1959. This is apparent from the total beta activities and from the $Sr^{90}$ and $Cs^{137}$ results in Table XXIX. While fallout activity in grass paralleled that in air and rain during the year, this correlation was not as obvious for soil or bottom silt. Thus, grass appears to be a more sensitive indicator for recent fallout than most other types of surface samples. As measured by the beta activities in grass, fallout was fairly uniform at all locations; samples collected from all locations at the same time contained similar concentrations of beta activity. The beta activities early in the year were the highest found since sampling was begun in 1952. A decrease by a factor of about ten in the last half of the year resulted in annual averages not greatly different from some of the earlier values.

The plutonium concentrations in grass (see Table XXIX) were small but measurable in three of the five samples analyzed. The plutonium presumably was derived from fallout. The plutonium content appears to be of the same order as the thorium concentration, and therefore the same conditions may permit, or discriminate against, the uptake of both elements. It is planned to obtain additional data on the occurrence of plutonium and thorium in grass.

Plutonium concentrations in a number of biological samples were reported by Isotopes, Inc. (Westwood, N. J.) in the October, 1959 Quarterly Statement on Fallout by the U. S. Atomic Energy Commission. This report gives plutonium concentrations ranging from 0.13 to 0.80 $\mu\mu c/g$ for alfalfa and wheat ash. In terms of the ashed sample, the results in Table XXIX range from 0.14 to 1.1 $\mu\mu c/g$, so the plutonium concentrations in the two groups of samples are of the same order of magnitude.

## G. Animals

Only a limited number of samples were collected and analyzed. Animal collection and sample preparation is time consuming, and with the facilities available the value of the results obtainable from animals was not believed to justify an extensive sampling program.

Two samples of fish, blue gills and bullheads, were collected from one of the ponds on the site in November. The samples contained 0.4 and 0.6 $\mu\mu c\alpha/g$ and 19 and 19.1 $\mu\mu c$ /g, respectively. Total activities in fish collected from 1953 through 1958 from the same pond ranged from 0.1 to 1.7 $\mu\mu c\alpha/g$ and 9.6 to 24 $\mu\mu c\beta/g$, and therefore there has been no long-term accumulation of activity in the fish population of the pond.

A field mouse obtained in September contained 0.25 $\mu\mu c\alpha/g$ and 20.9 $\mu\mu c\beta/g$. Mice collected in other years contained 0.1 to 0.3 $\mu\mu c\alpha/g$ and 6.8 to 12.4 $\mu\mu c\beta/g$. A white deer was obtained from the ANL site in February, and portions of the heart, lung, kidney, and liver were analyzed for total activity. The alpha activities varied from 0.1 to 0.9 $\mu\mu c/g$; the beta activities from 21 to 24 $\mu\mu c/g$. In 1957 the same organs from another deer on the site contained activities ranging from 0.3 to 0.8 $\mu\mu c\alpha/g$ and 2.5 to 9 $\mu\mu c\beta/g$. The alpha activities in the deer and mouse have not changed from earlier samples; the beta activities were significantly higher in 1959, but additional analyses will be required to determine if the increases were due to the $K^{40}$ content or to other activities.