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HEALTH PHYSICS DIVISION

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DATE ISSUED

OCT - 4 1960

Oak Ridge National Laboratory
Operated by
Union Carbide Corporation
For
U. S. Atomic Energy Commission
Oak Ridge, Tennessee

ANALYSIS OF WASTE DISPOSAL PRACTICE AND CONTROL AT ORNL

By

Yehuda Feige*, F. L. Parker and E. G. Struxness

Abstract

Liquid radioactive wastes at Oak Ridge National Laboratory are divided into three classes, high-, intermediate-, and low-level wastes. High level wastes are stored in tanks and allowed to decay. Medium level wastes are pumped to seepage pits where most of the activity is fixed on the soil. Low level wastes are passed to White Oak Creek with or without treatment depending upon the level of activity. Wastes discharged to the environment are daily grab sampled and beta counted, and composite samples are radiochemically analyzed monthly, which is adequate for health physics control. During 1948-1957 there has been an increase of strontium in White Oak Lake bed but since 1954 there has been more strontium, ruthenium, zirconium-niobium, and trivalent rare earths leaving White Oak Lake than entering. More cesium, however, has entered White Oak Lake than has left. The precision of these figures could be improved by using gamma spectroscopy. Wastes leaving White Oak Lake are diluted 1000 times by Clinch River water. Clinch River water has a calculated MPC less than 10% of ICRP values. Bottom sediments in the lake and in the river contain mostly cesium.

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Summary

Oak Ridge National Laboratory has been producing radioactive waste ever since the beginning of its operation in 1943. The amounts and the composition of the radioactive waste has varied with the expansion of the Laboratory's facilities. The methods of waste collection, waste treatment, waste storage, and waste release to the environment have often been revised.

Waste disposal problems have not been a central consideration in the planning and growth of ORNL. Waste problem had to be solved as they arose to enable safe operation of the various facilities.

This report correlates available information on the liquid radioactive waste system at ORNL with regard to beta and gamma emitting isotopes. Alpha emitters could not be included, as much of the necessary information for evaluating their fate is still classified and, thus, inaccessible to the author.

The monitoring system proved adequate in maintaining fission product concentration levels in the Clinch River below the MPC_w values recommended by NCRP and ICRP.

No balanced budget of the activity discharged through White Oak Creek could be obtained. The attempts to derive quantitative information about the curies of various radionuclides discharged, from the available data, presented many inconsistencies. The disagreements stem, in part, from an accumulation of small experimental errors, which are multiplied by large

flow values. The present monitoring system does not cover all the liquid waste contributors. Moreover, no differentiation is made between short-lived and long-lived isotopes in the routine daily samples. The samples collected at White Oak Dam, and aliquoted for monthly chemical analysis, are not taken on a proportional basis.

It is common practice to attach a positive value to the concentration of each isotope of interest. If the actual counts obtained are indistinguishable from the instrumental background count, an upper limit is given. This procedure is safe for Health Physics considerations, but it is misleading to use these data to arrive at a budget of the liquid waste system. It is obvious that these small concentrations, of the order of 10^{-9} μc/cc, when multiplied by the respective flow values add up to amounts which seem considerable; yet no real evidence exists that the isotope in question was actually present.

The following recommendations concerning monitoring methods might improve the accuracy and value of monitoring data:

(1) Install continuous monitoring at White Oak Dam.

(2) Differentiate between the short-lived and long-lived activity in the daily samples.

(3) Compose the monthly sample out of volumes proportional to the daily flow.

(4) Compare the samples obtained after radiochemical separation with a known standard of the specific isotope suspected. More correct values for μc/ml are then possible.

(5) Change to gamma spectroscopy for quantitative analysis wherever practical.

The tables in this report are consistent with the figures given by F. N. Browder.[3] They agree, within a few per cent, with the other sources of information used. The basic monitoring data discussed here were collected and summarized by H. H. Abee.[20]

Browder's report on Radioactive Waste Management at ORNL, Abee's Liquid Waste Monitoring Summary Techniques and Data (unpublished), Cottrell's report[23] on Radioactivity in Silt of the Clinch and Tennessee Rivers, together with the present report complement each other and should together present a comprehensive picture of the present status of low-level liquid waste disposal to the Clinch River.

1.0 Liquid Waste Handling at ORNL

1.1 Historical Background

A brief chronological review of the development of the ORNL liquid wastes disposal system is warranted, because the hazard of radioisotopes discharged to the environment is cumulative. It is necessary, therefore, to know the fate of all radioactive material discharged from ORNL in evaluating the present situation.

In 1943 the X-10 Graphite Reactor went critical, and the chemical separations plant associated with it began operations. Underground concrete tanks were constructed to store all the radioactive liquid wastes which were expected to accumulate during one year's operation - the then anticipated life of the Laboratory. Uranium liquid wastes were stored in special tanks.

Before long it became obvious that the capacity of the storage tanks was insufficient to contain the volume of waste produced. It was decided, therefore, to precipitate as much as possible of the radiochemical waste in the storage tanks by making the solution highly basic. The radioisotopes remaining in solution were decanted and diluted with the Laboratory's large volume of (nonradioactive) process waste water and discharged through White Oak Creek to the Clinch River. A portion of the precipitated radioisotopes remained as sludge in the storage tanks and is still there (1959).

At the end of 1943 a dam was constructed across White Oak Creek on White Wing Road to control the discharge of isotopes into the Clinch River. The artificial lake behind the dam covered an area of about 44 acres; its average depth was about 6 ft. Its storage capacity was about 10^8 gal. The lake was drained in 1956, and the lake bed is now being used for ecological studies. It might be used for temporary retention of a large volume of highly active liquid waste in case of an accident.

In July 1944 a settling basin of 1.5×10^6 gal capacity was put into operation (see Fig. 1) to provide more uniform dilution of the decanted radioactive waste with the large volume of process water. Radioactive particulates in the dilution water had more time to settle out and, therefore, the amounts and maximum concentrations of radioactive wastes released to the Creek were appreciably reduced. Further reduction in the activity released was achieved by storing the supernatant waste solution as long as possible (about 1 month) in storage tanks before decanting it to the settling basin so that much of the short-lived radioactivity decayed before discharge of the waste to the Creek. This mixing of the decanted high-level waste with the Laboratory's waste waters was abandoned when the evaporation process was installed.

Starting in June 1949, the highly active chemical waste was concentrated by evaporation before storage in tanks. This reduced the volume of radioactive wastes by a factor of about 30 which facilitated continued storing. The water boiled off by evaporation proved to be decontaminated by a factor of 700, on the average, and could thus be satisfactorily released.

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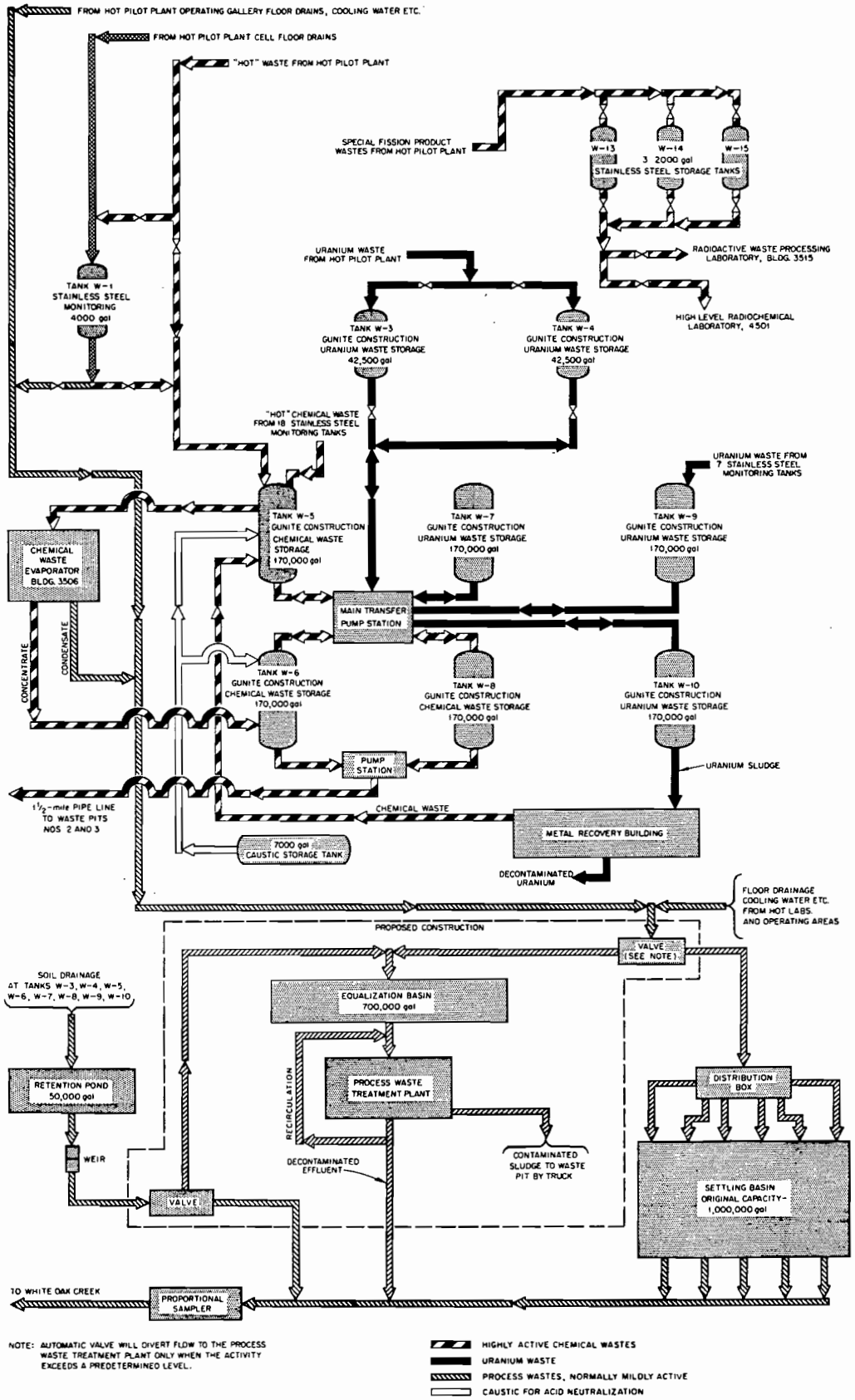

Fig. 1.

In 1952 a metal recovery plant began to process the separately kept liquid uranium waste. More than 130 tons of uranium have been reclaimed by the process. The waste from the recovery process has been added to the active chemical waste system (Fig. 1).

By 1954, following several years of experimental studies, the million gallon earthen seepage pits were used to receive the supernatant of the decanted chemical liquid waste. It was shown that cationic radioactive isotopes are adsorbed and retained by the clays in the Conasauga shale. The liquid was found to seep slowly through the shale towards White Oak Creek. The amphoteric radioactive ruthenium isotopes were not retained efficiently by the shale.[1,2]

1.2 Current Handling Methods

The present waste disposal system is shown schematically in Fig. 2. It is apparent that ORNL can dispose of its liquid wastes in three ways: (1) by storage and radioactive decay, (2) by fixation in the soil, and (3) by discharge through White Oak Creek to the Clinch River. ORNL's effort has always been directed towards retaining and containing as much as possible of the radioactive waste material by fixation in soil or by deposition as insoluble sludge in tanks and pits. Great care has been taken to maintain at low levels the amount of activity discharged to the Clinch River.[3,8]

Chemical Liquid Waste.- Most of the highly active chemical waste is produced in chemical processing buildings and chemical laboratories in an

average daily volume of about 7000 gal, containing between 3 to 50 mc/liter. This waste is neither chemically nor radiochemically analyzed. The waste is discharged first into 19 underground stainless steel monitoring tanks which are located near the waste producing buildings. The rate of waste accumulation in each of the monitoring tanks is continuously recorded. When a monitoring tank fills, the waste is pumped into three underground concrete central storage tanks. Most of the short-lived isotopes decay during the underground storage period. A major fraction of the activity is precipitated in the concrete tanks by making the solution highly basic. Similar tanks also receive the waste from the metal recovery building.

Two to three times each month about 80,000 gal of the supernatant are pumped out of the storage tanks into the seepage waste pits.[9] Representative samples of the solution pumped into the pits are routinely analyzed. They indicate that, in general, Cs^{137} contributes about two-thirds of the activity; Ru^{106} and the rare earths about 25%; about 5% of the activity is due to Sr^{90} - Y^{90}.

At present none of the highly active waste flow is <u>deliberately</u> dispersed into the uncontrolled environment. Prior to 1949, however, the supernatant from the storage tanks was mixed with the Laboratory's process waste waters and discharged through White Oak Creek. Until 1957 it was possible for leakage from the underground tanks to pass through a retention pond and into White Oak Creek without treatment. The volume of

chemical wastes discharged to the pits is shown in Table 1.

Table 1. Volume of Chemical Waste, 1950-1957.[9,10]

Year	Volume of Radioactive Waste 10^6 gal	Waste Discharge to Ground Pits: Volume 10^6 gal	Waste Discharge to Ground Pits: Beta Activity 10^3 curie
1950	2.2	--	--
1951	2.2	0.12	0.39
1952	2.1	0.04	0.95
1953	2.0	0.23	7.7
1954	0.7	1.0	7.2
1955	--	1.7	21.4
1956	--	2.7	35.0
1957	--	2.9	41.9

More than 100,000 curies have been discharged to the seepage pits. The external radiation level around the pits has reached levels of about 1000 mr/hr, which is not inconsistent with the gamma radiation to be expected from the Cs^{137}.

Low-Level Liquid Waste. - The mildly contaminated process waste water stream (700,000 gal/day) is water used mainly for cooling processes and is radiochemically clean. Most of the contamination in this water is the result of equipment failure, human error, or accidents. The average concentration of beta activity is about 0.5 μc/liter, with fluctuations up to a factor of 100.

This stream was discharged to White Oak Creek through the settling basin (Fig. 1). Beginning in August 1957 most of the contamination in this stream has been diverted to a waste water treatment plant,[14] before release into White Oak Creek. This treatment plant could not, of course, have had any influence on Clinch River contamination prior to 1957.

The drainage from monitoring pads underneath the storage tanks and some water seeping through the soil around the tank farm used to pass through the retention pond.[13] The pond served as a monitoring point for the detection of leakage in the active chemical waste piping and storage system. It has a capacity of about 50,000 gal; the average flow of water through the pond was about 26,000 gal/day. It was directly connected to White Oak Creek, bypassing the settling basin-monitoring station until 1957. Now the effluents of this drainage system is pumped into the equalization basin feeding the process waste water treatment plant.[3,12,13] This line provided a possible outlet for the high-level waste system into the river, in case of equipment failure. This actually happened several times, notably in 1953, when about 140 beta curies were discharged to White Oak Creek through the retention pond.[9]

The location of ORNL and the hydrology of the area[2,3] make it clear that practically all the activity reaching the Clinch River has to come through White Oak Creek. The creek constitutes the direct continuation of the process waste water flow (No. 3, Fig. 2). It also receives contamination released through the retention pond and seepage from the liquid waste pits.

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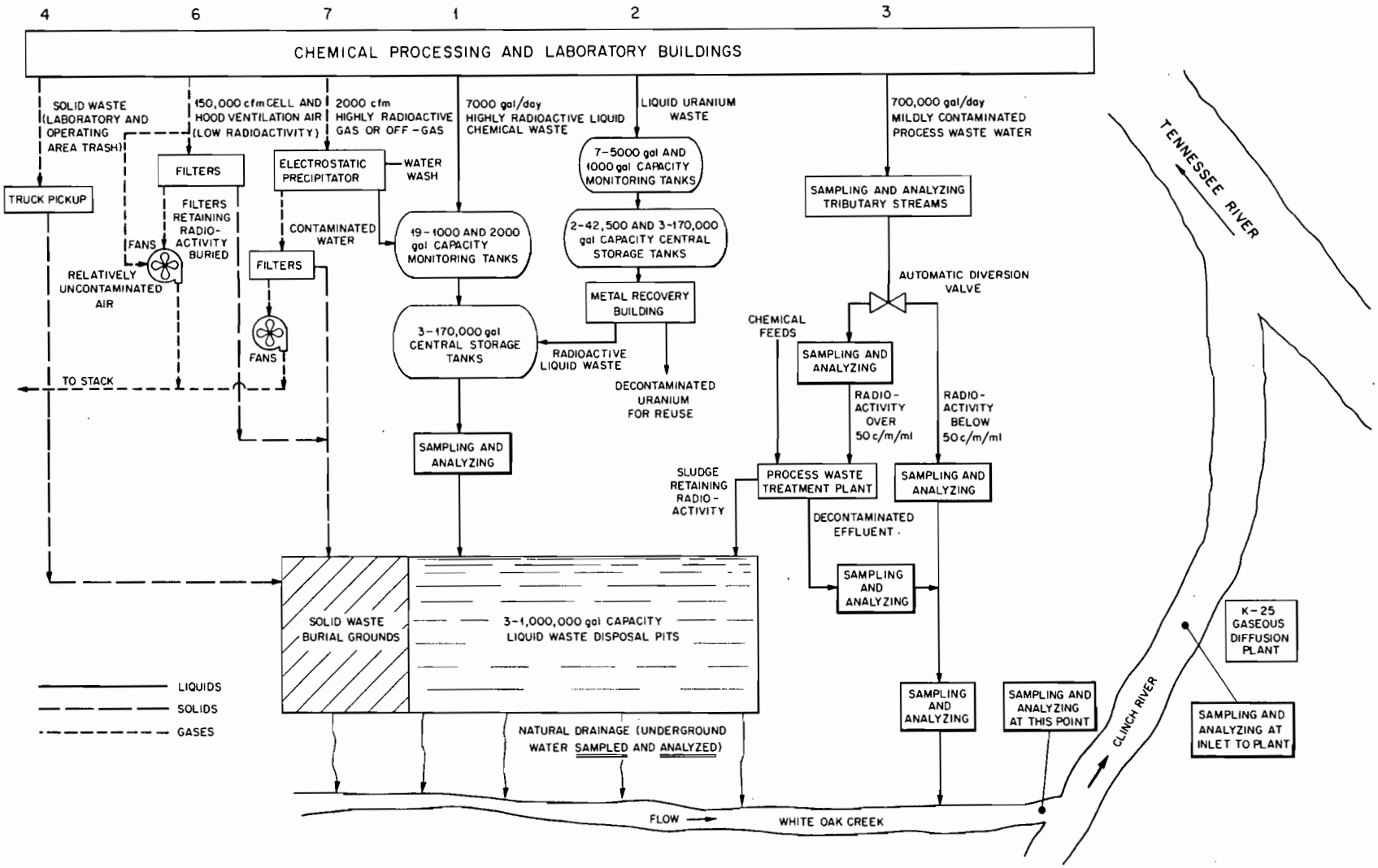

Fig. 2. Simplified ORNL Radioactive Waste Disposal Flowsheet.

In addition, the drainage of various reactor facilities located in Melton Valley (HRT, ART, etc.,) as well as cooling waters from the LITR and ORR are directly discharged to small tributaries of White Oak Creek. The gross beta and gamma count in these streams is usually below 100 c/m/cc.[3] Though much of this activity is due to short-lived neutron activation products of water impurities, such as Na^{24}, fission products also are often discharged.

More detailed information concerning the waste system at ORNL is presented in Browder's report.[3]

1.3 Discussion

One should always keep in mind that wastes discharged in the past may still be contributing significantly to the radiation level of the environment. In reviewing the history of this system, it is obvious that the main trend has been to alter the then existing waste system whenever it seemed inadequate. Minimum cost has been an important factor, so that new problems have usually been approached with a marked preference for existing methods and equipment. In retrospect, however, other approaches might have been more advantageous in the long run.

An illuminating example is the practice of diluting the decanted highly active liquid waste with mildly contaminated process waste water, practiced for several years prior to 1949. This mixing seemed logical as long as the dilution factor was sufficient and no additional treatment of the diluted waste was required. By 1949, however, even the mildly

contaminated process waste water was often difficult to handle. This diluted waste stream had to be retained to allow for settling of suspended solids and further treatment before release to White Oak Creek. Much effort (and money) could probably have been saved if the waste to be treated had not been diluted by the large volume of water from uncontaminated cooling systems. Such waste would have been more concentrated, significantly reduced in volume, and consequently easier to handle. Dilution as a last stage of treatment, instead of being applied at an earlier stage, would have offered many advantages.

The introduction of seepage pits might be cited to prove the broad usefulness of a new (unconventional) approach. The major portion of the radioactive material in the chemical liquid waste must be withheld from the environment. Permanent underground storage tanks have long been used for this purpose. As the amount and volume of the waste increased, precipitation and concentration by evaporation were tried. These refinements did not solve the problem of long-term storage of continuously accumulating waste.

The excavation of large shallow earthen pits, in areas where a fair knowledge of the geological structure and hydrology exists, led to almost inexhaustible possibilities. The evaporation process could be abandoned; a major reduction in volume of the waste could be achieved through seepage of the liquid into the ground.

2.0 Methods of Sampling and Analysis of Radioactive Waste Streams

To fully comprehend the validity of the figures quoted for this study of ORNL's waste system, it is necessary to understand the methods used to

monitor and analyze the liquid waste streams. First, the total volume discharged from the settling basin into White Oak Creek must be measured and, secondly, a representative sample must be collected and analyzed.

The settling basin was sampled by the "grab" sample method every 4 hrs during 1948-1950.[20] Flow measurements were made at the inflow to the settling basin by means of a box containing five V-notch weirs and a head measuring device. Beginning in November 1950, flow measurements were made by a single large V-notch weir at the outlet and a water level recorder in a stilling well. A trebler proportional sampler was installed in the weir box, and continuous proportional samples were collected.

White Oak Creek was sampled by taking an 8-gal grab sample at White Oak Dam once a day. More frequent samples were taken when significant changes in activity were anticipated.[20]

The flow at White Oak Dam was and is obtained by means of a water level recorder in a stilling well. Since the drainage of the lake in October 1955, the flows of White Oak Creek and that of the Melton Branch are measured separately, above the White Oak Lake basin, and summed. A correction factor of 1.16 is applied during precipitation periods to compensate for the runoff associated with the area between the gaging points and White Oak Dam.

The flow measurements, which constitute one factor in determining the daily release of activity, are conducted with standard approved methods, and should introduce but insignificant errors.

2.1 Gross Beta Determinations

Gross beta activity of the daily samples is determined by the Area Monitoring Section of the Health Physics Division in the following manner.[20]

(1) Measured volumes (5 - 10 ml) of the daily samples collected from the settling basin and at the Dam are evaporated to dryness on an aluminum dish.

(2) Each sample is counted with a shielded end-window type GM tube having a 3 mg/cm^2 mica window. At least 10^3 counts are obtained for each sample to attain reasonable counting statistics. The daily results obtained for the settling basin effluent fluctuate between 30 - 3000 c/m/ml; the corresponding values for White Oak Dam lie between 2 - 200 c/m/ml. These fluctuations insert a high degree of uncertainty to the White Oak Dam data, obtained by the grab sampling method.

(3) The gross beta activity in μc/cc is calculated from the net counts per minute, corrected to 10% geometry. The radiation from a known uranium disc, contributed essentially by the $Pa^{234}(UX_2)$(2.3 Mev) and Pb^{214} (RaB)(0.6 Mev) beta disintegrations is used for calibration. No corrections for self-absorption or back-scatter are made.

The daily gross beta curies discharged are calculated by multiplying the μc/cc values (derived in the above-mentioned way) by the total flow for the respective day. From these data the weekly (Table 1, ref. 20), monthly, quarterly, and yearly discharged beta activities are derived by simple summation.

2.2 The Monthly Chemical Analysis

About 140 ml of each daily sample, collected from the Settling Basin and White Oak Dam, are composited into monthly samples. Four liters of the composite sample are concentrated to approximately 100 ml by evaporation. The solution is centrifuged to remove the suspended solids. Radioactive isotopes from the solid are extracted with perchloric and hydrofluoric acids. This leach solution is added to the supernatant, giving a total volume of about 150 ml. This method assures a high extraction yield of the radioactivity from the monthly composite sample with relatively little inert solids, which helps in reducing self-absorption in the succeeding stages of the analysis.

The monthly samples are analyzed by Analytical Chemistry Division as follows:[18]

(1) About 2 ml of the concentrated solution are evaporated to dryness in an aluminum dish. The beta activity is counted by a lead shielded end-window GM tube at approximately 10% geometry. The results represent the total gross beta counts, expressed in net counts per milliliter. The average for the composite concentrated samples leaving the settling basin is about 4000 c/m/ml. The average for White Oak Dam effluent is about 600 c/m/ml. The background count of the shielded detection unit is about 25 c/m.

(2) Two milliliter samples are taken for radiochemical separation, using appropriate carriers.[11] Separations are made for the trivalent rare earth group, ruthenium, zirconium, niobium, cesium, strontium, iodine, barium, and cobalt.

After slow evaporation, the samples are counted for beta activity, as mentioned above. The results are corrected for recovery of carrier material and expressed in c/m/ml.

Adding up all the results of the various chemical separations gives the "Total Beta Activity Identified" in c/m/ml, which usually agrees within 15% with the value obtained for the total beta activity determined before (Tables 5-13, ref. 20).

(3) Cerium is extracted from a trivalent rare earth sample and counted separately for cerium isotopes (including Pr^{144}).

(4) Recently, the remainder of the composite solution has been used to extract strontium. Beta absorption measurements are made with this sample to evaluate the approximate Sr^{89} to Sr^{90} ratio.

3.0 Radioactive Material Discharged to White Oak Creek and the Clinch River

3.1 Results

Using the methods described in the last chapter for the determinations of the daily gross beta count and the flow of the waste streams into White Oak Creek and the Clinch River one obtains the gross beta flow shown in Table 2. Surprisingly, the inflow does not match the outflow, and since 1954 more curies have left White Oak Creek than can be accounted for by the inflow from the settling basin and the retention pond.

Because of the various contributors to White Oak Creek, which are not individually monitored, it is impossible to obtain a balance budget. But these contributors are usually quite small and so a reasonable balance should be possible.

Table 2. Gross Beta Curie Balance in White Oak Creek

Year	Curies Discharged into White Oak Creek: From Settling Basin	Curies Discharged into White Oak Creek: From Retention Pond	Curies Discharged into the Clinch River from White Oak Creek	Differences in Curies
1948	899		494	- 405
1949	1026		718	- 308
1950	172	15	191	+ 4
1951	169	3	101	- 71
1952	411	87	214	- 284
1953	289	140	304	- 125
1954	237	17	384	+ 130
1955	213	54	437	+ 170
1956	253	20	382	+ 109
1957	189		397	+ 208
1948-1957	3858	336	3622	- 572

The background count in the Analytical Chemistry Division laboratories, about 25 counts/min, is equivalent to roughly 2% of the total beta count of a 2-ml concentrated outflow sample $\left(\frac{25 \times 100}{2 \times 615}\right)$.

The background is about 0.3% $\left(\frac{25 \times 100}{2 \times 3880}\right)$ of the concentrated inflow sample counted. It follows that samples, separated for radiochemical analysis, which show activity of about 2% of the total gross beta count or less, have quite high statistical errors attached to them, and should be judged accordingly. Furthermore, no sample analyzed by Analytical Chemistry Division is counted for longer than a 10-minute period.

Iodine, cobalt and barium, which are also separated, were left out of Table 3 because of the low quantities detected, and thus, the total does not add up to 100 per cent. The values of zirconium and niobium, which have been presented, include probable errors exceeding 100%.

Comparisons of the inflow and outflow compositions of the monthly composite samples, as presented in Table 3, reveal that, on the whole, no large differences are found between the two sets of values. Significant differences appear only with ruthenium-rhodium and cesium-barium samples. About 1.5% of the average gross beta activity from the composite sample leaving the settling basin was found to be due to ruthenium-rhodium activity, whereas at the outflow samples of White Oak Dam 7.7% of the activity is due to ruthenium-rhodium, no doubt due to the ruthenium released from waste pits. Cesium 137 shows the reverse trend, 26.6% from settling basin effluents versus only 14.8% in White Oak Dam samples which follows from the known specificity of clays for cesium.

In trying to estimate the quantities of the various isotopes released to the environment one is easily led to multiply the values obtained by chemical analysis of the monthly composite sample by the values for the total curies discharged obtained by the daily gross count measurements. This method of directly combining the two sets of results has often been used.[3,8,12,13,16] The results reproduced in Table 4 call for many explanations.

Inspection of Table 4 shows that, with the possible exception of Cs^{137}, the contribution of the settling basin to the effluents of White Oak Creek is not the only contributing factor. Nine times more ruthenium leaves the Creek than is contributed by the measured ruthenium activity leaving the settling basin. It has been shown, by studying the wells around the waste pits, that ruthenium seeps out of the pits. This might account for the larger quantities at White Oak Dam. These same monitoring wells, however, have shown that no strontium has moved out of the pits thus far.[1] Thus,

Table 3. Percentages of Isotopes Entering and Leaving White Oak Creek

Isotopes	1953		1954		1955		1956		1957		1953-1957***	
	In*	Out**	In	Out	In	Out	In	Out	In	Out	In	Out
Sr^{89}, Sr^{90}	27.6	44.3	23.3	35.2	18.7	21.1	15.1	17.9	18.0	20.9	21.4	26.5
Zr^{95}	2.6	2.5	1.0	3.6	0.6	1.2	0.5	2.0	10.2	5.7	2.6	2.9
Nb^{95}	1.1	1.2	0.4	2.4	0.6	1.3	1.1	2.6	0.7	1.8	0.7	2.0
Ru^{106} + Rh^{106}	0.8	8.7	0.5	2.9	3.1	7.0	2.0	5.0	1.0	15.0	1.4	7.6
Cs^{137}	8.5	2.1	20.3	5.7	31.6	14.4	42.1	29.6	36.5	22.4	26.3	17.0
Ce^{141}, Ce^{144} + Pr^{144}	2.6	2.2	19.1	6.2	14.7	19.4	12.3	10.1	4.4	3.2	10.3	9.0
Y^{90}, Y^{91}, Nb^{144}, Pm^{147}, Pm^{149}	53.1	37.4	34.6	42.6	30.3	33.7	24.4	23.8	25.8	28.8	34.2	32.7
Total Gross Beta	96.3	98.4	99.2	98.6	99.6	98.1	97.5	91.0	96.6	97.8	96.9	97.7
Activity c/m/ml	1640	245	6900	940	4430	835	4260	710	2160	340	3880	615

In* - From Settling Basin

Out** - From White Oak Dam

*** - Weighted for total gross beta.

Table 4. Curies of Isotopes Entering and Leaving White Oak Creek

Isotopes	1953		1954		1955		1956		1957		1953-1957		
	In**	Out**	In	Out	In	Out	In	Out	In	Out	In	Out	Excess(Out-In)
$Sr^{89} + Sr^{90}$	80.0	134.7	55.2	135.2	39.8	92.2	38.2	68.4	34.0	83.0	247.2	513.5	266.3
Zr^{95}	7.5	7.6	2.4	13.8	1.3	5.2	1.3	7.6	10.3	22.6	31.8	56.8	25.0
Nb^{95}	3.2	3.6	0.9	9.2	1.3	5.7	2.8	9.3	1.3	7.1	9.5	34.9	25.4
$Ru^{106} + Rh^{106}$	2.3	26.4	1.2	11.1	6.6	30.6	4.1	19.1	1.9	59.6	17.1	146.8	129.7
Cs^{137}	24.5	6.4	48.1	21.9	67.3	62.9	106.5	113.1	69.0	88.9	315.4	293.2	-22.2
$Ce^{141}, Ce^{144} + Pr^{144}$	7.6	6.7	45.3	23.8	31.3	84.8	31.1	38.6	8.3	12.7	123.6	166.6	43.0
TRE*	154	113.7	82.0	163.6	64.6	147.3	61.7	90.9	48.7	114.3	411.0	629.8	218.8

*Trivalent rare earths excluding the cerium isotopes, including Y^{90} and Y^{91}.

**In - From Settling Basin
Out- From White Oak Dam.

another explanation is required to account for the apparent excess of strontium in the effluent of White Oak Creek. It has been suggested that strontium, which had settled down in White Oak Lake several years ago (when the inflow did exceed the outflow - Table 2), is partially being carried along with the silt washed out during periods of heavy rains and floods. There is little doubt that this could account for at least a part of the excess strontium detected. This explanation becomes improbable, however, in view of the findings discussed in the last chapter. Only very small amounts of Sr^{90} are actually detected in sediment samples of the Clinch River bottom and in the soil samples of White Oak Lake bed. There seems to be no significant settling down of Sr^{90} out of the stream. Moreover, it seems difficult to believe that the difference found with the shorter-lived isotopes like Zr^{95} (half life of about 2 months), and perhaps the rare earths too, can be due to a gradually washing out of previously settled activity.

3.2 Discussion

The estimated curies discharged yearly (Table 4) depend strongly on the data summarized in Tables 2 and 3. A discussion of the methods used, their scope and limitations, might resolve some of the apparent inconsistencies.

The large discrepancies between the recorded inflow and outflow activities of White Oak Creek are seen in Table 2. The grab sampling technique applied at White Oak Dam might not represent the average outflow concentration for the respective period. Another possible source of error is that no discrimination is made between long- and short-lived activities in the daily samples. As mentioned before, short-lived isotopes

are often discharged directly to the creek, bypassing the settling basin; they might well cause some of the activity detected at White Oak Dam. The monthly radiochemical data do not in themselves lead to inconsistent results. Still much improvement might be possible.

(1) Errors are possibly introduced by the counting statistics themselves. The Analytical Chemistry Division laboratories have a relatively high background for such low-level samples, and hence these minimum results are reported as less than a certain amount. These upper limit figures are the values quoted thenceforth, and, when multiplied by the flow, can be an appreciable quantity over the course of a year. Therefore, if one compares the minimum figures at the settling basin and the dam, a difference by a factor of 15 is introduced by their differences in flow alone. Other statistical errors inherent in the Analytical Chemistry Division laboratory are due to the relatively high background.

(2) It is apparent that there can be no simple way to obtain estimates of the μc/ml of various isotopes present from the counts per minute per milliliter values determined from the chemically separated samples. The half thickness layer for the absorption of the beta radiation from Nb^{95}, for instance, is about 3 mg/cm^2, which is about the same as the end-window thickness of the detecting GM Tube. Add to this the unknown self-absorption factor in the sample tested, and it becomes impossible to arrive at any correlation between the activity (in μc/ml) of a Nb^{95} sample and the net counts/minute detected by the above described method. Even relatively high activities will not help, as niobium is actually detected mainly through its Compton - scattered gamma ray from the lead shield into the GM counter and not by its weak beta radiation.

Because of the various beta energies involved in the composite sample, each isotope present has a different conversion factor to μc/ml from the detected counts/min/ml. Yttrium 90 and Sr^{89} are detected much more effectively by beta counting than is pure Sr^{90}. The detection efficiency, for beta radiations, of a GM tube increases quite markedly with the energy in the lower Mev energy region. The counter window thickness, the self-absorption in the dried sample, and the natural background counts set a practical threshold to detectability. Therefore, to obtain a good conversion factor one should compare the samples obtained after radiochemical separation with a standard amount of the specific isotope. By having the same mg/cm^2 on the aluminum plate, μc/ml might be obtained directly.

(3) A constant amount (about 140 ml) of each daily sample is included in the monthly composite sample. This monthly sample, thus prepared, represents, with all the above mentioned qualifications, the average monthly concentration of activity. It does not represent the average monthly activity itself. As this sampling does not take into account the daily flows, the method discriminates against days of higher flow. Relatively large amounts of activity are discharged through White Oak Dam to the Clinch River during floods, though the average concentration for these particular days might well be much below the average concentration at "normal flow" periods.

The curies released per year are derived by multiplying the yearly amounts of beta activity discharged by the per cent activity of the isotope found in the composite sample. It is difficult to determine whether these figures are correct. To make the analysis more exact the monthly

samples should be proportional to the volume. Daily representative samples should be added in volumes proportional to the daily flow.

(4) For more exact methods of analysis and to avoid the problems of backscatter, self-absorption, etc., gamma spectroscopy would be invaluable.

Most of these improved methods are already applied in many of the studies conducted within the Health Physics Division. So that this may not be construed as criticism of the Applied Health Physics Section, it should be stated that the purposes for which these analyses are made (i.e., in order to meet the NCRP criteria for discharge) the above procedures are adequate. For a more sophisticated study of the fate of the isotopes released, the above recommendations might be useful. These recommendations have been obvious to the Applied Health Physics and Waste Disposal Research and Engineering Sections for a long time and have been instituted wherever practical. Now that our interest of the fate of isotopes released is increasing, a more sophisticated approach throughout the analysis would be profitable.

4.0 Radioactivity in the Clinch River Water

White Oak Creek discharges into the Clinch River at C.R.Mile 20.8 (20.8 miles upstream from Clinch River's entry into the Tennessee River). The flow of the Clinch, which varies greatly, is regulated by the flood control and power dams at Norris and at Watts Bar. The mean annual flow is about 3×10^9 gal per day, which provides a dilution factor of about 10^3 for the contaminated water discharged from White Oak Creek.

The first user of the Clinch water for drinking purposes is the Oak Ridge Gaseous Diffusion Plant, located 7.6 miles downstream from White Oak Creek.

An additional average dilution factor of about seven is provided beyond the confluence of the Clinch with the Tennessee River. The first large city downstream which uses the Tennessee water extensively is Chattanooga. The estimated minimum time of flow for water leaving the mouth of White Oak Creek to Chattanooga is about twenty days.

4.1 Calculated Activity Concentration in the Clinch

A weekly calculation is made of the probable average concentration of gross beta curies in the Clinch River. The concentration is obtained by multiplying the average gross beta concentration at White Oak Dam by a dilution factor obtained from the ratio of the White Oak Creek flow to the Clinch River flows (about 10^{-3}) as derived from USGS flow measurements.[20] Immediate total mixing is assumed, which is known to be false at the mouth of the White Oak Creek, but is justified for all practical purposes when dealing with cross sections a few miles downstream.[29]

The calculated average concentration of the gross beta activity in the Clinch during 1948-1957 was 1.1×10^{-7} μc/ml. The weekly average concentration within a year ranged from 0.3 to 2.2×10^{-7} μc/ml. Calculated values for weekly concentrations above 10×10^{-7} μc/ml have occasionally been obtained.[20]

The average probable concentration values lie below the MPC value recommended for unidentified isotopes in the neighborhood of atomic plants, 10^{-6} μc/ml (see Appendix A). Quite frequently, however, they do exceed 1.3×10^{-7} μc/ml, the MPC value for Sr^{90}.

When weighted MPC values are computed, based on the percentage of specific nuclides identified radiochemically in the monthly composite samples from White Oak Dam, each yearly average concentration is below the weighted MPC values. As Sr^{90} - Y^{90} comprise about 18% of the estimated beta activity released to the Clinch, these MPC limits have often been closely approached. The absence of a reasonable safety factor between the computed concentration and the MPC limits induced the Laboratory to improve its liquid waste disposal procedures, i.e., draining White Oak Lake[3,14] and installing a waste water treatment plant.[8,12,13]

4.2 Activity in Clinch Water Samples

Water samples of the Clinch are routinely analyzed for beta activity at two locations downstream.

(1) The water is continuously samples at the Oak Ridge Gaseous Diffusion Plant water intake. The results of the radioanalysis are included in classified reports, which are inaccessible to the author. The concentrations of gross beta activity found are reported to be well below 10^{-7} μc/ml.

(2) A daily grab sample is taken from the Clinch at Center's Ferry near Kingston (C.R.M. 4) and composited for a quarterly sample. The composite sample is filtered. Radiochemical analysis of the concentrated filtrate, and separately of the suspended solids, is made.

It should be recognized that 10^{-7} μc/ml is 0.22 d/m/ml. For 10% geometry, and penetrating beta radiation, one still needs to evaporate quite large volumes before significant counts above background (25 c/m) are obtained. The comments made on the validity of analyzing composite

grab samples, discussed in the preceding chapter, will apply here as well.

4.3 Discussion

To comprehend how extremely low concentrations of the order of 10^{-8} μc/ml are, comparison with the gross beta activity values detected in rain water might prove useful. Activity concentrations in rain water were of the order of 10^{-6} μc/ml during 1957.[15] Rain water is not used directly for drinking; the water loses most of its activity by ion exchange when seeping through the soil towards developed ground water resources. Comparison with rain-out data in other locations of the USA show that average concentrations found in rain water in Oak Ridge are not exceptionally high. The rain-out originated probably from weapon tests rather than air-borne activity released by ORNL.

Though some fall-out may have contributed to the Clinch contamination, the activity actually detected in the water some 10 miles downstream is lower than the probable concentration calculated, assuming perfect dilution. The quarterly data indicate that, even if due allowance is made for the dilution of the additional inflow into the Clinch from the downstream drainage area, the actual concentration of activity in the water is lower, by at least a factor of 2, than the predicted concentrations.

5.0 Fate of Fission Products Discharged to White Oak Creek

5.1 Introduction

When liquid radioactive wastes are released to surface streams, other processes, besides dilution and physical dispersion, are known to occur which will influence the concentrations downstream. Among the various

mechanisms that tend to purify the water are: chemical precipitation, deposition by gravity of radioactive materials attached to silt particles, and uptake by aquatic organisms. These "self-purification" mechanisms of streams act differently for different elements and depend on the physical and chemical form of the isotopes. Stream conditions, such as depth, flow velocity, temperature, density of the biota, turbidity, etc., are important factors determining the extent of the water purification.

Although these self-purification processes generally lead to a significant reduction of activity in waters downstream, as compared to predicted values taking only dilution into account, the radioactive material is not destroyed, and occasionally might show up unexpectedly. Long-lived radioisotopes settled on the river bed might be suddenly released during unusual stream conditions, like floods and droughts. As mentioned before, activity washed out of White Oak Lake bed might have contributed as much activity to the Clinch as the settling basin.

Significant activity might be found in fish, which feed on algae, plankton, and other biota capable of reconcentrating specific isotopes to a high degree. Several survey programs have been initiated by the Health Physics Division to evaluate the relative importance of these factors. A brief review of reported results from the Clinch River and White Oak Lake studies is in order.

5.2 White Oak Lake

During the first year of White Oak Lake's existence (1943-47) most of the contamination discharged to the creek probably settled in the lake's mud. Until 1953 appreciable amounts continued to settle in the mud. By then

some sort of dynamic equilibrium seems to have been established between the settling of the inflow contaminants and the average radioactive decay and the carrying away of radioactive material by silt washed out through White Oak Dam.

No exact picture can be drawn, as very little data is available for the years prior to 1948. The recorded data after 1948 is incomplete in many respects: the effective half life of the isotopes was not determined; the actual amounts released are not certain, etc. About 70% of the radioactive isotopes discharged to the creek during the first ten years is believed to have settled there and partly decayed.[25] This contaminant was effectively withheld from the Clinch River.

Annual studies of the uppermost mud layers from the lake bed yield estimated values from 300 to 400 curies on White Oak Lake bed during 1948-1952).[26]

Recent studies of the soil 3 years after drainage of White Oak Lake, during which period considerable washout of activity may have occurred, in addition to some physical decay, indicate that the radioactive material still present in the lake bed (1959) might well exceed 400 curies.[17,24] The lake bed contamination was probably much higher before draining White Oak Lake. The radioactive material appears to be quite unevenly distributed across the lake bed. Mapping the gamma activity, 3 ft above ground by means of a GM detector, yielded different radiation zones, ranging from background reading up to about 50 mr/hr. The concentrations of activities might be related to differences in the soil properties of the various zones.[28] Most of the soil activity is due to Cs^{137}, which

constitutes about 55% of the activity identified radiochemically in upper soil samples. Cerium, rare earths, and cobalt follow in abundance. Only 1 to 7% of Sr^{90} was found in these soils.[17]

The discrepancy between the amounts of radioactive material stored in White Oak Lake bed based on the recent soil studies and the amounts estimated from the previous mud studies might be due to several factors:

(1) The differences in sampling methods applied in the two surveys and the different counting techniques used will undoubtedly influence the results.[16,26]

(2) The sample-preparing method in the mud surveys excluded most of the organic matter. The organic fraction, however, is capable of adsorbing considerably greater amounts of radioactive isotopes than the mineral fraction in the water and at the bottom of the lake.[27] The later soil samples do include the radioactivity contributed to the soil during the past few years from the decomposition of dead organic matter.

(3) One should also keep in mind the difficulties inherent in any method used to determine the activity in mud or soils. The high self-absorption forces us to leach out at least the pure beta emitters, if any accuracy is desired. Gross beta counts on dry soil samples are of little significance. The extent of leaching, however, depends on the chemical form of the isotope in the sample, which is seldom completely known. A compromise must be made between the attempt to leach out all the active material, on the one hand, and the need to exclude excessive inactive ingredients in the extracted sample. No generally accepted method has yet

been devised for a complete removal of the isotopes that will work with any kind of soil and for all organic matter too.

5.3 Clinch River Bottom Surveys

Annual surveys of the radioactivity on the bottom sediments of the Clinch (and the Tennessee) Rivers have been made each summer since 1951. About 10 gamma radiation measurements of bottom activity are taken across the Clinch every two miles. Sediment samples are collected from these locations, and the amount of cesium, strontium, cerium, trivalent rare earths, ruthenium, and cobalt is determined radiochemically for the composited sample of dry mud from each cross section.

The river study reports[21,22,23] contain a detailed description of the methods used, and the attempts made to overcome various possible sources of error; they reveal the limitations of the methods as well, and contain a detailed tabulation of the experimental findings. The results will not be reproduced here, and the following discussion is intended merely to serve as a link between the present report and the extensive river study data.[23,24]

A gradual increase in the bottom activity was found when moving downstream from the point of entry of the waste into the Clinch River (C.R.M. 20.8). Highest activity values, about twenty times the background count, have been encountered 10 to 12 miles below White Oak Creek's entry (0.1 mr/hr). The initial gradual increase in activity downstream might be explained by the flow conditions along the river. The higher velocity at the beginning slows the settling of silt particles, which adsorb much of the Cs^{137} activity. It might even wash out

occasionally some of the bottom sediments previously settled. The contaminated silt settles to the bottom gradually as the flow becomes slower and less turbulent.

Concentration of radioactive material in the sediments drops off markedly upon entering the Tennessee River, and continues to decrease thereafter. The distribution of activity through each cross section was found to be roughly proportional to the depth of water there (the deeper water usually has the lower velocity); many exceptions to this have been encountered, however.

5.4 Discussion

Table 5 contains the average values computed from the Clinch sediment analysis data.[23] Other concentration values, mentioned before, are included for comparison.

During the years 1953-1957, the gross beta activity leaving the settling basin was 245 μμc/ml. Cesium 137 and strontium 90 were present in almost equal amounts, over 20% each. The trivalent rare earths group was the most abundant, about 34%.

The average calculated Clinch River activity concentration was about 0.13 μμc/ml; the chemical composition, as determined from White Oak Dam data, revealed a higher percentage of Sr^{90} as compared to Cs^{137}. Ruthenium 106 concentrations were also relatively high. About 50% of the radionuclides detected in the water taken at Centers Ferry was due to Sr^{90}. The concentration in the water was probably less than half the calculated values.

Table 5. Fate of Radionuclides Leaving Oak Ridge National Laboratory

	Yearly Average	Gross Beta Activity 10^{-6} μc/g	Per Cent of Identified Activity							
			Cs^{137}	Sr^{90}	$Cs\text{-}Pr^{144}$	TRE	$Ru\text{-}Rh^{106}$	Co^{60}	$Zr\text{-}Nb^{95}$	Ref.
Leaving Settling Basin	53-57	245	26.3	21.4	10.3	34.2	1.4	--	3.3	(20)
Calculated Clinch River	53-57	0.13	17	26.5	9.0	32.7	7.6	--	4.9	(20)
Kingston Water Samples	57	< 0.07		~ 50						(19)
Clinch Bed Silt	56-58	265	72.9	2.1	9.2	4.6	2.9	8.3	0	(23)
White Oak Lake Soil	58	27,600	56.5	1.7	16.6	11.8	0.8	12.6	0	(17)

The Clinch River bed silt showed an average concentration of 265 μμc/g, which is 2000 times higher than the predicted probable water activity in the Clinch River. About 73% of the identified bottom sediment activity is due to Cs^{137}, 8% is contributed by Co^{60}, and only 2.1%, on the average, was identified as Sr^{90}.

White Oak Lake soil studies, conducted during 1958, yield an average concentration as high as 27,600 μμc/g, which is 100 times higher than the average concentration found on the Clinch River bed. For many years the major part of the radioactive material released to White Oak Creek settled on the lake bed. Here, too, 56% of the activity is due to Cs^{137}, while only 1.7% is due to Sr^{90}.

Although Sr^{90} and Cs^{137} are present in comparable amounts in the liquid low-level waste stream, most of the activity which settled is Cs^{137}. Therefore, one assumes, till additional data concerning Sr^{90} is received, that strontium remains in the water stream and is not removed significantly by adsorption on muds in the Clinch.

To estimate how much activity is actually stored by now on the river's bottom is difficult, before adequate information about the depth of the active silt layers is obtained. Neither do we know the total amount of long-lived activity actually released into the Clinch, which could serve as an upper limit value. Due to the routine monitoring used in the early days of the Laboratory, this lack of precise information will complicate the interpretations of any planned river experiment conducted in the Clinch River.

Attempts are being made to approach this problem by trying to correlate the gamma radiation measurements to the specific activity found in the corresponding mud samples. The basic formulae tried were derived by simplified assumptions and are given in Appendix C. In applying these formulae to soil data from White Oak Lake, one should remember that no backscatter is included, so one would expect higher gamma readings than predicted. The actual distribution of the radioisotopes in the sediment is unknown too.

As regards gamma measurements performed with the flounder in the river cross section studies, the build-up factors in the water (including backscatter from the bottom) is surely not negligible, and should be corrected for. As a first approximation, one might expect better agreement between the formulae and the monitoring data by comparing the ratio of the flounder gamma readings to the activity concentration in the mud of Cs^{137} alone (and not including the total beta activity). Accurate calibration is easy for this case. So the formulae could be tested and the build-up factors determined experimentally.

Though many interesting questions have yet to be answered, it should be emphasized that from a health physics point of view, the liquid waste disposal system is satisfactory. It has proven possible to keep concentration in the Clinch water below the MPC values recommended by the NCRP and the ICRP, for routine, normally supervised operation.

APPENDIX A

The Maximum Permissible Concentration of Radioisotopes in Water

The values of maximum permissible concentrations of the various isotopes in drinking water, recommended by the NCRP and the ICRP, serve as a guide for disposal of waste in the environmental streams. The average dilution factors one can count on have to be know, and also their seasonal variations. Then, if one is able to prove that the most hazardous isotopes are safely disposed of, all the others, in general, will be also. The standards for the maximum permissible occupational exposure rates from isotopes deposited in a person's body are:[4]

a) The weekly RBE dose to the bone should not exceed the dose delivered by 0.1 μc of Ra^{226} with its daughter products.

b) The maximum permissible weekly RBE dose is 0.6 rem to the skin and the thyroid gland; 0.1 rem to the gonads or the total body; 0.3 rem to any body organ, with the above mentioned exceptions.

The maximum permissible concentration of a radioisotope in water for occupational exposure (MPC_w) is defined as that concentration of the isotope in drinking water which, if maintained long enough, would cause a continuous (internal) exposure rate, to some body organ, at one of the above mentioned limits. (For the formulae based on the "Exponential Excretion Model" and for exceptions thereof.)[4]

Three sets of limits to the MPC of radioisotopes in drinking water have been proposed by the ICRP.

a) MPC for occupational exposure. - The assumptions are:

A 40-hr working period per week, 50 working weeks per year for a continuous work period of 50 years. The average rate of water consumption is 5.5 liters per 40-hr working week. The mass, effective radius of the various body organs, as well as their average chemical composition and other pertinent biological factors, are assumed to conform with the standard man.

b) MPC in the neighborhood of atomic energy plans, effecting a relatively limited population. - They are set to 1/30 of the above mentioned MPC_w limits. The assumptions are:

A weekly intake of 15.4 liters of water, during 52 weeks per year, which accounts for the reduction of the MPC by a factor of 3.

The M.P. internal exposure rates are reduced by a factor of 10; the weekly maximum permissible RBE dose to any body organ is 30 mrem with some exceptions (10 mrem to gonads and total body, 60 mrem to bone, skin, and thyroid).

c) MPC values for large portions of the population. - These are reduced by another factor of 10, and are set at 1/300 of the (MPC_w) values listed.

Various conflicting considerations (practical, human genetics, and many others) have led to the above mentioned classification. The (MPC_w) values - for occupation exposure - have gained almost universal recognition, and seem to be systematically applied.

Table I summarizes some information concerning the beta emitting isotopes which are most hazardous with respect to internal exposure.

Strontium 90-Yttrium 90 is by far the most hazardous, and it constitutes one of the most abundant fission products.

When Sr^{90}- Y^{90} and I^{129} are completely removed from the water, MPC values for unidentified radionuclides (MPCU) might be used, provided some alpha emitters are known to be absent too (see values at the end of Table I).

If all the radioisotopes listed in Table 1 are absent, a value of 0.6 μc/liter is safe for (MPC_w), for any combination of other isotopes ($MPC_w/30$ = 0.02 μc/liter). Above this concentration of the GI tract becomes the critical organ for many isotopes.

Table 1. Most Hazardous Beta Emitters[4]

Radionuclides	Half Life	Critical Organ	$(MPC)_w$ μc/liter	$(MPC_w/30$ μc/liter	Fission Yield (%) (5)
Sr^{90}- Y^{90}	28 y	Bone	0.004	1.3×10^{-4}	5.9
I^{129}	1.7×10^{7} y	Thyroid	0.016	5.3×10^{-4}	1.0
I^{126}	13.3 d	Thyroid	0.079	2.6×10^{-3}	--
I^{131}	8.05 d	Thyroid	0.088	2.9×10^{-3}	2.9
Cs^{134}	2.3 y	Total Body	0.25	8.1×10^{-3}	--
Ca^{45}	164 d	Bone	0.27	9×10^{-3}	--
I^{133}	20.8 h	Thyroid	0.32	1.1×10^{-2}	6.5
Ru^{106}- Rh^{106}	1.0 y	G.I.	0.34	1.1×10^{-2}	0.38
Ce^{144}- Pr^{144}	290 d	G.I.	0.34	1.1×10^{-2}	6.1
Sr^{89}	51 d	Bone	0.35	1.2×10^{-2}	4.8
Unidentified Isotopes			0.03	10^{-3}	

APPENDIX B

Fission Products in Liquid Systems

Fission products are the main contributors to the activity of the liquid-waste system at ORNL. Some of the more abundant fission products constitute most of the hazardous isotopes listed in Table 1.

The experience gained at ORNL, in handling and tracing the mixed fission products in the waste, might be of interest to any atomic plant. Their appearance in unexpected places (like cooling water, etc.) is often the earliest sign of an operational failure (rupture, leakage, etc.).

Table 2 lists some physical properties of the most important fission products. Only nongaseous isotopes with half lives between one day to 100 years have been included, provided their U^{235} fission yield exceeds 0.3%. Also listed are those isotopes likely to be present when trying to separate chemically the more important ones. Having isolated an element, it is often of major practical concern to get quantitative estimates of the various radioisotopes which are actually present.

The average energy of beta particles - column 6 - is generally listed as 1/3 of the maximum values appearing in column 5. In cases where more than a single beta transition occurs, the values listed are from Blomeke's report,[5] whenever the maximum energy values were in reasonable agreement with the "Table of Isotopes".[6] Whereas the maximum beta energy is important for detecting the isotope, the average energy is used for the calculations of MPC values.

Table 2. Properties of Important Fission Products[6]

Nuclide	Mass Number	U^{235} Fission Yield (%)[5]	Half Life	Beta Radiation: Maximum Energy Mev	Beta Radiation: Average Energy Mev	Half Thickness In Aluminum mg/cm^2	Most Abundant Gamma Ray: Energy Mev	Most Abundant Gamma Ray: Abundance %	Additional Gamma Present	Active Daughter
38-Sr	89	4.8	51 d	1.46	0.49	90				
	90	5.9	27.7 y	0.54	0.2	20				Y-90
39-Y	90	5.9	64.2 h	2.26	0.75	190				
	91	5.9	58 d	1.54	0.51	105				
40-Zr	95	6.4	65 d	0.4	0.13	17	0.76	49	+ +	Nb-95
41-Nb	95	6.5	35 d	0.16	0.05	2.8	0.765	100		
42-Mo	99	6.1	66 h	1.2	0.38	70	0.14	88	+	Tc-99
44-Ru	103	2.9	40 d	0.20	0.07	5	0.5	90	+	Rh-103
	106	0.38	1.0 y	0.04	0.01	< 1				Rh-106
45-Rh	103m	2.9	57 m			--	0.04	100		
	105	0.9	36.5 h	0.57	0.18	21	0.32	5		
	106	0.38	30 sec	3.53	1.05	380	0.51	22	+	
51-Sb	125	0.023	2.0 y	0.61	0.10	22	0.4	40	+ +	Te-125m
	127	0.25	88 h	1.57	0.4	100	0.67	50	+	Te-127
52-Te	129m	0.34	33 d			--	0.106	100		Te-129
	129	1.0	72 m	1.45	0.5	95	0.46	15	+	I-129
	132	4.3	77.7 h	0.22	0.07	4.6	0.231	100		I-132
53-I	131	2.9	8.05 d	0.61	0.19	25	0.364	80	+	
	132	4.4	2.3 h	2.12	0.43	170	0.78	100	+ +	
	133	6.5	20.8 h	1.4	0.45	90	0.53	94		X-133
55-Cs	137	5.9	26.6 y	0.514	0.19	18				Ba-137m
56-Ba	137m	5.4	2.6 m			--	0.661	100		
	140	6.3	12.8 d	1.02	0.27	50	0.53	40		La-140
57-La	140	6.3	40.2 h	2.15	0.49	170	1.6	88	+ +	
58-Ce	141	6.0	33 d	0.58	0.16	22	0.145	30		
	143	6.2	33 h	1.39	0.35	85	0.29	43	+	Pr-143
	144	6.1	285 d	0.31	0.1	8	0.134	20		Pr-144
59-Pr	143	6.2	13.7 d	0.93	0.3	47				
	144	6.1	17.3 m	3.0	0.97	300			+	
60-Nd	147	2.6	11.1 d	0.81	0.22	36	0.09	85	+	Pm-147
61-Pm	147	2.6	2.64 y	0.22	0.07	4.5				
	149	1.3	54 h	1.05	0.3	52	0.28	100		
62-Sm	151	0.5	93.y	0.08	0.02	< 1	0.02	100		
63-Eu	155	0.031	1.7 y	0.24	0.056	5.1			+	
	156	0.013	15.4 d	2.46	0.43	200	0.09	100	+	

+: Energetic and quite abundant additional gammas.

Column 7 contains the approximate thickness of the first half value layer, in aluminum absorber, for the beta particles, as derived from a curve.[7]

Many isotopes are more easily detected by their gamma rays. The most abundant gamma ray is also listed in Table 2, provided it exceeds more than 5% of the beta disintegrations. The most abundant gamma ray is not always the most energetic one,or the easiest to identify.

The active daughters are listed in the last column. When the daughter is of importance the pertinent information is included in the table later. In some cases the daughter product activity is responsible for the inclusion of its longer lived parent in the table, the half life of which controls the disintegration rate.

APPENDIX C

Formulae for Gamma Flux

I. Point Source

The flux ϕ at distance R from an isotropic point source of strength S, beyond a shield of thickness t is:

$$\phi = \frac{S \cdot B \cdot e^{-\mu t}}{4\pi R^2} \qquad (1)$$

where μ is the attenuation coefficient of the shield and B a build-up factor.[30,31]

II. Surface Source[32,33,34]

The uncollided flux (neglecting the build-up factor) from an infinite isotropic plane source of strength S_A at a depth t inside an absorbing medium with a linear attenuation coefficient μ is:

$$\phi_A = \int \frac{S_A \cdot e^{-\mu t}}{4\pi t^2} \, dA = \frac{S_A}{2} E_1 (\mu t) \qquad (2)$$

Equation (2) is easily obtained by integration of equation (1) after expressing the surface element dA in cylindrical co-ordinates.

Note that no build-up factor is included.

III. Volume Source[32,33]

Integrating equation (2) over h, and letting $\mu t = 0$, one obtains the flux ϕ_V at the surface of an infinite absorbing slab of thickness h,

absorption coefficient μ_s , containing a uniform source distribution of strength S_V.

$$\phi_V = \frac{S_V}{2\mu_s} \left[1 - E_2(\mu_s h) \right] \tag{3}$$

Useful approximations for the exponential integrals $E_n(x)$ for $x > 2$ are:[34]

$$E_n(x) \approx \frac{e^{-x}}{x + n} \tag{4}$$

Acknowledgments

I gladly acknowledge my gratitude to the Health Physics Division at Oak Ridge National Laboratory for the hospitality and the kind reception I always enjoyed with each of the sections.

Thanks are due to the U. S. AEC and the Israel AEC who arranged my trip to ORNL. This gave me the unique opportunity to acquaint myself with various aspects and developments in the field of Health Physics.

I wish to express my gratitude to Mr. E. G. Struxness, Head of the Waste Disposal Research and Engineering Section, for including me in his stimulating group and for his continuous personal interest and encouragement throughout this work.

I doubt if there was even one person in the Waste Disposal Research and Engineering Section whose experience and advice I neglected to consult - my thanks to each of them. I had frequent and lengthy discussions with Dr. F. L. Parker, Mr. R. J. Morton, Mr. K. E. Cowser, and Dr. S. I. Auerbach. I wish to thank them for their fruitful suggestions.

I was much impressed by the frank and co-operative attitude of Mr. H. H. Abee and Mr. W. D. Cottrell of the Applied Health Physics Section. This report uses their data extensively. Many possibilities for improvement in the sampling methods were first suggested by them.

I wish to thank Mr. F. N. Browder of the Chemical Technology Division for sending me a rough draft of his report prior to publication.

My thanks to Mr. E. J. Witkowski, Head of the Operations Division, for his interesting discussions concerning the liquid waste system at ORNL and his fruitful suggestions..

Last, but not least, I wish to thank Mrs. J. M. Davis for her untiring secretarial assistance.

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