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LOCKHEED MARTIN



**Attitudes and Practices Regarding
Disposal of Liquid Nuclear Waste at
Clinton Laboratories in the Very Early
Years: A Historical Analysis**

Stephen H. Stow

Environmental Sciences Division
Publication No. 4508

MANAGED BY
LOCKHEED MARTIN ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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Environmental Sciences Division

ATTITUDES AND PRACTICES REGARDING DISPOSAL
OF LIQUID NUCLEAR WASTE AT CLINTON LABORATORIES
IN THE VERY EARLY YEARS: A HISTORICAL ANALYSIS

Stephen H. Stow

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EXECUTIVE SUMMARY

Recent research in the Central Files of Laboratory Records at Oak Ridge National Laboratory (ORNL) has revealed a large number of previously unreferenced documents that demonstrate that the management and disposal of the liquid nuclear waste generated at Clinton Laboratories (which became ORNL after 1948) during the 1940s was performed with the highest degree of integrity and professionalism. The general perception is that these wastes were handled in a careless and haphazard fashion with little concern for human health and the environment; such was definitely not the case as shown by the information presented in this report.

Even prior to construction of the laboratories in early 1943, professionals were making plans for the "safe" disposal of waste through treatment and dilution at medically prescribed levels into White Oak Creek and the Clinch River. Medical professionals, working with scientists and engineers, led the establishment of the disposal procedures; concern for human health permeated all the disposal decisions. Throughout the war years, a remarkable degree of attention and responsiveness was devoted to ensuring that the wastes were released after treatment in amounts deemed safe. Application of chemical and physical treatment processes was paramount to removing as much of the activity as possible before release. Environmental and biological monitoring of the surface water systems into which the wastes entered was instituted very early in the disposal history. Information learned at Clinton Laboratories with regard to waste disposal was transferred to the Hanford site to help guide disposal operations there.

After the war, scientists, engineers, and medical professionals were still concerned about safe methods for disposal, and by the latter part of the 1940s, they were formulating fairly sophisticated research programs for management of liquid waste. Realizing that there were also concerns about the disposal of low-level solid waste, they also began research to address this problem.

The individuals who were charged with the management of the liquid waste were prominent scientists, engineers, and medical professionals, who held high positions of responsibility and authority at Clinton Laboratories and du Pont as well as in other branches of the Manhattan Project. Many had come from the Metallurgical Laboratory at the University of Chicago, having been hired for their expertise by Arthur Compton, and many left after the war to take prestigious positions elsewhere.

This historical analysis attempts to place the actions of the 1940s in proper perspective, drawing on the attentiveness and integrity of those who participated 50 years ago. Applying standards of the 1990s to actions in the 1940s must be done skillfully, carefully, and with the full realization that those

individuals were operating under extremely trying conditions and with minimal knowledge of radionuclide behavior. Nevertheless, they consistently maintained the highest level of responsibility in properly managing the liquid waste on the basis of the knowledge they had at the time.

PERCEPTIONS OF CARELESSNESS: AN INTRODUCTION

Disposal of wastes and other contaminated materials from the three plants on the Oak Ridge Reservation is a highly visible and dominant issue today. Indeed, many billions of dollars are being spent to correct what many perceive to be the mistakes of the past, the past meaning the decades from the mid-1940s through recent times. Many—indeed, perhaps most—people perceive that waste materials have been carelessly handled and have been disposed of in a haphazard and cavalier fashion with little or no regard for safety, human health, and the environment. Certainly in many instances care was not taken, as it should have been, and uncontrolled releases of contaminants to the environment have occurred. This fact is recognized, but documentation of those instances is not the objective of this report.

When people are queried about their perceptions of the disposal practices during the very early days of operation of Oak Ridge National Laboratory (ORNL), which was called Clinton Laboratories until 1948, the response is generally either that they know nothing about it, but presume that little professional attention was given to the issue, or they state outright that the wastes were probably “dumped in the woods, or in the river,” or words to that effect. A few individuals do hold the view that wastes were disposed of in a (semi) professional manner, but that position is rare and generally unsupported by facts.

Recent research in the Central Files of Laboratory Records at ORNL has revealed that very conscious efforts were made, even as the construction was started at Clinton Laboratories, to handle the liquid waste in a safe fashion on the basis of the scientific and medical knowledge of the time. This awareness continued through the war years, and afterwards. A group of insightful and influential individuals consistently sought safe methods of handling the highly radioactive and dangerous wastes and of disposing of them properly. Documentation of this position is the subject of this report.

This report deals almost exclusively with the liquid radioactive wastes with passing reference to gaseous wastes. Solid wastes are equally important, as we know today, but they did not receive the attention that the liquids did for many years; a discussion of solid wastes is included in this report, but they are a subject for further study. This investigation also does not attempt to determine rates of waste disposal and quantities disposed of; such data are fairly well presented in other documents referenced later. Finally, this present study does not deal with worker exposure and contamination incidents; these issues are beyond the present scope.

The objective of this report is to present information that demonstrates the awareness and surprisingly high level of conscientiousness in handling liquid wastes during the very early years of construction and operation of Clinton Laboratories, the years 1943 until after World War II. Less complete information will also be presented to substantiate attitudes and actions through the balance of the 1940s.

THE MANHATTAN PROJECT

To more fully understand and appreciate the significance of the waste handling issues during the war, at least a rudimentary knowledge of the Manhattan Project is important. Several books have been written on this topic and the reader is encouraged to investigate further for more details of the Project, including the activities at Oak Ridge (Hewlett and Anderson 1962; Hewlett and Duncan 1962; Gerber 1992; Kathren et al. 1994; Lyon 1976; Overholt 1987; Rhodes 1986; Smyth 1945; Weinberg 1994). The following is only a brief synopsis.

In 1938, German scientists determined that ^{235}U was fissionable and that the fission process, if occurring in a chain reaction, could yield immense amounts of energy. Nuclear physics calculations suggested that element 94 (mass 239), later named plutonium (Pu), was also fissile; experiments carried out in 1940 at the Metallurgical Laboratory (Met Lab) at the University of Chicago, where a small, crude, but effective nuclear reactor was soon built, actually produced microgram amounts of plutonium using a cyclotron. During that time period, President Roosevelt, having been contacted by Albert Einstein, authorized increased research on the production of gram quantities of plutonium and on methods to enrich ^{235}U (which is highly diluted by the nonfissile ^{238}U) for ultimate production of a nuclear weapon.

It was feared that the Germans might produce a nuclear weapon and time was limited, so many concurrent activities had to be undertaken. The ability to produce plutonium by neutron irradiation of ^{238}U in a reactor had been proven, but large quantities would be needed and large reactors would need to be built. The plutonium would have to be separated from both uranium and the many highly radioactive fission products that resulted from the breakup of uranium (and some plutonium), but little was known of the chemistry of plutonium. Materials for construction of the reactor were scarce, as was uranium, and the design of a massive reactor without pilot-scale testing was a formidable challenge.

In late 1942, the decision was made to use the Oak Ridge site for developing the technology to produce plutonium and to enrich ^{235}U ; the site that is now ORNL became known as the Clinton Engineer Works and later as Clinton Laboratories. Months before construction started, scientists realized that the potential dangers associated with large-scale production of plutonium were such that placing the production reactors near a highly populated area (Knoxville) was unwise, and the decision was made to construct them elsewhere; soon the Hanford, Washington, site, an arid and isolated place, was selected. Displeased and displaced

local residents lost their property as the federal government purchased thousands of acres of land for the research and production facilities. At the Clinton site in Bethel Valley, the du Pont Company began construction on February 2, 1943, with round-the-clock activity; by November 4 of that year, the graphite reactor (Building 105) was operating and producing plutonium (see Figs. 1 through 6 for the locations of buildings and the construction history through 1947). Shortly thereafter (December 14), the adjacent separations plant (Building 205) received the first slugs of irradiated uranium and began the pilot-scale operations for testing chemical methods for recovery of the plutonium. Although the reactor and the separations facility were the center of attention, major efforts were also under way in the Chemistry Division (Building 706A), where pioneering work was being conducted on the chemistry of radionuclides and methods of plutonium separation. Constant communications with Hanford about the construction activities there allowed changes in design of the facilities there, as new discoveries were made at Clinton Laboratories. From early 1943 into early 1944, 150 buildings, costing \$13 million, were constructed in Bethel Valley.

The organization of Clinton Laboratories paralleled that of the Met Lab. Originally, there was a Medical Division, with Biology and Health Physics Sections, a Separations Development Division, a Physics Division, a Chemistry Division, an Analytical Division, and an Engineering Development Section (later called the Technical Division). Figure 7 is an organization chart from February 1944, which generally reflects this structure and which contains the names of many individuals discussed in this report.

Because none of the different methods for enriching the ^{235}U was obviously superior, different plants were constructed that would use different technologies. Elsewhere at the Oak Ridge site, near the Clinch River, the gaseous diffusion plant, soon known as the K-25 Site, was being built at a speed similar to that seen in Bethel Valley. In Bear Creek Valley, another plant (Y-12) was developed for testing the electromagnetic method of uranium enrichment.

Efforts here, at Hanford, at Los Alamos (where the atomic bombs were designed and constructed), at Argonne National Laboratory, and at numerous universities and industrial research centers in this country and overseas, quickly led to the successful testing of the first atomic bomb in New Mexico on July 16, 1945. Only weeks later, the bombing of Japan swiftly brought an end to the war.

The Manhattan Project was remarkable in many ways. Not only did it require the top nuclear scientists in the free world to dedicate themselves



Figure 1: Early construction, March 1, 1943. Photograph was taken one month after construction started; view of the western part of the facility where administrative offices were located.



Figure 2: Progress as of April 15, 1943. A view looking northwest from above where the holding ponds would be built. The road in the center goes up hill where the pile (graphite reactor, Building 105) was located, out of the field of view to the right.



Figure 3: Progress as of June 27, 1943. This is the same perspective as in Figure 2. The steam plant is in the center and the foundation structure for the separations facility (Building 205) is seen on the right up the hill.



Figure 4: Progress as of August 31, 1943. The partially completed pile (Building 105) is seen in the center with the separations building under construction to the right. On the other side of the pile is the machine shop where the graphite was prepared. In the background are the six large graphite tanks, and on the north side of the road the two smaller tanks, all yet to be covered. Further away are the two retention ponds. The large building to the left (706A) is the Chemistry Division. The Physics Division building is on the hill to the right of the pile behind the steam plant.



Figure 5: Progress as of Summer 1944. At this time all the originally planned construction had been completed. The Chemistry building has a new addition on the front. The building between the pile and the Chemistry Division was the "Hot Laboratory" (706C). The settling basin, beyond the two retention ponds, has been completed. White Oak Creek flows just behind the settling basin.



Figure 6: Clinton Laboratories in 1947. The graphite reactor building had been painted white after the war and the "Hot Laboratory" expanded. The Physics building now had three wings. Just below the separations building was a new training school facility and quonset huts, which became the central machine shop, were under construction. There were no paved roads at this time.

Clinton Laboratories Organization Chart

Total Employees: 1099
Date: 2-25-44

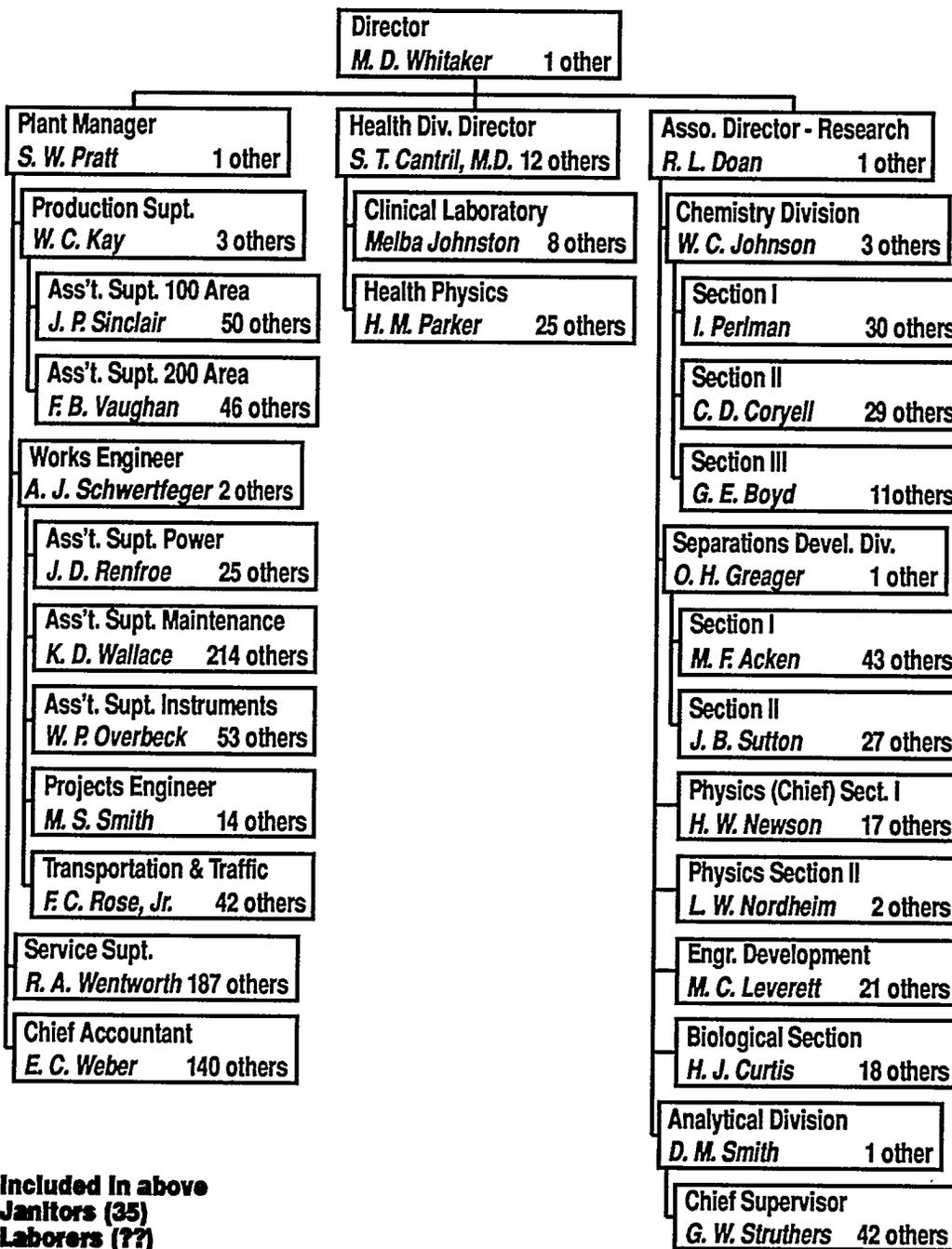


Figure 7: Organization chart for Clinton Laboratories, February 1944.

to deciphering nuclear physics and chemistry while in a crisis mode, but the decisions that had to be made with incomplete knowledge of a particular issue were innumerable. The rapidity with which the entire endeavor was completed would not be possible today.

LIQUID WASTE DISPOSAL AT CLINTON LABORATORIES

The topic of this report is liquid wastes, so emphasis will be placed on them, rather than on the solid low-level waste that has been placed in six burial grounds since January of 1944 or on the gaseous waste that was discharged to the atmosphere. For a detailed review of the liquid disposal operations, as well as those for solids and gases, the reader is referred to other publications that fully describe the diverse aspects of ORNL's entire waste disposal efforts over the years (Browder 1949, 1959; Coobs and Gissel 1986; Feige et al. 1960; Webster 1976).

Prior to construction of Clinton Laboratories, the necessity for properly handling the liquid waste was realized and plans were made for this, as shall be discussed later. Liquids were generated primarily from the separations plant (Building 205) and the Chemistry buildings (706 area); these wastes contained high amounts of uranium and fission products, some plutonium, nitrate, and other chemicals. To more fully understand some of the separations processes and the chemical nature of the waste materials resulting from them, refer to the Appendix, which has been excerpted from the daily notes of Glenn Seaborg (Kathren et al. 1994). The acidic wastes were neutralized with Na_2CO_3 or NaOH and most of the transuranic elements, as well as 90% of the fission products, were precipitated; the addition of CaCl_2 enhanced this precipitation, as we shall see later. Amounts of waste on the order of 30,000 gal/d were produced. If the total activity of these wastes was lower than 25,000 beta counts/ml/min, the liquid was sent to a 1.6 million-gal settling basin (see Fig. 5) where it was held to allow settling of the precipitates and decay of short-lived radionuclides. If the activity was above that limit, the waste was sent to the the tank farm area before further release.

The tank farm consisted of underground tanks constructed in the vicinity (206 area) of the Chemistry buildings (706 area) for retention of the higher activity waste; six of these tanks (the gunite tanks) had volumes of 170,000 gal each (Figs. 8 and 9); the others were smaller, and different types of liquid wastes were sent to specific tanks. In addition, underground tanks were associated with most of the buildings where radionuclides were handled, and waste entered these tanks before further treatment. At the tank farm, precipitation occurred and nuclides decayed. When the activity was low enough, the supernatant liquid was sent to the two 200,000-gal retention ponds, where the average holding time was approximately 3 days, before going to the settling basin pending release to the creek. Two smaller tanks were also located at the tank far across the street from the large tanks. The tank capacity had been designed for the planned 1 year of operation of Clinton Laboratories.

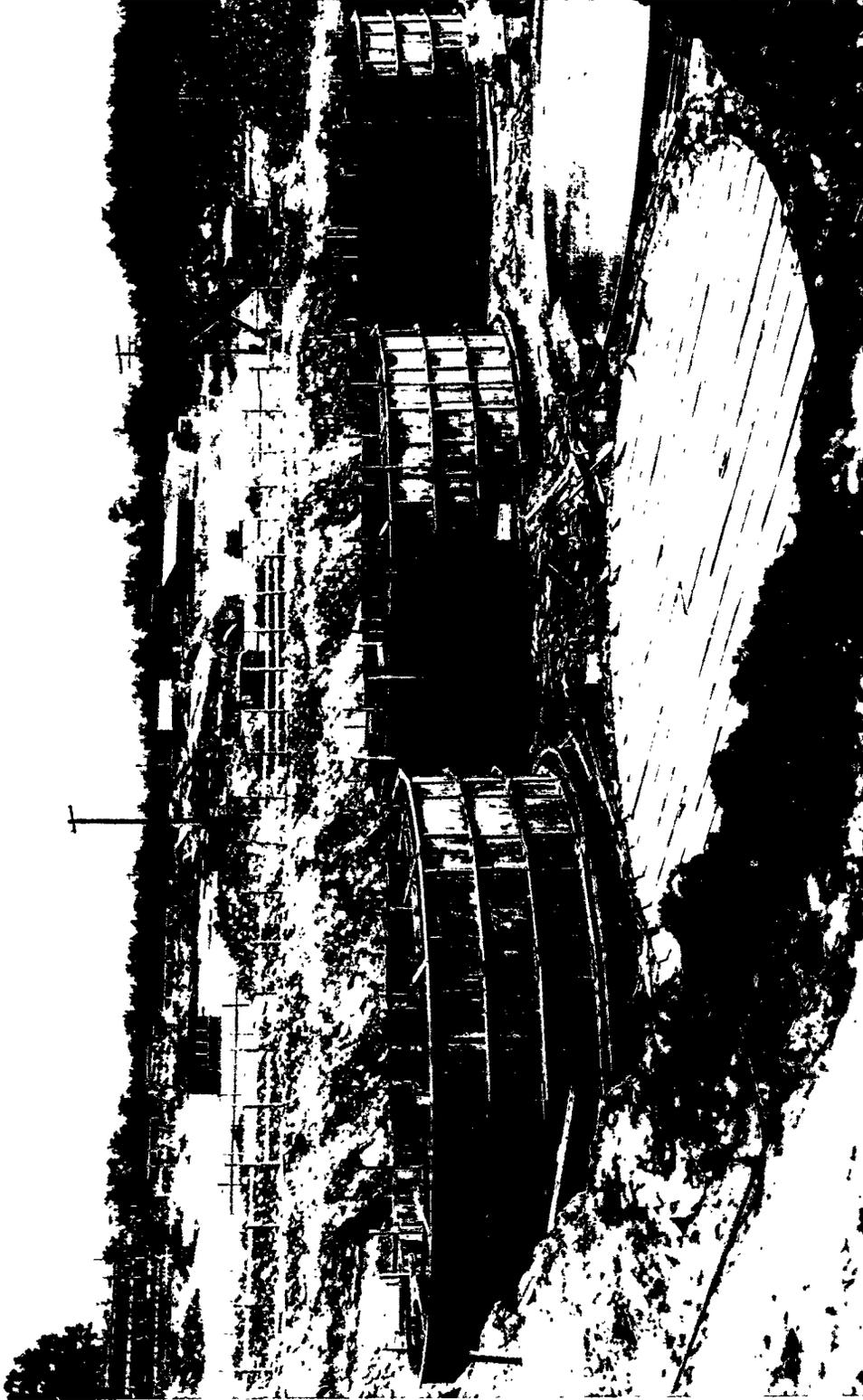


Figure 8: The gunite tanks in the spring of 1943. Construction is at an early stage on the six large gunite tanks.

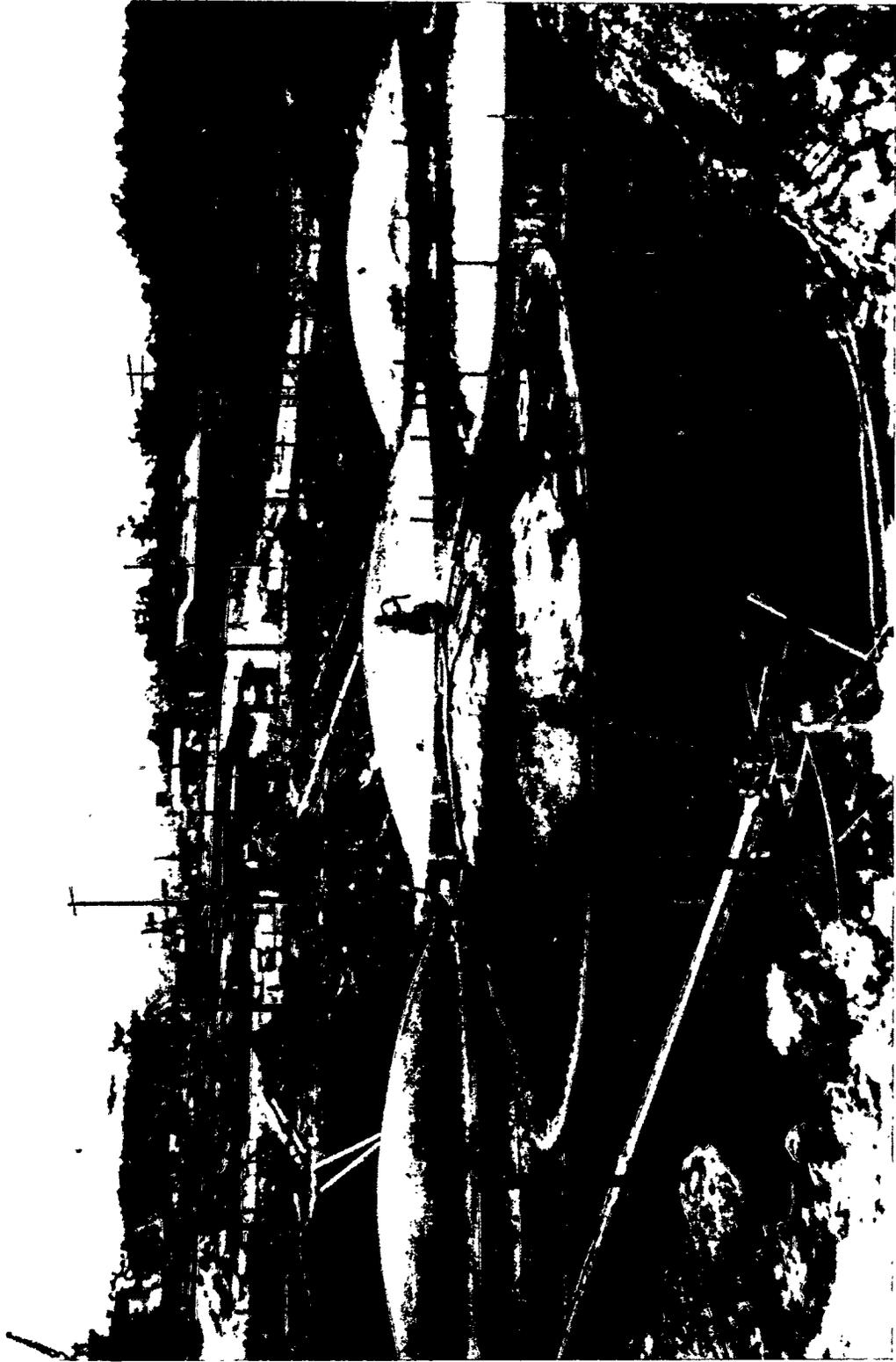


Figure 2: The gunite tanks, as of July 1943, nearing completion. The forms for pouring the concrete shield for the reactor can be seen in the left background.

Flow from the settling basin and the retention ponds was mixed with uncontaminated "process water" (ratio of about 1:35) and the mixture then released at regulated rates up to 5 Ci/d to White Oak Creek, which flowed into White Oak Lake, created in the fall of 1943 by construction of a dam at Highway 95 (Figs. 10 and 11). Releases from the lake to the Clinch River were then regulated, as described later. The dilution ratio in the Clinch River was approximately 500,000:1.

This general method of handling liquid waste prevailed until the late 1940s, when ORNL changed its disposal practice in response to the Atomic Energy Commission (AEC) policy that waste discharges to streams be eliminated or drastically reduced (Browder 1949). In 1949, an evaporator was constructed, and the volume of liquid waste was reduced by a factor of approximately 20. From the early 1950s until 1965, wastes were disposed of after chemical treatment by allowing them to seep into the ground through large pits and trenches in Melton Valley (deLaguna 1956; deLaguna et al. 1958; Coobs and Gissel 1986; Spalding 1987). This disposal practice was replaced in 1965 with the hydrofracture operation in which the liquids were mixed with cement and other additives and injected at a 1000-ft depth into a low-permeability shale (Weeren 1982; Stow and Haase 1986; Haase and Stow 1987). Since the early 1980s, the bulk of the wastes have been stored, awaiting development of treatment and disposal options.



Figure 10: White Oak Lake and Dam, 1943. The lake was created in the fall of 1943 by construction of the dam where Highway 95 crossed the creek. The gate structure can be seen on the right hand side of the dam.



Figure 11: The gates on White Oak Dam, 1943. View looking north with the lake itself to the right.

THE CENTRAL FILES: SURVIVING RECORDS

Folklore has it that the United States developed the atomic bomb without benefit of paperwork . . . but . . . the wartime atomic energy program must rank with the most thoroughly documented enterprises in history. In retrospect, the reasons are apparent. The atomic bomb was a two-billion-dollar gamble. If the wager were lost, Congress would demand an explanation. The men who had staked their reputations on the outcome wanted written evidence to justify their decisions.

The records have survived. Classified documents endure; they do not disappear from the files as souvenirs. (Hewlett and Anderson 1962)

In a review of archived historical documents, now declassified, innumerable references can be found that relate to disposal of wastes, to efforts directed at waste minimization and handling procedures, and to monitoring the impact of disposal on human health and the environment. Further searching of the archives will undoubtedly reveal a significant number of additional documents that will help substantiate the thesis of this report. Every reference to waste disposal in not intended to be covered in this report; those references that deal with health and safety concerns and those that demonstrate the actions and attitudes toward disposal practices are used.

The vast majority of the material comprising this discussion was acquired from the Central Files of ORNL Laboratory Records in the form of memos, letters, monthly reports, and other documents, all of which are unpublished and largely unknown. Some of the referenced material from the late 1940s that is used in this study was issued internally and distributed within the ORNL (or AEC) system. The Central Files at ORNL contain approximately 100,000 documents, over 50,000 of which were generated in the 1940s, and random searches would be highly unproductive. Fortunately, the early documents have been entered into a database that can be searched by author or keyword, and such a method was used to identify approximately 8000 potentially relevant titles for inclusion in this study. Of these, some 1000 that appeared most relevant were individually examined to determine if material was of use; this examination resulted in identification of the information base upon which this study is centered. Without doubt, many more documents could yet be identified and incorporated.

For reference purposes, the ORNL Central Files number is used in this report for documents from the unpublished information base. Each document has a unique number that consists of three parts: the first two-digit number represents the year in which a document was prepared (e.g., 44 for 1944); the second number refers to the month in which it

was logged in (1 for January, 2 for February, etc.), and the third number simply represents the sequence in which the document was received and filed in each month. Therefore, the number 44-5-139 would represent the 139th document filed in May of 1944.

In the following discussion, the original documents are extensively quoted; these quotes are presented verbatim, regardless of spelling, grammar, punctuation, or style. Throughout the text, activities in waste waters are quoted in a variety of units (curies/liter, counts/minute/milliliter, etc.). These have been taken verbatim from the references and no attempt has been made to convert them to one consistent unit; the units are summarized in Table 1 located near the end of this document.

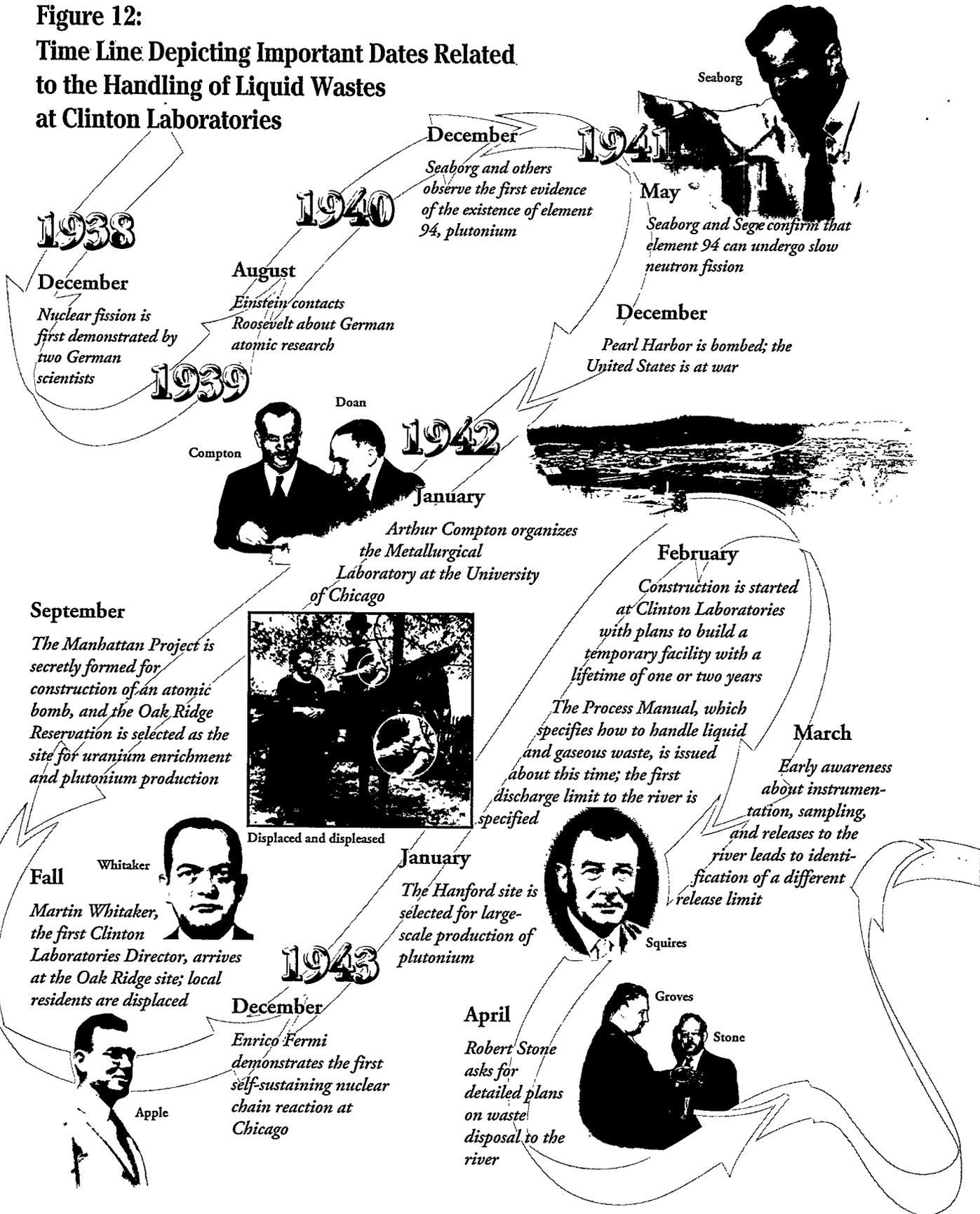
THE YEAR OF CONSTRUCTION

... APPROACHING PURE WATER

A detailed search of the Central Files records from late 1942 and very early 1943 revealed a dearth of references to liquid waste disposal, although some reports did show concern over the gaseous wastes, as will be discussed later. A series of early documents (43-2-42, 43-2-135, 43-3-2, 43-3-179, 43-3-203, 43-3-228, 43-3-297) all reference waste disposal in their titles, but they are missing from the files (perhaps as souvenirs after declassification). Indeed, in very early and rather thoughtful communications related to the design and construction of the Clinton Laboratories (42-12-22, 43-1-12), the issue of liquid waste disposal is notable by its absence, although the fact that wastes will be generated is noted on a flow sheet for a separations process (43-1-19). It is established, however, that the issue of safe waste handling was considered; discussions within the Chemical Engineering Group at the Met Lab on November 16, 1942, when different separations processes were evaluated, reflect that the question, "Can process wastes be handled safely?" was one of the factors considered, on the basis of notes taken by Glenn Seaborg (Kathren et al. 1994). Figure 12, a timeline that traces important events related to the following discussion, can be used for reference purposes.

A few early references that allude to disposal needs and surface water contamination do appear. One, a March 9 memo from G. (George) D. Graves, a du Pont engineer, to C. (Charles) W. J. Wende (43-3-85) was followed a few days later by a transmittal from Wende to M. (Martin) D. Whitaker, the Director of Clinton Laboratories. (Note that a "Process Manual," which is discussed shortly, was prepared in this same time frame and perhaps earlier.) Graves was writing to inquire "what water supply will be needed for the storage basins in which buckets of active uranium are to be stored at X in order to keep the activity in the basins down to a point so that the water can be drained to the river." ("X" refers to the Clinton Laboratories site.) He provides some insights on what assumptions must be made, and concludes with, "From all this and the volume of the basins I presume you can determine how much water should be kept moving through the basins without excessive contamination of the river." In the follow-up letter to Whitaker (43-3-124), Wende bases his calculation on rates of consumption of river water and absorption of nuclides by the body and concludes "a flow of 500 gallons per minute should reduce the activity to a tolerable level for a single dose of one gallon. A higher rate of flow would be required if the water is to be fit for continual drinking." He discusses both ingestion and immersion hazards, and a tolerance dose of "0.01 r per 8-hour day, assuming a man is to remain submerged for that time," is referred to. In

Figure 12:
Time Line Depicting Important Dates Related
to the Handling of Liquid Wastes
at Clinton Laboratories





Leverett



Peterson



Boyd

1944

December

Outside



Inside

The separations plant, building 205, comes on line, and Simeon

Cantril establishes the responsibility of the Medical Department for controlling releases to the river

November

The "pile" becomes fully operational; White Oak Dam is completed, and Howard Curtis

first proposes biological monitoring of the creek and river systems.

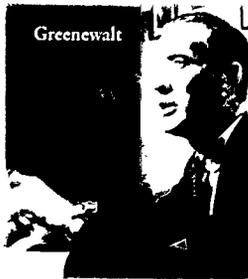
Whitaker informs Compton of the completion of the gunite tanks

A decision is made to change the separations process to one that uses bismuth phosphate

1943

June

S. W. Pratt and Lyle Borst question the proper disposal of wastes from the 706A building



Grenewalt

June

Cantril summarizes important data on contamination in White Oak Creek, and Curtis reports to Richard Doan on radiation of fish from the creek and river system; Parker and Hamilton continue to establish tolerance levels

July

The settling basin goes into operation on the 3rd, but by the 8th problems are noted; by the 24th, Kay documents operational changes to correct the problems, on the basis of suggestions from Miles Leverett

September

Heavy rainfall causes the dam to flood and overflow

May

Tests continue to determine if settling basin will function, and more laboratory work attempts to reduce waste; Whitaker informs Joseph Hamilton about disposal activities, and Hanford anticipates precipitation problems on the basis of Clinton's experience



Johnson

February

The first burial ground is established for solid wastes



Morgan

Karl Morgan writes Herbert Parker about background levels in White Oak Lake



Parker

April

Calcium chloride is added to waste stream to reduce activity (17th), and Overstreet and Jacobson survey creek (26th); waste discharge is stopped on the 27th and plans are made for a settling basin

March

This first recognized date of release of wastes to retention ponds and creek (6th) is followed by concern about precipitate forming; an initial survey of creek (15th and 16th) with recognition of a problem is followed by laboratory effort to reduce activity (18th-25th)



September

Wastes may have been released to the retention ponds this early, according to information from Oswald Greager



W. Smith

1944



Wigner

October

William Ray surveys the drainage system after the flooding; Medical Department authorizes a five-fold increase in the amount of activity allowed to be released



Briggs

November

Again, water tops the dam, and the procedure for cleaning the fish screen is changed; one of the gunite tanks overflows

February

A five-day period of uncontrolled releases over White Oak Dam is documented by Ray

July

The first radiation survey of the first burial ground takes place

1945

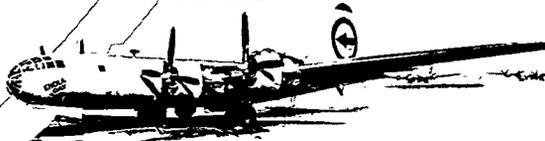
Early

The handling of the liquid wastes becomes much more routine and reporting on a (semi) regular basis begins to increase

1946

August

The war ends



Mid 1946

Morgan resists relaxing tolerance limits

April

Stone advises Hanford about groundwater monitoring wells

June

Health Physics reports are regularly issued

1947

January

Ray writes to James Lum regarding better methods of waste handling



Lum

March

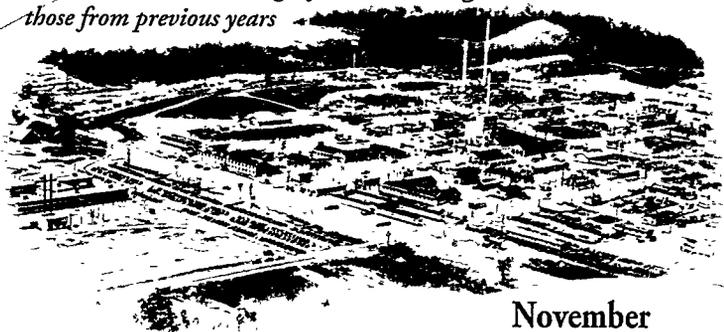
Cheka and Morgan report on sediment surveys performed in the White Oak drainage system, including those from previous years



Emlet

August

An early survey of the Clinch and Emory rivers is performed



October

Plans are made for another survey of the Clinch and Emory rivers

November

The AEC is advised that waste research should have a high priority at its facilities

1948

January

Questions arise regarding the correctness of disposal to the Clinch River, and new ideas on waste research begin to emerge

August

A conference is held with the AEC on liquid waste management

retrospect, this was an intriguing inquiry, as no further known documents refer to storage of uranium in this manner. After the war, Wende emerged in a prominent role with General Electric at Hanford and actively opposed certain reactor advances planned for Oak Ridge.

A weekly report in mid-March, entitled "Separation and Waste Disposal—TNX Manufacturing Division" (43-3-240), from J. D. Ellett, B. C. Nylen, and F. (Frank) B. Vaughan to W. (William) C. Kay, discusses the lack of available continuous radiation detectors for monitoring the waste waters and suggests the use of batch testing in the laboratory. The report states, "We shall seek approval of Chicago and other interested persons to this method." Likewise, the report also suggests that the testing of waste from certain tanks and from the ponds be done in batch-fashion with "a sample being taken from each pond and tested for sensitivity before discharging to the river." The "problems" area of this report points out that "The degree of sensitivity" (i.e., the degree of contamination) of contaminated wastes that can be sent to the ponds and "that can be discharged from the ponds to the river" is a problem. No explanation was offered, but certainly the awareness was there. At this same time, R. B. Smith wrote a lengthy memo detailing the need for radiation detection instruments for all aspects of work at Clinton (43-3-246); included in his list are "meters for measuring sensitivity of cooling water, water from hot metal storage pit, storage pond." At the end of the month, Ellett wrote to H. T. Daniels of the Design Division of du Pont (43-3-273) asking for provision of a means for continuously sampling "material entering the ponds" and Ellett, Nylen, and Vaughan reaffirm the batchwise sampling of the tanks and "continuous sampling of water entering and leaving the ponds to insure safe contamination limits" in communication with Kay (43-3-277); they note the further desire for continuous monitoring in communication to S. (Stuart) W. Pratt (43-4-115). In the letter to Kay, an early discharge limit is specified:

In the absence of definite specifications on safe or acceptable maximum contamination limits for water discharged from the ponds into the stream, volume relationships and a tentative limit of 10^{-4} curies/cu. ft. were given to W. R. Kanne to permit him to calculate sensitivity contents of the various flows. [Kanne (1913-1984) was a physicist at the Met Lab, later to work at Clinton, Hanford, and Savannah River.]

There was also concern about contamination of the Columbia River at Hanford. E. (Eugene) P. Wigner, at the Met Lab, wrote to C. (Crawford) H. Greenewalt in February (43-2-14), in reference to the generation of radionuclides in the river water as a result of irradiation of natural impurities (presumably after use for reactor cooling), pointing out that the level could be "about 50 times the permissible amount" if all nuclides were concentrated at one point. He proceeds to point out that

mixing and self-shielding will occur, concluding "it is apparent that not much has to be feared on this score, even if the total amount of impurities is higher than the old analysis shows." Wigner (1902-1995), of course, is renowned for his contributions to theoretical and nuclear physics and later served as Co-Director of Clinton Laboratories. Greenewalt (1902-1993), an engineer (MIT, 1922) and director of a research division in the Explosives Department of du Pont, was in charge of liaison with the Met Lab; he had served on the Lewis Committee and later rose as a prominent figure in AEC-related activities and was named President of du Pont in 1948.

An early reference to liquid waste disposal and health-safety issues, which introduces Robert Stone, is found in the minutes of an April 13, 1943, meeting of the Radiation Instrument Coordinating Committee (43-4-156), almost certainly held at the Met Lab. Two topics were discussed, "Waste Disposal at Site X" and the "Stack Meter for the Chemical Separation Plant (205)." These minutes note that "Dr. Stone requested a memorandum of the detailed plans for waste disposal at Site X with special reference to what will be put in the river." They go on to say that "It was Dr. Stone's opinion that the wastes going into the river must be kept at a very low level of radiation, approaching pure water." Also included is a description of the sampling protocol:

For the health and safety record each pond and tank should be sampled about every two hours and the hard and soft gamma and beta radiation fractions be determined. A 24-hour integrated sample was also to be tested for long time decay. The committee decided that two counters in the chemical control laboratory would be necessary for this waste sampling.

Robert F. Stone (1895-1966), whom we shall hear more from later, was a recognized authority on physics and radiation protection and served as Head of the Health Division for the Manhattan Project at the University of Chicago, having been hired in August 1942 by Arthur Compton [Project Director at the Met Lab and earlier (1927) a Nobel Prize winner] from his position as Chairman of the Radiology Department at the University of California at Berkeley. Stone supervised the medical and radiation protection activities at Clinton Laboratories and Hanford and was one of the few scientists with experience in applying nuclear physics to medicine; he had worked with Ernest O. Lawrence on the study of radiation hazards to humans. Stone, born in Ontario, received all his academic training from the University of Toronto (M.D., 1928) and worked in China and at the Grace Hospital in Detroit before going to California in 1925. He moved to Clinton Laboratories at an early point, as a "personnel list" for transfer, dated April 20, 1943, from Stone to Whitaker (43-4-168), lists him and his family, along with other familiar names, in a "family unit" here. After the war he returned to Berkeley.

CONSISTENT WITH THE PUBLIC HEALTH OF THE AREA?

At roughly the same time (April 9 and 27), the "Waste Disposal Flow Sheet for Site X," prepared by the University of Chicago Engineering Group, was issued (43-4-93, 43-4-218, 43-4-241). This was for the "wet fluoride" process, which was then the preferred method for separation of plutonium from the irradiated slugs [a bismuth process flowsheet had been issued in March (43-3-44)]. This April document contains a rather detailed analysis of the chemistry, volume, and fate of the entire waste stream with ultimate disposal in the waste storage tanks; no reference is made to disposal to the river. In June of 1943, a decision was made to use the bismuth phosphate process, at least in part, to replace the fluoride process at Clinton, because the bismuth process would be the one used at Hanford. In September, an equally detailed flow sheet (43-9-223) was issued for this newer process; again the tanks were the ultimate disposal method noted. At the time that decision was made, a detailed description of the waste handling process, with a preliminary flow sheet, was issued (43-6-151). The details contained in these flow sheets and the process description testify to the attention given to handling of the wastes. However, no reference is made in any of these documents to health and safety. The waste streams contained large amounts of uranium, which was very valuable of course, and which needed to be recovered for reuse, so a large part of the attention was for fiscal and conservation purposes. In a memo later in the year, from O. (Oswald) H. Greager to R. (Richard) L. Doan (43-12-46), reference is made to "plans to recover heavy material from the . . . wastes," and "every effort should be made to expedite recovery . . . and to return this storage capacity to use." Reference to these bismuth phosphate process documents is later found in the Chemical Engineering Section's September report (43-9-70), where a separate section appeared on "Waste Disposal."

Richard Doan (1898-1982) was Head of the Research Department at the time and had been Laboratory Director and Chief Administrative Officer at the Met Lab in 1942, having worked with Compton on study of cosmic rays; he was a member of Compton's "Planning Board," along with other notables, including Samuel Allison, Enrico Fermi, Norman Hilberry, Frank Spedding, Leo Szilard, John Wheeler, and Eugene Wigner. After coming to Oak Ridge, he served as Associate Director at Clinton Laboratories and the research divisions reported to him. He was a native of Indiana, had earned a doctorate in physics from Chicago in 1926, and had worked for Western Electric and Phillips prior to the war. Oswald Greager, born in Maryland in 1905, was a chemist with du Pont with a doctorate from the University of Michigan (1929). He had come to Clinton Laboratories in 1943 to supervise the shipment and reassembly of the bismuth phosphate processing equipment and was in charge of the Separations

Development Division in 1944 and reported to Doan; during the war he transferred to Hanford, where he occupied managerial positions for General Electric after the war ended.

An obvious concern about health and waste disposal was raised in June of 1943. In this case, it was in reference to the wastes from the 706-A Building in a letter from Pratt (per W. C. Kay) to Daniels on du Pont letterhead (43-6-112); Pratt (1899-1986) was a du Pont engineer transferred to Clinton as Plant Manager, and Kay was Production Superintendent, reporting to Pratt. Kay (1908-) held a D.Sc. degree from MIT (1937) and had been with du Pont since 1934. Attached to this is an earlier letter from L. (Lyle) B. Borst to Pratt regarding "Laboratory Wastes from 706A," in which he alludes to whether the "wastes from 706A will constitute a disposal problem." Following a list of quantities of waste materials, Borst's letter reads:

A preliminary examination and discussion with a sanitary engineer indicates that certain of these ions would be considered dangerous in a waste drainage rate of 1,000 gallons per minute. Dr. Whitaker tells me that he may not consult civilian engineers on this subject. We, consequently, hope that you will be able to find satisfactory recommendations within the duPont Company.

It is unclear why Whitaker was reluctant to seek advice; perhaps because of security issues. Pratt's letter forwards the chemical data and then says, "The Health Group at the University of Chicago has questioned whether the disposal of the products of this type and magnitude into White Oak Creek will be consistent with the public health in that area." The letter goes on to request a statement from the Engineering Department about consistency with "normal industrial practice in the general area." Within a few days, Daniels responded (43-6-147):

The resultant mixture discharged to Clinch River water from White Oak Creek should always contain 30 to 40 p.p.m. alkalinity. The KCN and CuO concentrations are acceptable. The proposed dam and retention pool at the outlet of White Oak Creek will assist greatly in averaging out the wastes toward the lower concentrations. [Note: the "CuO" has been corrected by hand to "CaO" but was originally "CuO" in Borst's letter, as well as in Pratt's letter.]

Although this answer was prompt, it was hardly scientifically defensible and almost certainly did not satisfy the original request from Borst. Lyle Borst (1912-), originally from Chicago, received a doctorate in chemistry from the University of Chicago (1941) and came from the Met Lab to Clinton Laboratories, where he remained until 1946; he left after the war to design the graphite reactor at Brookhaven National Laboratory and held a professorship at the MIT until 1951. He then held positions at

the University of Utah, New York University, and the State University of New York in Buffalo.

SETTING RELEASE LIMITS

A series of "process manuals" was issued in 1943 in which extremely detailed descriptions of various processes were presented. One of the manuals, "Waste Disposal Wet D Process," by W. (William) E. Kirst, was assigned to Whitaker, the Laboratory Director, in September of 1943. Exactly when this was prepared is not certain, although there is indication that it could have been as early as February 15, 1943; certainly it was prepared during the first few months of the year. Kirst (1892-1982) was a chemical engineer (Case School of Applied Science, 1918) who had been with du Pont since 1926. This manual, some 20 pages in length (a rather long document for those times), details the exact methods by which liquid and gaseous wastes will be handled, including detailed descriptions of the process lines, the underground tanks, the stack for the 205 Building, the chemistry of the various waste solutions, and step-by-step procedures for the handling of wastes, as well as methods and procedures for testing for leaks in the underground tanks. "The maximum allowable radioactivity that may be discharged into the river" is specified and defined:

such as to give a total exposure of .1 r based on 2 Mev gamma to a fish or animal immersed in it for 24 hours. The action of beta radiation of similar intensity would be about the same, except that the penetrating power is much less than gamma rays and consequently, as somewhat greater exposure can be tolerated.

A level of activity, " 8.1×10^{-10} curies per cc, or 2.3×10^{-5} curies per cubic foot," is defined for this discharge (compare this to the 10^{-4} curies/cu. ft. figure in the 43-3-277 memorandum) and it is specified, "The concentration is determined by counting a definite sample." [This figure of 0.1 r (roentgen) per day was originally established in 1934 by the U.S. National Committee on Radiation Protection and Measurements after the International Commission on Radiation Protection had established a limit of 0.2 r per day; Cantril and Parker (1945) provide an excellent historical analysis of tolerance limits.] A section on the need for holding waste in the retention ponds until it can be tested and found to be of low enough activity for discharge to the creek as well as a surveillance schedule "to insure the safe storage of radioactive wastes" are also found in the document. Martin Whitaker (1902-1960), a North Carolinian, had come to Clinton Laboratories in late 1942 as Director, having been one of Enrico Fermi's assistants at Chicago, where he helped in the design of an "exponential" pile; Whitaker was one of the team members (along with Greenewalt) present on December 2, 1942, when the first controlled

nuclear reaction was demonstrated and the experimental pile went critical at Chicago. He came from New York University where he chaired the physics department after earning a degree there in nuclear physics in 1935.

Regular weekly or monthly reports were issued from the different groups at Clinton Laboratories during this period. Reference to the bismuth phosphate process documents, noted above, is found in the September report (43-9-70) of the Chemical Engineering Section. Likewise, the Separations Development Group, under Greager, devoted part of its weekly report to this issue and noted in an October report (43-10-168) that the disposal tank (probably the one outside the 205 Building) is "filling very rapidly" and additional space will be required. A notation is made of the activity level (" 10^{-5} curies/cu.ft.") in cooling water from a set of experiments; the fact that this cooling water was sent "directly to the open settling basin" suggests that it was determined to have a low enough activity level to dispose of in this fashion. Greager's reference to the "settling basin" had to mean the retention ponds, because the basin was not yet constructed. This document, and others later in the year from Greager, raise uncertainties as to when liquid wastes were first released to the ponds and creek; later accounts clearly state that this did not occur until March of 1944. Even though irradiated slugs of uranium from the graphite reactor were not received in the 205 Building until mid-December, significant activity was underway; this separations facility was being used to treat irradiated uranium from the two small experimental piles at Chicago in order to recover minute amounts of plutonium for characterization and experimental work. Thus, liquid wastes were being generated.

The issue of proper disposal of the gaseous wastes, emitted primarily from the 205 stack, consistently appears in the early documents but much less frequently than does the issue of liquid wastes; reference to gas disposal is made in Kirst's Process Manual. A very early reference appears in a January 25th memorandum entitled "Site X Design Schedule" (43-1-26) from Kay to R. M. Evans (1898-1986, a du Pont chemist and manager who later managed Hanford). Kay states, "Dr. Whitaker mentioned the possibility of off-gas accumulating at the top of the stack to the point of being dangerous. It was pointed out that further consideration of disposing of this gas or controlling it in some manner is needed." Another early letter, dated January 30, from Whitaker to Compton, in which a January 28 letter from Greenwalt is included (43-1-12), acknowledges that "a substantial percentage of the fission activity will be released in the air . . . at the top of the stack." Estimates of the amount are requested. Assorted documents acknowledge that the gases could be harmful, but because those gases

that could enter the human body and cause harm (i.e., ^{131}I) were relatively short-lived, proper dispersal at a high elevation was deemed adequate disposal. "Cooling air gases leaving the Pile will be continually monitored and if abnormally high activity is found, the Pile should be immediately shut down," R. B. Smith, Secretary of the Central Safety Committee, wrote to Whitaker in April (43-4-193); dilution factors for the 205 Building emissions were approved in a February letter from (Lombard) Squires to Daniels (43-2-40).

Rather sophisticated calculations were performed to establish safe ("tolerable") levels for a variety of nuclides emitted to the atmosphere from tall stacks; for instance, a memo from K. (Katherine) Way and J. (John) A. Wheeler to K. Wyatt (43-4-196) presents tolerance concentrations for radioisotopes of xenon, oxygen, iodine, argon, and nitrogen. Wheeler (1911-) had joined the Met Lab group from Princeton, where he had worked with Niels Bohr on fission theory, and is the scientist who first proposed the term "moderator" for the material in a pile that slows neutrons down; his doctorate in physics was from Johns Hopkins University in 1933. After the war, Wheeler worked at Los Alamos and returned to Princeton. Way (1903-) had worked with Wheeler at the University of North Carolina, where she had received her doctorate in physics in 1938, and came to Oak Ridge in late 1942 from the University of Tennessee, where she had been since 1939. She was instrumental in the establishment of the Oak Ridge Institute for Nuclear Studies, now Oak Ridge Associated Universities (ORAU), and worked for the National Bureau of Standards and the National Research Council after the war, thereby continuing involvement with ORNL. In 1968, she took an adjunct professorship of physics at Duke University; she retired in 1988.

The Medical Department had previously (February 11) requested "that provision be made for sampling the off gases and the ventilating air at Site 'X,'" as reported in a letter from Squires to Daniels (43-2-39). The earlier referenced memo (43-4-156) contains a rather detailed description of the equipment to be used for monitoring the stack gases. There continued to be sporadic documents dealing with atmospheric monitoring, along with meteorologic studies of the Clinton site related to atmospheric circulation patterns. Efforts were directed at ensuring safety; an example is the "problem assignment," Disposal of Stack Gases at Site X (43-11-27), submitted by M. (Miles) C. Leverett. The stated reason for undertaking this research was "to make certain that the plant creates no personal hazard through atmospheric pollution." We shall hear a lot from Leverett (1910-), who came to Clinton Laboratories after heading the Engineering Group at the Met Lab in 1942. He had worked at Humble Oil Company, had participated in the early selection of the Tennessee site, and had worked for Compton and Wigner in the design of early pile reactors, having been hired by Compton at age 32 with ten

years of industrial experience. Leverett, a native of Illinois, held a Sc.D. degree (1938) in chemical engineering from MIT.

THE SACRIFICIAL "LAMBS"

In a definitive memo to Doan, dated November 2, from H. (Howard, "Bim") J. Curtis, Chief of the Biology Section and one of the first to study radiation effects on mice (43-11-44), the issue of biological monitoring of wastewater is suggested:

The waste water from the plant will flow into White Oak Creek, and at the point of entry into the creek it should be radiologically speaking, fit for human consumption. The Creek flows into White Oak Lake which will constitute an additional holding pond. The lake empties into the Clinch River, so the lake water will be many times diluted by the river water. The river water will be used as the water supply for several towns below the plant. In view of the uncertainties involved, both from radiological and heavy metals points of view, it is felt that it would be desirable to biologically monitor the water in the lake.

Curtis goes on to discuss the unknowns in establishing "tolerance concentrations" for elements in drinking water, pointing out that the heavy metals present an uncertain situation, "and it may well be that this is a greater hazard than the radiation." Tolerance doses will be determined from the biological monitoring program, "but it may be twelve to eighteen months before these data are complete." He proposes feeding pond water to 12 rabbits and 30 mice, taking blood counts periodically; the animals "will be sacrificed from time to time and carefully autopsied, both microscopically and radiologically." He notes that "all necessary materials are on hand" and that the experiment should continue for a minimum of 2 years. The records suggest that this proposal soon surfaced again. Howard Curtis (1906-1972) had received his Ph.D. in physics from the University of Michigan, his home state, in 1932 prior to teaching at Johns Hopkins and Columbia Universities. During the war, he was in charge of the Biology Section at Clinton Laboratories; after the war, he returned to Columbia for a year, then accepted the position as Head of the Physiology Department at Vanderbilt University Medical School. In 1950, he became Head of the biology program at Brookhaven National Laboratory. An eminent biophysicist, Curtis was a member of numerous national and international panels and review groups.

FOR SAFE DISPOSAL INTO THE RIVER

The handling of wastes from the separations plant (the 205 Building) remained a topic for careful study and thought. For example, on

November 6, Leverett, Chief for the Engineering Development Section in the Technical Division, wrote to Doan (43-11-79) and suggested three different methods for disposal, pointing out that the 14,400-gal waste tank at the building had capacity for about 6 months more waste and that the activity of the waste is "about 5×10^{-11} watts per cc, or about five times the tolerance activity." He points out that

The contents of the tank could be discarded into the river over a 16-day period without exceeding either the tolerance activity of the river water of 10^{-11} watts per cc or the tolerance concentration of uranium in drinking water of .1 ml per liter. [Note that this value of ".1 ml per liter" had to have been a typographical error in the original memo.]

Nevertheless, he goes on to recommend that the wastes be pumped into the gunite tanks in the 206 area because the wastes are volumetrically of small quantity. In a weekly report (November 20) for the Separations Development Division (43-11-152), Greager notes that analysis of a supernatant liquid in the disposal tank (presumably the one at the 205 Building) "is underway to determine whether it can be pumped out and discarded into the nearby creek. It is necessary that this waste tank capacity be recovered at this time if it is possible since the main plant waste tanks are not yet available." He follows with a December 13 report (43-12-108) in which it is noted that "the supernatant liquid, having an activity of about 0.10 r./24 hrs., has been sent to the retention pond"; this level was within the discharge limit defined in the Process Manual. Recall the earlier reference to Greager's notation about release of liquids to the ponds, and note the discrepancy in the dates.

The gunite tanks were, without question, the centerpiece for the waste handling activities and, without their proper functioning, activities could not proceed. They were part of the original design of the entire waste processing system (note previous discussion of the flowsheets and Process Manual) and their construction was carefully planned and undertaken; they were included in a January 25th "drawing list" (43-1-59); cover thicknesses were specified in April in a memo from Pratt, (per Vaughan) to Daniels (43-4-61). In an October 1943 report from Whitaker to Compton, entitled "Progress on Construction and Operation of Clinton Laboratories" (43-11-118), the status of the tanks is discussed:

The storage tanks for storing of process wastes have been completed according to original design and found to be watertight after testing. After the tests started a decision was reached to line the tanks after they had passed all inspections with a bituminous lining flexible enough to spread over hair-line cracks. This extra lining is to be covered over with an inside Gunite layer for the protection of the bituminous coat. In our

opinion, the original design of the tanks, plus the special lining which is being put in, serves to make the tanks as safe for their purpose as it is possible to make them.

This not only establishes safety as a concern, but it also raises a curious issue because Kirst's Process Manual, which was prepared earlier in the year (perhaps February), clearly showed that the "Gunitite storages painted internally with bitumastic paint" were planned in advance; yet Whitaker's communication suggests that the addition of the linings was an afterthought. Whitaker goes on to state, "Investigations have been initiated to determine the effect of heavy blasts on the tanks." These blasts were from the rock quarrying activities "six miles from the plant" (presently Kerr Hollow and Rogers quarries). He notes that seismographic equipment is being acquired to measure the amplitude of the vibrations at the plant but that his "experienced opinion is that the amplitudes resulting from the blasts are well above the value which might cause trouble." Clearly the importance of the gunitite tanks for the ultimate success of the Clinton mission was appreciated.

One of the most complete and detailed early documents dealing with the issue of properly handling the liquid wastes is entitled "Separation and Recovery of 49, Clinton Laboratories, Waste Disposal," by M. (Marshall) F. Acken, dated December 4, 1943 (43-12-55); Acken was in charge of Section I of the Separations Development Division and reported to Greager. (The term "49" was used to refer to ^{239}Pu , "25" was used to refer to ^{235}U , and "29" was used for ^{239}U .) In this report, Acken describes in considerable detail the assignment of different types of liquid waste to certain tanks and the sequence in which each tank will be filled and emptied. The objective was to hold the waste as long as possible to allow decay of activity; he states:

With this decontamination storage cycle, the minimum hold-up of decontamination wastes is ca. 225 days, during which time the activity will have decreased twenty-fold, therefore requiring at most 250 gallons per minute of dilution water for safe disposal into the river.

Acken describes seven different types of liquid waste (other investigators assigned different categories later) and proceeded to indicate the prescribed handling procedure for each. Repeatedly, in this nine-page (plus table) document, reference is made to assuring that the activity is low (by monitoring) "before discarding into the river." He presents calculations of the dilution factors required in order to lower the activity to acceptable levels "for safe disposal into the river." This report quite definitively demonstrates the fact that safety was a dominant concern in waste handling and disposal. Acken (1905-1985), who held a doctorate in chemistry from Pennsylvania State University (1929), had been with

du Pont and also later worked at Hanford; after the war, he remained at Hanford in a managerial position with General Electric.

RISING TO THE OCCASION

The year, 1943, drew to a close with added attention to human health. On December 30, a series of new problem assignment ideas dealing with "biological studies" and their application to Hanford's needs was issued by Doan's office (43-12-496). Although most of these proposed studies dealt with metabolism of fission products and exposure to radiation, one of them had as its objective "to make regular checks of the safety of the water in White Oak Lake from a human consumption point of view by feeding the water to animals under observation." It is pointed out, "This is of definite interest if the results can be translated into Hanford conditions as regards exposure, materials in solution in water, etc." Apparently, the Curtis proposal of November 2 for biological monitoring survived at least to this point, but it is still uncertain if the proposed experiments ever took place. Numerous other examples exist of Clinton Laboratories knowledge on waste handling that would be transferred to Hanford. For example, an early request was made to Doan by F. S. Chambers of the Technical Division of du Pont (43-11-56) to "evaluate the stability of this (carbonate) complex at its boiling point over a period of at least several months," expressing concern over the long-term storage of boiling liquid wastes at Hanford. Many more references to disposal at Hanford will surface.

A significant position was established on the 20th of December, 1943, in a memo from Simeon T. Cantril, Director of the Medical Division, to Whitaker, Laboratory Director (43-12-180). Cantril (1908-1959), with an M.D. degree from Harvard, was a recruit of Compton's to work at the Met Lab, having been Head of the Radiology Department at Swedish Hospital in Seattle after working at the Radium Institute in Paris (1936-1937) and at the Tumor Institute of Chicago; he came to Clinton Laboratories early in 1943, after working with Ernest O. Wollan on the study of radiation exposures to employees at the Met Lab. He writes, "This is a statement of the procedure which the Medical Department would like to formulate for the responsibility of emptying the waste-storage ponds and a lake backed up by a dam on the White Oak Creek." Cantril says that the Medical Division (called a "Department" earlier) would "hold the responsibility" for releases of water into the Clinch River after the activity had been determined and for inspection of the dam. Cantril indicates that locks will be placed on the dam structure so "it is not opened inadvertently by others." A report was to be issued to the Medical Department by Mr. Vaughn (Frank Vaughan) when the waste ponds were drained into the creek "so that we may have a record of the waste disposal into White Oak Creek and eventually into the Clinch River." A fence was requested

around the ponds to help control access. Finally, Cantril proceeds to say that a tolerance level ("100 mr/24 hr," a limit specified early in the year) must be adhered to and that if this level is exceeded by the pond water, no release will be authorized. This proposed responsibility was accepted by Whitaker and documented, apparently in correspondence from A. J. Schwertfeger to Wallace, January 26, 1944 (document unavailable). Frank Vaughan, born in Massachusetts in 1911, was a chemical engineer educated at MIT; he had worked with du Pont prior to joining the Met Lab, came to Clinton Laboratories in 1943, then transferred to Hanford where he was a chief supervisor in the separations plant. After the war, he returned to du Pont and retired in 1973.

Obviously, considerable attention had been directed by the end of 1943 to the proper management of the liquid waste and the protection of the public health. As the year of construction ended, not only was the major phase of construction completed, but the graphite reactor had gone critical almost 2 months earlier, and the separations plant had begun operation in December. With regard to liquid waste management, sophisticated process steps had been defined, release limits established, the importance of monitoring before (and after) release was understood, lines of authority had been drawn, and responsibilities for protection of the public health had been demarcated. The individuals who had stepped forward in establishing the waste management procedures were prominent scientists, engineers, and medical professionals in positions of authority. This point in time was still several months before any waste materials were to be released to the creek, on the basis of most accounts. In closing the year, it is evident that work schedules for all involved were tight and holidays were rare; on New Year's eve of 1943, Stone wrote to Compton a thoughtful letter (43-12-308) on "Health Physics at Site W" (the designation for Hanford) in which he expressed concern over the expertise available for the challenges ahead.

THE YEAR OF PRODUCTION

BACKGROUND VALUES

At the Met Lab, Glenn Seaborg's journal notes for January 1, 1944 (Kathren et al. 1994) reveal his anticipation and excitement as the new year unfolds:

The beginning of 1944 finds our Project deep in the problems of plutonium production, extraction, and purification. This vast involvement with a secret, synthetic element unheard of not much longer than two years ago and unseen until sixteen months ago in August 1942, would seem incredible to the outside world. Moreover, the means of producing plutonium in copious quantities—the chain-reacting pile—became operational just one year ago. I thought about these matters today when we received our first shipment of plutonium from Clinton Laboratories—1,500 micrograms! It equals almost the total amount of plutonium produced by all previous cyclotron bombardments. It is hard for me to remain nonchalant when I realize that before the end of February, production of plutonium will increase a thousandfold and gram quantities will then become available.

At Clinton, the year, 1944, started with the establishment of the first burial ground. It is relevant here to review the directive of January 5, 1944, (44-1-16) establishing the "burning ground" adjacent to White Oak Creek on the southern side of the Clinton Laboratories site for "disposal of actively contaminated broken glassware or materials not sufficiently clean to be used in other work." A brief memo from Cantril to R. B. Smith authorized this action. In stark contrast to the attention given to the liquid wastes, no reference is made to health or safety issues associated with this disposal decision and, indeed, it appears that the responsibility for this rested elsewhere than with the Medical Department, although Cantril does say that disposal of larger equipment should be taken up with the Health (not referred to as "Medical" here) Department. Quite obviously, little, if any, attention had been given to this disposal need, and it was evident that the understanding we have today of the magnitude of risk associated with low-level contaminated solids was not known at the time. The records reveal very little correspondence related to this burial ground; in one memo, dated April 27 (44-4-82), Greager issued a statement relative to the procedure for placing solid active wastes in cans at the 706A Building and to the transportation of the material to the burial ground. He does note, with regard to disposal of "heavily contaminated materials," special arrangements should be made to "make sure these operations are carried out safely." The safety issue here, however, is certainly one of worker exposure during the disposal operation, rather than one of preventing releases.

Simeon Cantril quickly began to exercise his authority. On January 7, he wrote (44-1-22) to Frank Vaughn (Vaughan) regarding proper disposal of "hot" material poured down "hot" drains in the Chemistry Division (706A Building); Vaughn was Assistant Superintendent for the 200 area and reported to Kay. Cantril writes:

It would seem to me however that in case of any proposed large dumping of wastes into the hot drains of 706A, which lead to the tank on the west side of the building, you should be notified of same so that either monitoring can be done before it is allowed to go to the cooling ponds or the present diversion to cooling ponds be held up until such monitoring is made. As I understand it the monitoring of this particular tank is under your jurisdiction.

It would seem to me that you and Mr. Johnson should have some formal understanding of the waste problem from 706A as it concerns the monitoring and emptying of the cooling water ponds or large waste tanks. Perhaps this has already been done but if so I am not familiar with the set up.

In a carefully crafted "P.S." to this polite admonition, Cantril says Mr. L. S. Parker has a very excellent map of the disposal system for 706A "in case you should need it for reference. S.T.C." The reference to "Johnson" was almost certainly to Warren Johnson, Division Director for Chemistry. Cantril had to follow up later in the year with regard to disposal in 706A stating in a memo to Doan (44-5-3), "it is becoming increasingly evident that the present system of waste disposal of hot material in 706-A is inadequate and is eventually going to run us into trouble."

The first few months of the year saw a diversity of activities related to the disposal issues—all the way from recognition of the need for instruments to waste minimization and monitoring. On February 2, Karl Z. Morgan, of the Health Physics organization, reported to Herbert M. Parker, Section Chief for Health Physics, the results of background counting rates for water from White Oak Lake (44-2-221); this was over 1 month before wastes would be released to the creek system, but they had, of course, been accumulating in the tanks (note the uncertainty discussed above about release in October of 1943). Morgan wrote, "the conclusion is that the contamination of the water is extremely low and too low to detect with a counter which can determine contaminations rather accurately as low as 3% of tolerance." Although only a few samples were taken, this foresight for establishing background levels is commendable.

It is interesting to note that Herbert Parker was a physicist from England who was attending a five-month school here to become knowledgeable of

health physics; he and about 15 others were being trained as part of Hanford's initial health protection and instrumentation team, arriving there later in the year, where he developed a long and very distinguished career in health physics (Kathren et al. 1986). At Hanford, he headed all health physics activities and, after the war, became Assistant Superintendent of the Medical Department and then Manager of Hanford Laboratories from 1956 to 1965; after General Electric left Hanford in 1965, Parker remained as a consultant to Pacific Northwest Laboratories until 1971, when he retired and formed his own company. Parker (1910-1984) had been educated at the University of Manchester (M.Sc., 1931) and did early research on radium therapy. In 1938, he joined Simeon Cantril to work on tumor therapy at the Swedish Hospital Tumor Institute in Seattle; it was Cantril who, in 1942, convinced Parker to join the Met Lab, which he did even though he was still a British citizen. Morgan (1907-), a North Carolinian, had arrived at Clinton in September of 1943, having completed his Ph.D. in 1934 on cosmic radiation at Duke University, prior to heading the physics department at Lenoir-Rhyne College and then working with Compton at Chicago; he spent the rest of his career at Oak Ridge, becoming head of the Health Physics Division in 1944 and retiring from ORNL in 1972. Morgan was instrumental in formation of the Health Physics Society (1955) and the International Radiation Protection Association (1966) and served as the inaugural president for both organizations.

F-PRODUCTS AND FISH

The issue of instrumentation for measuring activity in water and support for Hanford was raised at the end of March by Stone in a short memo to Doan (44-3-387). He states, "I wish to bring to your attention that to date a satisfactory instrument for calibrating the intensity of radiation in the water of the holding ponds has not been developed." He complains that the design of the instrument has been "in the hands of the Instrument Shop, that is 717B" for "several months." He emphasizes that Hanford will badly need this "apparatus" by June 30 and notes, "Each time a new list of dates of completion of apparatus comes out, the date of completing this is put off."

Reduction of the volume of waste, as well as lowering the activity in the waste ultimately released, were objectives that occupied the efforts of staff. In March, G. (George) E. Boyd proposed a problem assignment (44-3-254) for treatment of the wastes from the plutonium extraction system; the objectives of this chemical research were not only "much smaller waste volumes," but also removal of "by-products," presumably for recovery and reuse. Boyd (1911-), a native of Indiana, had been a professor at the University of Chicago, where he received his Ph.D. degree in physical chemistry (1937); he took a leave to join the Met Lab, where

he headed the Analytical Chemistry Section of the Chemistry Division working on detection of impurities in graphite and uranium. At Clinton, he headed a Section (C-III) devoted to development of analytical methods for alternate separations; late in the 1940s, Boyd became an Associate Director of the Chemistry Division and, in 1973, he accepted a position at the University of Georgia. Boyd's proposal was submitted to Warren C. Johnson, who had been a supervisor for plutonium chemistry work at the Met Lab, having temporarily left his professorship at the University of Chicago, and who was the Division Director for Chemistry at Clinton Laboratories. Johnson (1901-1983) was a native of Michigan and held a doctorate in chemistry (1925) from Brown University. Johnson forwarded the memo the next day to Doan with the recommendation that it be presented to the Steering Committee (44-3-222), adding that "This work is underway; as a matter of fact, the greater part of the program has been completed." On the bottom of this transmittal is a hand-written note from "RLD" (Doan) back to Johnson chastising him mildly for having done the work before approval. After the war, Johnson returned to the University of Chicago, but agreed to serve temporarily as Clinton Laboratories Director at the end of 1947 when there was difficulty in filling the position, and he is shown in this role on an organization chart. He never did fill the position, however; just before New Year's eve of that year, the AEC directed that all reactor research would be done at Argonne National Laboratory, and Johnson chose to stay at Chicago, where he eventually was named Emeritus Vice-President, retiring in 1967. He also served on a prestigious four-member committee, appointed by General Leslie Groves, to review the declassification of research results from the Manhattan Project.

An interesting and foresightful item is documented in a March 17 communication (44-3-267) from Stone to K. (Kenneth) S. Cole, who had been a biophysicist at the College of Physicians and Surgeons at Columbia University and who held a doctorate in experimental physics from Cornell University (1926). Cole had apparently moved to Clinton early in 1943 after establishing the Health Division of the Met Lab in 1942. Cole (1900-1984), a native of New York, whose research was directed at the toxicology of radioactive substances, had written Stone regarding the "distribution of radio-active materials in fish." Stone states: "Extension of work now being done and partially completed include the study of mixed F-products and the study of separated F-products on fish." He goes on to discuss the eating of fish, asking:

I wonder if you took into account that most of the activity is in the scales, skeleton and gills which are never eaten and considerably more is in the skin which is seldom eaten. For purposes of discussion as to whether fish would be above tolerance, we would need to know the exact amount in the

muscles which are the only organs eaten, with the exception of fish eggs.

Stone concludes with reference to experiments in Washington State, which "are being very satisfactorily carried out" on salmon. Results of this work are unavailable at present within the ORNL records, but the significance of it can not be overlooked. Gerber (1992) describes work done at the Applied Fisheries Laboratory at the University of Washington in 1943 and at Hanford on effects of radiation on fish, supported by the federal Office of Scientific Research and Development. After the war, Kenneth Cole was a professor of biophysics and physiology at the University of Chicago (1946-1949), Director of the Naval Medical Research Institution (1949-1954), with the National Institutes of Health until 1980, and then joined the Scripps Institute of Oceanography.

TECHNOLOGY TRANSFER TO HANFORD

By most accounts, wastes were first released from the tanks on March 6. In early April, W. (Waverly) Q. Smith, Section Chief in the Technical Division, wrote to Kay (44-4-47), pointing out that when the wastes from the separations plant (205) holding tanks were mixed with the pond water, a precipitate formed, which carried a large part of the activity out of solution. He hypothesized that the calcium in the water reacted with fluoride or phosphate in the waste to form this precipitate and pointed out that laboratory tests had shown that addition of calcium chloride would greatly reduce activity in the waste. He therefore proposed the addition of 500 lb/d of calcium chloride to the tank waste, indicating a decrease of activity by a factor of "5 to 10 in a week," saying that, "this procedure should greatly decrease the amount of active material being discarded and permit storage of only the concentrated sludge rather than the dilute solution." On April 12, the proposed action was forwarded (44-4-54) by Kay to Doan pointing out that this would "reduce the activity in the plant wastes discharged to White Oak Creek," and stating, "unless advised to the contrary, we plan to proceed with the proposal as outlined by Mr. Smith." The suggestion was implemented. We shall revisit this incident later in more detail. Waverly Smith (1912-1989), born in Virginia, held an M.S. degree in chemical engineering (1934) from MIT and had been with du Pont throughout his career.

At this same time, J. (John) N. Tilley, of the Manufacturing Division of du Pont, wrote to Whitaker (44-4-202) after a visit to the site and suggested "the possibility of using caustic soda for neutralization of the excess mineral acid in the metal waste." Tilley was a liaison officer at du Pont with the Explosives Department. His objective, clearly stated, was "to save us a considerable amount of metal waste volume under conditions at Hanford operations," and he asks that "Major Greagor's

group test this possible variation and if it looks favorable from the point of view of the semi-works" (Clinton Laboratories were referred to often by this term and this must have been a reference to Greager, whose name was frequently misspelled), "it might be desirable to have it confirmed by a change in the 205 building." He concludes, "if it is successful, it would be a very desirable change to be introduced in the next Hanford flow sheet revision." Incidents such as this, and others discussed elsewhere, are clear testimony to the environment that existed at the time—that of learning the processes as work proceeded and that of making decisions in the best way possible at the time. The issue of proper neutralization of the wastes had arisen earlier. Late in the previous year, Acken had proposed a problem assignment on neutralization during waste disposal and was informed of its approval by Doan (43-12-457). A progress statement appears in the Separations Development Division Monthly Report for March (44-3-273), and in April of 1944, a recommendation was made by Acken that a change be made in the neutralization procedure "and if successful be adopted as a standard procedure" (44-4-152). The use of the Clinton site for development of waste handling at Hanford is also demonstrated in a May 8 memo from R. (Richard) S. Apple to Kay (44-5-177) in which Apple suggests that the neutralization steps for Hanford wastes, which use NaOH and Na₂CO₃, resulting in a "significant reduction (about 25%) in the volume" be demonstrated "in the Clinton plant as soon as the change can conveniently be made."

Acken's work was apparently incorporated into the Hanford operations, as indicated by a June communication there (44-6-531) with copies to Clinton Laboratories. Squires writes about neutralization of the wastes from the 224 Building (the concentrations building in the separations area) and disposal into the 35,000-gal settling tank at Hanford, saying, in reference to data received from Acken, it "will materially reduce the hazard of discharging the wastes into the ground." Disposal of wastes from this building had earlier presented a problem, as stated by Squires in May (44-5-690):

The simplest solution of the problem would be to discharge the effluent from the 224 Building settling tank directly into the ground without dilution and in such a manner that the disposal system to the ground is completely covered at all times. It is probable that any product in the effluent discharge to the ground would be rather tenaciously absorbed in the ground and would not migrate. Tests to check this point both for product and by-product activity are under way at Clinton.

Squires describes also the experience at Clinton, referencing wastes from the 205 Building, with regard to formation of a precipitate that "settles out as a slime, in part in the retention basins and in part along the

bottom of the stream into which the wastes are discharged," and proceeds to say, "since the composition of the 224 Building wastes is quite similar to the low activity wastes discharged from the 205 Building at Clinton, and since the composition of the cooling water in the two places is also alike, a similar situation will probably develop at Hanford." His reference to "cooling water" is in the context of "dilution" water. He speculated that drying of the contaminated soil

will result in a serious dust hazard since sludge can be expected to dry and be picked up by the same winds which are responsible for the dust storms. This situation unquestionably represents a serious hazard and necessary design changes to overcome it should be made.

Lombard Squires (1906-), originally from New Jersey, had studied chemistry at the University of Kentucky (B.S., 1929), been an instructor at MIT, and joined du Pont in 1939. He served in a liaison role between the Met Lab and Clinton through late 1944, then transferred to Hanford. After the war, he returned to du Pont where he held various supervisory positions until he retired in 1969; he served on important AEC committees.

PRECIPITATED ACTIVITY PRECIPITATES ACTION

A definitive report, "Handling of Wastes from 205 Building," authored by Waverly Smith, was issued in early May of 1944 (44-5-101). He followed this report with a summary and updated letter to Whitaker on September 2 (44-9-54). The initial 15-page report contains the greatest detailed summary up to that time of the actual handling of the liquid waste that has been found, and the September letter complements it by adding considerable other detail. The report reviews the processing steps and presents data on the chemistry, volumes, and activities of the different types of liquid wastes. These two documents form the foundation for more fully understanding how certain decisions were made and for placing certain events in proper context. The following discussion draws on both.

Initial storage capacity for 1 year was planned for the waste on the basis of the use of the lanthanum fluoride separations process, but when the process was changed to the bismuth phosphate process in June of 1943, storage capacity shrank to 6 months because the new process generated much more waste than did the old one; the construction of the gunite tanks had progressed far enough that it was not practical to alter construction plans. However, it was felt if sufficient solids could be precipitated from the liquid waste that "it might be possible to dispose of the supernatant from the chemical wastes, thus increasing the life of the installed tanks to well over a year." In February of 1944, analyses of

the supernatant in the tanks demonstrated that "dilution with the available plant waste water to below tolerance levels would allow disposal of wastes at the rate of input into the tanks." This was calculated to be 25,000 gal from the tank to the ponds per day without exceeding the tolerance limit of 5×10^{-4} curies/cu. ft. of water." The discharge rate of "5,000 gallons per day seemed amply safe since further dilution would occur in White Oak Creek and the Clinch River." The first discharge from the tanks to the ponds was on March 6; although not specifically stated, Smith's description clearly implies that on or about this date discharges to the creek also occurred and "samples were taken of each pond before discharge to the creek in order to ensure that a body immersed in the water would not obtain more than .1 r in 24 hours."

Waverly Smith proceeds to describe observations following the discharge and says that within a few days "it was observed that a precipitate was collecting in the ponds and that a large fraction of it washed into White Oak Creek when the pond valves were opened." On March 15 and 16, samples from the pond bottom and exit ditch were taken and were found to have high beta (27,300 and 32,500 beta counts/5 cc, respectively) and gamma (900 and 410 counts/5 cc, respectively) activities; it was concluded that a large part of the activity was in the precipitate. Because the precipitate was being carried into and down the creek, water samples of the creek were taken (4—8 beta counts/5 cc) that same day (March 16). Laboratory tests conducted on March 18—25 "demonstrated that the addition of calcium chloride to the waste tanks would decontaminate the wastes by a factor of about 10." After review, approval was given for the addition of calcium chloride to the waste tanks, which started on April 17 and continued until April 27.

This activity ceased on April 27 because on the prior day (Smith incorrectly says three days prior) Roy Overstreet and Louis Jacobson, soil scientists, conducted a survey of mud in the White Oak Creek system (Overstreet and Jacobson 1944) and found "the material was found to be strongly active at a number of points and measurably active at the mouth of the creek where it empties into the Clinch River." Smith says:

if the activity found in the creek bed were of a species which was rapidly absorbed in the body or was one which had a long half-life, then continued discharge into creek might create a hazard. The elements responsible for the activity in the mud were not known nor were tolerance limits for the possible elements available.

On the basis of these observations, it was immediately proposed that a settling basin be constructed between the ponds and the creek "to effectively remove all precipitated solids." The holding time in the basin

was estimated to be about 30 hours and an "experimental program is underway to confirm this and the other bases for the design of the new pond." Smith continues in his May 1 report:

Since the remaining storage tanks can handle the chemical wastes for only two more months, design and construction of this settling basin will proceed without waiting for completion of all of these confirmatory experiments. It seems reasonably certain that the total activity leaving such a pond would not exceed one curie per day when handling wastes having the level of activity of the present materials. It is quite possible that the discharge can be held to a much lower volume than this by the introduction of additional adsorptive agents.

As shall be discussed later, the settling basin was constructed and put into operation on July 3 of that year.

In the September letter to Whitaker describing this sequence of events, Smith details some thoughts on discharge limits. He points out that the elements of most concern in the pond precipitates are barium, strontium, tellurium, and cesium, but that they represent "only a very small fraction of the total activity." He states:

J. G. Hamilton (letter to S. T. Cantril, 6/20/44) estimates that on the basis of this composition a tolerance limit of 1 curie per day would probably be a safe rate of discharge to White Oak Creek. H. M. Parker seems to think that this is conservative and that a tolerance limit of 5 curies might be permissible under certain conditions. This is about the average rate of discharge during the period of March 6 - April 27 when the active wastes were being added to the ponds.

Reference to the discharge limits suggested by Hamilton and Parker is mentioned again soon.

Thus, in summary, it is shown that as soon as this problem with the formation of the precipitate appeared, action was taken to characterize the situation and to alter the chemical processing of the waste to alleviate the problem. Once there was detailed knowledge of the extent of contamination of the creek system, immediate steps were taken to cease releases and to construct a new settling basin to correct the insult. Medical guidance, as we shall see later, continued to play a role in establishing release limits to protect the public health.

MONITORING ON THE INCREASE

During the latter part of May, a meeting was held to plan for further reduction in the activity of chemical liquid wastes from the separations

plant; present were Apple, Leverett, M. (Merlin) D. Peterson, Waverly Smith, Vaughan, and J. T. Weills. The minutes of the meeting, in the form of a memo from Smith to Kay (44-5-335), indicate that the settling basin is under construction and it is anticipated that only "500 to 1000 millicuries per day" will flow into White Oak Creek. All agreed that "a means should be sought for lowering this as much as practical," although unknowns prevent a more specific tolerance limit from being set. A "program" was agreed to. Leverett's group was to "continue settling tests" to "be sure that no unusual conditions will arise which will result in unsatisfactory removal in the new settling basin of precipitated solids." Apple's group was to seek ways to "decrease the gross activity of the basin overflow," using adsorptive agents (kaolinite, bentonite, zeolite, resins) or precipitating agents (i.e., water glass). It was noted that the "sludge" found below the ponds was enriched in silicon, aluminum, and fluorine, "suggesting that aluminum fluorosilicate might be the principal decontaminating agent." Finally, "this group" (it is uncertain which "group" is referred to)

will find what radioactive elements remain in the basin overflow and determine what happens to these on the further dilution which will occur in White Oak Creek and Clinch River. With information on the maximum concentration of each radioactive element in the water and in the precipitate formed by further dilution, it should be possible to decide first if there is really any hazard in discharging the fraction of a curie per day, and second what specific elements need to be removed.

On May 23, Whitaker sent a letter, "Waste Disposal from Site X" (44-5-360), to J. (Joseph) C. Hamilton of the Crocker Radiation Laboratory at the University of California in Berkeley. Hamilton (1907-1957), born in Massachusetts, served as a medical advisor to the entire Manhattan project. He worked with Seaborg in his search for new heavy elements by using the medical cyclotron at Berkeley for irradiation of plutonium; his formal training was through the Medical School at San Francisco, where he received an M.D. degree in 1936. Hamilton had visited the Clinton site earlier in the month. Whitaker points out that the new settling basin was under construction, although there had been some design changes since the visit, and that it should be ready in July. He describes some laboratory tests on establishment of dilution ratios for the waste and plant cooling water (1:35) and the formation of a precipitate, which removes "all but about 10% of the beta activity in the active waste." Whitaker points out that the supernate from the settling tests has an activity of only some "0.3 curie/day" when discharged. He proceeds to describe percolation tests on soil from the settling basin area designed to determine the hydraulic gradient ("0.07 cm/day"), saying, "This rate is so slow that there seems to be no reason for concern about the possibility that activity will seep through the walls of the basin." He continues with

reference to "the water that did seep through the bed of earth," pointing out that if all the wastes were treated (in the basin) "discharge from the plant would be reduced to approximately $60\mu\text{c}/\text{day}$." He presents analytical data on the now-famous precipitates and concludes with, "We are awaiting receipt of the results of the fission assay and a statement of the tolerable quantities of the materials found in the water discharged from our plant." Earlier in the month, Whitaker had written Hamilton about the distribution of fission products in the waste (44-5-128), summarizing the amount of waste generated per day ("5,000 gals/day") and the general handling procedures. Concern over the proper management of the liquid waste evidently received attention at the highest level.

As indicated earlier, there appears to be only sporadic documentation of issues related to disposal of solid wastes and, early in the year, the responsibility for tracking their proper disposal was unclear. In late May, Cantril wrote to Schwertfeger, who had been involved with the burial ground since its inception and who was in charge of Works Engineering, to report on discussions that had occurred in the "Activity Hazard Committee" meeting (44-5-402). It is interesting to note that, although passing reference is made to personnel safety (implied for exposure), there is still no consideration of dangers associated with release of contaminants. Because of access problems to the existing burial ground, Cantril suggested that a new one be sited "on the East end of the plant within the fence, which would obviate this necessity" (he refers here to the problem of getting a Material Pass from the Army Office to get to the existing burial ground) "and perhaps allow a larger area in which to place material later needed." The level of responsibility assumed by the Health Department and Cantril for the solid wastes at this time is still uncertain.

In June, while the settling basin was under construction, attention focused again on activity in the White Oak Creek system and its drainage off-site. Cantril summarized recent data on "mud" in the system moving from the holding ponds into the Clinch River (44-6-311), referencing some 19 samples of sediment and 7 of water taken from April 26 through mid-June and presenting the data in counts per gram; activity was found in the river sediments, although no special mention of this was made. In this report, data from Hamilton are also included for four of the sediment locations with specific elements (Ba, Sr, Zr, Cb, Ce, Y) noted; there are also data for a variety of nuclides for the overflow from a gunite tank (W-6), "pond precipitate," and "pond overflow." Cantril notes the cessation in discharges on April 27, saying, "the activity in the clay would seem to be reduced by a factor of 10 to 100 in counts per minute when one compares the analyses taken about two months apart. Further analyses will be made on both the water and clay in the

drainage system when the new disposal system begins operation." Another survey, "Activity of Mud from Pond Exits and White Oak Creek," was reported on July 7 by P. L. Eisenacher to Morgan (44-7-158). A total of 15 samples were collected from throughout the creek system, including downstream from the dam; activity was found in all samples. Eisenacher concludes:

It will be noted that these results vary considerably and do not agree with the last results all of the order of $10^{-5}\mu\text{c}/\text{gm}$. This may be due to change in activity of the mud caused by discharge of additional active material and location of samples, in the main, although numerous other factors are no doubt involved.

On June 16, Herbert Parker wrote Cantril in reference to "Radiation at White Oak Creek Dam" (44-6-279). He reported on an "atmospheric monitoring program" of J. S. Cheka, noting:

Cheka reported a sudden increase in activity beginning June 9, 1944, reaching a maximum on June 10, 1944 and thereafter diminishing slowly each day. Reference to the records of water level, to which Cheka did not have access, indicate that the dam was emptied between 2:30 PM June 9, 1944, and 3 PM June 10, 1944, and has been slowly refilling since. The correlation is perfect and enables one to state that Cheka is measuring the radiation from mud at the bottom of the pond. There are more direct ways of doing this, but the matter is brought to your attention . . . to emphasize the need for such dam measurement, which have been held up since January by failure to obtain a satisfactory portable counter.

The issue of monitoring at the dam will surface again soon, but it will a different type of monitoring. One wonders why the measurements had been needed since January, as wastes were not released until March, according to most accounts. Perhaps they were intended to establish background levels.

At this same time, Curtis reported to Doan on biological monitoring (44-6-271), saying, "There has been considerable speculation here as to the condition of the fish living in the waters receiving the waste from the separations plant. I feel you should be advised of the facts insofar as we know them at the present time." He presents activity data on five fish and one crayfish from the Clinch River, both below and above White Oak Creek, and from White Oak Lake. Activity was determined on each specimen, as well as on selected organs of the fish. Those from the lake all had activity, with a catfish being most contaminated, and the highest specific activity was found in the hearts. Fish from the river above the plant "had zero activity," but the crayfish collected below the confluence of the creek and river did show contamination. Curtis cautions that the

data are preliminary and concludes with "Tolerance doses of these elements" ("elements" are not specified) "for fish are not known, but making certain guesses one might conclude that one of these fish was receiving somewhat more than a tolerance dose of radiation while the rest were receiving less than tolerance."

THE USE OF A NOXIOUS AGENT

A July 1 report from Apple to Kay (44-7-68) confirms what had been tried earlier with calcium chloride, pointing out, "no treatment has been found which is more effective for decontamination." This, combined with dilution with raw water, should result "in a lowering of activity by one thousand-fold, (but) this value has not been adequately checked." Secondly, he references a June 20 communication from Hamilton to Cantril regarding discharge limits to White Oak Creek, quoting:

In view of the fact that both analyses of the W-6 wastes done at site and here at Berkely show small amounts of Sr, Te, Cs, and Ba, together with observation here that none of these four elements are present in significantly large quantities in the mud, it is my opinion that the tolerance problem is very considerably simplified. Moreover preliminary data in hand indicate that a very large proportion of the activity from these four elements is presumably fixed on the clay, which should significantly reduce their absorption from the digestive tract.

Since the bulk of the activity is made up of radio elements that are not absorbed from the digestive tract . . . it would appear to me that the maximum daily amount of activity that can be released into the creek can be increased with safety to at least 250 millicuries a day and probably to one curie per day.

Apple reports that on the basis of this one-curie limit, decontamination of the wastes beyond that achieved by calcium chloride treatment and dilution "would not be required." He concludes the report with a fairly detailed recommendation on how to best handle the waste among the four active gunite tanks, emptying the oldest first "as rapidly as the capacity of the settling basin or the activity of the effluent to White Oak Creek will permit," and:

we recommend that data on the settling rates in the new basin, the gross activity discharged from the settling pond and from the waste tanks, and the composition of the activity in the effluent from the pond be obtained for at least two weeks. We also wish to follow the activities in any of the buried waste tanks that are treated with CaCl_2 .

It is unclear why the 2-week period was specified instead of a lengthier or continual period. Richard Apple (1909-1985), a native of Mississippi,

held a doctorate in inorganic chemistry from Iowa State College (1936) and had been with du Pont since 1935; he had worked at the Met Lab prior to coming to Clinton Laboratories and then worked at Hanford (1944-1946). After the war he became a section head at du Pont's Burnside Laboratory.

In July, Herbert Parker prepared a rather rambling report entitled "Review of Water Monitoring Procedures at Clinton Laboratories" for the Met Lab, submitting this to Compton, Stone, Whitaker, and Cantril (Parker 1944). In this document, he offers a detailed and sophisticated discussion of the establishment of tolerance limits, beta-gamma counting considerations (geometry, energy levels, self-absorption, etc.), and various aspects of discharge and exposure scenarios associated with the waste in the settling basin and White Oak Creek system. Setting the scene in his opening paragraph, he says:

The methods used for water monitoring were set up at a time when it was anticipated that the water activity would be low at all times. It was expected that sufficiently accurate information for extrapolation to the W levels would be obtained and that no significant levels for health hazard at Clinton would arise. It was further assumed . . . that the limiting hazard would be that of external radiation to a body immersed in the water, rather than that of ingestion.

Parker presents argument that the discharge from the settling basin should be "1 to 2 $\mu\text{c}/\text{liter}$, or for a daily discharge of 600,000 gals, a daily discharge of 2-1/2 to 5 curies. On the average no more than this can proceed through the dam a day." Relying on flow rates (average and low) in the Clinch River, he calculates the average concentration of " 10^{-4} $\mu\text{c}/\text{liter}$ to 5×10^{-2} $\mu\text{c}/\text{l}$," respectively, in the river. He points out, "In fact, relatively little of the activity goes through [over] the dam," as most is retained in the clay sediments behind the dam. This sediment poses a hazard, he adds: (1) radiation from the sediment; (2) biological uptake by plants, animals, and humans; (3) absorption by fish; and (4) discharge to the river by flood. The first two are controlled by fencing and the third by the fish screen at the dam. If all the 1000—2000 Ci in the lake were discharged at once into a flooding Clinch River, the concentration would be " 10^{-2} $\mu\text{c}/\text{liter}$;" if all this "accumulated at one place in the river bed it would not present a hazard in a river 6 feet deep. The risk of ingestion by a fish and subsequent consumption as food would be real." He compares his permissible discharge limit of "5 curies per day" with a more conservative one established by Hamilton (discussed earlier), "inadvertantly based on the minimum Clinch flow." Releasing the activity to the Clinch River is better than trying to fence off a large area, he says, and "the maximum waste discharge is therefore

governed largely by security since an elaborate fence and patrol system outside the plant site might excite interest." Finally he suggests that a "noxious agent" might be added to the water "to discourage drinking by pastured animals and swimming," but dilution in the Clinch River would have to render the water "palatable." This report represents an interesting blend of sophisticated science, with regard to the physics of accurately counting and establishing tolerance limits, with other less familiar (at least to Parker) aspects of environmental protection and waste management.

A COMPROMISE BETWEEN IDEAL DESIGN AND GETTING THE JOB DONE

At the plant site, concern was mounting about the new settling basin. Put into operation on July 3, it was not fulfilling expectations. In a July 8 memo to Doan (44-7-167), Leverett wrote, "We have received some information from M. D. Peterson" (Section Chief for Chemical Development in the Technical Division, 45-1-65) "and other sources which indicates that the 1,600,000 gallon settling basin recently put into operation is falling short by a factor of 3 to 5 of the desired and predicted decontamination." He offers a "number of suggestions" to remedy the situation. First, he points out that the dilution ratio "is probably considerably over 100 to 1," recalling earlier laboratory work that showed that 35 to 1 was optimal, and noting that too short a settling time resulted because of the excessive amount of water flowing through the basin. Secondly, he recommends a separate basin be dug for storage of the precipitated "sludge," much of which is accumulating in shallow water "at the edge of the basin . . . which is an undesirable condition since there is activity associated with the sludge." Third, he recommends that the two 200,000-gal retention ponds, which were already built, be used "as sedimentation basins in which most of the sludge would be settled out." Finally, he makes additional suggestions related to adjustment of weirs and addition of baffling to control flow and mixing in the basin. Apparently, many of these suggestions were in the original discussions and plans for the basin, as pointed out several times by Leverett, but they had not been ultimately incorporated. Recall that it was Leverett's group that had the responsibility in May to make certain that no "unusual conditions" would hinder the operation of the settling basin.

Later in July (24th), Kay responded to Leverett (44-7-20) with specific comments on each item. A constant head weir box would be used to regulate the dilution ratio. He indicates that there was neither time nor experience to have installed the separate basin and "complications in piping and operations" hindered the use of the holding ponds as settling basins. Finally, the construction of baffles would have delayed start-up

and would have been of "doubtful benefit." A most revealing comment is made by Kay in which he says, "we proceeded with the basin in a manner which was certainly a compromise between ideal design and the necessity for getting the job done as quickly as necessary." Such a situation almost certainly applied to virtually everything that went on during those hectic months and years, and here it is openly acknowledged. Kay concludes by noting that the release rate to the creek is "about 1 curie of activity per day, and the Health Division informs me that they believe the decontamination is adequate at the present time." William Kay transferred to Hanford as a process engineer at about this time and resumed his original job with du Pont after the war, retiring in 1960.

The composition of the material in the settling pond was the subject of a lengthy memo later in the year from W. Singlevich to Morgan (44-11-318). He lays out rather detailed procedures for finding out "exactly how much product was present in the settling pond at S-x, both from an economical as well as a health hazard point of view." The term "product" referred to plutonium. This procedure established a "satisfactory procedure for the quantitative determination of product in waste liquors in the settling pond."

Oversight and guidance from the Health Division seemed to dominate the next several months with regard to waste disposal. In late July, Stone wrote a letter to W. (Walter) O. Simon (1906-), the Manager of the Hanford Engineering Works (44-7-319), in which he makes recommendations regarding waste disposal there. He suggests that Hamilton, Overstreet, and Jacobson (the latter two of whom had done the April creek survey at Clinton Laboratories):

review the waste disposal problem at Hanford, particularly the chemical waste disposal from the 200 Areas. The main point in their studies would be a study of the soil into which the chemical wastes are likely to discharge and the type of soil in the Columbia River into which any activities entering the river might become attached.

Drs. Overstreet and Jacobson are soil chemists and have been making a study of the soil of California, much of which resembles the soil of the Richland Area, at least superficially. Since 1942 they have been working with Dr. Hamilton on the metabolism of fission-products. They visited Clinton Laboratories and made valuable suggestions to us on the question of waste disposal here.

In mid-August, Simon (via his Technical Superintendent, S. J. Bugbee) responded (44-8-212) to Stone, saying that Overstreet and Jacobson would visit on August 16 and "we will advise you of the outcome of their visit." The response is yet unavailable, but there are other interactions

with Hanford during the year. Stone also had raised the issue earlier in the year (44-5-439) of monitoring stack gases at Hanford. He says:

I find out something that carelessly escaped my attention down here, namely that Mr. Kanne's monitoring was taking care of the gases from the Dissolver Cell only and was not monitoring the gases coming from the other cells of the Separations Plant. I hope that in the plans for monitoring gases going into the stack at W, that all of the gases are taken into consideration and not just those from the dissolver.

Stone also offered guidance to Argonne National Laboratory on disposal, this time for solid waste, in a communication with J. J. Nickson (1915-1985) on the 22nd of the month (44-7-336). He indicates a "special lined disposal pit" is not necessary, and "burial in the ground is sufficient, provided the area will be marked off for a long time to come and fenced in." Marking should indicate where equipment is buried that they may wish to exhume for future use.

The Health Division acquired responsibility for monitoring the 205 stack gas emissions in September, some 9 months after rising to the occasion and taking on the monitoring responsibilities at White Oak Dam; this is documented in a short memo from Doan to Leverett (44-9-170) in which Doan responds to Leverett's July 12 suggestion and says, "turn the equipment over to the Health Division for operation. You are therefore authorized to work with the Health Physics people and turn the responsibility for continuing the observations over to them at such a time as is mutually agreeable." Health Physics was a part of the Medical Division.

The Overstreet and Jacobson (1944) report on contamination of White Oak Creek was issued officially in September, although the work had, of course, been conducted in April of that year. The report had widespread impact, both at the time that the work was done—as we have seen earlier—and also when it surfaced again later. It is interesting to read the peer-review comments made on this report in a communication from Whitaker to Stone [44-9-59 (also 44-9-876)], dated on the 2nd of the month. The report (apparently in draft form) had been sent to Whitaker by Hamilton at Berkeley, and Whitaker had Laboratory staff review it. It got a quite critical review and, comparing the review comments with the final version received at Clinton on the 21st of the month, it appears that Overstreet and Jacobson responded to most of the comments in a highly professional manner. The comments pointed out factual errors, as well as misleading statements. One reviewer took issue with the third sentence of the report, objecting to use of the words "strongly" and "appreciably" in describing the activity in the creek mud;

the word "appreciably" was changed to "measurably" (see earlier discussion).

NO DAM RELEASES

The ability to change procedures in response to new events and situations is demonstrated with the "Revision of the Plan for Operation of Gates at White Oak Creek Dam," which was discussed in a September 26 memo to Whitaker (44-9-893) from J. (John) E. Wirth, now Director of the Medical Division (Cantril was on loan to Hanford, having followed his colleague, Herbert Parker, there; Cantril remained at Hanford to work with Parker and later served on the interim medical advisory committee of the AEC). Wirth references Cantril's December 20, 1943, letter in which the Medical Department stepped forth to take the responsibility for the dam and the verification of such on January 26, 1944. Noting "that there has been such a rainfall that the procedure outlined in the above mentioned letters is impractical," Wirth states that water has topped the dam on at least two occasions even though "the upper gate has been open continuously for the last two months." The reason for this is that the fish screen clogged up with debris and caused the water level to rise, topping the structure. He proposes a more frequent cleaning schedule for the screen or "construction of additional screening system to prevent the present fish screen from being plugged so that water cannot run through it continuously." Wirth is careful to point out that the debris, which is released to the creek below the dam when the screen is cleaned, is of very low activity and that "repeated water samples have never given a reading much above that obtained from drinking water." He also emphasizes that the effluent from the plant area into the creek is monitored "four times a day for beta activity and once a day for gamma activity. This information will warn us of any higher than usual activities. The Health Physics Section in addition makes a weekly check on the activity of the water at the dam." In a precautionary way, he says:

it is conceivable that a dilution factor might be desirable at some time. With the above arrangement such a dilution factor by hold-up at the dam would be impossible. It is therefore our desire to keep the water at the dam at a level with the junction of the upper and lower gates so that if such a dilution factor is necessary the upper gates may be closed for any desired period to allow for this dilution.

He concludes by stating, "This will require a reversal of the procedure previously set up to the extent that the Medical Department will notify Maintenance when it is necessary to stop the flow at the dam." The file copy of this memo has "O.K. by MDW, 9/28/44" handwritten at the end, indicating approval by Whitaker. The significance of this procedural

change should not be overlooked; it demonstrates the level of attention to detail and concern that apparently dominated the issue of preventing uncontrolled releases of contaminants.

Shortly after the flood, on October 6, a letter report was issued by William H. Ray to Morgan, entitled "Health Aspects of the High Water of Sept. 29, 1944 at Clinton Laboratories" (44-10-100). This report contains the details related to what occurred during the flood and the analytical information on activity in the settling "pond" (basin), the creek mud, and water samples in the creek and lake taken before, during, and after the event; document 44-10-221, dated on the 16th, contains corrections to the original report with regard to activities in some of the samples. The bottom line is, "no downstream health hazard resulted" and "the results of this monitoring by the Health Section showed no reason for alarm downstream. A survey . . . September 30, 1944 at White Oak Lake found normal conditions. Mud where the water had receded was less active than usual," Ray states, perhaps overlooking the fact that the contaminated mud had been washed downstream. Perhaps the most interesting fact found in this report is that apparently the downstream bank of the settling basin was almost lost during the flood; the report reads, "The dike erected by a bulldozer on the upstream side of the pond [basin] stopped the flow across before the downstream banks had eroded a few inches. This eliminated a flushing out of the active materials settled on the bottom of the pond." He also refers to "withholding of waste by 205 after noon of the 29th," clearly indicating that there was no place for the waste to go if released because of the flooding within the entire system.

Toward the end of October, Wirth again wrote to Whitaker (44-10-320) to follow up on the change in procedure for cleaning the fish screens at White Oak Dam. He states:

Continued monitoring of the water mud samples and debris collecting at the fish screen of White Oak Creek dam indicates that the present policy in regard to waste disposal is satisfactory. In the letter noted above it was stated that it was satisfactory to continue to dispose of the debris in front of the fish screen at the dam by allowing it to pass on down the White Oak Creek to the Clinch River. This debris is not believed to represent any health hazard. It is necessary, however, to point out to you that this debris having an activity varying from $1.4 \times 10^{-3} \mu\text{c/gm}$ to $6.5 \times 10^{-5} \mu\text{c/gm}$ may have a security or legal significance if by chance it should be detected along the Clinch River by someone not connected with the project.

This was not the end of problems with the fish screen, however. At the end of November, Wirth wrote a note to the files (44-11-375) in which he

recounts the fact that dead fish continue to be found clogging the screen and that the regular weekly cleaning visit was missed on November 18. On the 20th, "water was flowing over the top of the spillway and the screen was covered with more debris than usual." After that, daily cleaning took place.

THE RELEASES INCREASE

The allowable level of activity released from the settling basin needed to be increased at the end of October. Wirth wrote to Nylén, who worked in the 200 area (44-10-355):

to confirm our previous conversation to the effect that the Medical Department agrees that the effluent water from the settling pond may be increased in activity by a factor of 5 over the former rate of discharge to allow for a more rapid discharge from W-5 and W-6 tanks, begun on October 14, 1944. The above increase is based on calculations of the amount of activity previously discharged in to the White Oak Creek, monitoring of mud, water, and debris throughout White Oak Creek and the Clinch River, and discussion by J. G. Hamilton in his letter to R. S. Stone of October 11.

Three days later, A. W. Frankenberry, from the 200 area, wrote to Wirth (44-10-362) confirming the guidance received from Medical and establishing a new activity limit:

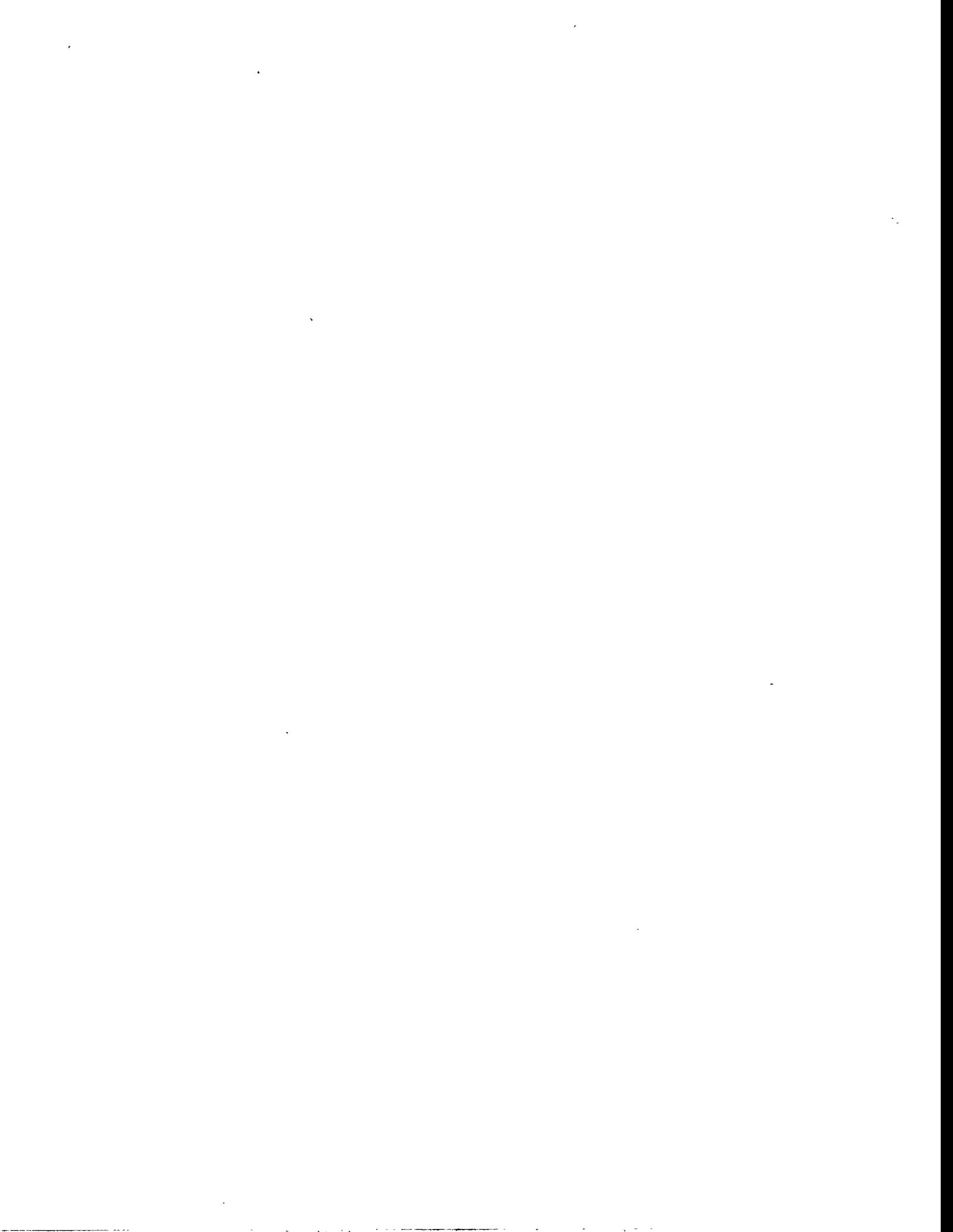
It has been our aim in the past to maintain a limit of 100 cts/cc/min. on this water. For future operations we now set up a procedure to read as follows: 1. Discharge of waste to the settling basin will be made at such a rate that the effluent water will not show an activity of more than 200 cts/cc/min. 2. In case at any time the activity should rise to 400 cts/cc/min. or above the waste discharge will be stopped immediately and the Health Department notified. Further discharge will be made only after the water activity has again become 200 cts/cc/min. or less.

The "factor of 5" increase authorized by the Medical Department must have referred to *total* activity released, whereas the two-fold increase simply referred to the allowable activity in water at any one point. It is curious to note that no guidance is provided for water with activities between 200 and 400 cts/cc/min.

Toward the end of the year, one of the gunite tanks (W-11) overflowed, as described in a communication from H. (Harrison) S. Brown to Doan (44-11-342). The Chemistry Division had assumed responsibility for monitoring the liquid level in the tank. Apparently, on November 21, the instrument used for this observation (a manometer) had malfunctioned and had not been properly fixed. That evening the

Health Physics Group saw that the tank was overflowing. Brown estimates that "about 90 gallons left the tank." He ascribes the incident to "(1) negligence on our part and (2) poor instrumentation, a situation which is also largely our own fault." He concludes by noting that the inspection procedures will be tightened and a "float type" liquid level indicator will be installed, as on the other tanks. Brown (1917-1986), born in Wyoming, came as a chemist from the Met Lab in 1943 and was Assistant Division Director in Chemistry in 1944, having worked on volatilization methods for separation of plutonium from uranium under the direction of Seaborg. He held a doctorate in chemistry from Johns Hopkins University (1941); after the war, he returned to work in the Institute for Nuclear Studies at Chicago and, in 1951, moved to the California Institute of Technology where he worked on trace elements in meteorites.

By the end of 1944, great strides had been taken to manage the safe disposal of the liquid waste at Clinton Laboratories. Full scale production was achieved in the graphite reactor and the separations building, and considerable activity was occurring in the 706 area; therefore, wastes were being generated at a rapid rate. The attentiveness and responsiveness of those individuals charged with managing "safe" waste disposal was reflected in their actions throughout the year, and waste disposal was still under the authority of the highest levels of Laboratory management.



THE YEAR OF DESTRUCTION

IF HANFORD, WHY NOT CLINTON?

During the last months of 1944, and certainly in 1945, there was a distinct change in the communications associated with the liquid waste disposal issues at Clinton Laboratories. The learning curve had been steep in the first 8 or 9 months of 1944, as evidenced by the communications and the challenges faced by staff. Apparently, the frequency of problems, and the associated documentation, began to sharply decrease during the last months of 1944, and the records do not reveal any sort of significant resurgence of problems in 1945. The ORNL Central Files database substantiates this observation; for example, the following number of documents had the word "waste" in the title for the years 1943—1947: 1943 (39), 1944 (90), 1945 (20), 1946 (17), and 1947 (88). The dramatic increase in 1947 reflects an increasing amount of work done at Clinton in support of Hanford's needs as well as more frequent reporting from the Health Physics organization. By the end of 1944, of course, Hanford had become operational and the initial reason for the existence of the Clinton Laboratories had been fulfilled, resulting in uncertainty concerning the mission here. In mid-1945, Monsanto took over operation of the Laboratory.

The handling of the liquid wastes had become somewhat routine, and much more regular reporting on waste discharges at all points became the norm in 1945. Further searches of the records may clarify this view, but it is doubtful that it will be altered significantly. In addition, staff began to look toward the future more than they had during those first 2 years and memos reflect the knowledge, later in the year, that the war was nearing closure. More personal thought takes place in the wording of memos, with concern about staff and working conditions.

In February, attention was directed at the two holding ponds, which had been part of the waste management system since its inception in 1943. Nylén, the Superintendent in the 200 Area, wrote Leverett to update the current status of the ponds (45-2-290):

When clay samples taken from White Oak creek revealed considerable active contamination early in 1944, agreement was reached with the Health Group to discontinue bottom outlet discharge from the ponds. Since that time the levels have been maintained to cover the sludge deposits and flow through the East pond has consisted only of cooling water. Routine checks of the overflow from this pond have been well within the beta tolerance set for water discharge with no evidence of sludge disturbance. No flow has passed through the West pond.

The above information is presented for guidance in future operations. Recent checks made around the outer edges of the East pond, at a time when natural leakage had lowered the level somewhat, showed that appreciable decay had lowered the activity to $4\mu\text{r/hr.}$ or less. No check was made at the time for alpha activity, however, it is suggested that the levels be closely watched to minimize drying of the cake deposit and possible spread of active dust. At some future date it would seem advisable to fill in the old ponds with earth.

Presently, some 50 years later, the ponds (now one large pond) are still unfilled.

Laboratory management began to make certain decisions regarding off-site air monitoring in the spring. Wirth informed Whitaker (45-4-150) that "on March 31, 1945, all the off area and the on area X-22 chambers for monitoring atmospheric radiation were discontinued." He states that the "experience of the last year and a half . . . has given us sufficient information to confirm the fact that no radiation hazards exist off the plant site by virtue of the operation of the plant." He points out that they now have enough information to establish a background "which could be used for a comparative basis in case of an accident at the plant." Citing "the expense of their upkeep" as a basis for the decision, he does say that the GM Counters, north of Bethel Valley road, and "other monitoring devices on the plant will be sufficient for the present and future contemplated operations of the plant." Note that this referred, it appears, only to atmospheric monitoring, rather than water monitoring.

Also during April, Stone sent an interesting letter (45-4-293) to Hamilton following a visit to "Site W." He says, in reference to Hanford, "the waste disposal system is already completely installed," and changes will not be made

unless tests of waters from various wells indicate that contamination is occurring. Very little active material is being put anywhere except in the storage tanks and the amount of inactive water that is being disposed of is more than enough to dilute all of the usual active wastes, that will get into the ground, to well within tolerance limits. Consequently the only chance of undue contamination would be an accident of some kind.

Up to the present time there has been objection to the procedure of digging new wells in order to test the water running from the 200 area towards Cold Creek and Richland, and the 300 Area. I am hoping the company and the Army will agree to the sinking of one or more wells as monitoring areas, but the absolute need for these has not been established and they would be used only as monitoring methods.

Stone concludes by saying that the information from these wells is "not likely to be put into practical use unless some unforeseen accident occurs," and it does "not seem wise to continue these studies on any long time or more thorough basis." He calls for a final report. The irony of this naivete is clear today as we assess the remediation challenges at Hanford; it would be informative to know more of why Stone made the decision that he did. It is interesting also that groundwater monitoring wells were installed at the arid Hanford site as early as 1945, but were not constructed at Oak Ridge until 1950, where the magnitude of rainfall was great and where population centers were close. The disposal of massive amounts of liquid waste to the ground at Hanford (Gerber 1992), a practice not used at Oak Ridge until the 1950s, must have led them to see the need for some sort of groundwater monitoring. In contrast, burial grounds were not established at Hanford until well into 1945: the Special Hazards Bulletin No. 3 (45-4-538) says, "there will be one burial ground in each Area," and that it (they) will "be under Operations supervision."

BEGINNING TO LOOK TOWARD THE FUTURE

The files indicate that monthly reports from the Health Physics Section, under Morgan, began mid-year (45-6-2), but the regularity of them at this early stage appears to have been sporadic. The majority of the reports deal with radiation exposure, clean-up of contaminated areas, etc., rather than with waste disposal, and no attempt is made here to review each monthly report of the Health Physics Section. The June report, cited above, states, "a new burial ground has been opened east of the plant site. Initial observation indicates that it may become as messy and offer the same radiation problems as did the old one [burial ground 1] unless more careful planning is carried out." This statement was in reference to burial ground 2, which was on the hill across from what is today the 4500 complex. In the monthly report for August (45-8-292), the activity of water discharged from the settling basin is reviewed, noting, "it had varied considerably during the past four months. It is seen, therefore, that the total radioactive discharge ranged between zero and ~14 curies/day during this period. This activity was diluted before it left the settling basin with ~900,000 gal. of water/day."

During 1944 and early in 1945, the amounts of radioactive wastes generated from the separations plant were very large. However, production of plutonium slowed at the Clinton Laboratories at this time, and some of the waste disposal issues associated with the 205 Building changed as the barium separations process was adopted early in 1945. Activities continued in the Chemistry Division and wastes were still produced from the 706 complex. For the barium separations building

(706-D), to more effectively handle the "large amount of active waste discharged from 706-D Building during the first production run," R. B. (Beecher) Briggs of the Technical Division [Corrosion Group Leader (44-5-423) and later Section Chief (45-1-65)] wrote to Leverett, Division Director (45-6-70), offering a number of suggestions regarding the transfer of wastes among tanks in the 206 area, stating that "because the permissible activity of the outlet from the Settling Basin has not been exceeded at the given discharge rate, it is believed that an average of 10 to 15 curies per day can be received from 706-D operations." For efficient operation of the new procedure, he asks for a daily forecast of the amount of activity to be discharged. Briggs, an engineer, was significantly involved with reactor design after the war. Several days later, Wirth sent a memo to Leverett (45-6-184) in which he explained a number of points regarding the difference between wastes from the 200 area and those from the 706-C and D Buildings, helping to explain the need for procedural changes, as suggested by Briggs. Although communications of this type are relatively rare during 1945, they testify to the attention still given to discharge limits and the proper handling of waste materials.

At this point, the records signify that the direct involvement of Miles Leverett in waste disposal issues waned, although he is later involved. After the war he resigned his position as Division Director and was instrumental in the design of new reactors, leading the activities for the Air Force at Oak Ridge on applying nuclear energy to aircraft propulsion; in 1951, he joined General Electric, where he headed a group on aircraft propulsion. His position as Director of the Technical Division was filled by Merlin Peterson (1910-1995), a physical chemist originally from Utah, who had been at the Laboratory ever since its founding and who came originally from du Pont, having taken his graduate degree under Willard Libby at Berkeley in 1936. Peterson had previously been involved with issues related to the handling of the wastes, as we have seen. He held this position for a year until he left to assume the Chemistry Department Chairmanship at Vanderbilt University in 1949, where he stayed until 1957 when he joined Columbia University. In 1960, Peterson moved to Argonne National Laboratory; he retired in 1973.

As it became clear that the war would end, staff began to look toward the future. Morgan crafted a statement to Stone in June entitled "Some of the Problems Confronting Health Physics if Fission Research and Development Continues" (45-6-105). In this, he identified and discussed some 12 separate, but related, issues of concern to the health physics community. Most are not directly related to the issue of waste disposal, but he does touch on the topic twice. First, he notes the need for meteorological study to assess "the extent and magnitude of expected airborne radioactive dusts and gases." Secondly, he discusses one issue

entitled "make a long time study of radioactive waste disposal problems":

A careful study should be made of the topography and geology in the vicinity of each proposed fission plant. The soil surface should be carefully studied. The weather history of the site should be carefully reviewed in order to forecast and avoid serious consequences of floods and erosion.

Apparently, this is primarily in reference to solid waste disposal, rather than liquid waste. Indeed, during this time frame and the following years, the evidence shows an increased awareness of, and concern over, issues related to contamination from the burial of low-level contaminated materials.

Nevertheless, the hazards associated with the disposal of liquids were not ignored in the least. In August, for instance, Morgan wrote to Stone on "The Past and the Future Health-Physics Programs of Clinton Laboratories" (45-8-263). In this document, Morgan describes the responsibilities and functions of the three sections that comprise the Health Physics "effort": Personal Monitoring, Survey, and Research and Development. The Survey Section "is to take meteorological data and determine the magnitude and extent of radioactive materials in the area surrounding the plant," he says, in reference to atmospheric monitoring. He suggests that two men with the proper "GM tubes" could conduct the monitoring "should a necessity ever arise again for such measurements." He continues:

Another responsibility of this Off-area Group has been the measurement of the activity of the discharged water and mud below the Plant. There is every reason to believe that the off-area monitoring has been accomplished satisfactorily. In no case has the radioactivity level of the water or air reached a level that could be expected to cause any damage to man, animal or plant.

This last statement is one of the earliest references to the detrimental impact of radionuclide releases on the environment.

In a broader communication, Brown wrote to Doan (45-7-305) just before the end of the war, saying:

Although it is too early at the present time to make definite prognostications as to the future of Clinton Laboratories in relation to the post-war nucleonic development program, it is clear that all of us must seriously consider the possibility that our Laboratory will continue after the war on a permanent basis. I believe we would all agree that should Clinton Laboratories continue on a permanent basis, it is in the national interest that

it be a strong and vigorous research and development organization.

Brown continued with a more detailed discussion of issues related to retention of staff, research and development conditions and opportunities, need for enhanced salaries, and improvement in living conditions. Nowhere within this rather forward-thinking document addressed to Doan is any reference made to waste disposal. It might be interpreted that the focus of the staff was now on challenges other than those associated with waste disposal, although we shall see in later years that the need for sophisticated research on wastes is strongly voiced. This is the last we will hear of Doan. Richard L. Doan left the laboratory shortly after the war and was heavily involved in reactor development for the Phillips Petroleum Company at Idaho Falls; in 1964, he joined the AEC as Director of Reactor Licensing and in 1968 he served as Assistant Director of Special Projects. Brown became a leader in the Federation of Atomic Scientists, an organization directed at transmitting scientists' views on nuclear energy to the public and to Congress.

AFTER THE END: ENHANCED AWARENESS

THE TAKING OF CHANCES IS UNWARRANTED!

Attention continued on monitoring and regulating the releases of radionuclides at the war's end and during the following years, but as shown earlier, the absolute number of documents bearing on this subject diminished from previous years, at least in 1946. One, which is a detailed recounting of "The Dumping of Wastes of Unusual Activity January 17 to January 25, 1946" (46-2-277), by Ray to Morgan, contains some very powerful language related to the protection of surface waters; Ray served as a Section Chief in the Health Physics Department (later called a Division) and reported to Morgan. The opening paragraph of this report serves as a summary:

Operation of the 206 (Tank Farm) area without sample reports from the Fission Product Laboratory led to the discharge of liquid wastes of abnormal activity over a period of five days to such an extent that even with careful manipulation of the White Oak Lake reservoir the discharge of water contaminated nearly to the tolerance level into the Clinch river was inevitable.

A combination of factors seemed to lead to this occurrence. Not only were analytical reports on the effluent from the settling basin delayed, but new and inexperienced operators were on hand, and heavy rainfall also contributed. Ray notes that for several days the effluent activities were in excess of the 400 ct/min/ml discharge limit and some "samples reported were fifteen times this value." The high rainfall caused White Oak Lake to top the "steel piling of the dam," and "it would appear that some water left White Oak Lake contaminated slightly above tolerance with Strontium."

This was checked by obtaining samples from three different down stream locations along the Clinch river on the following day. No positive gross counts were observed from the samples with a counting probable error of +/- 2 counts per min per ml. The return of White Oak Lake to normal values after purging by three inches of rain fall warranted opening of the upper gate of the dam on February 4th. However, the draining of activity held up in the creek had increased the contamination of White Oak Lake so that on February 8th it again became advisable to close the upper gate. A 1.65 in. rain fall on February 9th and 10th again caused the dam to overflow and resulted in the return of activity to its usual levels so that the upper gate could be opened for normal operation.

Ray states emphatically:

That this event was experienced without serious consequences should not lull us into complacency but should spur us to maintain increased vigilance toward preventing any such accident, the magnitude of which might easily have been greater. That the protection of the drinking water systems of the Tennessee and Mississippi river valleys depend upon the correct handling of our liquid wastes cannot be over emphasized for those responsible for their control. The taking of chances is unwarranted!

INTOLERANCE FOR TOLERANCE CHANGES

Repeated considerations were given to reestablishing, altering, and adhering to the tolerance levels for discharges from the plant. In April of 1946, Morgan contacted F. R. Ward (46-4-83), apparently in response to an inquiry related to the discharge limit into the river, to reaffirm the discharge level for "once through the pile" water at "0.5 $\mu\text{c}/\text{l}$." Morgan suggests that "water from the new pile run directly to the Clinch river by pipe instead of mixing it with the F.P.'s [fission products] in White Oak Creek," and he cites a spot a quarter mile above or below the confluence of the creek and river as an entry point. The advantage of this would be that the settling pond would not have to be used (chemical wastes would be treated in the normal fashion), and Morgan says, "if this water should enter the river with an activity $> 0.5\mu\text{c}/\text{l}$, a small fishtrap could be built around its outlet so that fish could not reach the above tolerance water." He recommends "sufficient holdup or dilution (or both)" be applied so that the limit is not exceeded. Finally, it is noted that the highest activity found in the river below the confluence with the creek is " $\sim 10^{-2}\mu\text{c}/\text{l}$," whereas the highest value in the lake is "about $0.2\mu\text{c}/\text{l}$, approximately the ingestion tolerance value for water." The reactor water, as far as can be determined, was not discharged directly to the river.

Shortly after the communication with Ward, Morgan again responded to a request (46-5-446) that a tolerance limit be changed, answering W. A. Rodger, Chief Supervisor for Technical Operations in the Technical Division under Leverett (46-5-510), "relative to the possibility of increasing the tolerances levels of the water in the settling basins." Because of the changes in the chemistry of the wastes, with much less coming now from the separations plant, and more short-lived iodine and strontium with less longer-lived zirconium and columbium, Rodger had suggested that the tolerance level be raised. On the basis of high oral absorption rates for iodine and strontium, Morgan states, "the greater absorption of the isotopes now present in the settling basin just about

compensates for any gain we might have sustained due to the shorter half life." The request is therefore politely denied.

In early July, Wirth contacted Peterson (46-7-10) in response to an inquiry as to whether "the tolerance amount of plutonium allowed in the plant waste per day" from the waste tanks would be exceeded; he indicates that Peterson's figure (600 micrograms of "49" per day) "will be well within the tolerance figure." He goes on, however, to caution Peterson that the gross beta-gamma activity that will be added to the system from the tanks might cause that tolerance level to be exceeded; he suggests some laboratory tests and concludes, "the settling basin will have to be watched very carefully." Finally, a Ray to Morgan memo in July (46-7-337), entitled "Drinking Water Tolerances Concentration of Ru and Cs," deals with calculations for the settling basin discharges.

Yet another communication on tolerance levels occurred in late July; Peterson (per W. H. Baldwin) wrote to Leverett (46-7-272) to suggest that tolerance levels be established not at the exit from the settling pond, but elsewhere:

The tolerable limit for activity discharged from the settling basin has been set as 500 beta counts/min./ml. (A. C. Vallado). However, this limit is for relatively new activity, and Dr. Wirth has indicated that for such all-long-lived material as the metal waste supernate a several-fold lower tolerance would be set, such as 100 cts/min./ml. Therefore, discharge of the supernate liquid . . . at a rate of 1 tank per 2 months, would itself give approximate tolerance activity in the plant exit water from the settling basin (i.e. tolerance for all-long-lived activity), while the newer activity now being discharged from the basin actually runs 50 to 100% of the allowed tolerance. Discharge of the metal waste tank supernate must therefore be closely correlated with the basin exit activities, and must be expected to require about six months per tank, with the present tolerances. Closing in White Oak Creek, like the settling pond, and specifying activity tolerances at the exit of the creek enclosure instead of at the exit of the settling pond, may safely allow several-fold faster discharge, because of Creek dilution, etc.

No record of a response from Leverett to this suggestion has been found, but later documents indicate that the tolerance limits were still established at the exit from the settling basin.

The "old burial ground" was the subject of a survey in July of 1946 (46-8-78), documented in a memo from Ray and D. M. Davis to Morgan. Seven samples of soil were taken to check for alpha contamination, and very low levels were detected in some of the samples. The concern was for plutonium-contaminated dust, and it was recommended that the

burial ground be seeded and marked "with permanent monuments to guard against excavation or defacing which would promote erosion."

IT HAS SERVED ITS PURPOSE WELL

Early in 1947, Ray wrote to J. (James) H. Lum [47-1-163 (also 47-1-332)] on the subject of "Radioactive Waste Disposal Rates Anticipated for Future Developments at Clinton Laboratories." Lum (1903-), who had come from Monsanto with a background in chemical engineering (Ph.D., Yale, 1932), and Wigner served as Co-Directors of the Laboratory starting in the summer of 1946, when Martin Whitaker, a leading advocate for proper disposal techniques, resigned to assume the Presidency of Lehigh University. Ray presents two alternatives for waste disposal (liquid and gas). The first alternative is simply the continuation of wastes discharged after dilution with the "trust that no natural phenomena will selectively reconcentrate any of the active matter to a degree that it could cause damage." The second is to "remove all radioactivity from plant effluents, and store the active matter . . . in concrete vaults where its location will be known forever." He concentrates his comments on liquid wastes from a "high flux or power pile" and gaseous waste from the chemical processing of fuel. In reference to the latter, Ray says:

The chemical processing of fuel, however, also releases appreciable radioactivity in gaseous and vapor form. I, Xe, and Kr are the elements considered of major significance. The 205 pilot plant discharged as much as 100 curies up a 200 ft. stack during a 6 hour period. It is believed that the proposed fuel reprocessing plant can achieve this average rate of discharge although it may be necessary to hold back activity to spread the discharge over a longer period.

Ray proceeds with a discussion of the use of activated carbon for retaining certain gases and the release of the noble gases "since fixation in the body seems improbable." He notes the "discharges of cooling water containing activated impurities" from the high flux pile and says:

plans call for ponds to hold back this effluent for a period to effect decay to the discharge activity of $1/2$ μ curie per liter. This rate of flow at tolerance concentration would be diluted by the Clinch River minimum of flow of 150 cu. ft./sec. If the coolant problem (both air and water) could be solved in terms of closed systems and heat exchangers, no radioactivity need be discharged from future plants. Developments to achieve this are still over the horizon.

Ray definitely was looking toward the future as he sought new and better ways for disposal of wastes. The opening paragraph in this analysis reads:

Present practice, from the viewpoint of the writer, may be tolerable, but now that the pressure of war upon operation has relaxed, a revision should be effected to handle the liquid borne wastes in a more ideal fashion even though it may not be necessary to reduce the activity released to permit the discharge of additional activity into the Clinch River.

In March of 1947, Cheka and Morgan issued a quite comprehensive and informative report on "Radioactive Fission Product Contamination in the Mud of White Oak Drainage System." This review summarizes most of the different surveys made on the activity in the drainage system, starting with the Overstreet and Jacobson work of April 1944, and including data acquired by Hamilton (June 10, 1944); D. M. Black (April 14, 1945; August 15, 1945); J. E. Hudgens, a Group Leader for Radiochemical Analysis in the Analytical Division (February 14, 1946); and H. R. Craft (October, 1946). An earlier report, October 1946, from Hudgens to Cheka (46-10-340) compared analytical techniques on mud from the creek. Cheka and Morgan present data on the total number of curies suspected to have been released into the system, saying:

Morgan and Western indicate that a total of about 520 curies were discharged from the beginning of operations of Clinton Laboratories until the time of the April 1945 general mud survey. Thus, about 13% of the activity discharged into White Oak drainage system during a year and a half of operation still remained in the mud in April 1945. During the seventeen months between this and the October 1946 survey, an estimated additional 890 curies were discharged into the system, making a total of 1410 Beta curies since the beginning of operation. Of this quantity about 7% was detected during the October 1946 survey.

The report contains enlightening historical information:

A sharp rise in activity appeared in the creek during January, 1945, when the Plutonium Separations Building was cleaned up after the cessation of separations operations. This radioactivity subsided in a short time, and values remained fairly constant at the December level until the Barium separations began in volume in Buildings 706C and 706D. The radioactivity in the Clinch River mud at the mouth of White Oak Creek began rising early in 1945, and has followed somewhat erratically the fluctuations in the radioactivity of the mud in White Oak drainage system.

Cheka and Morgan explain that the wastes from the barium process are different than those from the plutonium separations process, being less subject to precipitation. Therefore, "about half or more of the curie content of the supernate that is jetted out of tank W-6 passes through the settling pond and into White Oak Basin." They explain that in June

of 1945, when 706D was in full operation, the mud activity increased by "a factor of about 10 throughout the drainage basin."

The report concludes:

In an overall evaluation of the White Oak Creek basin as a part of the waste water decontamination process, one might conclude that it has served its purpose well. With the proposed changes of process, and consequent changes in the nature of the chemical wastes, further changes in the decontamination of wastes may be required in order to keep the White Oak Lake effluent within safe limits. In general, it seems that slowly flowing drainage system like White Oak Creek and lake with a clay bottom is an economical and rather efficient means of removing fission products from plant waste water.

Later in the year, T. (Thomas) H. J. Burnett also issued a preliminary report on the "Efficiency of White Oak Creek" (47-11-554). This involved sampling the creek system at 7 locations and he concluded, "The decrease of activity observed from the Settling Basin to White Oak Dam is largely due to dilution," although decay of shorter-lived isotopes also contributed. He points out that slow flow rates are beneficial, suggesting that a lower dike be reestablished and that the "controlled addition of silt for adsorption purposes" and that continuous instrument monitoring at the settling basin exit and the dam be instituted.

The following year, yet another assessment of the efficiency of the White Oak Creek and Lake system was prepared by Lloyd R. Setter (48-12-293), in which data are presented by month for radioactivity entering and leaving the "White Oak System." A series of floods during 1948 had resulted in unusual releases of activity from the lake:

The floods cause a "slugging" of the Clinch River for short intervals and impaired the effectiveness of the White Oak Creek disposal system. Thus 70.4 to 81.4 percent of the wastes are removed during dry weather months, 55.5 to 68.9 percent are removed during moderately wet months and negative removals are obtained during flooding periods for an overall eleven-month removal of 55.4 percent of the added wastes.

In August of 1947, a summary report was issued from Burnett to Ray (47-8-68) on the "Activity of the Clinch and Emory Rivers" in which data from 9 Clinch River, 8 Emory River, and 1 Norris Dam (background) samples are reported. Burnett reports:

Activity carried by particles in suspension is seen to be in three instances about five times average background or four times maximum background data. The greatest distance this magnitude of activity occurs is 19 river miles from White Oak Dam, at mile 2,

Emory River. In general, the level of water activity may be considered as probably satisfactory, but there can be little question of the importance of systematic and periodic survey of these areas. Additional background data would also appear to be in order.

Obviously, the concern over proper and safe disposal of the liquid waste at the laboratory persisted into the years following the war.

REQUESTING PERMISSION

The year, 1947, seems to be the first time that routine periodic reports were issued on discharges from the settling basin and the lake, although this information had been documented in a less regular fashion in previous years. The "Liquid Waste Disposal" report for March of 1947, a transmittal from M. A. Buford to Morgan (47-4-3), contains data on routine water surveys "on alternate days at White Oak Dam and the Settling Basin." This particular month "no mud samples have been taken . . . due to lack of laboratory facilities." The content of the reports was rather consistent, and the reports were very frequent, especially during the latter part of the year, but the authorship varied considerably; for instance, for July, Buford reported the data to Burnett (47-8-441); whereas for August Burnett reported to Ray (47-9-168); and for September (two reports), November, and December, Burnett reported to R. H. Firminhac (47-10-99, 47-10-104, 47-12-89, and 48-1-87). Even in 1948, Burnett reported to J. H. Roberson (48-2-123) and W. D. Cottrell to J. C. Hart (48-3-153, 48-4-174), for example; Cottrell was a Group Leader and Hart was a Section Chief in Health Physics, having come to Clinton with Morgan and Parker in 1943. A series of (semi) regular reports on air monitoring (e.g., 48-3-210) were also written, in which monitoring results, meteorological data, instrumentation needs, etc., were discussed.

The monthly Health Physics Reports, from Morgan to E. J. Murphy (e.g., 47-10-190, 47-11-346, 48-11-103, 48-11-297, 48-12-304), were much more detailed, up to ten pages in length, and reviewed many issues beyond waste disposal. In the section on "mud and water surveys," appearing in the 1947 reports, data on daily and monthly discharges are presented or referenced with Central Files numbers. The October report indicates:

discharge of activities exceeding 500 c/m/ml are permitted only with the permission of the Health Physics Department, so that the water leaving White Oak Dam can be carefully monitored and, if desirable, the dam gate can be closed until the specific activity of the water can be reduced.

Recall that in late 1944 (and then in February of 1946 with the Ray report on the "dumping of wastes of unusual activity"), the discharge

limit was established at 200 cts/cc/min and that the Health Department was to be notified and the discharge stopped immediately if the activity rose to 400 cts/cc/min; this limit had been raised to 500 by July of 1946, according to the Peterson to Leverett memo (46-7-272), but the record of this decision is unclear. Table 1 contains a summary of the various "discharge limits" that have been referred to in this analysis. Many are in agreement with each other, although one must question the mixture of units and the use of absolute and relative amounts, a situation that almost certainly must have led to some level of uncertainty.

The October report also indicates that "another survey of the activity in the Clinch and Emory Rivers will be conducted in the near future" (apparently following up on the August recommendation) and that there is a cooperative effort with K-25 "in analysis of wastes characteristic of their operation . . . obtained at the same time that samples are obtained for our study." This report indicates a collaboration between sites relative to the river survey. The November monthly report refers to small-scale construction work at the tank farm to "prevent further contamination of the environment," an early use of the word; also in this document are data on "a survey of the burial ground for radioactive materials" and confirmation that new samples had been taken from the Clinch River and at Harriman and Oakdale (presumably from the Emory River). The 1948 reports contain relevant information under the heading "Activities," where updates on the Clinch River work, nuclide sorption studies, discharge studies at the dam, planned ecological studies of White Oak Creek, and development of field instrumentation are reported. One gradually sees more references to the burial grounds; the hiring of Paris Stockdale, a geologic consultant, for "proposed geological studies particularly as they might throw light on the underground flow of water in this vicinity" accentuates this new thinking. Core drilling is referred to. As one tracks these reports throughout the months, they become more detailed and complete.

The fact that laboratory management was conscientiously attempting to dispose of wastes properly is proven by some 1947 correspondence related to disposal of barrels of dissolved uranium, uranium slugs from the reactor (pile), exposed materials containing plutonium from the reactor canal, and other items. The record is not complete, but the message is clear. The correspondence referred to here is (1) Murphy, Assistant Research Director at Clinton Laboratories (previously a Major with the Corps of Engineers), letter to Col. W. (Walter) P. Leber of the AEC, dated May 21, 1947 (Disposal of Waste X Metal, 47-5-350); (2) L. (Logan) B. Emler (General Superintendent of the Operations Division and later Vice-President for Union Carbide Nuclear Corporation) memo to Murphy, dated August 14 (Disposal of Waste "X" Metal, 47-8-179); (3) Murphy

Table 1: Discharge Limits Referred to for the Liquid Waste Disposal System at Clinton Laboratories, 1943—1947

(Consult the text for specific applications.)

Discharge to:	Discharge Limit	Reference
River	0.1 beta r/24 hrs	Kirst 1943
River	8.1×10^{-6} curies/cc (2.3×10^{-5} curies/ft ³)	Kirst 1943
River	0.01 r/8 hrs	Wende, 43-3-124
Creek	10^{-4} curies/ft ³	Ellett et al., 43-3-277
River	10^{-11} watts/cc	Leverett, 43-11-79
Creek	0.1 r/24 hrs	Greager, 43-11-152
Creek	100 mr/24 hrs	Cantril, 43-12-180
Creek	5×10^{-4} curies/ft ³	Smith, 44-5-101
Creek	0.25—1 curie/day	Apple, 44-7-68
Creek	500—1000 millicuries/day	Smith, 44-9-335
Creek	0.1 r/24 hrs	Smith, 44-9-54
Creek	1—2 microcuries/liter	Parker 1944
River	10^{-4} — 5×10^{-2} microcuries/liter	Parker 1944
Creek	5 curies/day	Parker 1944
Creek	1 curie/day	Hamilton, see Smith, 44-9-335
Creek	5-fold increase	Wirth, 44-10-355
Creek	100 cts/cc/min	Frankenberry, 44-10-362
Creek	200 cts/cc/min (400 max)	Frankenberry, 44-10-362
Creek	400 cts/min/ml	Ray, 44-2-277
River	0.5 microcuries/liter	Morgan, 46-4-83
Creek	500 cts/min/ml	Peterson, 46-7-272
Creek	500 cts/min/ml	Morgan, 47-10-190

letter to James C. Stewart of the AEC, dated August 21 (Disposal of Waste X Metal, 47-8-230); and (4) Emlet letter to P. (Prescott) Sandidge (Assistant Executive Director of Clinton Laboratories) dated November 8 (Disposal of Radioactive Contaminants, 47-11-130). Since October of 1946, the laboratory management had been requesting permission from the AEC to dispose of the items noted above; the uranium is "contained in approximately 200 wooden barrels which are rapidly decaying . . . stored in the vicinity of the Settling Basin," writes Murphy. He points out that "the ground near the barrels was becoming contaminated," provides supporting data, and says, "we again recommend that this health hazard be eliminated as soon as possible by the most practical method; namely, burial in the area designated for this purpose." Emlet questions the efficacy of the AEC using the laboratory for disposal of waste from other facilities, stating, "if it is decided that we will take care of all project garbage, I should like to make a thorough study of the problems involved and forward the comments to you on what additional facilities would be necessary." [An October 23 memo from Stewart, Chief of the "Clinton Laboratories Division," to Sandidge of Monsanto at Oak Ridge (47-10-560) details the amounts of liquid wastes from Argonne National Laboratory to be disposed of at Clinton Laboratories.] Emlet's statement refers primarily to solid waste that was put in the burial ground, but he also questions, "if it is desirable to dispose of too large a quantity of radioactive wastes in one location because of the selective absorption that the mud along the river banks has for several of the activities," referring now to the liquids (and perhaps releases from the burial grounds, although this is unspecified). These documents reflect a certain level of frustration with the AEC, and the tensions between the laboratory and the AEC at this time were well documented (Hewlett and Anderson 1962). AEC responses have not yet been found. Emlet was a native of Pennsylvania and had received an M.S. degree in chemistry in 1936 from the University of Pennsylvania; he had a long career in atomic pile construction and operation and had supervised the construction of the graphite reactor at Clinton after working at the Met Lab in 1943.

UNLESS WE GET YOUR SUPPORT AND INTEREST . . .

The issue of research directed at waste disposal began to surface in 1948. Certainly "research" had been conducted in previous years on ways to minimize the volume and activity of the liquid waste, as we have discussed earlier, but no organized research program existed at that time. As the Health Physics Division grew, a much enhanced research flavor evolved with respect to nuclide behavior in biota, mineral surface interactions with contaminants, etc. This is reflected in a January communication from Burnett to Firminhac, "Activity Values in the Body of a Wildfowl" (48-1-368). A dead bird had been found near the dam

and was highly contaminated; individual organs were studied. In reference to human consumption of contaminated fowl, Burnett says in a precautionary fashion:

while it is a most remote improbability that humans would ever thus be damaged, it is in principle an undesirable situation, representing dispersal of activity without control, and incompatible with the precept that the ideal is zero exposure at all times to all people. Other species of migrating duck, etc., feed on White Oak Lake too, so the direction of dispersal is varied. The results obtained are, while interesting, not conclusive from the viewpoint of being only a single examination. However, there is little reason to doubt that they are indicative of the proper order of magnitude of activity, energy and exposure.

He relates all of this to waste management practices, reminding the reader that the concept of dilution and dispersion of nuclides assumes that there is no mechanism for reconcentration. Yet, uptake in the food chain through algae and plankton into fish and fowl constitutes a reconcentration process. Perhaps other waste disposal practices should be further considered, such as enhanced water purification through the use of ion exchange systems, enhanced precipitation, or adsorption on clay followed by centrifugation. He offers some initial calculations on treatment volumes per day and concludes:

The present efficiency of our system leaves much room for improvement as pointed out in . . . "Preliminary Report - Efficiency of White Oak Creek." It is contemplated that the Waste Disposal Group will begin pilot experimental studies at once of possible improvement along one or more lines of inquiry mentioned.

A seven-page report from Burnett to Forrest Western, Assistant Director of the Health Physics Division, entitled "Waste Disposal Research Problems" (48-1-369), outlines a series of waste-related issues for improving the management of gaseous and "liquid active" wastes. Three approaches are presented: (1) removing "the maximum possible amount," (2) dilution of residual activity, and (3) measurement techniques. This appears to be an internal-type research statement, rather than one directed at the AEC. However, later in 1948, a much more outspoken position was established with regard to the AEC taking on a major role in fostering basic research related to liquid waste disposal.

In August, a conference on "Liquid Waste Disposal" was held; the minutes of this conference were published (ORNL 1948). A summary of the meeting is in the Central Files (48-10-343). The undertaking was in response to the AEC, and the introduction, taken from the minutes, says:

A committee consisting of representatives from Dow and Monsanto Chemical Companies, Argonne National Laboratory, and Division of Engineering, AEC, Washington, has been formulated to study the AEC-wide problem of disposing of process waste solutions. The members are: S. Lawroski, Argonne, Chairman (W.A. Rodger, Alternate); J. Grebe, Dow Chemical Company; L. A. Mathison, Dow Chemical Company; W. Hurschkind, Dow Chemical Company; M. N. Haring, Monsanto Chemical Company; F. C. Mead, Monsanto Chemical Company; J. H. Hayner, Division of Engineering, AEC, Washington (H. Noble, Division of Engineering, AEC, Washington, Alternate). The committee, in its investigation of the problem, plans to visit the various sites which are confronted with disposing of process waste solutions and which have experience in handling them.

The purpose of the meeting was "to ascertain the nature of the waste disposal problems confronting the Oak Ridge plants" (all three were represented) "and to determine the scope and nature of the research and development related to the problem."

A total of 20 ORNL staff attended, including F. N. Browder, Burnett, Emlet, Hart, Morgan, Western, and others. The AEC was represented by 10 attendees. A series of presentations was made: Liquid Waste Disposal System, by E. J. Witkowski and P. B. Orr; Effectiveness of Our Present Waste Disposal Program, by Morgan; Burial Grounds for Solid Waste, by Orr; Related Research on Ion Exchange, by J. A. Swartout; Rare Earth Separations Studies, by B. H. Ketelle; Precipitation and Scavenger Techniques, by R. W. Stoughton; Summary of Technical Division on Liquid Waste Disposal Problems, by F. L. Steahly; Ion Exchange Development in the Technical Division, by R. E. Blanco and I. R. Higgins; Development of an Evaporator System for Oak Ridge National Laboratory Liquid Waste Disposal, by C. E. Winters; Design of an Evaporator System for Oak Ridge National Laboratory Liquid Waste Disposal, by F. L. Culler, Jr.; The Waste Disposal Program Undertaken by the Health Physics Division, by Western; and Proposal for a Central Burial Ground for Oak Ridge Operations, by J. Deal.

Although much of the specific material from these talks is beyond the scope of this present report and was quite technical, some of it is highly relevant. Note that issues related to liquid, as well as to solid, wastes are intertwined. Morgan, in his presentation, said:

recognizing some of the limitations of the system we are using . . . we have taken some steps to better orient ourselves. Eighteen months ago we began efforts to get together a group to study our waste disposal problems. Sometime toward the close of 1947 we got a group together composed of men from the United States Public Health Service, Tennessee Valley Authority, United States

Weather Bureau, University of Tennessee, and Vanderbilt University.

With regard to solid waste, Morgan states, "as yet we have not made a study of the geology of this territory," and "our burial ground is in about the worst possible place, because of its proximity to a fault. Solid waste gets into ground water and may show up many miles away." This, a reference to burial ground 3, is noted here because the issue of groundwater has equal relevance to liquid wastes, but was not associated with their disposal at the time, although clearly Morgan recognized the importance of groundwater with respect to solid wastes. The minutes reflect titles for lantern slides and (apparently) comments about them. One is a "picture of White Oak Dam and fish gates," with the curious comment "we maintain that a person could drink the water in White Oak Lake for the rest of his life without damage. The level of activity must be so low that persons or fish could swim in water for rest of life without damage." Morgan concludes:

We do not claim that our waste disposal system is ideal. Our system was set up during the war during which our principal effort was to get a pilot plant into operation. Now we recognize its limitations. Some of the necessary changes in our pile arrangement and waste disposal system are underway. It will be some time before we have all the answers to our problems of waste disposal.

Little did Morgan realize at the time how prophetic his last sentence was. Also, he does not seem to take appropriate credit for the magnitude of the efforts expended during the war years to properly manage the wastes.

In his presentation, Orr reviewed the solid waste disposal system, stating, "the treatment of solid wastes is very poor at best." He mentions the burial of "alpha material" (i.e., liquids), stating that "the location of the alpha burials is not very satisfactory," and noting, "it should be covered with concrete the same day to prevent any of the waste from floating to the top of the hole in case of rains. This has occurred in the past." This elicited a question about putting the waste in containers, to which Orr replied, "the material is in containers, but once or twice the containers have burst and black liquid has floated out of the ground." Other questions ensued, mostly related to the location of the burial ground. One person, however, asked, "do you bury much liquid?" The answer was, "yes," but if it is "very hot," it goes to the tank farm, rather than burial. (After the war certain alpha wastes, generally in liquid form, were routinely buried in containers under cement in the burial grounds). Morgan chimed in with a statement that the system for disposal "is becoming more and more dangerous because of the long life hazard." The session ended with a plaintive statement (acknowledged to no one

in particular) obviously directed at the committee and the AEC: "unless we get your support and interest we cannot make further investigations along the lines outlined above."

Western comments, "with the development under the AEC" and "with relief of wartime urgencies" there are "increased tendencies to question the wisdom of continued dumping of significant quantities of radioactive wastes into the river," adding that they had "no adequate information upon which to base a good evaluation of the hazards involved." He makes a pitch for an "extensive, systematic study of the behavior of radioactive materials released into the drainage system which combined with . . . biochemical research, would provide a much better basis for such evaluation." He uses an expression "of a cautious attitude" by the AEC about waste disposal to help promote research at ORNL, emphasizing that "radioactive conditions" have been established "in White Oak Creek, the Clinch River and possibly, the Tennessee River" that "may aid us in obtaining answers to many of our questions." A rather extensive listing of waste disposal research topics is included to end his talk. Indeed, many of these topics were the focus of waste-related research of the Health Physics Division in the ensuing years.

ORNL was making a major attempt to land new research activities related to waste disposal. The openness with which problems were discussed and portrayed—perhaps even to an extreme—is testimony to the fact that staff saw the need for fundamental research in this area. Although significant waste-related research activities did, indeed, emerge at ORNL in the coming years, reflecting many of the suggestions Western made in his presentation, exactly to what degree this meeting influenced those initiatives is unclear. The issue of having the AEC involved in such research continued.

THE AEC SHOULD . . .

In November, excerpts from a report of the "Liquid Process Waste Disposal and Reclamation Committee" were issued (48-11-310) following a committee meeting the previous month at Argonne National Laboratory. Contained in this document is summary information on the handling of liquid wastes at Argonne, Hanford, ORNL, and Mound Laboratory. A number of insightful observations came from the committee: (1) waste disposal problems are not as attractive for research targets as are more fundamental topics; (2) solutions to waste problems often are not completed because of demands made on the "production" side; (3) "contractors" often offer only minimal and temporary solutions to waste problems; (4) no "systematic" approach has been applied to solution of waste problems; (5) waste disposal has been, until recently, of quite low priority at most sites; and (6) "management has failed to recognize that

the problem of waste disposal is comprehensive and involves the entire technology of Pile operation." Certainly many of these statements are not entirely true, as shown by the documented activities that occurred at Clinton Laboratories during the previous years.

The recommendations are challenging. First and foremost, the "AEC [should] establish a long range work program for studies on liquid waste disposal," with the scope to include "fundamental research on the chemistry" of wastes, and development of pilot-plant and full-scale operations. Information exchange and periodic review and coordination of the program are recommended; each site should "critically examine the present practice," and future contracts (presumably between the AEC and the sites) "should embody the requirements of handling the waste disposal problems." The statement, "waste disposal should be given a good priority and cooperation should be obtained between AEC, U.S. Public Health Service, geological and water supply agencies," is important. At ORNL, later records reflect that there was such collaboration.

Whether or not the following reference relates to the October committee meeting is unclear, but contained in the November monthly report of the Health Physics Division (48-11-297) is the following, which at least reiterates the AEC position:

During the week of November 8, O. R. Placak worked with others in the Division to restudy the Liquid Waste Disposal Research Program of the Division and prepared an outline of the three major projects of which the program is composed. This was part of the preparation of a consolidated report on Waste Disposal Research in all Divisions of ORNL requested by the Oak Ridge office of the A.E.C. preparatory to a Commission-wide research program on Liquid Waste Disposal. [Note: Placak came from the U.S. Public Health Service in Cincinnati (47-9-618).]

The summary outline on the ORNL program appears shortly (48-11-193), including identification of projects, brief descriptions, their applicability, status, estimated time from start to completion, physical facilities required, personnel available, and budgets for fiscal years 1949 and 1950. With the exception of projects dealing with a survey of the disposal systems at ORNL, evaporator and spray dryer designs, and instrument development, the other seven projects had as their objective chemical studies of the wastes.

A final relevant document, dated November 30, 1949, "Organization, Objectives, and Program of ORNL Chemical Development Steering Committee" (49-11-283), by Boyd (now Associate Director of the

Chemistry Division) and others, contains a telling statement. Under the section on "liquid chemical waste disposal," it says:

within the Laboratory, the liquid waste disposal problem appears well in hand, not only with respect to ORNL wastes but also those at Hanford. With regard to disposal problems for future sites or processes, the logical procedure will be to consider waste disposal as an integral part of each separations process. Therefore, the committee will limit its review of waste disposal to current situations with the understanding that considerations of all future chemical processes will include disposal of wastes.

Obviously, as the decade drew to an end, the issue of waste disposal was evident and integrated into the fundamental studies of the Chemistry Division.

Certainly, more materials could be presented to document the attitudes and practices related to liquid waste disposal at Oak Ridge, but at this point the dialog of archival review will come to an end. What follows is an attempt to place relevance on the material that has been presented.

ISSUES OF RELATIVITY

The history described in this paper represents much more than an anecdote that is of ephemeral interest—it has relativity as much more than a simple footnote to the Manhattan Project. The documents discussed reveal a remarkable degree of integrity displayed by those who were leaders in Clinton Laboratories and waste management at the time, operating under extreme conditions. Placing those early disposal practices in perspective should help us to understand the level of care and dedication those early leaders exhibited. This level of care may not come as a surprise to some, but to most it will. Long overdue and generally posthumous acknowledgment and recognition are deserved, not only for the obvious personal interest and reward, but also for scientific and medical documentation of what occurred.

In no way does the revelation that commendable disposal practices were exercised over 5 decades ago diminish the fact that large amounts of contaminants were released to the environment. These observations neither remove one curie of activity from the list of those that have been generated and released, nor diminish in any way the technical and fiscal challenges that we face today in the world of environmental restoration as we apply standards of the 1990s to practices of the 1940s. However, one of the social challenges faced today is that of dealing with public perception and in fostering understanding and acceptance of the historical ways in which wastes were handled. Without doubt, the prevailing opinion is that wastes of all kinds—and perhaps especially the liquids—were handled carelessly. The present U.S. Department of Energy, a successor to the AEC (which of course, did not even exist during the war years), is continually facing criticism and scrutiny for real—as well as perceived—historical mistakes. Perhaps information of the type contained in this study can help to frame a slightly mollified and more receptive reaction on the part of those who are critical.

The fact that conscientious decisions were made in the disposal of the wastes in the 1940s, as documented herein, should not lead one to believe that the *entirety* of activities directed toward waste disposal was commendable. Without doubt, conscious—and perhaps deliberate—steps were taken to dispose of materials in an unauthorized fashion, and there certainly were accidents, totally undocumented, in which materials were inadvertently released. Because of this lack of documentation the records fail to reveal much of this; those incidents that have been found documented have been presented. Numerous additional records deal with laboratory contamination and issues of personnel exposure, but such have not been the focus of this study.

In evaluating the actions from 50 years ago, one must do so in light of what was known at the time and in relationship to what the practices of the time were. It is in this perspective, perhaps, that the greatest admiration is deserved. Although probably not fully recognized at the time, the waste materials generated at the Clinton Laboratories can be said to have represented the greatest disposal challenges within the entire Manhattan Project. True, much greater amounts were generated at Hanford, but the toxic mixture of fission products, uranium, plutonium, transplutonium elements, nitrate, and hazardous metals in mobile liquid form at a location with rainfall and surface water used for drinking—coupled with nearby population centers—certainly elevated the risk at Clinton Laboratories. Facets of this are woven throughout the historical documents. Remember also, that the Clinton Laboratories were originally expected to operate for about 1 year and the tanks were so designed. When separation processes were changed and when the facility operated for extended times, these factors obviously stressed the waste management plans accordingly. During the 1940s, little was known of the health effects of nuclides, especially their behavior in the environment, and inhalation and ingestion were deemed to be the exposure scenarios; hazards from chronic exposure to radiation were not well recognized. Finally, instrumentation was primitive; gross beta and gamma (and alpha) counts could be obtained, but specific knowledge of the pertinent nuclide was either impossible to get or occurred weeks after a sample was taken. To blindly apply present standards to past practices is not only improper, but it is misleading and reflects ignorance of the facts.

Today, the thought of releasing 1 or 2 curies of activity—much less 5!—per day into the creek and river system is an unpardonable sin. Yet, the release limit was carefully calculated, monitored, and accepted 50 years ago. Medical knowledge at the time deemed this to be an acceptable release scenario based on the dilution that would occur in the river and on scant information related to biological uptake of radionuclides. It was readily acknowledged that adequate information was unavailable to completely assess the danger, but conservative thinking was applied to arrive at this release limit; those who set this limit can not be faulted for lack of knowledge or for misdirected intent. It almost goes without saying that the other contaminants with which we concern ourselves today (metals, nitrates, organic compounds) were largely not even recognized as dangerous then. Really, the only one that was monitored at all was lead, a known toxicant. Interwoven throughout this is the fact that the concern was strictly for human health and not for environmental impact, although environmental systems (fish) were used to measure the extent of contamination.

Throughout this report passing reference has been made to the attention given to the disposal of the contaminated solid waste, with recognition that the treatment of such was much less insightful than that for the liquid wastes. In light of the facts that the liquid wastes were obviously much more contaminated, were in a mobile form, and that burial of solid waste of all sorts below the ground surface was the standard practice everywhere, it is not surprising that this distinction occurred. During the 1940s, little was known about groundwater and its potential for transporting contaminants; indeed, in the technical literature for groundwater hydrology, scant reference occurs to groundwater systems at this time, much less to contamination thereof. Even though knowledge of groundwater systems was also important for the proper management of liquid waste, recognized here in the 1950s, one notes that groundwater is not mentioned at all in health and safety considerations for the liquid wastes during the 1940s; its importance with regard to the burial grounds was noted before its relevance to the liquid wastes was realized. Indeed, knowledge of subsurface hydrologic systems and their importance simply did not exist then. It is incorrect to criticize this "oversight" today.

However, one might legitimately raise the question that if things were done so conscientiously at the time and done by individuals of high professional calibre, such as we have discussed, then why are we today faced with such great costs to rectify their actions, and why do we automatically presume that things were done carelessly 50 years ago? The answer to the first part is simple. Our knowledge base today is orders of magnitude greater than it was then, and we much more fully understand parameters that control contaminant movement in the environment, as well as the potential dangers associated with contaminants of all kinds, not just the radionuclides. Add to this the directly relevant fact that the standards have changed drastically over the years, and clearly the situation we face today is a result of our own genius and creativity, the product of the natural evolution of scientific endeavors. Why the presumption of guilt? Perhaps because so many of those pioneers who led the way during the war left shortly thereafter, and there was simply less attention directed at the waste disposal challenges, many of which were perceived to have been addressed. Attrition of staff and new directions of programs quickly took their toll, and no written history was left behind. In the absence of a record, recent revelations related to cleaning up the "sins" of the past—revelations by individuals who have no first-hand knowledge of what transpired 50 years ago—automatically spawn thoughts that waste disposal must have been done incorrectly if it is such a problem today. Obviously, this leads one to ponder what we may be doing "correctly" today that will provide a costly or dangerous legacy for our descendants.

Weinberg (1994) addresses this historic issue of waste disposal, reflecting on the fact that standards have changed and pointing out that there should have been more scientific attention given to the waste disposal challenges in the early years. In principle and in hindsight, one finds it difficult to disagree with this observation; however, it is even harder to envision exactly how a more structured waste disposal research effort might have been mounted during the war years in light of the urgency of the time and recalling the simple fact that the detrimental environmental impact of radionuclides was not nearly as well understood then as it is now. After the war, certain staff at ORNL (and certainly elsewhere) did, indeed, strive to develop a more formal research program to address this issue, but exactly what level of active support and leadership was received from the AEC is unclear.

THE FACTS SPEAK: A CONCLUDING STATEMENT

Certainly the perception that liquid wastes were handled in a haphazard or careless fashion during the earliest years of existence of Clinton Laboratories can not be substantiated on the basis of the material presented in this analysis. In fact, the evidence is irrefutable that the highest level of professional concern was devoted to the proper disposal of wastes, on the basis of knowledge at the time and considering the wartime environment in which all had to operate. These facts should speak for themselves. This is not withstanding the speculation that there were improper disposals, however.

Essentially all aspects of the waste disposal challenges faced today were effectively handled during the war. Problem areas were identified and research was proposed to address the problems; waste minimization and treatment, establishment of release limits, and monitoring were all evident. Establishment of authority, policing, and castigation were demonstrated in a highly responsive way.

The individuals with the responsibility for management of the wastes were no light-weights. Many had been recruited because of their expertise to work at the University of Chicago Met Lab, and to later transfer to Clinton Laboratories. Here, they were in positions of authority and responsibility, as were others who arrived via alternate routes. After the war ended, most left and many took prestigious positions elsewhere, whereas others continued their professional careers at Oak Ridge making contributions to the nuclear sciences. In retrospect, it can be said that Clinton Laboratories—and today ORNL—immeasurably benefited from the professional integrity that had been instilled in these leaders; if they had had a mentality oriented toward pure production, the environmental insults could have been much greater, and we could today be facing an ever greater restoration challenge.

The nexus between intent and action has been shown for any number of incidents, although many still deserve more investigation. The realization of this waste disposal effort can be appreciated only by viewing the entirety of the evidence, including the day-to-day actions, rather than considering individual pieces of documentation. No single document has been found that serves as the "Rosetta Stone," alone unlocking the thoughts and actions of the times. A full and complete review of the ORNL Central Files archives, plus interviews with individuals personally knowledgeable of the practices at the time, would be desirable to complete the history. Perhaps this can be accomplished in the future.

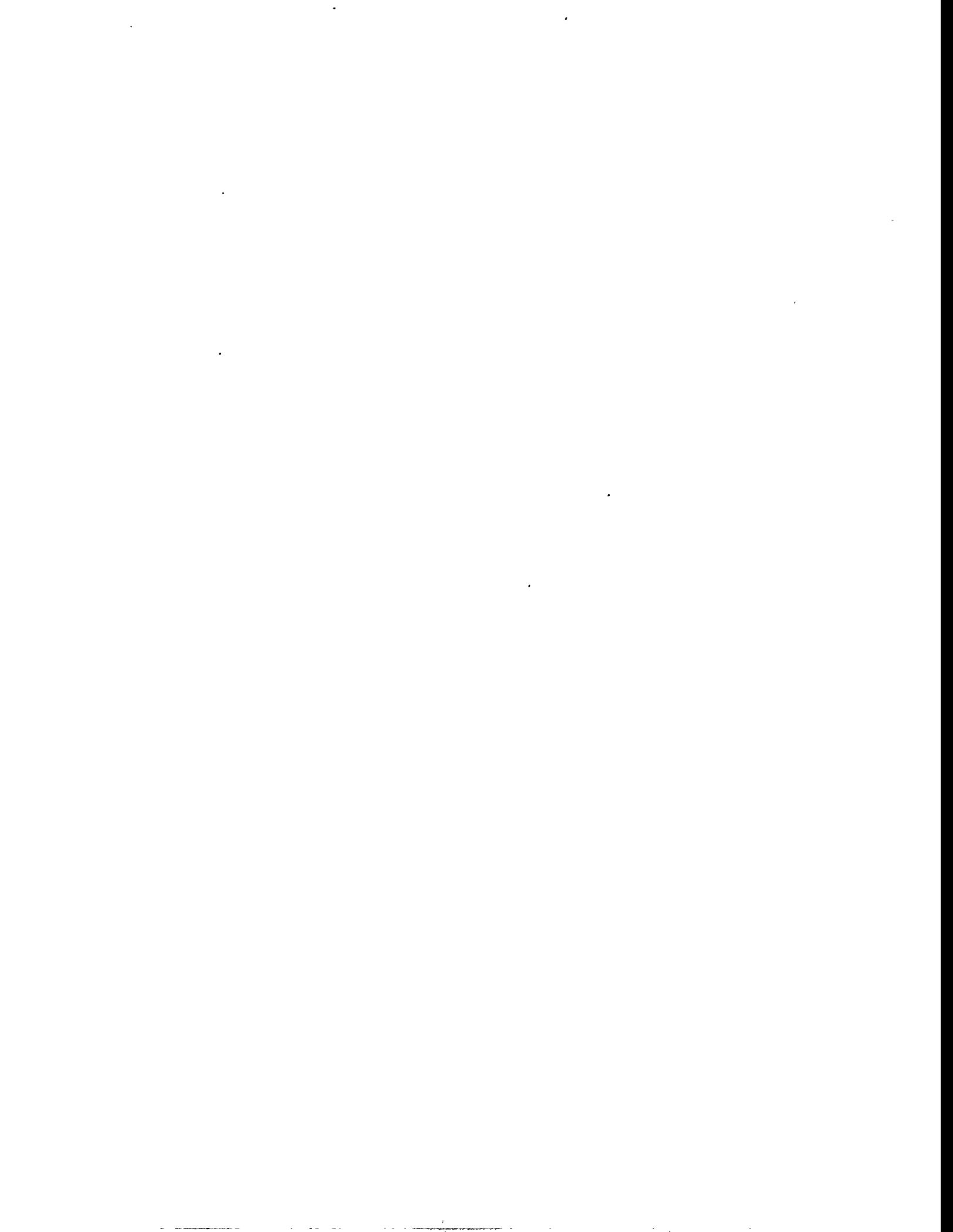
Regardless, the attitudes and achievements of those scientists, engineers, and medical professionals mentioned in this study are deserving of high recognition and acknowledgment. Their integrity and foresight, although perhaps not as visible as that of their counterparts who actually pioneered, designed, and produced the nuclear weapons to end the war, can be viewed to be of equal importance as we look back 5 decades.

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Finally, I wish to thank Virginia Dale, whose imagination was the catalyst that spawned my curiosity to seek out this story. Virginia originally came up with the idea of a presentation on the history of the earth sciences here for the Environmental Sciences Division's 1995 Information Meeting and asked that I prepare such a talk. In doing so and in innocently seeking out a few early records, it quickly became evident that there was more to be told about the attitudes and practices related to waste management than we had previously realized. Thus, this document has emerged.



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APPENDIX

The following material has been excerpted from the daily notes (Wednesday, February 3, 1943) taken by Glenn Seaborg at the Met Lab (Kathren et al. 1994) and is offered to provide information on the chemical processing that was undertaken to separate plutonium from other elements. This description is for laboratory separations, but eventually the bismuth phosphate process was undertaken in the separations buildings at Clinton Laboratories and Hanford. The separation steps were done after the aluminum-clad slugs of irradiated uranium had been dissolved in acid. The notes read:

It is important to ascertain whether 94 [plutonium] can be oxidized in the presence of HF as a part of the wet fluoride separation process. Today Davidson tested the oxidation of 94 in 10% UNH [uranyl nitrate hexahydrate], 0.5 N HNO₃, 2 N HF, and 0.02 M dichromate ion at 65-70 degrees for a half-hour. He finds consistently that 50-60% of the 94 is not oxidized under these conditions.

Today I sent a memorandum (MUC-GTS-12) to C. H. Greenewalt summarizing methods alternative to the Wet Fluoride Method for extracting 94. I indicated that our extraction development group under the supervision of J. E. Willard is working on a number of alternative methods to the Wet Fluoride Method for extracting 94 and listed them as: (1) Sodium Uranyl Acetate Method, (2) Bismuth Phosphate Method, (3) Adsorption Method, (4) Crystallization method for concentrating 94 and (5) Iodate method for separating 94. I described the methods briefly as follows.

1. Sodium Uranyl Acetate Method. Sodium acetate and sodium nitrate are added to an 11% UNH solution to precipitate all of the uranium as sodium uranyl acetate with the 94 remaining in solution in its reduced state; less than 1% of the fission activity comes down with the precipitate. The second step involves oxidation of the 94 which is then precipitated with about 5% of the amount of sodium uranyl acetate which has precipitated in the first step. Over 98% of the 94 and only about 1% of the fission activity is precipitated in this step. The advantages of the method are that it effects a good concentration of the 94 relatively free of the fission activity with very little corrosion hazard and it recovers the uranium in relatively decontaminated form. The disadvantages include the necessity to precipitate and separate all of the uranium before the separation of 94, and the method involves the use and disposal of relatively large amounts of reagents.

2. Bismuth Phosphate Process. Here the 94 in reduced form is coprecipitated with bismuth phosphate from 20% UNH solution with a

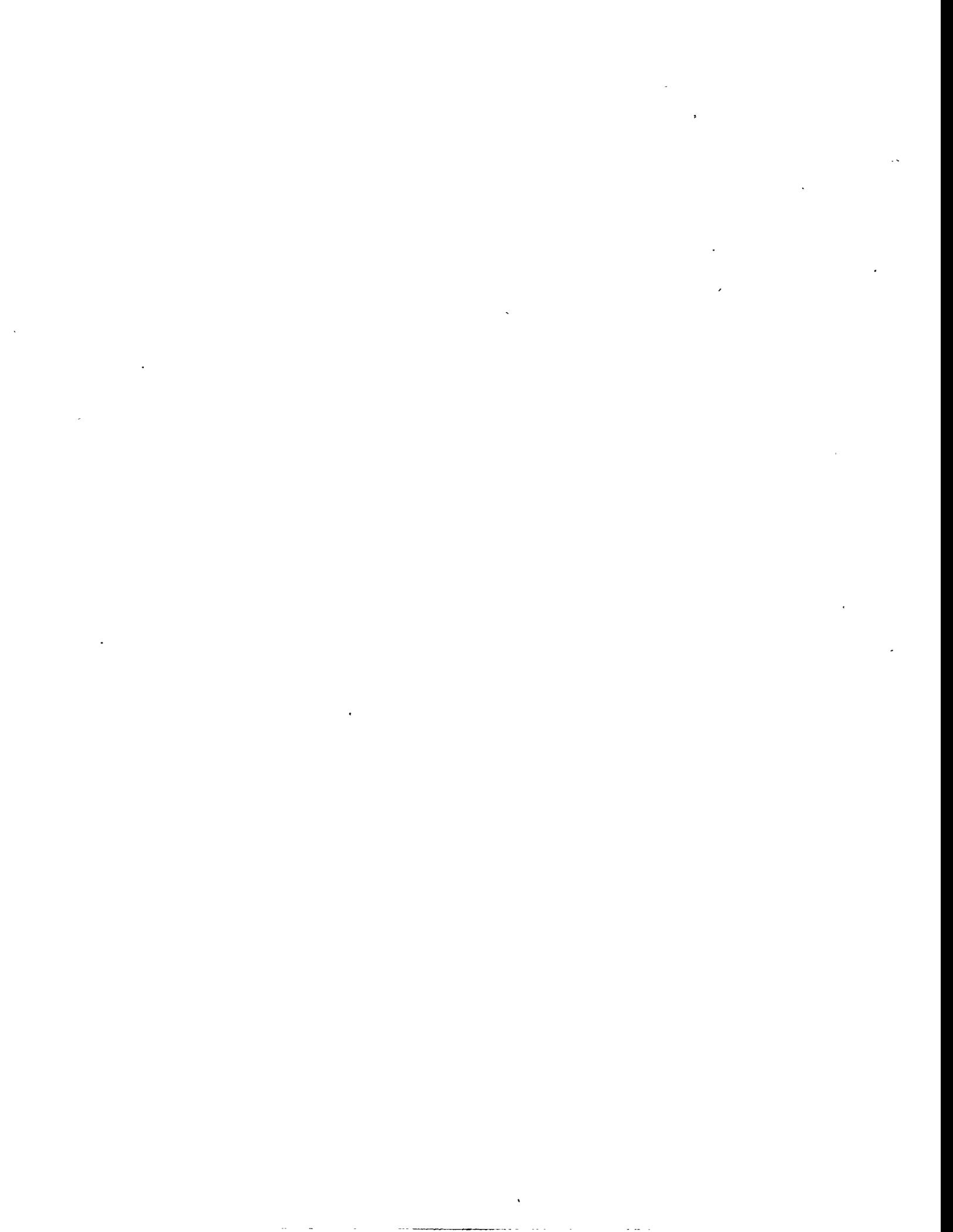
yield of about 98% of the 94 and carrying with it less than 1% of the uranium and about 7% of the gamma activity four days after shutdown of a 40-day bombardment. The precipitation is done at 70 degrees C or above from a solution 1 N in H_2SO_4 . After separation of the bismuth phosphate by filtration or centrifugation, the bismuth phosphate may be dissolved in HCl or HNO_3 . From the HCl solution the 94 may be precipitated with lanthanum fluoride and a wet fluoride oxidation-reduction cycle may be carried out. When the bismuth phosphate is dissolved in HNO_3 , this may be followed by an oxidation-reduction cycle involving bismuth phosphate, a process which is yet to be worked out. The advantages include the fact that the bismuth phosphate precipitate is small and can be readily filtered or centrifuged and the solutions are non-corrosive; also the precipitation of the bismuth phosphate takes place from relatively concentrated solution of UNH so that an oxidation step in the presence of the bulk of the uranium is avoided.

3. Adsorption Method. The 94 in reduced form in 10% UNH solution is passed through a column of diatomaceous earth. This treatment removes all of the 94 and about 5% of the uranium and about 15% of the fission product gamma activity corresponding to one week after shutdown of a 40-day neutron bombardment. The second step consists of washing the column with 5 N HNO_3 during which the leading edge removes the 94 together with the adsorbed uranium and less than 2 % of the original fission product gamma activity. This procedure has worked well when small columns are used, but difficulties have been encountered in attempting to scale up to 100 cm diameter columns.

4. Crystallization method for concentrating 94. In this method the original UNH solution containing 94 in the reduced state with an excess of HNO_3 at 60 degrees C is cooled to room temperature with stirring, resulting in the crystallization of UNH. Under these conditions about 85% of the 94 concentrates in the liquor and by a three-stage counter-current process 99% of the 94 can be concentrated in 8-12% of the uranium. The fate of the fission products has not yet been determined. Methods of separating the crystallized UNH have not been perfected, and the method has not yet proven itself with respect to reliability.

5. Iodate method for extracting 94. The 94 in reduced form is coprecipitated with thorium iodate from a 10% UNH solution. The precipitate carries more than 95% of the 94, less than 5% of the uranium and less than 20% of the fission product radioactivity. The thorium iodate precipitate is separated by centrifugation and is readily soluble in dilute HCl. After solution of the precipitate, the 94 is oxidized by means of dichromate ion and a second thorium iodate precipitation is made which leaves 94 in solution in the oxidized state together with about 5% of the fission product gamma activity and less than 5% of the uranium.

Problems with the method include the use of corrosive HCl. Attempts to use HNO₃ as the dissolving agent lead to problems with the precipitation of uranyl iodate.



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