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ENVIRONMENTAL RADIOACTIVITY AT ARGONNE NATIONAL LABORATORY

Report for the Year 1958

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TABLE OF CONTENTS

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I. SUMMARY

The radioactive contents of several types of materials collected and analyzed during 1958 are given in this report. The materials were obtained from the ANL site, and from locations approximately 10, 25, and 100 miles from the Laboratory. The 10 and 25-mile locations are referred to as "near ANL," while the 100-mile locations are termed "reference sites." The principal purpose of this work was to determine if any radioactivity in the environment originated from operatives at Argonne. In order to evaluate this, it was also necessary to estimate the contributions of natural and fallout activities to the total activities.

The sampling locations are given in Part II. Since ANL waste water is discharged into Sawmill Creek, which in turn empties into the Des Plaines River, special emphasis was placed on sampling these streams. Most of the results were obtained by counting the total alpha and beta activities after suitable preparation of the sample. Some of the samples were analyzed for specific elements and nuclides. The average total activities in samples of surface water, lake and stream beds (bottom silt), soil, plants, and air filters are given in Figures 1 to 3. For comparison purposes, the results obtained for similar samples from 1952 through 1958 are given in Figures 4 to 8.

Fission product activity from nuclear detonations was found in most samples from all locations. Fallout activity was greatest during the spring and fall, and was particularly noticeable in air, precipitation, and plant samples. The total long-lived beta activity in air filter samples was primarily due to fission product fallout and has increased regularly since 1954 from about 0.5 $\mu\mu c/m^3$ to approximately 3 $\mu\mu c/m^3$ in 1958. In 1953, the first year of air sampling, the total beta activity averaged 1.4 $\mu\mu c/m^3$. The total alpha activity has remained essentially constant over this same period of time.

Radioactivity originating at Argonne was detected in water and bottom silt collected from Sawmill Creek below the waste water outfall. At this location the average total alpha activity was 13.4 micromicrocuries ($\mu\mu c$) per liter, or about 10 $\mu\mu c/l$ higher than Sawmill Creek water obtained upstream from the Laboratory site. This difference is attributed primarily to natural uranium added to the stream in Argonne waste water. As in previous years, other alpha-emitting nuclides made only small

Figure 1

AVERAGE RADIOACTIVITY IN SURFACE WATER, 1958

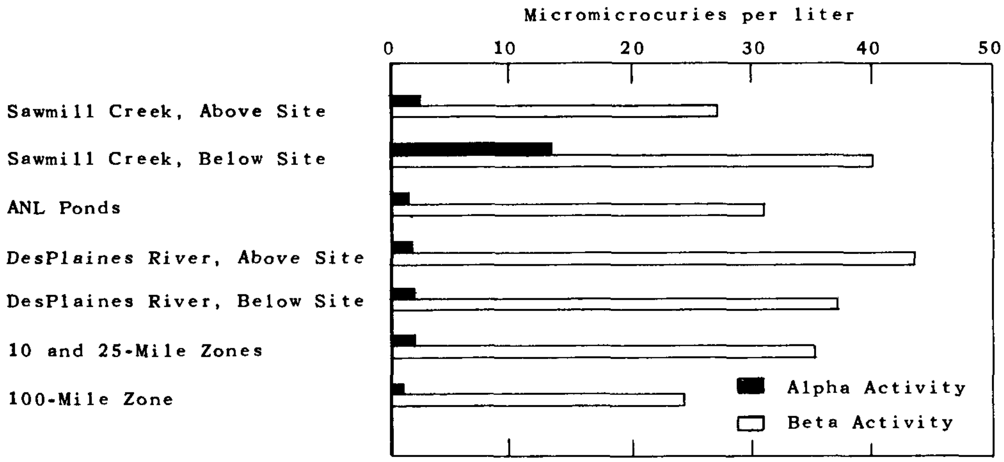


Figure 2

AVERAGE RADIOACTIVITY IN BOTTOM SILT, 1958

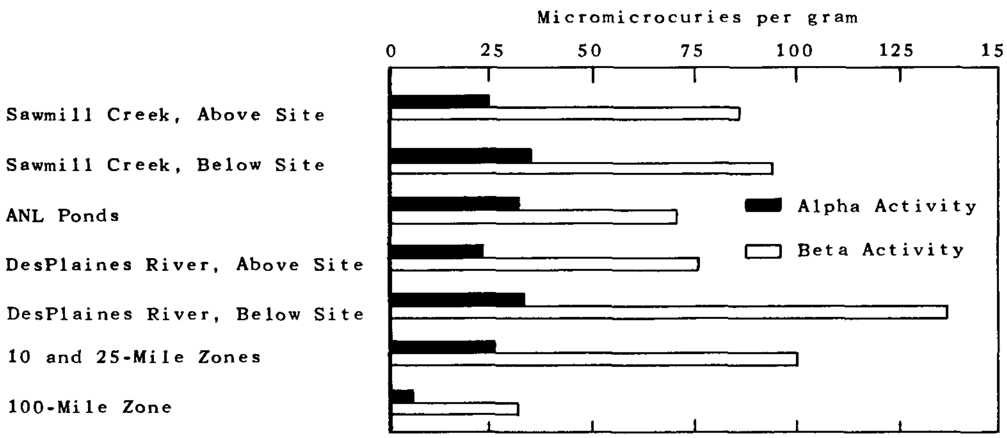


Figure 3

AVERAGE RADIOACTIVITY IN SOIL AND PLANTS, 1958

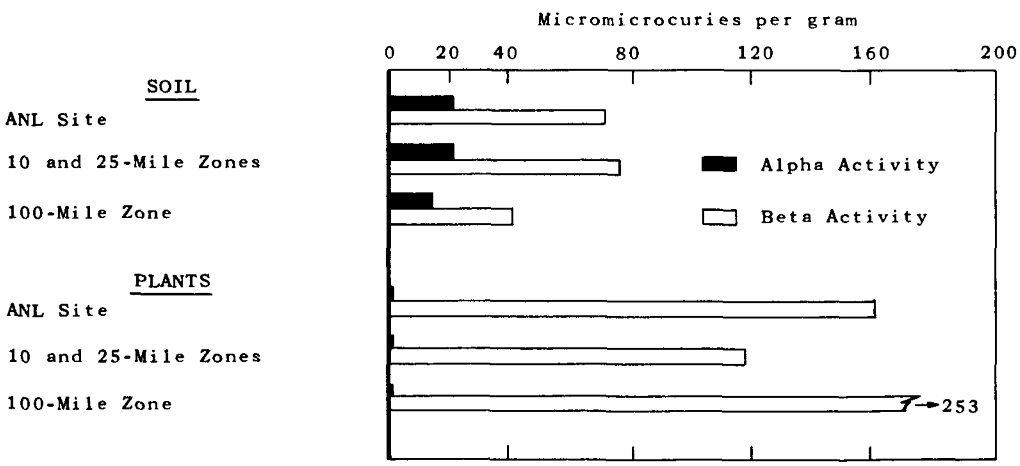


Figure 4

AVERAGE RADIOACTIVITY IN SURFACE WATER, 1952-58

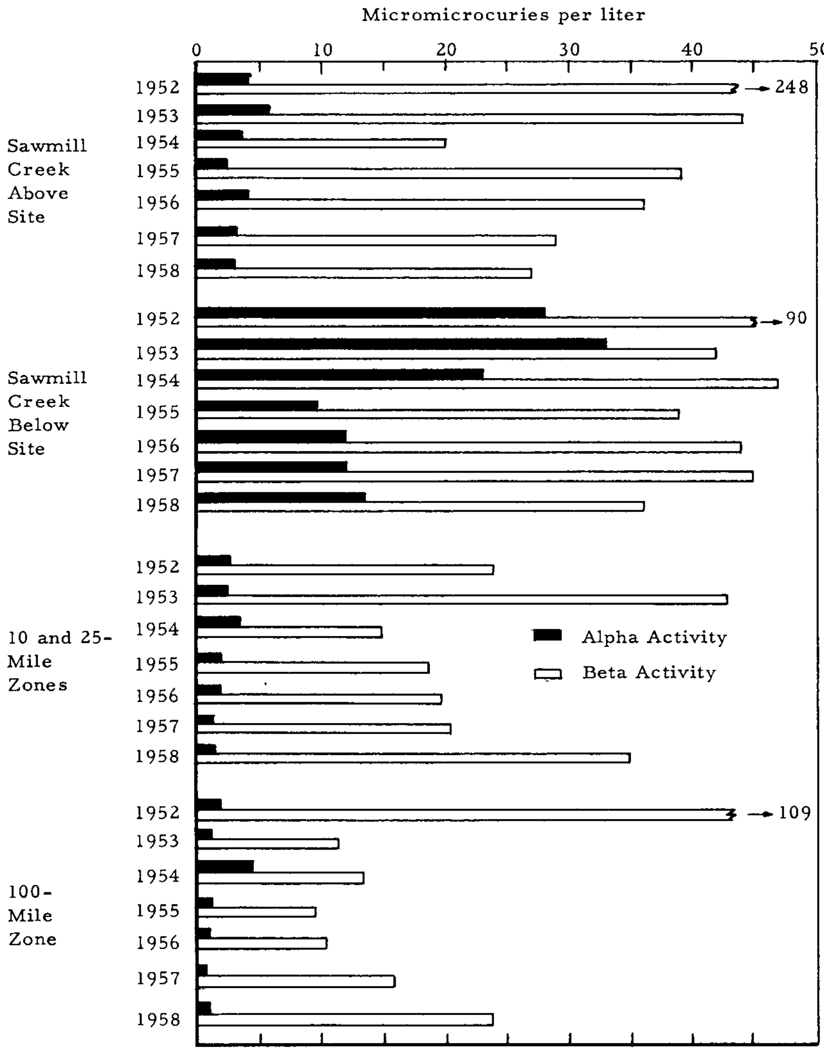


Figure 5

AVERAGE RADIOACTIVITY IN BOTTOM SILT, 1952-58

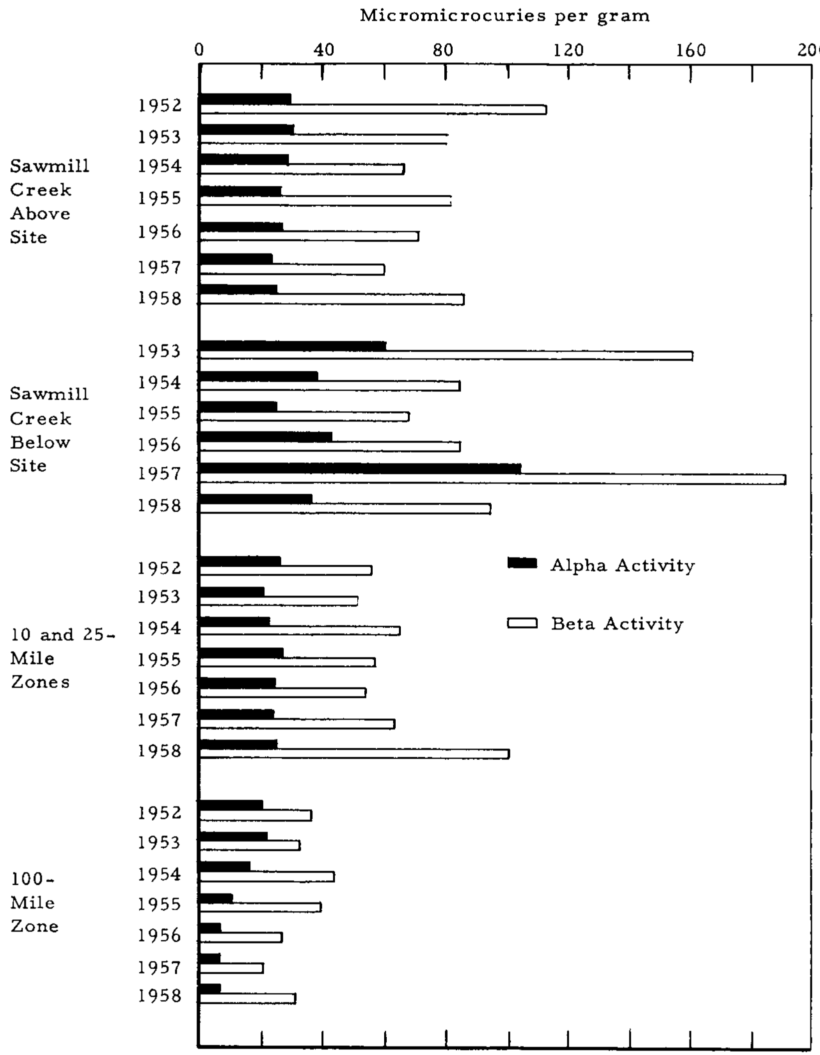


Figure 6

AVERAGE RADIOACTIVITY IN SURFACE SOIL, 1952-58

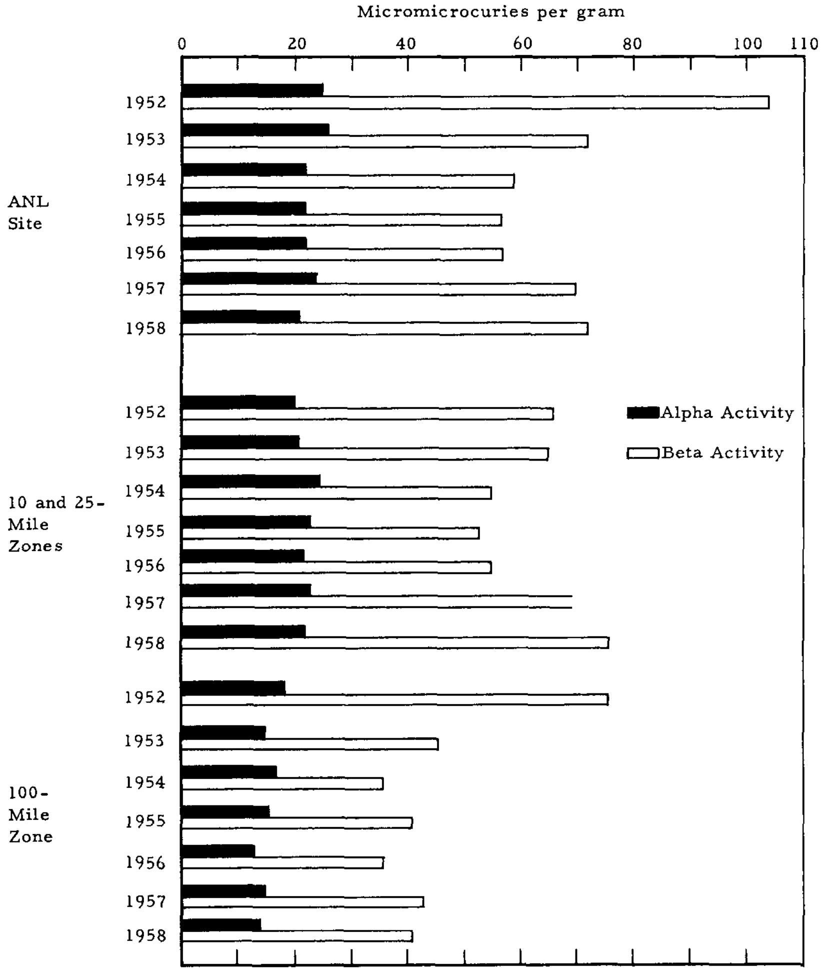


Figure 7

AVERAGE RADIOACTIVITY IN PLANTS, 1952-58

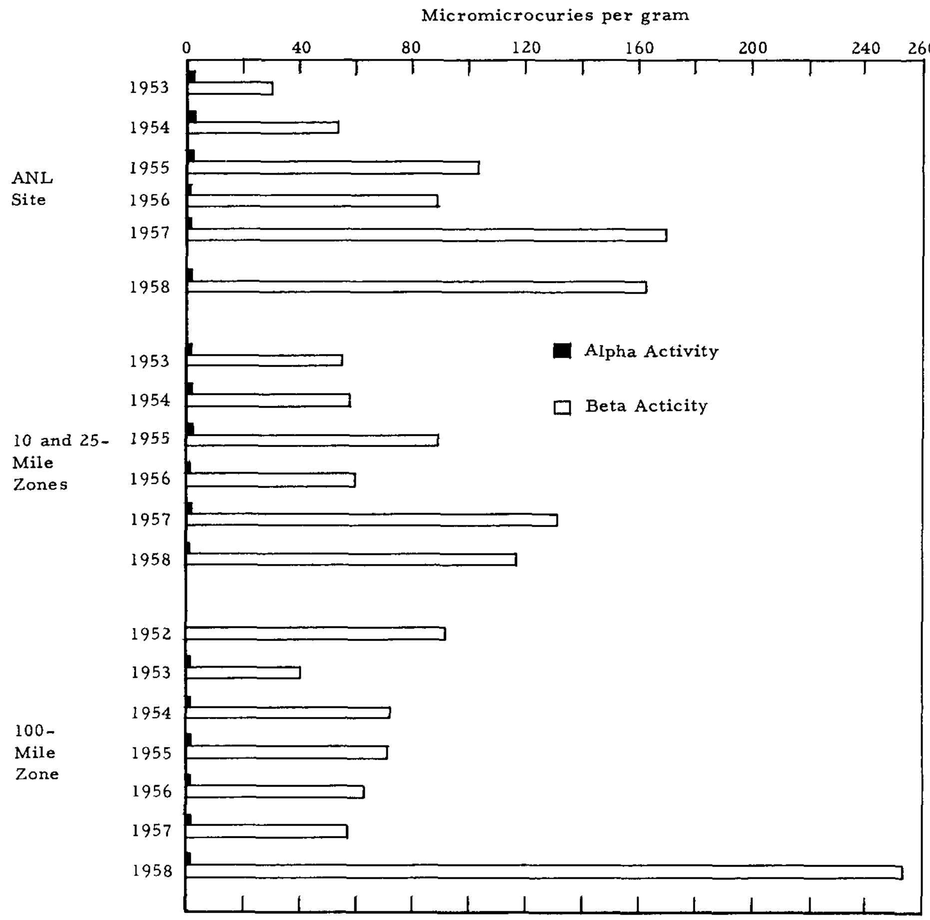
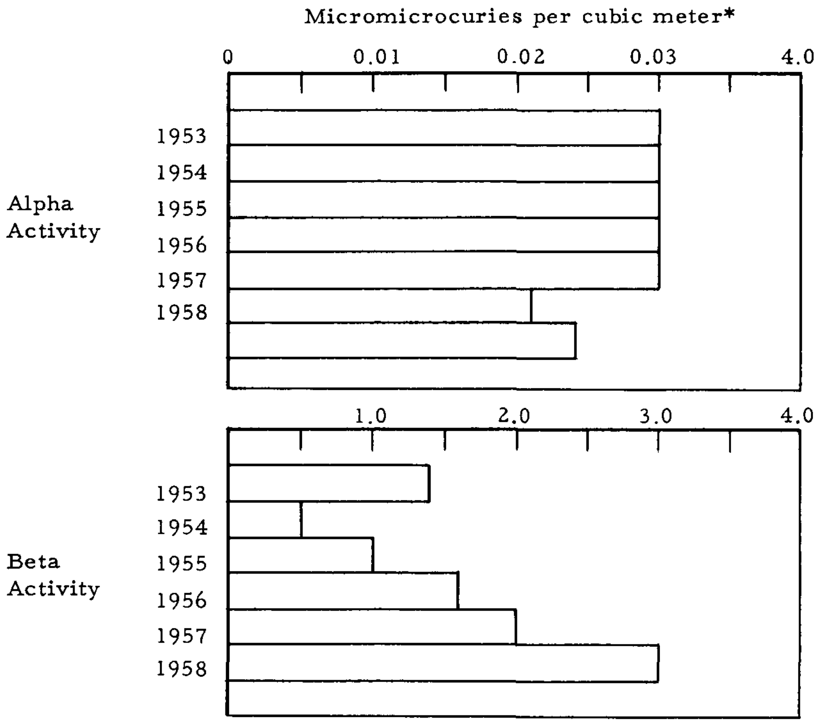


Figure 8

AVERAGE RADIOACTIVITY IN DAILY AIR FILTER SAMPLES
ON ANL SITE, 1952-58



*Activity remaining three days after end of filtering period

contributions to the total alpha activity. The average uranium concentration in those below-outfall samples analyzed for uranium was 21 $\mu\mu c/l$, or 0.3% of the maximum permissible concentration (MPC).* Most, but not all, of the below-outfall water samples contained concentrations of uranium above normal. Small amounts of plutonium, also evidently present in ANL waste water, were also found in about half of the below-outfall water samples analyzed for plutonium. The average plutonium concentration, 0.4 $\mu\mu c/l$, was only 1.8% of the MPC. Beta activity due to contamination in Laboratory waste water was found in Sawmill Creek below the outfall, and fission products from nuclear detonations were found both above and below the outfall. During the first three months of the year the beta activity below the outfall was about 30 $\mu\mu c/l$ higher than above-site water. During most of the remainder of the year, fallout activity increased, with the result that the annual average beta activity below the outfall, 36 $\mu\mu c/l$, was only about 10 $\mu\mu c/l$ higher than the average above-site. Small

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*The concentrations of radioactivity attributable to Argonne operations were quite low, but it may be of interest to compare them with some published maximum permissible concentrations. The MPC's used in this report are those given in Title 10, Part 20, of the Federal Register and apply to the dissemination of radioactivity to unrestricted areas by installations licensed by the AEC. For most nuclides the values given in the Federal Register are 10% of those given in National Bureau of Standards Handbook 52, and, therefore usually agree with recommendations made by the International Committee on Radiological Protection. The MPC's for water apply to drinking water only, and it should be emphasized that Sawmill Creek water is not used for human consumption.

concentrations of Sr^{90} from ANL waste water were detected in about 90% of the below-outfall samples. The average concentration, 2.5 $\mu\mu c/l$, amounted to 3% of the MPC. Water collected from Sawmill Creek above the site contained an average of only 0.4 $\mu\mu c/l$ of Sr^{90}, and this nuclide was found less frequently at this location than below the outfall. However, the shorter-lived fission products, e.g., Sr^{89} and I^{131}, were found in greater concentration above the outfall. This was apparently the result of dilution of below-outfall water with Laboratory waste water containing smaller concentrations of these nuclides than natural Creek water.

The activity added to the Creek in ANL waste water was also detected in bottom silt. Some of the samples obtained below the outfall contained above-normal concentrations of total alpha and beta activity, uranium, plutonium, and Sr^{90}. This contamination was confined to the first 20 yards of the bed below the outfall.

The radioactivity in Sawmill Creek does not add significantly to the radioactivity in the Des Plaines River, apparently due to dilution. The average alpha, beta, and uranium concentrations in River water were essentially the same above and below the mouth of Sawmill Creek. Beta activity due to fallout was detected in the Des Plaines River as well as in most types of samples from all locations.

A storage lagoon for contaminated waste water on the ANL site contained several times the normal concentrations of total activity as a result of the use made of this lagoon. Several of the samples contained 50 to 1000 $\mu\mu c/l$ each of uranium, plutonium, and several long-lived fission products. The radioactivity in the natural lagoons on the ANL site was similar to that found in lakes and streams near the Laboratory, and no evidence of activity originating at Argonne was found in any of these water samples on or off the site. The alpha activities in surface water were low and normal both on and near ANL. As a result of fission product fallout, the beta activities were 10 to 15 $\mu\mu c/l$ higher than in recent years, and averaged about 33 $\mu\mu c/l$ in 1958. Water from the reference sites also contained above-average concentrations of fallout activity in July, but the alpha activities remained normal. The beta activity in Lake Michigan samples collected since 1950 are summarized in Part III. The normal beta activity is 3 to 4 $\mu\mu c/l$. Temporary increases to about twice this value were found in 1954, 1955, and 1958. The highest activities were found in May 1953, when two samples contained 29 and 34 $\mu\mu c/l$, respectively.

Except for the storage lagoon on the ANL site and Sawmill Creek, bottom silt from all locations contained only natural and fallout activities. A significant increase in fallout activity was noted in many of the samples collected near the Laboratory. At these locations the average beta activity in bottom silt was 101 $\mu\mu c/g$, about 40 $\mu\mu c/g$ higher than found earlier. A survey of the bed of the Des Plaines River above and below the mouth of

Sawmill Creek did not disclose any radioactivity attributable to Argonne waste water. Samples from two locations, the Du Page River at Naperville and a pond on the ANL site, contained 50 to 100% more activity than is normal for this area, although the values were normal for these particular locations. The additional activity was apparently due to the thorium chain, since the samples contained normal concentrations of uranium, about 2 $\mu\mu c/g$, but approximately twice the normal concentration of thorium nuclides, about 5 $\mu\mu c/g$.

Surface soil collected near a uranium storage shed on the ANL site contained up to five times the concentrations of alpha and beta activities normally found in the Chicago area. The additional activity was found to be due to uranium, apparently from the storage shed. The total activities in the other soil samples from all locations on and off the site were similar to those found earlier. Most samples contained 20 to 30 $\mu\mu c/g$ of alpha activity and 30 to 80 $\mu\mu c/g$ of beta activity. Fallout activity was not as great in soil samples as in the other types of samples. A soil sample collected near the Du Page River at Naperville contained about twice the normal alpha activity. As in the case for bottom silt at this location, the additional activity was due to natural thorium isotopes, and the uranium concentration in this soil was normal.

Except for grass samples collected near the uranium storage shed, no evidence of activity originating at Argonne was found in plants during 1958. The samples obtained near the storage shed contained about 4 $\mu\mu c/g$ of alpha activity as opposed to normal concentrations elsewhere of 1 to 2 $\mu\mu c/g$. The additional activity was due to uranium. The beta activity in plant samples from all locations was higher than usual. Samples collected on and near the ANL site contained an average of 163 $\mu\mu c\beta/g$ and 110 $\mu\mu c\beta/g$, respectively, about the same as in 1957, but one and one-half to five times greater than found from 1953 through 1956. The average beta activity in the reference site samples was 253 $\mu\mu c/g$, three to five times greater than in any previous year. These increases were the result of abnormally high concentrations of mixed fission product activity from fallout in samples collected in July and November.

Soil borings were collected near a plot of land formerly used by ANL as a burial grounds for contaminated waste. The results were normal, and there was no evidence that any of the activity had moved from the plot to the surrounding area.

II. PROGRAM AND PROCEDURES

This report presents the results of the environmental monitoring program at Argonne National Laboratory for the year 1958. The purposes of the program are to measure the natural radioactive content of the ANL site and its environment, and to determine the magnitude 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 counting on the types and number of samples given in Table I.

TABLE I

SAMPLES COLLECTED IN 1958

Type	Number
Water	275
Precipitation	82
Soil	63
Bottom Silt	146
Plant	57
Air Filter	689
Animal	2

Since the most probable means of dispersal of radioactive contamination are by water and air, most of the samples were of these types. Samples were collected from the Argonne site and from locations approximately 10, 25, and 100 miles from the Laboratory. Sampling locations on the ANL site are shown in Figure 9. The sampling locations within 25 miles of the Laboratory ("near" the Laboratory) are given in Figure 10. The latter samples should indicate the extent and direction of contamination in the event that significant amounts of activity are released at Argonne. Samples from the 100-mile locations (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 measurements of alpha activity, but because of the widespread occurrence of fallout from nuclear detonations these samples have served primarily to indicate the extent and magnitude of fallout activity over a large area. Since fallout activity is present in most samples, beta activities at the levels found in the environment have been more difficult to interpret, in terms of origin, than alpha activities. However, by making the proper choices of sampling location and type of analysis, and by comparing results obtained on-site and off-site, it was usually possible to distinguish between beta activity from nuclear explosions, Argonne operations, and natural sources.

Total-activity measurements were made by counting thick samples (5 to 75 mg/cm^2) after the minimum of sample preparation. The samples were mounted on two-inch-diameter stainless steel planchets and counted in nylon-window (0.1 mg/cm^2 thick) proportional counters. Water samples were acidified with nitric acid immediately after collection to prevent hydrolysis; the residue obtained after evaporation was used for total-activity counting. The residue was flamed and ground in a mortar, then spread evenly over the surface of a weighed counting planchet with carbon tetrachloride. The planchet was reweighed to obtain the weight of the residue. Soil and bottom silt were dried at 110°C, ground in a mortar, and

Figure 9

Sampling Locations on the Site of Argonne National Laboratory

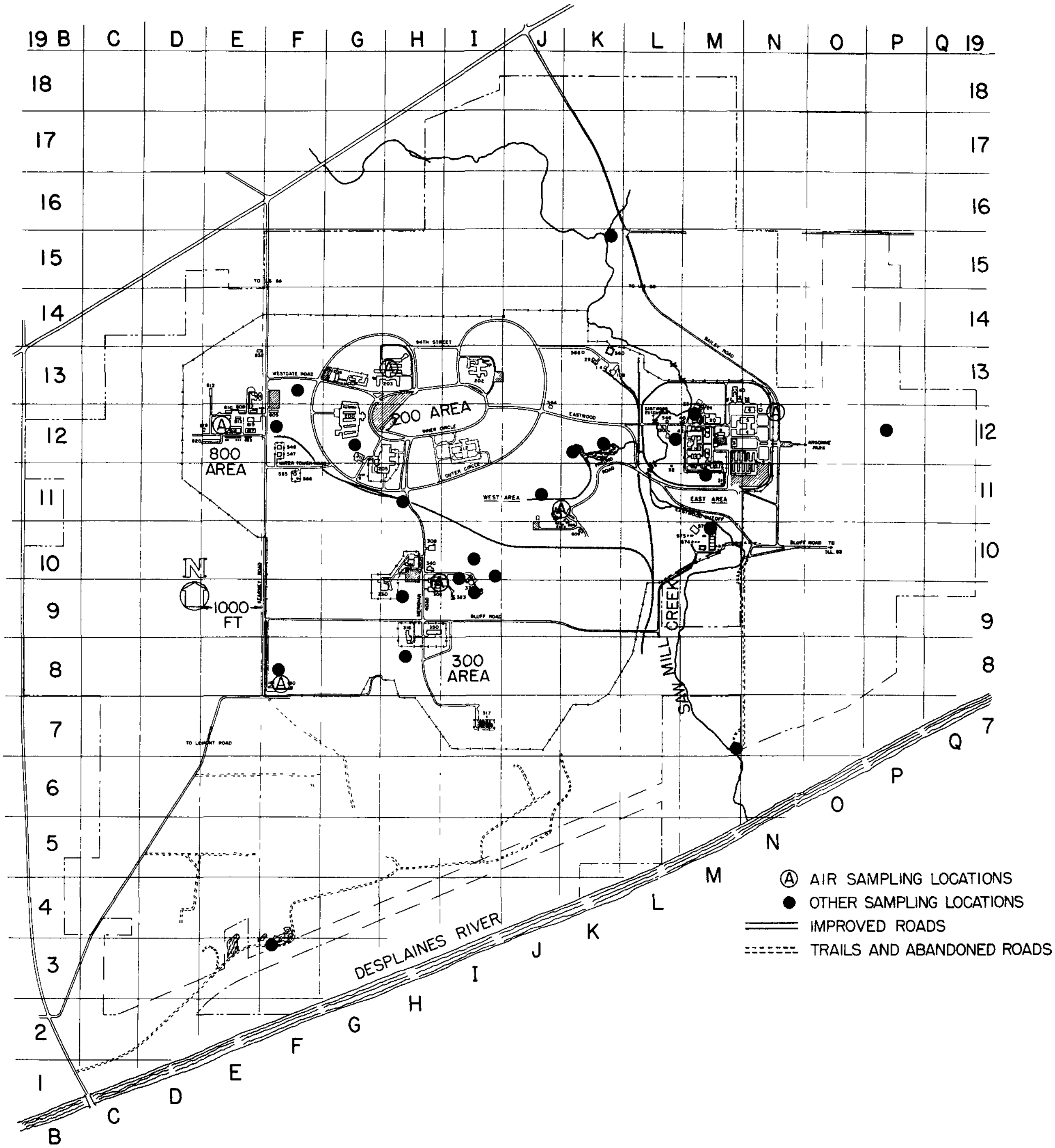
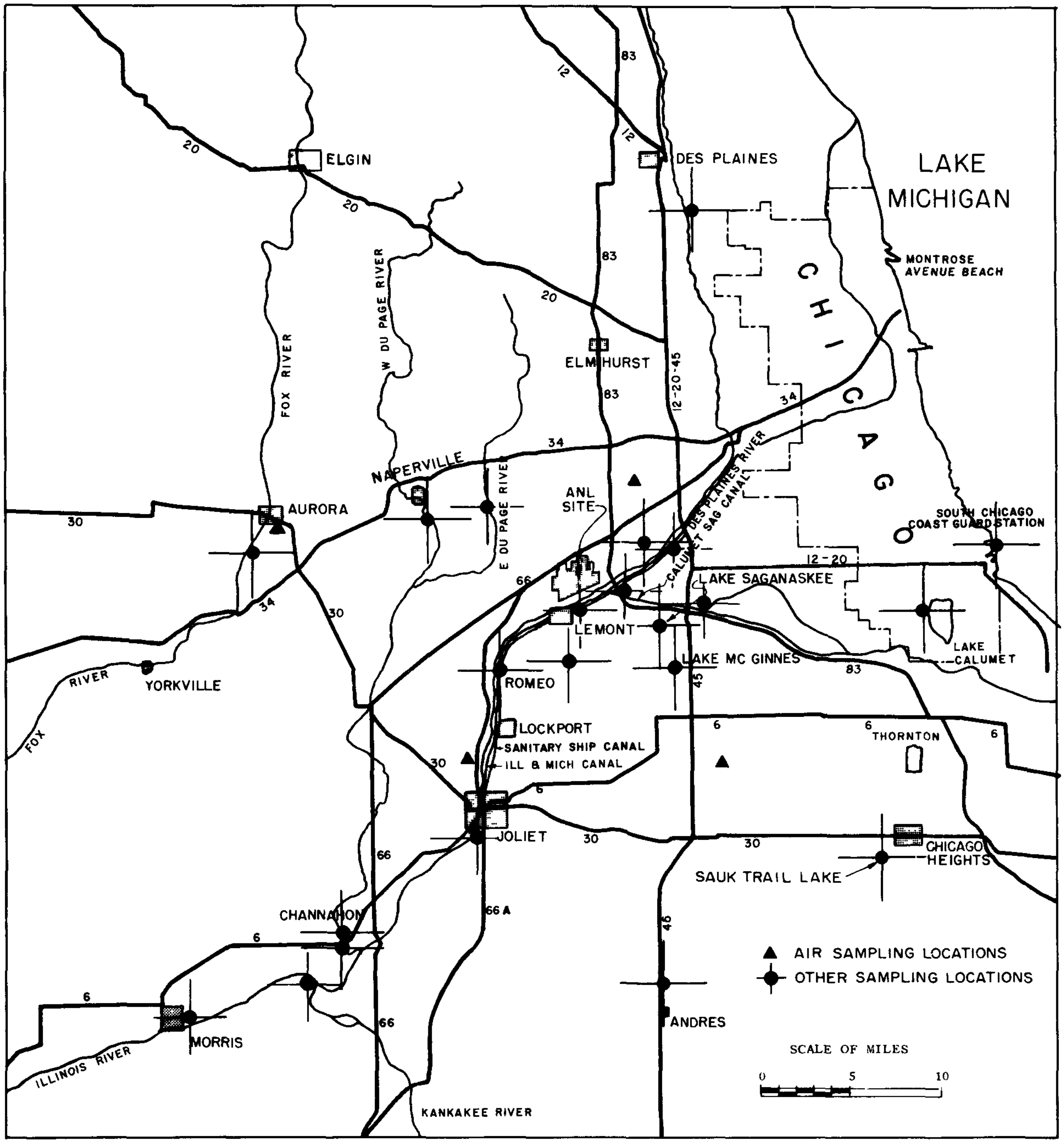


Figure 10

Sampling Locations near Argonne National Laboratory



a weighed portion spread on a counting planchet. Plant and animal samples were dried, ashed, and a weighed portion of the ash mounted for counting. 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. The methods of determining the correction factors to convert the counting rates of thick samples to disintegration rates are given in the previous reports in this series (see below).

Specific fission product analyses were made by separating the desired element with carrier added and counting in an anti-coincidence shielded beta counter. Counting rates were converted to disintegration rates by applying experimentally determined counting yields for each fission product. Uranium analyses were made with a fluorophotometer designed at ANL, and the results converted to micromicrocuries, assuming the natural isotopic composition of uranium. Plutonium and thorium analyses were made by extraction with a solution of thenoyltrifluoroacetone in benzene. The two elements were separated from other alpha emitters and from each other by adjusting the acidity of the aqueous phase and the oxidation state of the plutonium.

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

III. RADIOACTIVITY IN ENVIRONMENTAL SAMPLES

A. Precipitation

The rain collector was arranged to collect two successive one-gallon portions when the rainfall was sufficiently large. From January through July the area of the rain collector was 16.2 sq ft, and each one-gallon portion was equivalent to 0.1 inch of rain. In order to collect a larger fraction of the total rainfall, a collector with an area of 3.36 sq ft was used beginning in August. With this collector, each one-gallon portion amounted to 0.48 inch of rain. The latter collection system is capable of collecting over 90% of the rain, and also provides a sample of sufficient size for analysis from rainfalls of only a few hundreths of an inch. When two successive portions were obtained from one rainfall, they were analyzed and tabulated as separate samples. In most cases the first portion contained the larger amount of both radioactivity and solids, although occasionally the reverse situation was found. Samples for total-activity analysis were evaporated to a small volume in a beaker. If the residue was small, the remaining slurry was transferred to a two-inch-diameter stainless steel counting planchet, dried, flamed, and counted. When the sample contained a large amount of solid material, the slurry was transferred to a porcelain evaporating dish, dried, and flamed. The ignited salts were ground in a mortar and spread uniformly on a counting planchet.

The total alpha and beta activities found in precipitation on the ANL site during 1958 are given in Table II. Analyses of selected samples showed that the alpha activity was primarily due to naturally occurring radioactivities, while the bulk of the beta activity was due to fission products, presumably from atomic detonations. The beta activities were low until the end of March. From March 28th to the middle of August the average beta activity increased to several thousand micromicrocuries per liter. From the middle of August to the middle of October the average beta activity dropped to 600-700 $\mu\mu c/l$. During the remainder of the year the average activity again increased to several thousand micromicrocuries per liter. The age of the beta activity in most of the March, April and May samples, as calculated from a $T^{-1.2}$ decay rate, showed that the fission products originated during the latter part of 1957. This was unusual since activities as high as several thousand $\mu\mu c/l$ have been generally associated with fission products less than 60 days old. During June the beta decay rates of most of the rain samples indicated the presence of fission products originating in March and April. April 28 was announced as the date for beginning of a series of nuclear detonations, and the June samples evidently contained fission products from the 1958 and from the 1957 detonations. During July, August, and September the decay rates indicated the presence of fission products produced in explosions occurring

TABLE II

NONVOLATILE RADIOACTIVITY IN PRECIPITATION AT ANL, 1958

Month	No. of Samples[a]	Alpha Activity ($\mu\mu c/l$)				Beta Activity ($\mu\mu c/l$ x 100)				mc/mi^2
		After 1 day decay		After 1 week decay		After 1 day decay		After 1 week decay		
		Max	Avg	Max	Avg	Max	Avg	Max	Avg	
January[b]	2	-	-	8.2	4.2	-	-	1.29	0.78	1.0
February	2	-	-	6.6	4.9	-	-	4.62	3.8	2.4
March	2	18.0	9.4	18.0	0.9	67.0	37.3	57.0	32.0	19.3
April	10	32.7	7.5	16.1	4.8	100.6	25.8	80.9	22.3	89.6
May	6	80.0	38.1	17.0	7.3	46.4	24.6	42.4	22.2	81.5
June	16	17.7	4.9	6.3	1.6	32.8	15.4	27.2	13.7	129
July	12	33.9	8.0	5.7	1.7	136.0	32.4	124.0	27.1	203
August	6	8.3	2.9	0.8	0.2	35.8	7.8	32.8	7.1	66.7
September	8	30.0	7.4	2.5	0.6	12.2	7.0	11.5	6.3	75.2
October	6	60.0	16.0	6.7	1.5	269.4	56.2	124.0	29.0	221
November	6	6.0	2.7	2.3	1.4	38.7	22.4	35.4	20.1	146
December	5	23.2	12.9	8.5	6.3	35.0	24.7	31.8	22.3	67.9

[a] Two successive portions were collected for heavy rains and are listed as separate samples (see text).

[b] Samples collected in January were counted four weeks after the end of precipitation. All other samples were counted as indicated in the table headings.

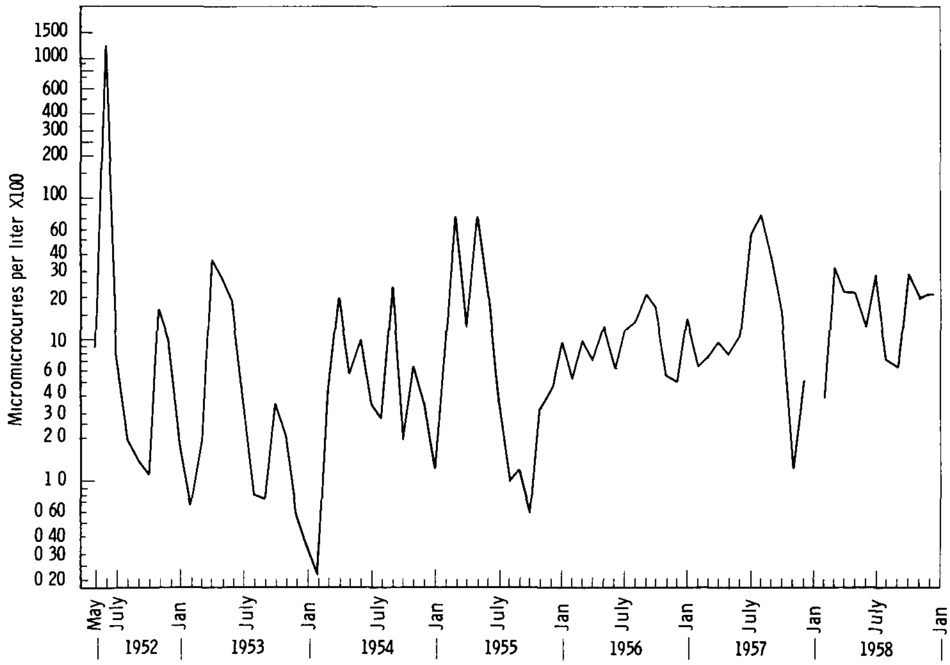
during May, June, and July. From October to the end of the year the decay rates indicated fission products originating in the latter part of September and in October. This corresponds to a series of Russian test explosions announced as beginning September 20. During October, the beta activities were of very recent origin. Five of the six samples during that month give calculated ages ranging from 8 to 20 days.

The total amount of beta activity in the precipitation samples collected during 1958, as calculated from the "one-week" beta activity, was approximately 1100 millicuries per square mile. This was essentially the same as was found for 1955 and in 1957. For 1953, 1954, and 1956 the total beta activity carried down by rain and snow during each year was about 600 mc/mi^2, while in 1952 the corresponding value was 11,000 mc/mi^2. Thus, with regard to mixed fission products only, i.e., mixtures with average half-lives ranging from days to months, the amount carried down by rain and snow has increased by a factor of two since 1953, but has not approached the high activities carried down in June, 1952. This conclusion is not affected by the change in area of the rain collector during 1958, since the change only served to increase the amount of activity collected this year. The fallout of long-lived fission products cannot, of course, be determined from the total beta activity only.

The general trend in mixed-fission product fallout can be seen from Figure 11, where the average monthly "one-week" beta activity is

Figure 11

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



plotted, beginning with the first samples collected. The samples obtained in January, 1958, are not included, since they could not be counted until four weeks after collection. The highest fallout occurred in June, 1952, and the levels found then have not been approached in any subsequent rains. Since June, 1952, the monthly averages have varied between 20 to 8000 $\mu\mu c/l$, although the activities have generally been higher during the past three years. This is probably due to the fact that bomb testing from 1956 through 1958 was conducted over a larger fraction of the year than was the case earlier, and thus there was less opportunity for the activities to decrease for an extended period of time in recent years.

Since rain activities are a sensitive indication of the amount of fallout, the results are very valuable in interpreting changes in beta activity in other types of samples.

B. Air Filters

Air-borne particulate matter was collected at five locations on the Laboratory site and at four locations off the Laboratory site. The locations are shown in Figures 9 and 10. The off-site sampling locations and their locations relative to ANL are: Stateville Penitentiary, 10 miles southwest; Suburban Cook County Tuberculosis Sanitarium, 6 miles northwest; Tinley Park State Hospital, 13 miles southeast; and the Illinois Department of Public Health Office in Aurora, 20 miles west. These sampling locations were obtained through the cooperation of the Illinois Department of Public Health. Samples were collected on Hollingsworth-Vose No. 70 filter paper at an air flow rate of 30 cubic meters per hour. The filter papers were changed and counted at regular intervals. At all the off-site locations and at four of the on-site locations the papers were changed and counted at weekly intervals. Daily samples were collected at one of the on-site locations to obtain short-term changes in air activities. Air sampling was continuous except for occasional equipment failures and for the few minutes required to change filter papers. The weekly on-site samples were counted on the first day after the sampling period to obtain the radon and thoron concentrations, and then after three and seven days to obtain the long-lived activities. The radon and thoron concentrations will be reported at a later date. The off-site air samples could be counted only after three and seven days. The daily samples were counted once after three days. Additional details on the sampling and counting techniques are given in reports ANL-5808 and ANL-5934.

The long-lived alpha and beta activities for 1958 are summarized in Tables III and IV for the on-site samples and in Table V for the off-site samples. The alpha activities, which were due primarily to naturally occurring active nuclides present in dust, averaged essentially the same both on and off the Laboratory site, although individual samples at both locations deviated from the average by factors of two and three. The long-lived alpha

TABLE III

LONG-LIVED RADIOACTIVITY IN 24-HOUR AIR FILTER SAMPLES COLLECTED ON ANL SITE, 1958

Month	No. of Samples	Alpha Activity ($\mu\mu c/m^3$)		Beta Activity ($\mu\mu c/m^3$)	
		Max	Avg	Max	Avg
January [a]	29	0.014	0.007	1.33	0.77
February [a]	28	0.041	0.006	1.97	0.93
March	28	0.052	0.016	5.95	1.87
April	27	0.120	0.045	13.21	6.73
May	29	0.150	0.042	8.50	5.55
June	30	0.050	0.020	7.50	3.34
July	29	0.195	0.032	9.40	2.60
August	29	0.052	0.023	3.80	2.27
September	28	0.129	0.031	3.11	1.61
October	28	0.060	0.031	11.23	4.34
November	27	0.124	0.032	17.70	4.86
December	29	0.033	0.009	5.85	1.89
Summary	341	0.195	0.024	17.70	3.04

[a] These samples were counted 4 to 30 days after collection. All other samples were counted 3 days after the end of the collection period.

TABLE IV

LONG-LIVED RADIOACTIVITY IN WEEKLY AIR FILTER SAMPLES COLLECTED ON ANL SITE, 1958

Month	$\mu\mu c/m^3$ after 3 Days Decay					$\mu\mu c/m^3$ after 7 Days Decay				
	Samples Counted	Alpha Activity		Beta Activity		Samples Counted	Alpha Activity		Beta Activity	
		Max	Avg	Max	Avg		Max	Avg	Max	Avg
February	4	0.0047	0.0033	0.75	0.71	5	0.0032	0.0028	0.77	0.70
March	12	0.0087	0.0040	4.94	1.37	12	0.0038	0.0027	4.58	1.27
April	13	0.018	0.009	6.75	4.58	16	0.015	0.005	5.90	4.24
May	17	0.023	0.008	7.44	4.17	17	0.0073	0.0047	4.94	3.67
June	17	0.011	0.006	3.88	2.84	17	0.0044	0.0027	3.84	2.64
July	15	0.010	0.005	3.96	1.72	16	0.0042	0.0021	2.59	1.48
August	17	0.010	0.006	2.56	1.76	17	0.0033	0.0025	2.46	1.67
September	16	0.021	0.008	1.81	1.23	17	0.0040	0.0031	1.73	1.08
October	17	0.023	0.010	7.42	3.19	17	0.0063	0.0032	4.76	2.66
November	14	0.020	0.009	6.99	5.27	15	0.0069	0.0040	5.73	4.33
December	12	0.015	0.007	4.25	2.90	14	0.011	0.004	3.69	2.60
Summary	154	0.023	0.007	7.44	2.83	163	0.015	0.003	5.90	2.52

TABLE V

LONG-LIVED RADIOACTIVITY IN WEEKLY AIR FILTER SAMPLES COLLECTED NEAR ANL SITE, 1958

Month	After 3 Days Decay					After 7 Days Decay				
	Samples Counted	$\mu\mu c\alpha/m^3$		$\mu\mu c\beta/m^3$		Samples Counted	$\mu\mu c\alpha/m^3$		$\mu\mu c\beta/m^3$	
		Max	Avg	Max	Avg		Max	Avg	Max	Avg
January	0	-	-	-	-	7	0.014	0.008	0.77	0.56
February	2	0.0022	0.0021	0.75	0.76	9	0.0058	0.0036	1.11	0.68
March	5	0.0031	0.0023	0.92	0.69	8	0.0059	0.0026	1.85	0.80
April	10	0.015	0.008	10.00	5.59	13	0.0080	0.0049	9.33	4.90
May	14	0.018	0.011	9.42	5.26	16	0.0094	0.0055	8.90	4.80
June	13	0.010	0.005	3.92	2.77	14	0.0044	0.0031	3.61	2.50
July	8	0.011	0.005	4.11	2.38	10	0.0067	0.0031	4.90	2.55
August	15	0.010	0.005	2.75	1.79	15	0.0048	0.0025	1.43	0.94
September	9	0.014	0.008	1.61	1.23	10	0.0090	0.0041	1.73	1.19
October	13	0.019	0.006	8.86	3.79	13	0.012	0.0043	7.65	3.10
November	10	0.021	0.009	9.06	5.86	13	0.010	0.0053	8.83	5.01
December	6	0.013	0.008	5.60	3.01	8	0.0085	0.0044	5.24	2.78
Summary	105	0.021	0.007	10.00	3.37	136	0.014	0.004	9.33	2.73

activity depends to some extent on the amount and type of dust in the air, and thus variations with time and location are to be expected. However, no significant or consistent differences could be found between samples collected on and off the site. The decrease of about 0.004 $\mu\mu c/m^3$ in alpha activity from the third to the seventh day indicates that some thoron daughter activity still remained on the filter paper after three days.

The long-lived beta activities were also similar on-site and off-site, averaging 2.83 and 2.52 $\mu\mu c/m^3$ on-site after three and seven days, respectively, and 3.37 and 2.73 $\mu\mu c/m^3$ off-site after three and seven days. The differences of 0.2 to 0.5 $\mu\mu c/m^3$ between the two locations are small compared to the week-to-week variations observed at the same location, and it is evident that ANL operations did not contribute significantly to the beta activity found in any of the samples. The bulk of the beta activity remaining three days after the end of the filtering period was due to fallout from atomic detonations. The rate of fallout was evidently quite uniform over the area covered by the air samplers, since the activities varied uniformly at all locations. The gamma-ray spectra of selected samples showed the typical mixture of fission products expected from fallout, primarily, Zr^{95}-Nb^{95}, Ru^{106}-Rh^{106}, and Ce^{144}.

The concentrations of beta activity in air did not fluctuate as widely as those in rain, but there was qualitative correlation between the air and rain activities. Like the rain samples, the beta activity in air was low in January and February, increased in the spring and fall, and decreased during the summer. Also, as with the rains, the increase in the spring was due primarily to fission products from detonations occurring in 1957, while the fall increase was due to current explosions. The average beta activity in the daily air filter samples was generally about 25% higher than that in the weekly samples, because of the shorter time interval between collection and counting for the daily samples. The week-to-week variations were similar for both types of samples. There were no significant differences in activity between the various sampling locations on the ANL site.

The average beta concentrations since sample collection was initiated are given in Table VI. The steady increase in beta activity since 1954, after a decrease from 1953 to 1954, is apparent. The alpha activity has remained quite constant, varying by only about 30% from year to year, while the beta activity has increased by about a factor of six since 1954 and by about a factor of two since 1953. These types of variations are consistent with the conclusion that the alpha activity is primarily due to natural activities while the beta activities are due to fission products from bomb testing. However, the fission product beta activity still remains on the average only a few per cent of the radon daughter beta activity.

TABLE VI

AVERAGE ANNUAL THIRD DAY BETA ACTIVITY IN AIR ON ANL SITE

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	2.1	0.021	2.0
1958	0.007	2.8	0.024	3.0

The alpha activities in the daily samples are greater than in the weekly samples, probably because the same self-absorption corrections were used for both types of samples, and the daily samples were considerably thinner and, therefore, required less correction. The results, however, are self-consistent and comparisons among the same type of sample are valid. The beta counting rate is not as sensitive to variations in the amount of dirt collected on a sample.

Beginning in April the Experimental Boiling Water Reactor (EBWR) was operated with a simulated defective fuel element in the core. This experiment was conducted by the Reactor Engineering Division of ANL. As part of the environmental air-sampling program, additional air samples were placed in operation in and near the EBWR building in an attempt to detect any activity leaving the reactor stack during this work. Three air samplers were placed from 300 to 400 feet from the reactor, since the greatest amount of fallout of activity leaving the stack was expected at this distance. The air in the duct leading to the reactor stack was also sampled by drawing it through a Millipore filter and a dry ice-cooled charcoal trap in series, and by bubbling the air through water and sodium hydroxide. The Millipore filter papers and the water and sodium hydroxide traps contained very little beta activity. In addition, the sodium hydroxide traps were analyzed for iodine, and none could be detected, although radioiodine was found in the reactor water itself. All of the activity on the Millipore samples could be attributed to natural radon daughter activity. Significant amounts of activity could be found on the charcoal traps, however, and barium, strontium, cesium, and cerium fission products were found in these samples. The strontium fractions constituted the bulk of the total activity on the charcoal; they contained small amounts of Sr^{90}. The distribution of fission products among the types of samples collected in the duct was consistent with previous experience with this reactor. The only fission product elements that appear to leave the water in the reactor are the inert gases. These can be adsorbed on cold charcoal, where they decay to solid element descendants. The daughters were not detected on the Millipore samples because there was apparently not enough time for significant amounts of the inert gases to decay before reaching the sampler. No activity attributable to EBWR could be detected in the air samples collected outside the building. Although some of the inert gases and their daughters probably left the stack, the quantities were so small that, after dilution with the outside air, they were undetectable in the presence of the much larger amounts of natural and fallout activity normally present in the air.

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 grounds and after the waste water outfall. During part of the winter and during dry periods the stream was not flowing at the waste water outfall; at such times the below-outfall water sample consisted essentially of Laboratory waste water. When water was flowing in the stream the sample was collected after the waste water had mixed with the natural Creek water. During December and early January of this year there was no water in the Creek above the Laboratory site.

The alpha activities found in Sawmill Creek water are summarized in Table VII. The average total alpha activity in water collected from the Creek above the Laboratory site was 2.8 $\mu\mu c/l$. The average alpha activity in all samples collected below the waste water outfall was 13.4 $\mu\mu c/l$, and the below-outfall water samples, collected only on days when above-site water was also obtained, averaged 13.7 $\mu\mu c/l$.

TABLE VII

NONVOLATILE ALPHA ACTIVITY IN SAWMILL CREEK WATER, 1958

Month	Alpha Activity				Uranium			Plutonium				Thorium			
	Location[a]	No of Samples	μμc/l		No of Samples	μμc/l		No of Samples	Samples >0 1 μμc/l			No Samples	Samples > 0 1 μμc/l		
			Max	Avg		Max	Avg		No	Max	Avg		No	Max	Avg
January	A	3	2 4	2 1	3	2 5	2 3	2	0	–	–	2	0	–	-
	B	11	71 6	17 9	5	69 5	39 [illegible]	4	3	1 6	1 4	4	3	0 8	0 4
February	A	4	2 5	2 4	4	2 9	2 1	4	0	–	–	4	2	0 2	0 2
	B	11	51 7	17 7	6	48 0	27 4	4	2	1 5	0 8	4	3	0 3	0 2
March	A	4	2 7	2 2	4	2 3	1 7	2	0	–	–	2	0	–	–
	B	13	45 2	10 4	5	36 3	20 8	3	2	0 8	0 8	2	2	0 5	0 3
April	A	5	3 5	2 0	5	1 9	1 5	2	0	–	–	2	0	–	–
	B	12	18 1	9 2	5	15 2	11 9	2	1	0 2	–	2	0	–	–
May	A	4	2 9	1 9	4	1 6	1 4	2	0	–	–	2	1	0 1	–
	B	12	16 9	5 8	4	10 4	6 7	2	2	0 7	0 6	2	0	–	–
June	A	4	2 6	2 0	4	2 8	1 6	2	0	–	–	2	1	0 1	–
	B	13	144	35	9	149	42 2	2	0	–	–	2	0	–	–
July	A	5	2 8	2 0	5	1 9	1 7	3	0	–	–	3	1	0 3	–
	B	14	39 8	10 8	6	36 6	13 6	4	0	–	–	4	0	–	–
August	A	4	2 5	2 2	4	3 2	2 0	2	0	–	–	2	2	0 5	0 4
	B	13	21 9	6 7	4	7 8	5 2	2	2	0 9	0 7	2	2	0 4	0 3
September	A	4	3 8	3 1	4	2 3	2 2	3	0	–	–	2	1	0 4	–
	B	11	10 6	5 2	4	6 1	3 7	3	1	0 5	–	3	0	–	–
October	A	5	5 0	4 1	5	3 7	2 8	3	0	–	–	3	0	–	–
	B	14	15 6	9 5	5	16 3	13 6	4	2	2 7	2 2	4	0	–	–
November	A	4	8 2	6 3	4	6 3	4 2	4	0	–	–	4	1	0 1	–
	B	12	27 3	11 4	4	30 0	16 4	4	1	0 2	–	4	1	0 5	–
December	A	–	–	–	–	–	–	–	–	–	–	–	–	–	–
	B	11	41 1	22 4	5	44 5	24 7	2	1	0 2	–	2	0	–	–
Summary	A	46	8 2	2 8	46	6 3	2 1	29	0	–	–	29	9	0 5	0 1
	B	148	144	13 4	54	149	21 0	36	17	2 7	0 4	35	11	0 8	0 1

[a] Location A is above the Laboratory Site   Location B is below the waste water outfall

The additional alpha activity below the outfall, about 10 $\mu\mu c/l$ on the average, is attributed to activity added to the Creek in Laboratory waste water. Additional analyses indicated that the principal contaminant in Laboratory waste water was normal uranium. The total alpha activity in below-outfall water varied widely, from activities essentially the same as above-site water to a maximum of 144 $\mu\mu c/l$. For those samples containing above normal amounts of alpha activity, uranium accounted for essentially all the alpha activity.

The average uranium concentration in those samples analyzed for uranium was 21 $\mu\mu c/l$, only 0.3% of the maximum permissible concentration. The average uranium concentration was greater than the average alpha activity because most of the samples analyzed for uranium contained above-average concentrations of alpha activity. The average uranium concentrations on only those days when samples were collected from both locations was 2.1 $\mu\mu c/l$ above-site and 13 $\mu\mu c/l$ below the outfall. Small amounts of plutonium were also found in below-site water, and since concentrations greater than 0.1 $\mu\mu c/l$ were not found in above-site water, the plutonium was evidently added to Sawmill Creek in Laboratory waste water. Seventeen of thirty-six below-outfall water samples analyzed contained more than 0.1 $\mu\mu c/l$ of plutonium. The average and maximum plutonium concentrations were only 0.4 and 2.7 $\mu\mu c/l$, respectively. These values are 0.4% and 1.8% of the MPC. Concentrations of thorium isotopes were low, averaging 0.1 $\mu\mu c/l$ at both sampling locations, and consequently this element did not contribute significantly to the total alpha activity at either location.

The alpha activities in Sawmill Creek water during 1958 followed the same pattern observed during earlier years. That is, below-outfall water contained, on the average, more alpha activity than above-site water, the difference ranging from an average of about 7 $\mu\mu c/l$ in 1955 to 27 $\mu\mu c/l$ in 1953. In all cases, the additional activity was due primarily to natural uranium. Contributions to the total activity by plutonium, thorium, and radium have been small and infrequent.

The beta activities found in Sawmill Creek water are given in Table VIII. In addition to the natural beta activity in the stream, fission product fallout was detected at both sampling locations. Fission products, UX_1 and UX_2 originating in Argonne waste water were found in below-outfall water.

During January, February, and March, below-outfall water contained an average of 30 $\mu\mu c/l$ more beta activity than above-site water (47 $\mu\mu c/l$ more, comparing only samples collected from both locations on the same day), and this difference is attributed primarily to contamination in Laboratory waste water. Fallout activity was also apparent in the Creek during this period as shown by the results of fission product analyses.

TABLE VIII

BETA ACTIVITY IN SAWMILL CREEK WATER, 1958

Month	Total Activity				Sr^{90}				Sr^{89}				Ce^{141}				Ce^{144}				I^{131}				Ba^{140}			
	Loc.[a]	No. of Samples	μμc/liter		No. of Samples	Samples >1 μμc/liter			No. of Samples	Samples >1 μμc/liter			No. of Samples	Samples >3 μμc/liter			No. of Samples	Samples >3 μμc/liter			No. of Samples	Samples >3 μμc/liter			No. of Samples	Samples >3 μμc/liter		
			Max	Avg		No.	μμc/liter Max	Avg		No.	μμc/liter Max	Avg		No.	μμc/liter Max	Avg		No.	μμc/liter Max	Avg		No.	μμc/liter Max	Avg		No.	μμc/liter Max	Avg
January	A	3	7.3	6.9	2	0	-	-	2	2	9.0	5.6	2	0	-	-	2	0	-	-	2	0	-	-	2	0	-	-
	B	10	85.6	45.9	3	3	2.2	1.8	3	0	-	-	3	0	-	-	3	0	-	-	3	0	-	-	3	0	-	-
February	A	4	53.0	22.0	3	2	2.2	1.7	3	3	8.1	6.2	3	2	12	11	3	1	4	-	4	0	-	-	4	0	-	-
	B	11	134	50	5	5	15.2	6.4	3	3	18.4	8.5	4	2	4.0	3.5	4	3	6	4	4	0	-	-	4	0	-	-
March	A	4	14.2	9.3	4	0	-	-	3	1	3.0	-	2	0	-	-	2	0	-	-	4	0	-	-	4	0	-	-
	B	13	104	31	4	4	10.5	4.2	4	2	7.4	6.2	2	1	25	-	2	1	3	-	4	0	-	-	4	0	-	-
April	A	5	25.7	14.2	4	3	2.1	1.7	4	3	7.1	3.6	2	0	-	-	2	0	-	-	4	0	-	-	4	1	5.9	-
	B	12	45.6	19.1	4	4	2.3	1.6	4	2	2.1	1.7	2	0	-	-	2	0	-	-	4	0	-	-	4	1	8.8	-
May	A	4	13.1	11.0	4	3	1.7	1.4	4	4	5.0	3.7	2	0	-	-	2	0	-	-	4	0	-	-	4	2	4.9	3.7
	B	12	43.4	20.9	4	4	3.4	1.9	4	4	4.5	2.0	2	0	-	-	2	0	-	-	4	0	-	-	3	0	-	-
June	A	4	47.1	38.9	4	0	-	-	4	4	32.2	22.5	3	2	17	11	2	1	3	-	4	2	17	13	3	1	18	-
	B	13	86.2	41.5	4	4	4.5	3.2	4	4	20.6	10.0	2	0	-	-	2	0	-	-	4	1	6	-	2	0	-	-
July	A	5	57.7	35.5	5	0	-	-	5	5	34.6	25.1	3	1	18.7	-	3	1	6.4	-	5	0	-	-	4	1	12	-
	B	13	366	63	5	4	5.9	2.4	5	5	27.5	11.0	2	0	-	-	2	0	-	-	4	0	-	-	5	0	-	-
August	A	4	36.2	31.6	4	1	1.4	-	4	4	24.3	17.8	2	1	14.2	-	2	1	3.8	-	4	1	4.3	-	4	1	2.9	-
	B	13	143	41	5	5	5.6	3.0	5	5	12.1	6.8	2	0	-	-	2	0	-	-	4	0	-	-	5	0	-	-
September	A	4	53.6	41.1	4	2	1.4	1.4	4	3	17.5	13.4	2	0	-	-	2	0	-	-	4	0	-	-	4	0	-	-
	B	11	53.1	25.5	4	3	4.7	3.0	4	4	20.1	13.6	2	0	-	-	2	0	-	-	2	0	-	-	2	0	-	-
October	A	5	63.0	36.1	5	3	1.9	1.4	5	4	20.2	13.6	2	0	-	-	2	0	-	-	5	2	11	8	4	2	6.8	5.7
	B	14	42.3	25.2	5	3	2.5	2.1	5	4	9.0	5.4	2	0	-	-	2	0	-	-	4	1	10	-	4	1	3.5	-
November	A	4	59.0	43.4	4	2	1.1	1.1	4	4	14.0	10.8	2	0	-	-	2	0	-	-	4	1	6	-	4	1	3.0	-
	B	11	46.2	30.7	4	4	2.0	1.7	4	4	9.0	4.4	2	0	-	-	2	0	-	-	4	0	-	-	4	1	5.0	-
December	A	0	-	-	0	-	-	-	0	-	-	-	0	-	-	-	0	-	-	-	0	-	-	-	0	-	-	-
	B	11	74.6	40.0	5	4	6.0	2.8	5	5	9.0	4.0	2	0	-	-	2	0	-	-	5	2	5	4	5	0	-	-
Summary	A	46	63.0	26.9	43	16	2.2	0.4	42	37	34.6	11.4	25	6	18.7	3	25	4	6.2	0.7	44	6	17	1.3	41	8	18	1.5
	B	144	366	36	52	47	15.2	2.5	50	42	27.5	5.8	27	3	25	1.2	27	4	6	0.6	46	4	10	0.5	45	3	8.8	0.4

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

Both strontium and cerium nuclides were found at both locations, but the longer-lived activities, Sr^{90} and Ce^{144}, were detected more frequently and in greater concentrations below the outfall. This indicated that the two locations contained fission products of different origins. Beginning in April, and continuing for the rest of the year, the average difference in beta activity between the two locations became appreciably smaller, and frequently above-site water contained more beta activity than below-outfall water. This is more apparent when only samples collected from both locations on the same day are compared. For example, during July all below-outfall samples contained an average of 63 $\mu\mu c/l$ compared to 36 $\mu\mu c/l$ for the above-site samples. However, the below-outfall samples collected on the same days as the above-site samples averaged only 33 $\mu\mu c/l$. The increased contribution of fallout activity to the beta activity in Sawmill Creek beginning in April was to be expected, since the fallout activity in air and rain also increased sharply in April, and remained high during most of the remainder of the year. Beta activity originating in Argonne waste water could be detected from the results of fission product and uranium analyses, and from the rates of growth or decay of the total beta activity in the Creek during periods of high fallout. The presence of larger Sr^{90} concentrations and greater Sr^{90}/Sr^{89} ratios below the outfall indicates that fission products entered the stream in the waste water. Increased beta activity from uranium daughters also accompanied increases in uranium concentrations below the outfall. The rate of beta decay of samples containing small amounts of uranium was also useful in distinguishing between fallout and waste water activity, and samples obtained below-site showed beta decay rates equal to or less than samples collected above-site and elsewhere on the same day. Therefore, the fission products in Argonne waste water were generally of equal or older ages than the fallout activity.

Because of the wide day-to-day variation in beta activity in the waste water and because of the dilution of the stream water by waste water, it was possible for below-site water to contain smaller total beta activity concentrations than the above-site water. It was also possible to observe higher concentrations of the shorter-lived fission products predominant in fallout (Sr^{89}, Ba^{140}, etc.) in above-site water, while the Sr^{90} activity was greater below-site.

Sr^{90} was detected in a large fraction of the below-outfall water samples, but the individual concentrations were low. This nuclide was found in greater than 1 $\mu\mu c/l$ concentrations in 47 out of 52 samples analyzed. The average concentration was 2.5 and the maximum was 15.2 $\mu\mu c/l$. These concentrations are 3 and 19%, respectively, of the MPC. Sr^{90} was also found in 16 out of 44 above-site water samples. The average and maximum values, 0.4 and 2.2 $\mu\mu c/l$, respectively, were only 0.5 and 2.8% of the MPC. The Sr^{90} found above-site was derived from fallout only. Valid comparisons with previous years cannot be made because concentrations less than 3 $\mu\mu c/l$ were not determined earlier. Sr^{89} derived from fallout was found during all months, and in a somewhat greater fraction of the samples than in earlier years. The average concentration, about 11 $\mu\mu c/l$, however, was similar to that found earlier.

The average contribution of fallout activity to the total beta activity in the stream was 15 to 20 $\mu\mu c/l$ above-site and about 10 $\mu\mu c/l$ below-site, averaged over the year. The natural activity in the stream is believed to be from 5 to 10 $\mu\mu c/l$, approximately 5 $\mu\mu c/l$ from natural potassium-40 and the remainder from uranium daughters. The average contribution of Laboratory waste water to the Creek below the outfall was 15 to 20 $\mu\mu c/l$. The annual average activities, about 27 $\mu\mu c/l$ above and 36 $\mu\mu c/l$ below the Laboratory site, were approximately the same as found previously. From 1953 to 1957 the above site averages ranged from 20 to 44 $\mu\mu c/l$, while the below-site averages ranged from 39 to 47 $\mu\mu c/l$. Thus, there has not been any increase in the total beta activity in Sawmill Creek since sample collection was begun.

2. Des Plaines River

Sawmill Creek empties into the Des Plaines River, and the river was sampled above and below the mouth of Sawmill Creek to determine if the activity in the Creek water had any effect on the radioactivity in the Des Plaines River. The total alpha and beta activities, as well as the uranium concentrations found in the Des Plaines River at two locations, are given in Table IX. The total alpha activities and uranium concentrations were uniformly low and essentially the same at both locations. The average concentrations (1.9 $\mu\mu c\alpha/l$ and 1.5 $\mu\mu cU/l$), as well as the ranges of the individual results, were similar to those observed during previous years. Consequently, long-term as well as short-term variations have been quite small. The sample-to-sample variations in beta activity were large and can be attributed to variations in the amount of fission product fallout. The total beta activity increased in May, when increased fallout was found in air and rain, and remained relatively high for the remainder of the year. Some of the samples were analyzed for radiostrontium, barium, cerium, and zirconium. The relative amounts of fission products at both locations were the same and typical of fallout activity, while Sawmill Creek water below the waste water outfall contained relatively greater concentrations of the longer-lived fission products. Therefore, Sawmill Creek did not contribute significantly to the beta activity in the Des Plaines River. This is evidently the result of dilution of the Creek water in the River. Additional evidence that increases in beta activity in the River below the mouth of Sawmill Creek were not due to Creek water was the fact that increases in beta activity were not accompanied by increases in alpha activity, although Sawmill Creek below the outfall showed increases in both uranium and fission products.

TABLE IX

NONVOLATILE RADIOACTIVITY IN DES PLAINES RIVER WATER, 1958

Date Collected	Location [a]	Alpha Activity ($\mu\mu c/l$)	Uranium ($\mu\mu c/l$)	Beta Activity ($\mu\mu c/l$)
February 19	A	1.7	2.2	12.3
February 26	B	3.2	1.2	23.3
March 19	A	2.4	1.8	8.9
March 19	B	2.2	1.8	10.3
April 3	A	2.1	2.4	14.4
April 3	B	2.2	1.6	13.7
May 21	A	3.7	1.8	39.3
May 21	B	2.5	2.1	20.4
June 18	A	2.2	1.6	32.1
June 18	B	2.3	1.5	32.6
July 16	A	3.2	3.0	122
July 16	B	2.1	2.6	61.9
August 22	A	2.0	1.0	89.5
August 22	B	1.3	0.8	63.1
September 17	A	1.1	1.2	64.9
September 17	B	1.6	1.0	65.0
October 15	A	0.8	0.9	49.8
October 15	B	1.7	1.2	90.4
November 19	A	1.0	0.9	56.7
November 19	B	1.3	1.2	61.2
December 19	A	2.3	1.6	30.7
December 19	B	2.1	2.1	36.6
Average	A	1.9	1.5	43.4
	B	1.9	1.4	39.9

[a] Location A is 0.5 mile downstream from the mouth of Sawmill Creek. Location B is 2.5 miles upstream from the mouth of Sawmill Creek.

The samples collected in July and October are noteworthy in that the beta activities for each pair differed by a factor of about two, and in all four samples the bulk of the activity was due to fallout. Thus, large differences in fallout activity may occur in locations relatively close together. The level of total beta activity in the Des Plaines has increased slowly since 1954 from about 15 $\mu\mu c/l$ to about 40 $\mu\mu c/l$ in 1958. The general level in 1958 was approximately the same as it was in 1953. The highest beta activity found in the river was 500 $\mu\mu c/l$ in 1953, when a large amount of short-lived fission products from continental tests was detected.

3. Other Waters

The total activities in water from the ponds on the ANL site are given in Table X. Except for the storage lagoon for contaminated

TABLE X

NONVOLATILE RADIOACTIVITY IN PONDS ON ANL SITE, 1958

Date Collected	No. of Samples	Alpha Activity ($\mu\mu c/l$)		Beta Activity ($\mu\mu c/l$)	
		Max	Avg	Max	Avg
April 3	4	260[a]	66	2270[a]	582
	3	1.7[b]	1.5[b]	23.6[b]	19.7[b]
May 28	3	1.7	1.3	24.9	20.9
August 1	4	146[a]	37	631[a]	170
	3	1.2[b]	0.9[b]	55.8[b]	46.7[b]
November 24	4	5.4[a]	3.4	60.6[a]	42.8
	3	4.2[b]	2.8[b]	63.5[b]	36.8[b]
Average		29		156	
		1.6[b]		31.0[b]	

[a] Storage lagoon for contaminated waste water.
[b] Results excluding storage lagoon.

waste water, the alpha and beta activities were in the range normally found in these ponds and in surface water collected off the Laboratory site. The alpha activities in the natural ponds ranged from 0.8 to 2.2 $\mu\mu c/l$ and averaged 1.6 $\mu\mu c/l$. The corresponding values for the beta activities were 14.1 to 55.8 $\mu\mu c/l$ and 31 $\mu\mu c/l$. About two-thirds of the beta activity in the more active samples was due to fallout and as a result the annual average, 31 $\mu\mu c\beta/l$, was 10 to 20 $\mu\mu c/l$ higher than the averages since 1953. However, off-site samples during the year also showed this increase due to fallout. The storage lagoon for contaminated waste water contained abnormal amounts of both alpha and beta activities as a result of the use made of this lagoon. The April and August samples contained 100 to 200 times the normal alpha activity and 10 to 40 times the normal beta activity. These samples contained 50 to 1000 $\mu\mu c/l$ each of uranium, plutonium, Ce^{144}, Cs^{137}, and Sr^{90}. Shorter-lived fission products were absent, or were present only in small concentrations, and the rate of beta decay was considerably less than for the more active beta samples obtained from the other ponds. The alpha activities in the storage lagoon samples did not show any decay.

The results of total activity measurements made on water collected within twenty-five miles of ANL are given in Table XI and summarized in Table XII. The alpha activities ranged from 0.5 to 4.5 $\mu\mu c/l$, and none of the samples showed any significant change from previous years. The range of total beta activities in water collected near ANL was similar to that found previously. However, a large fraction of the samples, particularly those collected in July, contained 20 to 100 $\mu\mu c/l$ more fallout activity than usual. As a result the average beta activity, 35 $\mu\mu c/l$, was 10 to 20 $\mu\mu c/l$ higher than any year except 1953, when the average was 43 $\mu\mu c/l$. The July samples decayed rapidly, reaching one-third to one-fourth their original value in six months. The increased fallout activity in July surface waters is reasonable in view of the large amount of fallout activity in rains collected during the month.

TABLE XI

NONVOLATILE RADIOACTIVITY IN LAKES AND STREAMS NEAR ANL, 1958

Lake or Stream	Location	May		July		November	
		μμcα/l	μμcβ/l	μμcα/l	μμcβ/l	μμcα/l	μμcβ/l
Des Plaines River	31 st St., Brookfield	-	-	-	-	1.4	65.6
Des Plaines River	Willow Springs	3.7	39.3	3.2	122	1.0	56.7
Des Plaines River	Lemont	2.5	20.4	2.1	61.9	1.3	61.2
Des Plaines River	Romeo	2.4	30.4	2.7	60.6	0.9	46.9
Lake Michigan	98th St., Chicago	-	-	-	-	0.5	8.0
Calumet Lake	111th St., Chicago	-	-	-	-	1.0	31.1
Confluence, Des Plaines, Kankakee & Illinois Rivers	McKinley Woods	1.2	8.2	1.3	30.6	1.7	16.3
DuPage River	Naperville	4.5	18.4	2.6	39.5	1.4	26.0
DuPage River	Channahon	-	-	1.5	26.7	0.7	32.6
Saganaskee Slough	104th Ave. & Sag Canal	0.6	12.4	-	-	-	-
Flag Creek	German Church & Wolf Rds., Willow Springs	1.1	19.8	-	-	0.6	18.6
McGinnis Slough	U.S. Rt. 45 & Ill. Rt. 7	-	-	0.9	55.7	-	-
Cal-Sag Canal	U.S. Rt. 83 & 86th Ave.	-	-	0.8	18.3	-	-
Illinois River	Morris	-	-	0.8	32.9	2.0	20.4
Fox River	Aurora	-	-	-	-	0.8	20.9
Sauk Lake	Park Forest	-	-	1.0	63.6	-	-
Sanitary & Ship Canal	Lemont	-	-	-	-	1.3	43.4
Long Run Creek	135th St. & Archer Ave.	-	-	-	-	1.9	11.5
Salt Creek	Wolf Road, Western Springs	-	-	-	-	2.1	35.8

TABLE XII

NONVOLATILE RADIOACTIVITY IN LAKES AND STREAMS NEAR ANL, 1958

Month	Distance from ANL (miles)	No. of Samples	Alpha Activity (μμc/liter)		Beta Activity (μμc/liter)	
			Max	Avg	Max	Avg.
May	10	5	4.5	2.5	39.3	22.1
July	10	5	3.2	1.9	122	59
November	10	8	2.1	1.4	65.6	39.9
Average	10		1.8		40	
May	25	3	2.4	1.8	30.4	19.3
July	25	5	2.7	1.5	63.6	42.9
November	25	7	2.0	1.1	46.9	25.2
Average	25		1.4		29.9	
Average	10 & 25		1.6		35	

Table XIII gives the results of total activity measurements made on water collected from locations 100 miles from the Laboratory. The alpha activities were uniformly low and showed no significant change from earlier years. Beta activity from fallout was present in all samples, although the July samples contained, on the average, about 20 $\mu\mu c/l$ more than normally found. The latter samples decayed at about the same rate as those collected in July near ANL, and apparently the fission products in both sets of samples had the same origin.

TABLE XIII

NONVOLATILE RADIOACTIVITY IN WATER FROM REFERENCE SITES, 1958

Location	July		November	
	$\mu\mu c\alpha/l$	$\mu\mu c\beta/l$	$\mu\mu c\alpha/l$	$\mu\mu c\beta/l$
Illinois River, Starved Rock, Ill.	1.9	32.1	1.5	18.3
Lake Delavan, Delavan, Wis.	0.4	50.0	0.7	26.3
Lake Geneva, Geneva, Wis.	0.4	24.7	-	-
Lake Michigan, St. Joseph, Mich.	0.2	15.4	0.6	11.2
Pipestone Lake, Mich.	0.2	29.5	0.4	12.6
Shafer Lake, Monticello, Ind.	1.1	28.6	1.8	20.4
Fox River, Oak Point State Park, Wis.	-	-	1.8	19.1
Tippecanoe River, Monticello, Ind.	-	-	1.8	13.1
Average	0.7	30.1	1.2	17.3

Because of the large population that regularly uses Lake Michigan water for drinking purposes, the activities in this lake are of special interest. Samples have been collected since 1950, and the beta activities are summarized in Table XIV. Since the samples collected in 1950 and 1951, before the detonation of fusion bombs, were analyzed considerably later, they essentially contained only natural activities. The beta activities in subsequent samples fluctuated and have returned to normal values, 4 $\mu\mu c/l$ or less, after temporary increases due to fission product fallout. The highest beta activities were found in May, 1953, when two samples contained 29 and 34 $\mu\mu c/l$, respectively. Abnormally high concentrations of fallout activity were also found in all water samples collected in May, 1953, from the Chicago area, while the sample collected in St. Joseph in May, 1953, contained only 5.6 $\mu\mu c/l$. Changes in beta activity have occurred at both sampling locations independently, and fallout activity may change at any one time on one side of the Lake only. As might be expected because of its large size, Lake Michigan has not shown as large increases due to fallout as smaller bodies of water. The average from May, 1953 through 1957 was 4.1 $\mu\mu c/l$. A small increase was noted in 1958, when the three samples ranged from 8 to 15.4 $\mu\mu c\,\beta/l$. However, in 1958, samples were only collected in July and November, when fallout activity was above normal, and the average activity for the entire year cannot be inferred from such samples alone. The total beta activities in all above normal samples decayed to the normal range in 1 year or less.

TABLE XIV

NONVOLATILE BETA ACTIVITY IN LAKE MICHIGAN WATER, 1950-1958

Year	Location	No. of Samples	Beta Activity ($\mu\mu c/l$)	
			Range	Avg.
1950	St. Joseph, Mich.	1	-	4.4[a]
1951	St. Joseph, Mich.	1	-	3.3[b]
1952	Chicago, Ill.	3	3.8- 5.4	4.4
1953	Chicago, Ill.	8	0.9-34.0[c]	9.6[c]
	St. Joseph, Mich.	4	0 9- 5.6	3.7
1954	Chicago, Ill.	2	1.8- 2.6	2.2
	St. Joseph, Mich.	2	1.6- 7.7	4.7
1955	Chicago, Ill.	4	2.6- 6.4	4.2
	St. Joseph, Mich.	2	6.8- 8.3	7.6
1956	Chicago, Ill.	2	2.7- 3.4	3.1
	St. Joseph, Mich.	2	0.4- 4.6	2.5
1957	Chicago, Ill.	2	4.3- 6.4	5.4
	St. Joseph, Mich.	2	3.7- 4.6	4.2
1958	Chicago, Ill.	1	-	8.0
	St. Joseph, Mich.	2	11.2-15.4	13.3

[a] Collected in August 1950 and analyzed in May 1951.

[b] Collected in June 1951 and analyzed in August 1951.

[c] Excluding two samples collected in May 1953, the range was 0.9 to 4.5 and the average was 2.3 $\mu\mu c/l$.

The alpha activities in Lake Michigan water varied from 0.1 to 1.5 $\mu\mu c/l$, and averaged 0.4 $\mu\mu c/l$. There was no apparent correlation between the alpha and beta activities. All variations in alpha activity were due either to natural causes or to the experimental error in measuring the very low alpha activities often found in Lake Michigan.

D. Bottom Silt

Concentrations of activity in bottom silt are of interest because they indicate the extent to which activity in water contaminates the bed of the lake or stream, wherein the bed conditions are appropriate for removal of activity from water, and where plant and animal life are exposed to higher concentrations of activity than is expected from water activities alone. In addition, bottom silt, by concentrating activity, may show low-level stream contamination when water analyses do not.

The total activities in bottom silt from the Des Plaines River and Sawmill Creek are given in Table XV. About one-half of the Sawmill Creek samples obtained below the waste water outfall contained 15 to 30 $\mu\mu c/g$ more alpha activity than above-site samples collected on the same day. The additional alpha activity below the outfall is attributed to radioactive nuclides entering the stream in Argonne waste water and accumulated by the bottom silt. This is confirmed by the data given in Table XVI. The samples collected from Sawmill Creek 20 yards or less from the outfall contained from 5.6 to 38.8 $\mu\mu c/g$ of uranium and from 7.6 to 25.6 $\mu\mu c/g$ of plutonium, while samples collected elsewhere contained approximately 2 $\mu\mu c/g$ of uranium and less than 0.1 $\mu\mu c/g$ of plutonium. The thorium concentrations in Sawmill Creek bottom silt were quite uniform, 2.0 to 2.9 $\mu\mu c/g$, except for the sample collected 30 yards below the outfall. The latter sample contained several times less thorium,

TABLE XV

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM SAWMILL CREEK AND DES PLAINES RIVER, 1958

Date Collected	Sawmill Creek			Des Plaines River		
	Location[a]	$\mu\mu c\alpha/l$	$\mu\mu c\beta/l$	Location[b]	$\mu\mu c\alpha/l$	$\mu\mu c\beta/l$
February 26	A	24	76	A	23	95
February 26	B	47	108	B	27	96
March 19	A	24	78	A	26	63
March 19	B	29	86	B	22	75
April 3	A	20	62	A	19	85
April 3	B	29	82	B	23	80
May 21	A	23	68	A	24	104
May 21	B	49	146	B	24	205
June 18	A	26	107	A	22	81
June 18	B	20	72	B	36	192
July 16	A	24	118	A	23	63
July 16	B	20	39	B	30	165
August 22	A	22	75	A	19	60
August 22	B	29	77	B	23	138
September 17	A	25	96	A	18	89
September 17	B	42	146	B	24	140
October 15	A	24	77	A	22	83
October 15	B	57	132	B	23	142
November 19	A	27	100	A	19	51
November 19	B	18	35	B	27	151
December 19	A	25	91	A	-	-
December 19	B	40	107	B	-	-
Average	A	24	86	A	22	77
	B	35	94	B	26	138

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

[b] 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

uranium, and total activity than any of the other Sawmill Creek samples, and is evidently composed of material that does not adsorb activity well. These results are consistent with the alpha activities found in Creek water, since the water also contained above normal concentrations of uranium and plutonium but only normal amounts of thorium. Uranium and plutonium contamination below the outfall has also been found in other years, and usually in larger amounts than in 1958. The average alpha activity above-site, 24 $\mu\mu c/g$, was essentially the same as found in earlier years and is considered the normal value for the stream bed.

TABLE XVI

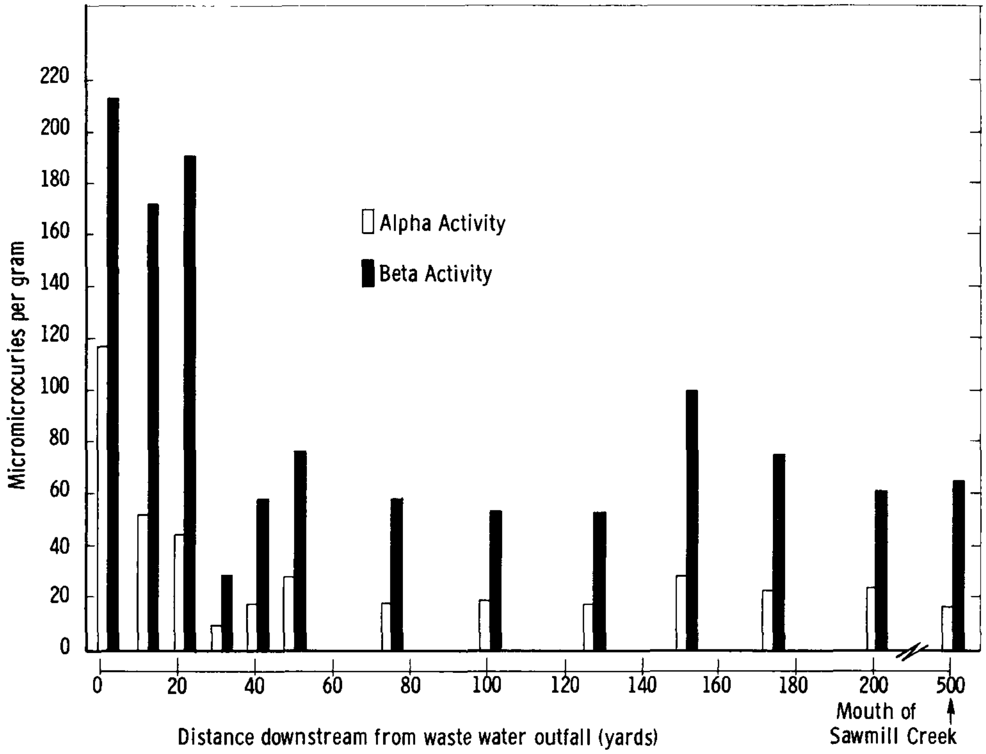
RADIOACTIVITY IN BOTTOM SILT, 1958

Location	Date Collected	Micromicrocuries per Gram				Total Activity	
		U	Th	Pu	Sr^{90}	$\mu\mu c\alpha/g$	$\mu\mu c\beta/g$
Sawmill Creek at waste water outfall	Sept. 24	38.8	2.9	25.6	5.5	118	213
Sawmill Creek, 10 yards below waste water outfall	Sept. 24	5.6	2.0	7.6	1.2	52	171
Sawmill Creek, 20 yards below waste water outfall	Sept. 24	6.9	2.4	10.8	1.0	44	189
Sawmill Creek, 30 yards below waste water outfall	Sept. 24	1.1	.44	<0.1	<1	9.2	29
Sawmill Creek, 20 yards below waste water outfall	Oct. 15	9.7	2.4	9.4	1.0	57	132
Sawmill Creek, above ANL site	Oct. 15	2.2	2.5	<0.1	<1	24	77
DuPage River, Naperville	Nov. 21	2.2	5.4	<0.1	<1	42	144
Pond, ANL site	Nov. 24	1.8	4.4	<0.1	<1	68	82

Beta activity originating in Argonne waste water was also found below the outfall, and fallout activity was apparent at both locations. The normal beta activity in the bed of the Creek is approximately 70 $\mu\mu c/g$. Activities greater than 70 $\mu\mu c/g$ are usually due to fission product fallout or to contamination in Argonne waste water. The presence of contamination from the waste water can be detected in the presence of fallout activity by comparing the relative concentrations of certain fission products. For example, Sr^{90} concentrations greater than 1 $\mu\mu c/g$ was detected only below the outfall, and the concentration decreased from 5.5 to <1 $\mu\mu c/g$ in the first 30 yards. The rate of beta decay also aided in distinguishing between natural, fallout, and waste water activities. On this basis, above normal activity in the below-site samples collected in February, April, May, and October was due primarily to waste water contamination, while in September and November the excess beta activity was due to fallout. To determine the extent to which the bed of the Creek was contaminated by ANL waste water, a survey of the bed was made in September. The results are given in Figure 12. Both the alpha and beta contaminations were confined to the first 20 yards below the outfall. After 20 yards both types of activity were in the normal range. Changes in alpha and beta activities generally paralleled each other, indicating that the nature of the bottom silt material is important in determining the concentrations of both natural and non-natural activities.

Figure 12

NONVOLATILE RADIOACTIVITY IN THE BED OF SAWMILL CREEK, SEPTEMBER, 1958



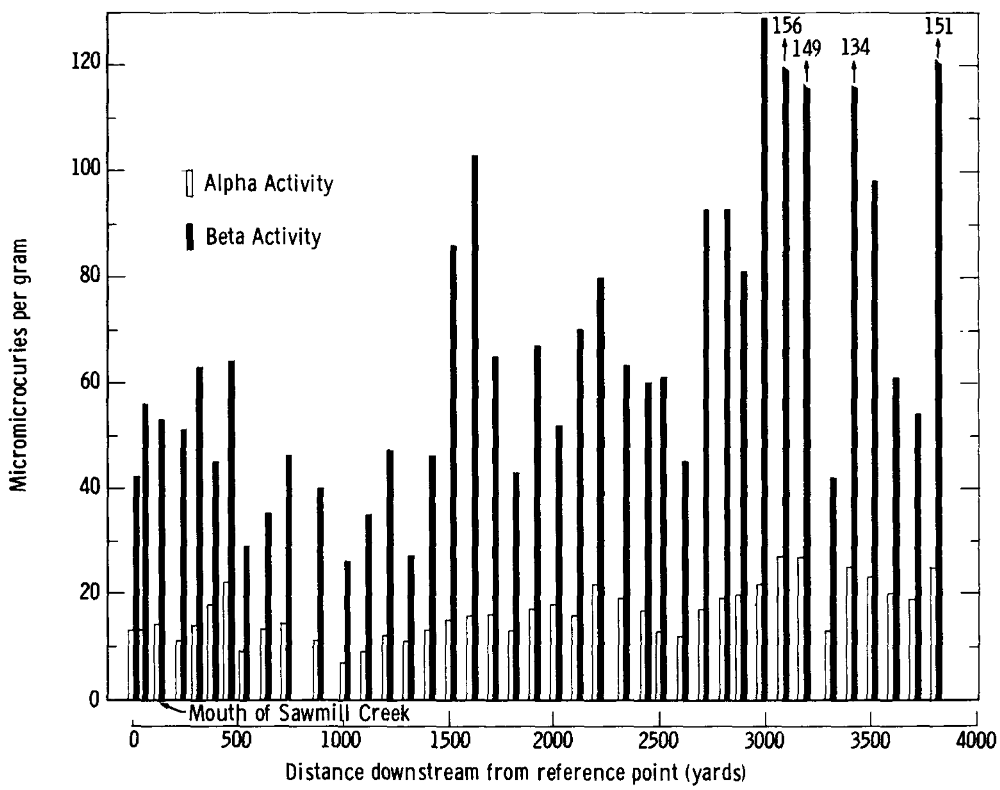
The total activities in bottom silt from two locations in the Des Plaines River are also given in Table XV. The average alpha activity below Sawmill Creek, 26 $\mu\mu$ c/g, was slightly higher than the average above the Creek, 22 $\mu\mu$c/g, primarily as a result of two below-Creek samples containing 30 and 36 $\mu\mu$c/g, respectively. However, all of the samples were in the range previously found in the Des Plaines River bed, and there was little, if any, alpha activity in the bed below Sawmill Creek that originated in ANL waste water.

The beta activities below Sawmill Creek were essentially the same as those above the Creek during the first three months of the year. Beginning with the sample collected on May 21, the beta activities below the Creek were from 1.5 to 3 times greater than those found farther upstream, and this additional activity was the result of fallout activity and did not originate in Sawmill Creek water. Gamma-ray spectra of samples from both locations were similar in that they contained the same relative concentrations of the fission products normally found in fallout. The increase in beta activity may be related to increased fallout activity in air and rain. It was not observed in the sample collected on April 3, and there were only two abnormally beta-active rains in 1958 prior to that date.

The total activity in precipitation before April 3rd was only 43 mc/sq mi, while 60 mc/sq mi was carried down in rain from April 3 to April 30, and larger amounts were carried down in the remaining months of the year. The increased beta activity was not observed in the River above the Creek, probably because the bed material at that sampling location does not have strong adsorptive properties. In this respect the results of a survey of the bed of the Des Plaines River made in September are of interest. The bed was sampled at frequent intervals from 150 yards above to 3800 yards below the mouth of Sawmill Creek. The results are plotted in Figure 13 as a function of distance from an arbitrary reference point. Although there is some fluctuation from point to point, the activities were significantly lower in the first part of the survey. The average activities in the first 3000 yards were 14 $\mu\mu c\alpha/g$ and 56 $\mu\mu c\beta/g$, while the next 900 yards contained an average of 22 $\mu\mu c\alpha/g$ and 108 $\mu\mu c\beta/g$. Similar differences have been observed in other surveys of the same portion of the River and indicate that the absorptive and adsorptive properties of the bed vary considerably. In general, sandy samples contain less activity than samples consisting primarily of clay. The bed at and near the mouth of Sawmill Creek contained only normal concentrations of activity, although it might be expected that any activity due to Sawmill Creek water would have a higher probability of being found in that location than farther downstream.

Figure 13

NONVOLATILE RADIOACTIVITY IN THE BED OF DES PLAINES RIVER, SEPTEMBER, 1958



Since both alpha and beta activities from waste water were found in Sawmill Creek, increases in the Des Plaines River from this source should be found in both types of activity. This is additional evidence that the beta activities in the River were due to fallout. As was the situation with Des Plaines River water, Sawmill Creek water is apparently sufficiently diluted so that the Creek has a negligible effect on the radioactivity of the Des Plaines River bed.

TABLE XVII

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM PONDS ON ANL SITE, 1958

Month	No of Samples	Alpha Activity ($\mu\mu c/g$)		Beta Activity ($\mu\mu c/g$)	
		Max	Avg	Max	Avg
April	4	891[a]	243	1217[a]	355
	3	31[b]	27[b]	74[b]	67[b]
May	3	51	37	68	63
July	4	462[a]	132	798[a]	256
	3	22[b]	21[b]	86[b]	75[b]
November	4	1070[a]	297	2002[a]	561
	3	68[b]	39[b]	102[b]	80[b]
Average		187		325	
		32[b]		71[b]	

[a] Storage lagoon for contaminated waste water

[b] Excluding the storage lagoon

Total activities in bottom silt from ponds on the ANL site are given in Table XVII. The storage lagoon for contaminated waste water contained approximately 10 to 40 times the normal alpha and beta activities found in bottom silt. The active nuclides found in these samples were the same as those found in water from this lagoon (see Section C-2). The shorter-lived fission products predominant in fallout (e.g., Sr^{89}, Ce^{141}, $Zr\text{-}Nb^{95}$) were absent or present in much smaller concentrations than the long-lived fission products. One of the ponds on the site contained abnormal concentrations of natural activity, and these results will be discussed below. All other samples from the ANL ponds contained concentrations of alpha and beta activities similar to those found earlier. All alpha activities were normal, while a few of the samples contained 25 to 50 $\mu\mu c\beta/l$ of fission product fallout.

The alpha activities in bottom silt from lakes and streams near ANL (Tables XVIII and XIX) were normal for each particular body of water. The average beta activities were about 40 $\mu\mu c/g$ higher than found in other years, due to high concentrations of fallout activity in a large fraction of the samples. Since all samples were collected during periods of relatively high fallout, the average beta activity is abnormally high. The July sample from McGinnis Slough (240 $\mu\mu c/g$) contained the second highest concentration of beta activity found this far off the Laboratory site. The highest sample, collected in August, 1952, from the Des Plaines River below the ANL site, contained 248 $\mu\mu c/g$. The high samples collected in 1958, all decayed

TABLE XVIII

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT
NEAR ANL, 1958

Lake or Stream	Location	May		July		November	
		$\mu\mu c\alpha/g$	$\mu\mu c\beta/g$	$\mu\mu c\alpha/g$	$\mu\mu c\beta/g$	$\mu\mu c\alpha/g$	$\mu\mu c\beta/g$
Des Plaines River	31st St, Brookfield	-	-	-	-	23	111
Des Plaines River	Willow Springs	24	104	23	63	-	-
Des Plaines River	Lemont	24	205	30	165	-	-
Des Plaines River	Romeoville	26	112	23	162	18	47
Lake Michigan	98th St, Chicago	-	-	-	-	5	29
Calumet Lake	111th St, Chicago	-	-	-	-	27	102
Confluence of Des Plaines, Kankakee & Illinois Rivers	McKinley Woods	-	-	23	113	-	-
Du Page River	Naperville	55	96	37	94	42	144
Du Page River	Channahon	-	-	35	164	25	82
Saganaskee Slough	104th Ave & Sag Canal	13	45	-	-	-	-
Flag Creek	German Church & Wolf Roads	26	66	-	-	-	-
McGinnis Slough	U S Rt 45 & Ill Rt 7	-	-	23	240	-	-
Illinois River	Morris	-	-	2	19	8	22
Fox River	Aurora	-	-	-	-	5	33
Sauk Lake	Park Forest	-	-	19	70	-	-
Sanitary & Ship Canal	Lemont	-	-	-	-	-	-
Long Run Creek	135th St & Archer Ave	25	80	-	-	31	193
Salt Creek	Wolf Road, Western Springs	-	-	-	-	24	90

at a relatively rapid rate, 15 to 20% during the first month. Except for the Des Plaines River at Naperville, there was no evidence of any source other than fallout for the above-normal beta activities in these samples.

As in the past, bottom silt from the DuPage River at Naperville contained up to twice the normal concentrations of radioactivity. A similar situation was found in a natural pond on the ANL site (Table XVII). Additional analyses on some of these samples are given in Table XVI. These samples are of interest in that they contained normal concentrations of uranium, but approximately twice the normal concentration of thorium isotopes. The thorium fractions appeared to consist primarily of Th^{232} and Th^{228} from the thorium chain, although the presence of other thorium

TABLE XIX

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM LAKES AND STREAMS NEAR ANL, 1958

Month	Distance from ANL (miles)	No. of Samples	Alpha Activity ($\mu\mu c$/gram)		Beta Activity ($\mu\mu c$/gram)	
			Max.	Avg.	Max.	Avg.
May	10	5	55	28	205	99
July	10	4	37	28	240	125
November	10	4	42	30	193	135
Average	10		29		118	
May	25	3	26	24	112	96
July	25	5	35	25	162	106
November	25	5	27	15	102	53
Average	25		21		83	
Average	10 & 25		25		101	

isotopes cannot be excluded on the basis of results obtained thus far. Additional analyses on larger samples are planned to further identify the nuclides present.

Total activities in bottom silt from the reference sites are given in Table XX. All results were normal; the average values, 5.5 $\mu\mu c\alpha/g$ and 32 $\mu\mu c\beta/g$, as well as the individual values, were similar to those found previously at these locations.

TABLE XX

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM REFERENCE SITES, 1958

Location	July		November	
	$\mu\mu c\alpha/g$	$\mu\mu c\beta/g$	$\mu\mu c\alpha/g$	$\mu\mu c\beta/g$
Lake Delavan, Wis.	5.6	38	10	28
Lake Geneva, Wis.	2.0	18	-	-
Fox River, Oak Point State Park, Wis.	-	-	4	31
Illinois River, Starved Rock, Ill.	1.3	3.9	-	-
Lake Shafer, Monticello, Ind.	4.8	41	10	55
Tippecanoe River, Monon, Ind.	-	-	9.8	62
Lake Michigan, St. Joseph, Mich.	4.6	24	-	-
Pipestone Lake, Mich.	2.5	20	3.2	26
Average	3.5	24	7.4	40

E. Surface Soil

The total activities in surface soil collected on the ANL site are given in Table XXI. Except for the samples collected near a uranium storage shed, the alpha and beta activities were very similar to those found on the site in previous years and to those found near ANL this year. Some of the samples contained 10 to 40 $\mu\mu c/g$ of fallout activity. The fallout activity, however, was no greater than found in some of the other years.

TABLE XXI

NONVOLATILE RADIOACTIVITY IN SURFACE SOIL ON ANL SITE, 1958

Month	No. of Samples	Alpha Activity ($\mu\mu c/g$)		Beta Activity ($\mu\mu c/g$)	
		Max.	Avg.	Max.	Avg.
April	5	23	21	77	67
May	4 3	32[a] 23[b]	24 22[b]	91[a] 71[b]	72 66[b]
July	5	75[a]	48[a]	202[a]	140[a]
November	6	137[a] 26[b]	40 20[b]	508[a] 89[b]	151 78[b]
Average		34 21[b]		111 72[b]	

[a]Near uranium storage shed.

[b]Excluding samples collected near storage shed.

The area adjacent to the uranium storage shed contained up to five times the normal concentrations of alpha and beta activity. As indicated by the date in Table XXII, this additional activity is primarily due to uranium. Normal uranium concentrations in surface soil are about 3 $\mu\mu cU/g$, while two samples of soil near the shed contained 43 and 116 $\mu\mu cU/g$. This contamination of the soil near the shed has been found in other years. Thorium, plutonium, and strontium concentrations in these samples were in the normal ranges.

TABLE XXII

RADIOACTIVITY IN SOIL, 1958

Location	Date Collected	Micromicrocuries per Gram					
		U	Th	Pu	Sr^{90}	Total Alpha	Total Beta
ANL[a]	Aug. 1	43	3.2	<0.1	<1	75	202
ANL[a]	Nov. 24	116	2.9	<0.1	<1	137	508
ANL	Nov. 24	3.3	2.9	<0.1	<1	26	77
Naperville	Nov. 21	3.3	8.4	<0.1	<1	63	87

[a] Near uranium storage shed.

The total activities in surface soil collected off the ANL site are given in Tables XXIII and XXIV. The alpha activities in samples collected off-site were essentially the same as found earlier at the same location. Within 25 miles of the Laboratory the average alpha activity was 22 $\mu\mu c/g$, and the annual averages found from 1952 through 1957 ranged from 20 to 25 $\mu\mu c/g$. Except for one sample containing 63 $\mu\mu c\alpha/g$, the

TABLE XXIII

NONVOLATILE RADIOACTIVITY IN
SURFACE SOIL NEAR ANL, 1958

Month	Distance from ANL (miles)	No of Samples	Alpha Activity ($\mu\mu c/g$)		Beta Activity ($\mu\mu c/g$)	
			Max	Avg	Max	Avg
April	10	2	24	21	61	54
May	10	4	25	21	71	62
July	10	4	30	20	87	76
November	10	6	63	30	96	85
Average	10		24		73	
May	25	3	22	21	64	62
July	25	4	24	18	66	56
November	25	5	27	20	247	111
Average	25		20		80	
Average	10 & 25		22		76	

range of activities was also normal, from 11 to 34 $\mu\mu c/g$. The alpha activities in the reference site samples averaged 14 $\mu\mu c/g$, compared with earlier annual averages ranging from 13 to 18 $\mu\mu c/g$. The abnormally high alpha sample (63 $\mu\mu c/g$) was obtained near the DuPage River where bottom silt alpha activities were also above normal. Additional analyses of this sample are given in Table XXII. As was the case for the bottom silt samples, the additional activity was due to thorium isotopes and their daughters. The thorium activity in this sample was 8.4 $\mu\mu c/g$, while all the other soil samples analyzed contained only about 3 $\mu\mu c/g$. The uranium concentration was normal, 3.3 $\mu\mu c/g$.

TABLE XXIV

NONVOLATILE RADIOACTIVITY IN SURFACE SOIL
AT REFERENCE SITES, 1958

Location	July		November	
	$\mu\mu c\alpha/g$	$\mu\mu c\beta/g$	$\mu\mu c\alpha/g$	$\mu\mu c\beta/g$
Delavan, Wis	16	50	16	46
Geneva, Wis	5	33	-	-
Oak Point State Park, Wis	-	-	14	48
Starved Rock State Park, Ill	30	30	22	54
Monticello, Ind	12	47	13	37
Monon, Ind	-	-	17	60
St Joseph, Mich	8	33	20	51
Pipestone Lake, Mich	5	26	2 4	19
Average	13	37	15	45

The average beta activity in soil collected near ANL was slightly higher than found previously. This increase was due primarily to increased fallout activity in the November samples, although all samples contained some fission products. The average beta activity in 1958 was 76 $\mu\mu c/g$, compared with averages ranging from 53 $\mu\mu c/g$ to 70 $\mu\mu c/g$ from 1952 through 1957.

Increased fission product activity was not found in most of the reference site samples, and the average value 41 $\mu\mu c \beta/g$ was similar to averages obtained since 1953. The highest beta activities in the reference site samples were found in 1952, when the average was 76 $\mu\mu c/g$.

F. Plants

Plant sampling was limited to grass because it could be found at all sampling locations and, thus, intercomparison of results were more reliable. The results obtained for grass collected in 1958 are given in Tables XXV, XXVI, XXVII, and XXVIII in terms of the oven-dried plant. The alpha activities at all locations were low and quite similar; the average on-site was 1.0 $\mu\mu c/g$ while the off-site average was 0.8 $\mu\mu c/g$ near ANL and 0.9 $\mu\mu c/g$ at the reference sites. These averages are approximately two-thirds lower than has usually been found at each location in the past. Only four samples in 1958 contained alpha activities greater than 2 $\mu\mu c/g$ and none contained more than 6 $\mu\mu c/g$. In other years a larger fraction of the samples contained more than 2 $\mu\mu c/g$, although the range of alpha activities was about the same in 1958 as earlier.

TABLE XXV

NONVOLATILE RADIOACTIVITY IN GRASS ON ANL SITE, 1958

Month	No of Samples	Alpha Activity ($\mu\mu c/l$)		Beta Activity ($\mu\mu c/l$)	
		Max	Avg	Max	Avg
April	4	1 5	1 3	101	86
May	4	0 7	0 6	97	83
July	4	5 0[a]	2 1	178[a]	163
November	6	3 0[a]	1 0	410[a]	268
Average		4 0 1 0[b]		163 103[b]	

[a] Near uranium storage shed

[b] Average excluding samples collected near storage shed

Two samples collected near the uranium storage shed contained above average concentrations of alpha activity, 3 and 5 $\mu\mu c/g$. As was the case for soil from the same location, the additional alpha activity was due to uranium. The July sample obtained near the shed contained 4.7 $\mu\mu c U/g$, while the other samples analyzed for uranium contained from 0.06 to 0.16 $\mu\mu c U/g$. The latter values are in the range previously found in grass both on and off the ANL site.

The beta activities in grass on the ANL site and from locations near ANL averaged 163 and 119 $\mu\mu c/g$, respectively, and were approximately the same as in 1957 and 1955, but several times greater than in

TABLE XXVI

NONVOLATILE RADIOACTIVITY IN
GRASS NEAR ANL SITE, 1958

Month	Distance from ANL (miles)	No of Samples	Alpha Activity (μμc/gram)		Beta Activity (μμc/gram)	
			Max	Avg	Max	Avg
April	10	2	1 3	1 1	65	64
May	10	4	1 0	0 5	79	68
July	10	4	0 9	0 5	169	142
November	10	4	1 1	0 9	330	209
Average	10		0 7		129	
May	25	3	0 3	0 2	63	59
July	25	4	5 6	1 7	170	130
November	25	5	1 9	0 7	268	127
Average	25		0 9		111	
Average	10 & 25		0 8		119	

TABLE XXVII

NONVOLATILE RADIOACTIVITY IN GRASS
FROM REFERENCE SITES, 1958

Location	July		November	
	μμcα/g	μμcβ/g	μμcα/g	μμcβ/g
Delavan, Wis	-	-	0 80	202
Geneva, Wis	0 14	86	0 17	51
Oak Point State Park, Wis	-	-	0 9	250
Starved Rock, Ill	0 57	176	5 2	196
Monticello, Ind	0 35	158	0 48	496
St Joseph, Mich	0 50	160	0 72	679
Average	0 39	145	1 5	340

TABLE XXVIII

RADIOACTIVITY IN GRASS, 1958

Location	Date Collected	Micromicrocuries per Gram				
		U	Sr^{89}	Sr^{90}	Total Alpha	Total Beta
ANL	May 28	0 16	1 5	1 1	0 6	95
ANL[a]	July 31	4 7	17 5	1 1	5 0	178
ANL	Nov 24	0 07	14 8	0 8	0 5	364
104th Ave and Sag Canal	May 23	0 06	2 0	1 1	0 3	79

[a]Near uranium storage shed

other years. The average beta activity in grass from the reference sites, 253 $\mu\mu c/g$, was about twice as great as in the other 1958 samples and three to five times greater than samples from the same locations in other years. All abnormally high beta activities from all locations, including those collected near the uranium storage shed on-site, were due to fallout. The presence of fission product activity in the samples was confirmed by gamma-ray spectroscopy, fission product analysis, and total decay rates. The highest beta activities were found in samples collected during July and November; smaller concentrations of fission products were present in the April and May samples. This is consistent with the beta activities found in air and rain. Because reference site samples were collected only in July and November, and other locations were sampled earlier in the year as well, direct comparison of annual averages are misleading. During July all three groups of locations contained about the same amount of activity. In November, however, fallout activity in grass was greater at the reference sites than at locations on and near the Laboratory. In spite of the relatively large amount of mixed fission products present in many of the samples, the Sr^{90} concentrations, about 1 $\mu\mu c/g$, were not much greater than was found in earlier samples. The shorter-lived Sr^{89}, however, was present in greater concentrations than usual, and this indicates fallout activity of relatively recent origin.

Except for the small amount of UX_1 and UX_2 presumably present in grass near the uranium storage shed on the ANL site, there was no evidence to indicate that any of the beta activity in the on-site grass samples originated at ANL.

From the results given, it appears that grass tends to concentrate fallout activity and is thus a more sensitive indicator of fallout than soil, water, and possibly air, when only total beta counting is used. For example, soil samples from the reference sites did not show abnormal beta activities, although grass from the same locations at the same times were obviously contaminated with fission products.

G. Animals

The value of the results obtainable from animal samples was not believed to justify an extensive sampling program similar to that undertaken for the other types of samples. Animal collection was limited to fish from the ponds on the ANL site, since these ponds drain a significant fraction of the site. Two samples of bullhead and bass collected in September each contained 0.1 $\mu\mu c\alpha/g$. The alpha activities in earlier samples from the same ponds ranged from 1.7 $\mu\mu c/g$ in 1953 to 0.1 $\mu\mu c/g$ in 1957, and, thus, there has been no long-term accumulation of alpha activity in the fish population. This was expected from the uniformly low alpha activities found in water samples from the ponds. However, it is possible that contamination at a very low level might remain undetected in the water, while the fish could indicate such a situation by concentrating the activity. Evidently this has not occurred.

The beta activities have also shown no definite change in recent years. The beta activities in the 1958 samples were 24 and 15 $\mu\mu c/g$, respectively. Earlier results for fish from these ponds were 16 $\mu\mu c/g$ in 1953, 23 $\mu\mu c/g$ in 1954 and 1956, and 11 $\mu\mu c/g$ in 1957.

H. Site A Samples

Samples of soil were collected in November near an area formerly used as a burial grounds for contaminated waste. The burial grounds, designated as Plot "M," is located at Site A of ANL, about 4 miles east of the main ANL grounds. The burial plot, a rectangular area about 125 feet square, was capped with concrete in 1954. The concrete covered the surface of the plot and extended four feet down into the ground around the entire perimeter. The purpose of the concrete was to prevent disturbance of the active material remaining in the plot. The ground north and east of plot M slopes downhill, and an intermittent stream flows in a northeasterly direction around the plot and eventually drains into the Des Plaines River. This creek and the soil near the plot were sampled in 1954 prior to the capping to determine if any activity had been leached from the plot by ground or surface water. No evidence of any movement of activity was found. The results are discussed in ANL-5446. Soil borings down to a depth of five feet were collected this year from a point 15 feet northeast of the plot. The alpha activities in these samples varied from 23 to 30 $\mu\mu c/g$ and the beta activities from 54 to 66 $\mu\mu c/g$. The values did not change in any regular manner with depth. These results were all normal and not significantly different from the samples collected in 1954 from the same location. The latter samples contained 20 to 28 $\mu\mu c\alpha/g$ and 58 to 71 $\mu\mu c\beta/g$. A sample of surface soil collected from the east slope, between the plot and the creek, contained 26 $\mu\mu c\alpha/g$ and 77 $\mu\mu c\beta/g$, and these activities are also normal. The creek draining the area was dry at the time of sampling, but a sample of the bed collected downstream from the plot also contained normal concentrations of radioactivity, 17 $\mu\mu c\alpha/g$ and 54 $\mu\mu c\beta/g$. Thus, no evidence of movement of the activity from the burial grounds was detected five years after the initial sampling. The area will be sampled periodically in the future.