LAKE MICHIGAN STUDIES

Special Report Number LM 6

RADIOCHEMICAL INVESTIGATIONS

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U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service
Division of Water Supply and Pollution Control
Great Lakes-Illinois River Basins Project

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INTRODUCTION

An investigation of the existing radioactive contamination of Lake Michigan was begun in April of 1962. Potential sources of radioactive contamination have been identified, information has been assembled on levels of radioactivity in tributaries to the Lake, and samples from Lake Michigan have been collected and analyzed.

This report includes results of samples collected and analyzed by the Great Lakes-Illinois River Basins Project from April 1962 until February 1963. Also included are selected data on radioisotope users on the tributaries to Lake Michigan which were provided by the states of Michigan and Wisconsin.

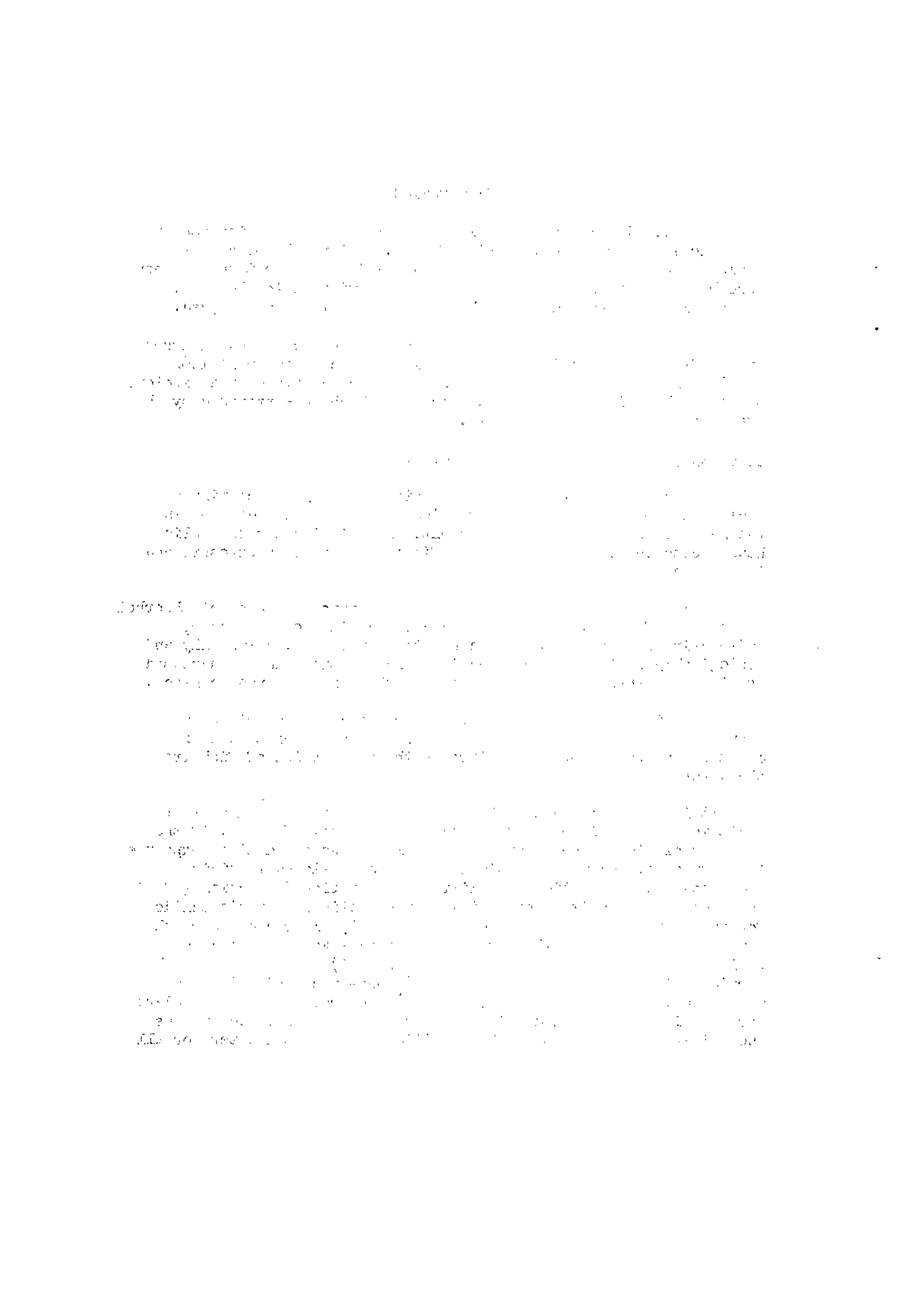
Significance of Radiological Contamination

Expanding production and use of atomic energy and continued nuclear weapons testing are increasing the amount of radiation in the general environment. As a result, there is increasing public health concern over the long term effects of radiation exposure and in radioactive contamination of the environment.

Radioactive wastes discharged to the environment are not absorbed in harmless fashion. Even though decay and dilution may occur, radioactive wastes may be reconcentrated physically, chemically and biologically so that the radioactive concentration can be increased as it passes through the environment to the point of human contact.

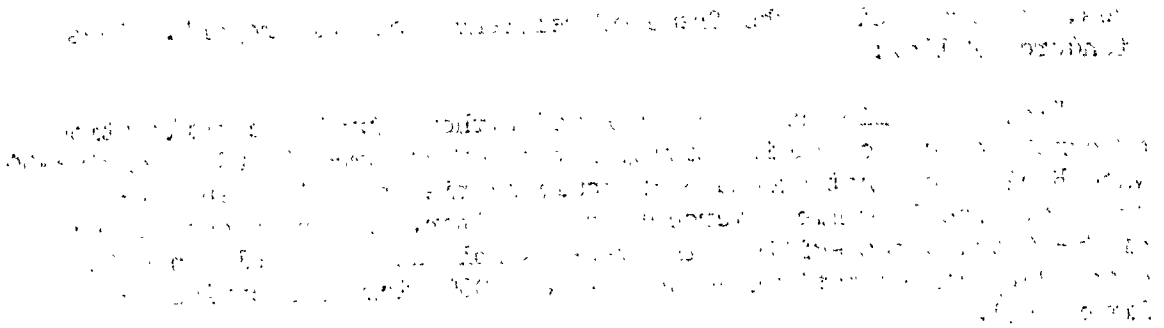
Contamination of surface water by radioactive materials can result in human exposure through the use of the water as a source of municipal water supply and through the consumption of fish from the water.

It is generally agreed that all unnecessary human exposure to radiation is undesirable and should be prevented. However, if the benefits of atomic energy are to be utilized, some radiation exposure is inevitable, and various guides have been developed to minimize the exposure. The National Committee on Radiation Protection (NCRP) has recommended maximum permissible concentrations of radionuclides for occupational exposure for many years. (1) In recent years the Federal Radiation Council (FRC) has provided a Federal policy on radiation exposure for the general public. (2) The Public Health Service Drinking Water Standards of 1962 have received widespread use by various regulating agencies. (3) The radioactivity standards for drinking water included in the above are generally accepted as the criteria for evaluating the condition of untreated water for all



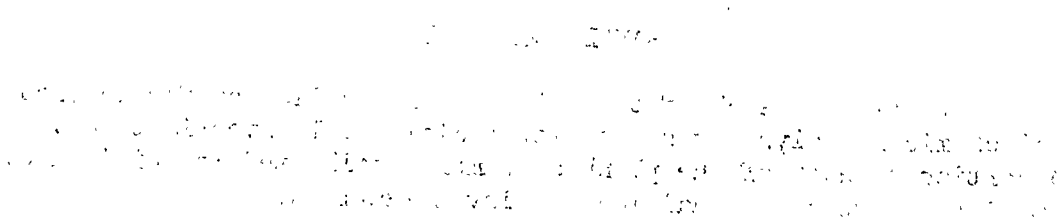
uses, and are used as the frame of reference for this report. These standards follow:

"Water supplies shall be approved without further consideration of other sources of radioactivity intake of Radium-226 and Strontium-90 when Ra-226 does not exceed 3 micromicrocuries per liter and Sr-90 does not exceed 10 micromicrocuries per liter. In the known absence of Sr-90 and alpha emitters the water supply is acceptable when the gross Beta concentrations do not exceed 1000 micromicrocuries per liter" (3).



SAMPLE COLLECTION

A portion of each of the water samples collected for physical and chemical analyses was used for radiological investigations. A portion of each of the plankton samples collected for biological studies was used for radiological investigations.



LABORATORY PROCEDURE

Radiological determinations were made at the Project laboratory. The determinations were made under the supervision of an experienced radiochemist. Analytical procedures followed the methods described in Standard Methods for the Examination of Water and Wastewater (4) and the Radionuclide Analysis Laboratory Manual of the Public Health Service (5).

Water samples were prepared on two inch diameter cupped aluminum planchets. Plankton samples were ashed and prepared on similar stainless steel planchets. Suspended solids were separated by membrane filter, transferred to a planchet and burned with ethyl alcohol. Dissolved solids were obtained by careful evaporation of the water sample on a hot plate kept below the boiling point, then complete transfer of the solids to a planchet. All planchets were dried and fixed when necessary with lucite in acetone.

All of the samples were counted at least 59 days after the date of collection. This delay in counting insures that any fallout in the samples is at least 59 days old. In comparing results of analyses, this rules out variations which could be caused by the effect of fresh fallout products with short half lives.

Counting was done in a windowless internal proportional counter (Nuclear Chicago D48) with a two inch lead shield and automatic sample changer (NC Model C210 Special), combined with an NC Model 202 Scaler and Hewlett Packard 560A Digital Recorder. All samples were counted for thirty minutes (three 10 min. counts) and corrections were made for geometry (G), backscatter (B), self-absorption (A), sample volume (V), and background using the general equations:

Net cpm/GBAV 2.22 = gross radioactivity, $\mu\mu c/l \pm$ C.E.

where: C.E. ($\pm\mu\mu c/l$) = $1.96\ (cpm_s/t_s + cpm_b/t_b)^{\frac{1}{2}}/GBAV\ 2.22$

where: C.E. = counting error at 95% confidence level

cpm_s = counts per minute sample

cpm_b = counts per minute background

net cpm = $cpm_s - cpm_b$

t_s = counting time; sample

t_b = counting time; background

SOURCES OF RADIOACTIVE WASTE

Potential sources of radioactive wastes which could reasonably be expected to find their way into Lake Michigan can be grouped into these categories:

A. Nuclear Reactors

B. Fallout from Nuclear Weapons Testing

C. Radioisotope Users

These are in addition to the background radiation from naturally occurring radionuclides in the earth's surface.

A discussion of each of these sources of radioactive wastes follows:

A. Nuclear Reactors (Big Rock Point)

At the present time the Big Rock Point Reactor is the only reactor located on Lake Michigan or its tributaries. The reactor is located on the south shore of Little Traverse Bay, Lake Michigan, five miles northeast of Charlevoix, Michigan (see Fig. 11). The immediate vicinity of the reactor is heavily wooded and sparsely populated and the shore line is rocky and barren. Big Rock is a 48,000 KWE[1], 157,000 KWT[2] high power density, oxide fueled, direct cycle boiling water reactor. The plant is owned and operated by the Consumers Power Company of Michigan and was constructed by General Electric Company. The plant was completed in 1962 and became critical in September of 1962. Full power operation is expected early in 1963.

Liquid wastes generated by the reactor are collected and routed to a liquid radioactive waste system for appropriate treatment, monitoring and release under batch control. Cooling water and service water can become radioactive only as a result of a leak in the heat exchanger. This water can be released continuously but the release is monitored. 50,000 gpm of cooling water is available for diluting radioactive wastes.

The plant discharge is limited by AEC and Michigan rules and regulations (6) (7). Waste containing an unidentified mixture of

1 KWE = Kilowatt electrical
2 KWT = Kilowatt thermal

radionuclides may be discharged if the concentrations averaged over 365 consecutive days do not exceed 10^{-8} μc/ml above that of plant intake water from Lake Michigan. If the absence of Ra-226 and Ra-288 is demonstrated by appropriate analysis, the above limits may be raised to 10^{-7} μc/ml. All wastes are released to a discharge canal which empties into Lake Michigan.

Accidental release of primary reactor water is the worst potential source of radiological contamination which might be available. If such liquid were drained to the waste collection tanks and accidentally pumped to discharge at the maximum pumping rate available, the concentration of the discharge canal would be on the order of 0.1 μc/ml. Very large dilutions would occur within a short time after release and combined decay and diffusion would limit the region of significant contamination to a relatively small area. Any other surface or underground leakage would be controlled by diking and by the low permeability of the soil (8).

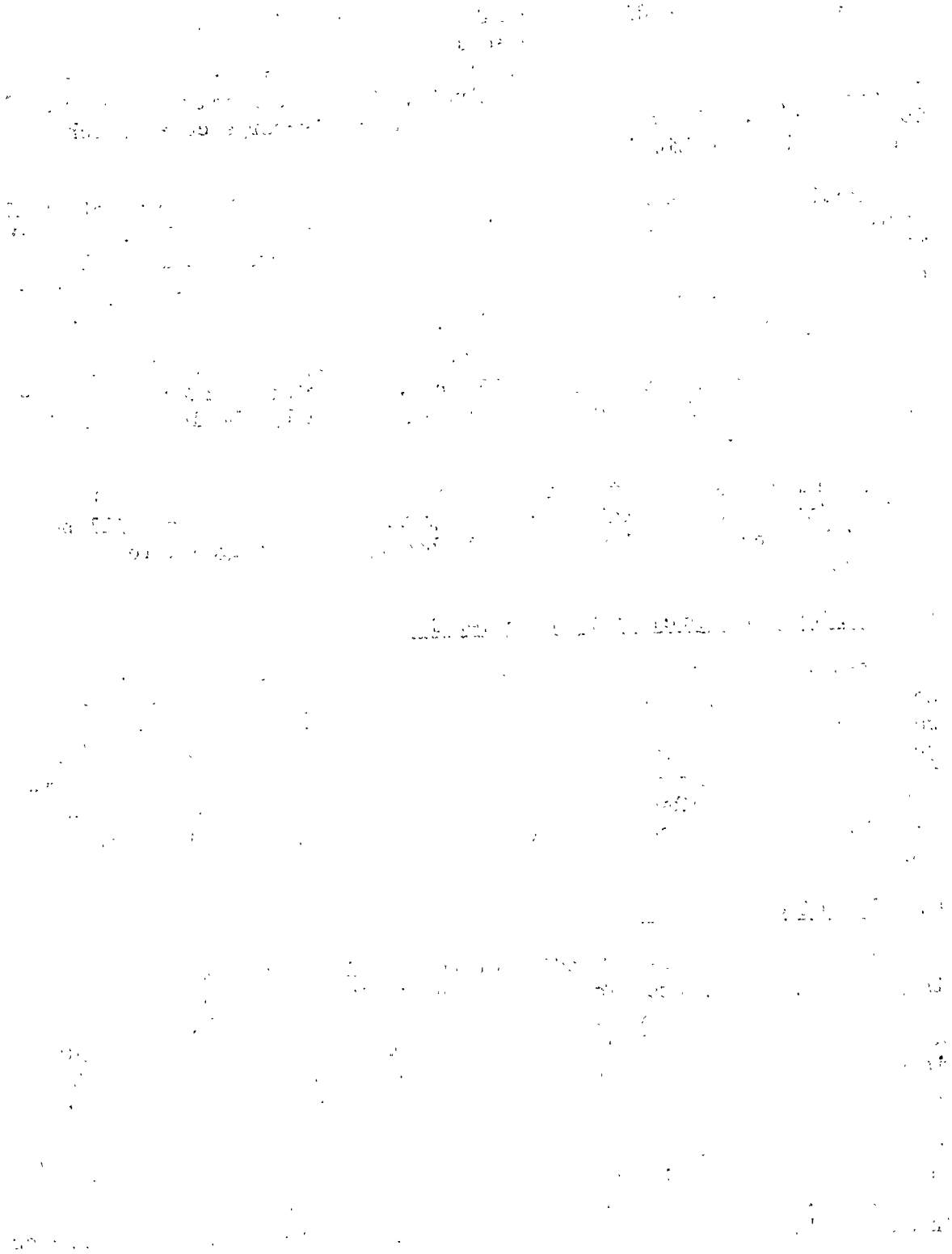
It can be expected that under normal operating conditions the radioactivity added to Lake Michigan by the Big Rock Reactor will be well within acceptable limits established by the Michigan Water Resources Commission (7).

B. Fallout from Nuclear Weapons Testing

The contribution from fallout is probably the most significant source of radioactive pollution in Lake Michigan. Besides the fallout which occurs directly on the lake surface, additional and probably greater amounts are collected by surface runoff to the tributaries to Lake Michigan and from thence into the lake. In addition to the fallout, each of the tributaries may carry additional amounts of radioactivity from the permitted discharge of the licensed radioisotope users on the watershed.

C. Radioisotope Users

Production and use of radioisotopes has increased steadily since they were first released for general use in 1946. The radioisotopes are licensed for use and regulated by the AEC for medical, educational and industrial users. Much of the total activity shipped by the AEC is used as sealed sources (9). Sealed sources are designed so that leakage of material is prevented. The bulk of remaining isotopes in use have half lives of less than 30 days and are used chiefly in medical diagnosis and therapy as well as in industrial development and research. Some of this material can be expected to find its way into the sewers; however, the amount would be small. Of 263 licenses in Michigan, 15% are located on the Lake Michigan watershed. Of 124 licenses



in Wisconsin 65% are located on the Lake Michigan watershed. Roughly 77% of the Michigan and 23% of the Wisconsin licenses are for material in sealed sources. A summary of unsealed sources in these states appears in Table 1. A few more sources are undoubtedly located on the relatively small area of the Lake Michigan watershed which is in Illinois and Indiana but these sources were not pinpointed. It is not possible to determine from this information how much of this activity reaches the sewer system since much of it is either utilized or stored and is either discharged to the sewer over a period of time or after a period of significant decay.

RESULTS OF ANALYSES

Lake Michigan Water Samples

Results of gross radioactivity analyses of Lake Michigan water samples are shown on Figures 1 through 5. Five cruises on Lake Michigan from April, 1962 to October, 1962 are shown on these figures.

Gross beta radioactivity analyses in suspended solids and in dissolved solids were made separately but the results were combined in presenting these figures in order to simplify the presentation of data. Gross alpha analyses were also made on these samples but so few alpha results were above 3μμc/liter that it was not meaningful to plot the results. 99% of the total alpha results were below 3μμc/liter. The few alpha results which were above 3μμc/liter are shown in Table 2. Each cruise is presented separately on these figures even though some of the same stations were collected on different cruises. A comparison of the data from these stations make it readily apparent that considerable variation in the radioactivity concentrations occurred between the dates of these cruises. Since complete coverage of the lake requires several months to obtain, it is difficult to present the results of these cruises as a whole in an effort to establish an overall pattern of radioactivity in the lake. Similarly, since each cruise was on a different area of the lake (except for Cruises 1 and 3) there was insufficient sampling of the same stations to draw any conclusions regarding seasonal variations.

Figure 1 shows the results of gross beta activity analyses for total solids on samples collected in the deep water in the southern half of Lake Michigan during April and May 1962. To make interpretation of the data as uncomplicated as possible all results of less than 10 micromicrocuries per liter (10 μμc/l) are indicated by an open square on the graph. All results between 10 μμc/l and 20 μμc/l are indicated by a shaded square and numerical values are entered directly in the square for all results above 20 μμc/l. The depth in meters at which the sample was collected is indicated by the small number directly beneath each square. It can be seen by a fairly rapid scan of this figure that roughly half of the gross beta results fell below 10 μμc/l and 37% of the results are in the range of 10 to 20 μμc/l. Values in excess of 20 μμc/l (namely 20, 22, 23, 23, 24, 26, 27 and 39) occurred in 8 locations which are readily discernible. There seems to be a random distribution of these high results both as to location and in depth.

Figure 2 shows the results of gross beta activity analyses on samples collected in the northern half of Lake Michigan during June and July 1962. The data is presented in the same manner as described for Figure 1. A rapid scan of this figure reveals approximately the same percentage distribution of 10 μμc/l and 20 μμc/l results as in Figure 1. Five values were in excess of 20 μμc/l.

Figure 3 shows the results of gross beta analyses on samples collected in the same area as in Cruise 1 (Figure 1) but collected three months later.

A comparison of the results on these cruises (Figures 1 and 3) proves very interesting. In Figure 3, 75% of the results are less than 10 μμc/l, compared to 49% in Figure 1; all of the results in Figure 3 are less than 20 μμc/l whereas 14% of the results in Figure 1 are in excess of 20 μμc/l. These results will be referred to again later in discussing the plankton analysis. Figures 4 and 5 show the results of gross beta analyses on inshore samples collected on the west side of the lower lake during August and September and on the east side during October of 1962. A study of these data shows that 40% of the samples near the west shore are above 10 μμc/l whereas only 14% of the samples on the east shore are above 10 μμc/l. This percentage on the west shore compares favorably with the percentage distribution found on deepwater study (Figures 1 and 2). But the results near the east shore and in Cruise 3 were in a class of their own. Two high values of 23 μμc/l and 112 μμc/l are found on Figure 5. These results both occurred quite close to the mouth of two of the major tributaries, the Muskegon River and the St. Joseph River and could be expected at these points as a reflection of the levels of radioactivity frequently encountered in streams. High values of 47 and 53 found on the south shore off Tremont, Indiana, and 25 at the Indiana-Illinois Line are not so readily explained however, nor is the high result of 87 μμc/l found offshore from Highland Park, Illinois.

Plankton Studies

Figures 6 to 10 show the results of plankton studies in Lake Michigan on the same cruises and same dates as described in Figures 1 to 5 for gross beta analyses of water samples. The plankton results are presented in a slightly different manner. The radioactivity levels in plankton are higher due to the concentrating effect in the plankton. The plankton sample is obtained by a tow from near the bottom of the lake to the top surface. This provides a rough composite sample of all depths, and allows the alpha and beta results to be presented graphically side by side since only two basic symbols are needed.

The alpha results are presented as an open circle for all results less than 3 μμc/g and by numerical designation inside the circle for all results greater than 3 μμc/g. The beta results are presented beside the alpha results by use of a square. An open square is used for results less than 10 μμc/g, a quarter of the square is shaded for results less than 25 μμc/g (but more than 10 μμc/g). Half of the square is shaded for results more than 25 but less than 50. The range 50 to 75 is shown by three quarters of the square being shaded and from 75 to 100 by shading the entire square. All results greater than 100 are indicated by showing the numerical result above the shaded square.

A percentage distribution comparison of both the water and plankton beta radioactivity results is made in Table 3. A study of this table reveals that (as was pointed out earlier) from Cruise 1 to Cruise 3, which were both deepwater cruises in the lower half of the lake but three months apart, the percentage of the water samples above 10 μμc/l dropped from 50% to 25% and the percentage above 20 μμc/l dropped from 14% to zero. Now, looking at the plankton results it is found that 93% of the Cruise 3 plankton results are greater than 100 μμc/gram as compared to only 4% of the Cruise 1 plankton results. A comparison of the cruises on this table also reveals that the highest radioactivity results in the plankton samples (Cruise 3) were obtained simultaneously with the lowest radioactivity levels found in the water samples. These results indicate that the known ability of plankton to concentrate radioactivity may be responsible for the above observation.

Comparing percent distribution of the plankton results of Cruise 2 and 3 with the other cruises suggests increased radiation levels per gram of plankton in June and July. However these samples were collected in different areas of the lake so nothing more than a general observation can be made, which may or may not be supported by further study.

The geographic distribution of high beta radioactivity levels in plankton seems to be entirely at random. A study of the Figures 6 through 10, reveal the high results to be scattered without any pattern. Cruise 3 which had high results from 100 to 700 on nearly all stations, exhibited a uniform distribution of the highest and lowest values.

High alpha results in plankton also exhibit the same random geographical distribution as the beta results, although the alpha results are only a fraction of the beta results and are not as noteworthy. It is interesting to see, however, that on Cruise 3 when the plankton beta results were at their highest levels the alpha remained the same or perhaps less than on the other cruises.

SUMMARY AND CONCLUSIONS

The contribution from fallout is probably the most significant source of radioactive pollution in Lake Michigan. In addition to fallout which occurs directly on the lake surface, fallout collected by surface runoff is also contributed to the lake by the tributaries. These tributaries also carry additional amounts of radioactivity from the permitted discharge of licensed radioisotope users. Radioactive wastes are also discharged to the lake from the recently completed Big Rock Point nuclear power reactor, the only reactor on the lake or its watershed at present. Under normal operating conditions the radioactivity from this source is expected to be within acceptable limits.

Water sample radioactivity analyses reveal only a few moderately high results. The few alpha results found which were greater than 3 μμc/l were all located in the southern half of Lake Michigan and all except one were more than ten miles from shore.

The few moderately high beta results found (20 to 112 μμc/l) were evenly distributed between deepwater and inshore samples and randomly distributed throughout the entire lake.

Studies of radioactivity in plankton samples also demonstrate the same random distribution of high results throughout the entire lake for both alpha and beta results. The highest radiation levels in plankton samples were obtained in the summer, simultaneously with the lowest results found in water samples. The ability of plankton to concentrate radioactivity may have been responsible for this observation.

Results of the radioactivity in the effluent from the Chicago sewage treatment plants were presented in the Report on the Illinois River System, Part II, Table V-14. The radioactivity found in the effluent was no higher than some of the levels found in the lake, therefore it does not appear that the return of this effluent to the lake would have any appreciable effect on the radioactivity levels in Lake Michigan.

REFERENCES

1. Handbook 69. National Bureau of Standards, U.S. Government Printing Office (June, 1959).

2. Background Material for the Development of Radiation Protection Standards. Staff Report No. 1, Federal Radiation Council, U.S. Government Printing Office (May 13, 1960).

3. Public Health Service Drinking Water Standards. U.S. Department of Health, Education and Welfare, Public Health Service, Washington 25, D.C. (1962).

4. Standard Methods for the Examination of Water and Waste Water, 11th Edition. American Public Health Association, American Water Works Association, Water Pollution Control Federation (1960).

5. Radionuclide Analysis of Environmental Samples. Technical Report R-59-6, Robert A. Taft Sanitary Engineering Center, U.S. Public Health Service, Cincinnati, Ohio (April 1962).

6. Atomic Energy Commission Rules and Regulations Title 10, Code of Federal Regulations, Chapter 1, Parts 20 and 100.

7. Order of the Michigan Water Resources Commission in accordance with Act 245, Public Acts of 1929 as amended by Act 117, Public Acts of 1949 (January 1961).

8. Hazards Summary Report, Big Rock Point Reactor, Consumers Power Company (December 1961).

9. Hearings Before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy, Industrial Radioactive Waste Disposal, I:140, 708; III:2488, U.S. Government Printing Office (1959).

TABLE 1
Unsealed Isotope Use
Lake Michigan Watershed

Type of User	Number of Users	Amount of Activity on Hand (Curies)	Isotopes Used
I State of Michigan			
Industry	2	1.8	Atomic No. 3-83*, H-3, C-14 Na-22, P-32, S-35, Cl-36
Medical	6	1.0	Ca-45, Cr-51, Co-58, Fe-59 I-131, Ir-192, Au-198
Educational	1	10.3	Po-210, V-233, Am-241
Total (Michigan)	9	13.1	84
II State of Wisconsin			
Industry	9	5.0	Atomic No. 3-83* H-3, C-14, Na-22, Na-24
Medical	39	12.9	P-32, S-35, Cl-36, K-42 Ca-45, Cr-51, Fe-55,
Educational	1	1.8	Fe-59, Co-58, Co-60 Ni-63, Br-82, Br-83 Rb-86, Sr-85, Sr-89, Sr-90, Y-90, Ag-110 Ag-111, I-125, I-131, Ba-135 Au-198, Hg-203, Tl-204
Total (Wisconsin)	49	19.7	81
Grand Total	58	32.8	84

*Broad licenses permitting use of any radioisotope between atomic numbers 3 and 83 within quantity limitations of the license (usually for research purposes).

TABLE 2
Lake Michigan Water Samples
April to October 1962

Samples which had Alpha Radioactivity
in Excess of 3 μμc/l

Cruise	Station Number		Depth (Meters)	α Activity of Total Solids (μμc/l)
	Latitude	Longitude		
1	422300	863300	50	3.2
	422300	872500	5	6.9
	422300	872500	75	3.3
	422300	870000	5	4.8
	422300	870000	125	3.2
	443900	861700	5	3.7
3	433600	864700	110	3.5

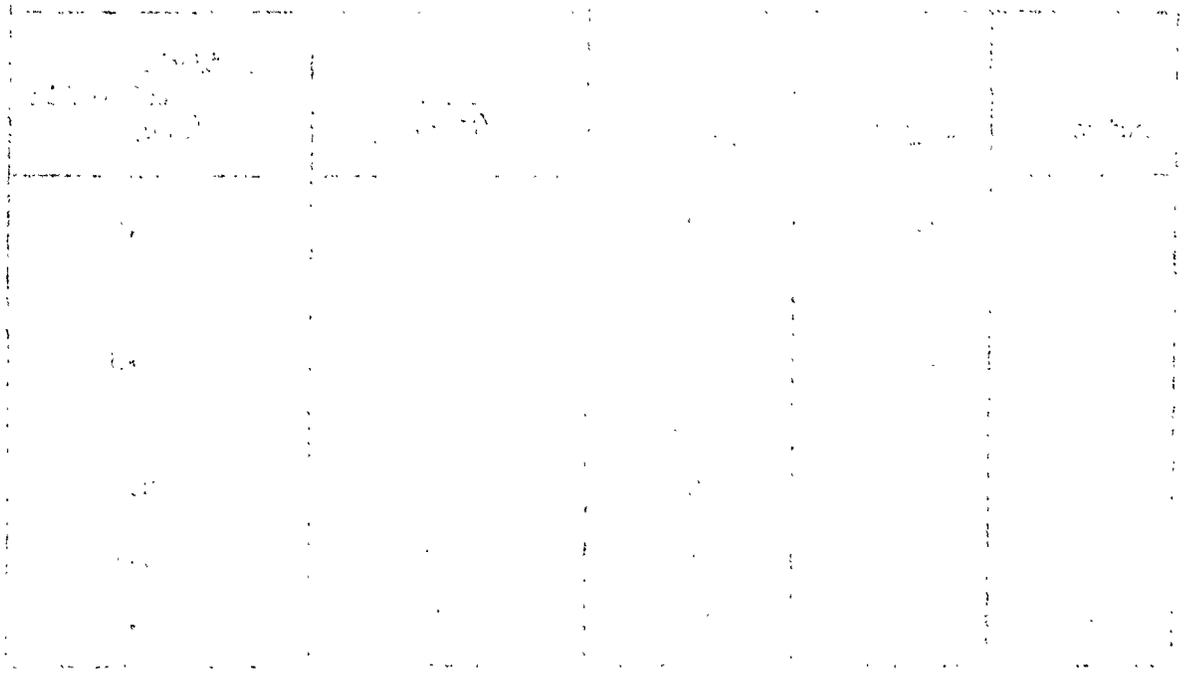


TABLE 3
Lake Michigan Radiological Studies
April-October 1962

Percentage Distribution of Beta Radioactivity
In Water and Plankton Samples

Cruise No.	Date of Cruise	Percentage of Water Samples Having Gross Beta Activity In Micromicrocuries/Liter		
		< 10	> 10 < 20	> 20
1	April-May	49	37	14
2	June	56	38	6
3	July	75	25	0
4	Aug.-Sept.	60	35	5
5	Oct.	86	11	3

Cruise No.	Date of Cruise	Percentage of Plankton Samples Having Gross Beta Activity In Micromicrocuries/Gram					
		< 10	> 10 < 25	> 25 < 50	> 50 < 75	> 75 < 100	> 100
1	April-May	4	0	48	30	13	4
2	June	0	0	4	26	15	55
3	July	0	0	0	3	3	93
4	Aug.-Sept.	11	13	42	18	5	11
5	Oct.	5	0	5	20	16	54