

A Short History of Hanford Waste Generation, Storage, and Release

R. E. Gephart

October 2003

Prepared for the U.S. Department of Energy under Contract DE-AC06-76RL01830



DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor Battelle Memorial Institute, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or Battelle Memorial Institute. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

PACIFIC NORTHWEST NATIONAL LABORATORY

operated by

BATTELLE

for the

UNITED STATES DEPARTMENT OF ENERGY

under contract DE-AC06-76RL01830

A Short History of Hanford Waste Generation, Storage, and Release

Roy E. Gephart

October 2003

Pacific Northwest National Laboratory Richland, Washington 99352

Prepared for the U.S. Department of Energy Under Contract DE-AC06-76RL01830

A SHORT HISTORY OF HANFORD WASTE GENERATION, STORAGE, AND RELEASE

Roy E. Gephart¹

Pacific Northwest National Laboratory Richland, Washington, USA Publication PNNL-13605

To understand today you have to search yesterday.

-- Author Pearl S. Buck

Summary

Nine nuclear reactors and four reprocessing plants at Hanford produced nearly two-thirds of the plutonium used in the United States for government purposes². These site operations also created large volumes of radioactive and chemical waste. Some contaminants were released into the environment, exposing people who lived downwind and downstream. Other contaminants were stored. The last reactor was shut down in 1987, and the last reprocessing plant closed in 1990. Most of the human-made radioactivity and about half of the chemicals remaining onsite are kept in underground tanks and surface facilities. The rest exists in the soil, groundwater, and burial grounds. Hanford contains about 40% of all the radioactivity that exists across the nuclear weapons complex. Today, environmental restoration activities are under way.

Hanford Reactors and Reprocessing Plants

Nothing like this had ever been attempted before...the great risk involved in designing, constructing, and operating plants such as these without extensive laboratory research and semi-works experience simply had to be accepted.

--General Leslie Groves (Groves 1962)

In the early 1940s, scientists and government officials expressed concern about the hazardous nature of new types of contaminants to be generated from the industrial-scale production of plutonium³. This was a public and worker safety problem facing the Manhattan Project, which was building the world's first nuclear weapon industry in the shortest time. Typical pilot-scale engineering steps normal in constructing a prototype chemical factory were circumvented for expediency. General Leslie Groves, Army

¹ Email address is roy.gephart@pnl.gov

² Hanford has five reprocessing plants. However, only four (T, B, REDOX, and PUREX) were used to reprocess irradiated uranium for plutonium recovery. The fifth (U Plant) was used to recover uranium from tank waste.

³ The half-lives of specific radionuclides mentioned in this paper are listed after the references.

commander of the Manhattan Project, and his staff were also concerned about safety and security, should a major accident happen releasing large quantities of radiation. Because of such issues, the first plutonium facility, Hanford, was located in an isolated, scarcely populated area in the western United States, away from population centers on the East Coast. Hanford also offered other advantages, including availability of electricity, cool river water, a railroad line, sand deposits, and a mild, dry climate. In March 1943, the federal government acquired 1650 square kilometers (640 square miles) in southeastern Washington State⁴. Residents were given about three weeks to leave. Nearly 50,000 workers were hired, and construction began (Gerber 1997).

The first nuclear reactor (B Reactor) and reprocessing plant (T Plant) were built and operating in late 1944—less than 2 years after Enrico Fermi and his team of physicists at the University of Chicago proved that a controlled nuclear chain reaction was possible. Plutonium used in the world's first nuclear explosion at Alamogordo, New Mexico, in July 1945 and in the second atomic bomb dropped on Japan (Nagasaki) in August came from Hanford.

Most of the uranium metal shipped to Hanford was prepared at Fernald, Ohio, and Weldon Spring, Missouri. Further milling, metal cladding, and fuel preparation were completed in the southern part of Hanford in the 300 Area. Nearly 20 million uranium fuel slugs were prepared at Hanford⁵. These resembled short metal cylinders about 23 centimeters (9 inches) long and 3.5 centimeters (1.5 inches) wide. Those used in the last reactor built (N Reactor) were larger.

Between 1943 and 1963, nine reactors were built along the shore of the Columbia River in the 100 Area. Hanford reactors were graphite-moderated because their interior cores, or "piles," were constructed from tens of thousands of finely machined and tightly stacked graphite blocks. Graphite slowed down neutrons, so they were easily captured by the uranium atoms in the fuel.

The reactors were of different sizes and power levels. According to Miller (1976) and Miller and Steffes (1987), between 200 and 390 metric tons (220 and 430 short tons) of uranium fuel were inserted into 1000 to 3220 aluminum tubes running through the reactors' core. Reactor piles measured 7 to 11 meters (23 to 36 feet) on each side. The first reactor had an initial power level of 250 megawatts (Du Pont 1946). Power levels increased to 4400 megawatts when Hanford's largest reactors (KE and KW), built in the mid-1950s, were upgraded in the 1960s.

Water was pumped from the Columbia River, treated in purification plants, and passed through sealed pipes in the reactors to cool them⁶. Chemicals were added to adjust pH,

⁴ Today, the Hanford Site covers 1517 square kilometers (586 square miles)--nearly half the size of Rhode Island.

⁵ Most uranium was unenriched or only slightly enriched: unenriched uranium contained the same 99.3% (by mass) uranium-238 and 0.7% uranium-235 abundance found in nature. Enriched uranium contained slightly more uranium-235.

⁶ The fission of uranium-235 atoms and the resulting radiation releases heated the reactor water to near the boiling point.

prohibit algal growth, remove dissolved solids, and lessen metal corrosion (Foster et al. 1954). The concentration of dissolved and suspended minerals was reduced by filters and chemical coagulation. Helium gas was circulated through the air-sealed pile to remove moisture and reactor-generated gases absorbing neutrons. Later, carbon dioxide was added to the helium flow to reduce radiation-induced swelling of the graphite core.

In the first eight reactors built (B, D, F, H, DR, C, KE, and KW), heated cooling water was stored temporarily in nearby basins before returning to the Columbia River. These were called single-pass reactors because water made one trip through the core. Most radionuclides released in this water were activation products--created when elements dissolved in the water captured neutrons. When the metal cladding surrounding the fuel ruptured, fission products were sometimes released into the river before the water was diverted to a nearby liquid disposal site⁷.

The irradiated uranium fuel was dangerously radioactive and thermally hot. It was stored underwater for a period of time and later transported to reprocessing plants located in the central part of Hanford called the 200 Area.

Originally, 60 to 65 days were considered adequate for uranium fuel to cool down before reprocessing (Cooper 1943; May 1944). This allowed many of the short-lived radionuclides to decay, or at least reach lower concentrations, before the fuel was dissolved. When pressure mounted for plutonium production, fuel was sometimes reprocessed within 30 to 60 days after irradiation (Goldberg in Hevly and Findlay 1998). There is inferential evidence that in 1945 some fuel may have been reprocessed less than three weeks after irradiation. By the late 1950s and early 1960s, the average storage time had increased to between 200 and 250 days (DOE 1997a).

Plutonium was separated from unwanted radionuclides and chemicals using various chemical precipitation and solvent extraction⁸ techniques. This took place inside large rectangular concrete buildings called reprocessing plants or "canyons." Five were built in the 200 Area. Starting in late 1944 and 1945, T and B Plants used a bismuth phosphate batch processing technology. Higher efficiency and safer solvent extraction technologies were relied on in both the REDOX (Reduction and Oxidation) and PUREX (Plutonium and Uranium Extraction) Plants beginning in the 1950s. The fifth plant, U Plant, was operated from 1952 to 1958 to recover uranium from tank waste. It did not reprocess spent fuel.

These reprocessing facilities were up to 305 meters (1000 feet) long, 50 meters (160 feet) wide, and 30 meters (105 feet) tall. Twenty to 40% of their height was underground. The lower level contained the remotely operated chemical reprocessing cells where spent fuel and their metal coatings were chemically dissolved in hot solutions of sodium hydroxide and nitric acid, and the usable plutonium, uranium, and other desired radionuclides separated from waste solutions.

-

⁷ 2000 fuel elements ruptured during the 43 years Hanford reactors operated.

⁸ Solvent extraction is the separation of materials of different chemistries by solvent action. This works because some materials are more soluble in one solvent than another, and therefore, they can be separated.

Some 96,900 metric tons⁹ (106,600 short tons) of uranium were reprocessed at Hanford. Approximately 2 metric tons (2.2 short tons) of fuel were reprocessed each day of operation through T and B Plants. This increased to as high as 33 metric tons (36 short tons) per day in PUREX (Gerber 1993). Nearly 70% of the Hanford's uranium was reprocessed through PUREX (Napier 1992).

Plant	Fuel Reprocessed (metric tons)	Operating History
T and B Plants REDOX Plant PUREX Plant	8100 (8%) 22,400 (23%) 66,400 (69%)	1944-1956 1952-1967 1956-1972, 1983-1990
Total	96,900 (100%)	

Located in the 200 West Area, the Plutonium Finishing Plant made weapons-grade plutonium metal from plutonium-containing solutions shipped from Hanford's reprocessing plants. It operated from 1949 to 1989.

About 67 metric tons¹⁰ (74 short tons) of plutonium were recovered from the uranium fuel reprocessed at Hanford (Usdin 1996). This was 65% of the nation's supply of nearly 103.5 metric tons (114 short tons) of plutonium created inside government reactors¹¹. Hanford plutonium consisted of 54.5 metric tons (60.0 short tons) of weapons-grade plutonium and 12.9 metric tons (14.1 short tons) of fuel-grade plutonium¹². Most of the remaining 35% (36 metric tons or 40 short tons) of plutonium came from the Savannah River Site.

Waste and Nuclear Materials

At the Hanford Engineer Works, the ground disposal of low- and mid-level atomic wastes and the underground tank storage of high-level wastes were first conceived as temporary, wartime expedients. However, increasing volumes of liquid process wastes generated during the ensuing twenty years presented new and unexpected difficulties to site scientists and officials.

--Historian Michele Gerber (Gerber 1997)

⁹ If combined, 96,900 metric tons of uranium metal would form a cube 70 feet per side.

¹⁰ If combined, 67 metric tons of plutonium would form a cube 5 feet per side.

¹¹ Plutonium production in the United States came from 14 government-owned reactors (nine at Hanford and five at Savannah River) and six reprocessing plants (four at Hanford and two at Savannah River). Only four of Hanford's five reprocessing plants recovered plutonium.

¹² Weapons-grade plutonium is made of 94% or greater (by mass) plutonium-239. The remaining 6% is plutonium-240 plus minor amounts of other plutonium isotopes. Fuel-grade plutonium contains 82% to 94% plutonium-239.

Highly radioactive waste from reprocessing was piped to underground tanks. Uncontaminated to slightly contaminated liquids and cooling water were pumped to ditches and ponds. Contaminated water discharged from the reactors was pumped to the nearby soil as well as into the Columbia River. Solid waste was buried in shallow trenches or stored inside facilities. Gaseous effluents were released into the air. More than 1600 waste sites have been identified on Hanford (DOE 1999).

Reprocessing generated the largest volume of waste—more than 85% of the radioactive waste generated during the production of material for nuclear weapons (DOE 1997a). The amount reduced as reprocessing facilities became more efficient and waste management practices improved.

During the 1940s and 1950s, the bismuth phosphate process used in T and B Plants generated an average of 30 cubic meters (8000 gallons) of waste for each metric ton of spent fuel reprocessed (Agnew 1997). Initially, the hexone solvent extraction process used in the REDOX Plant produced 15 cubic meters (4000 gallons) of waste per metric ton of fuel. Process improvements reduced this to 2 cubic meters (500 gallons). Depending on the fuel reprocessing campaign, the tributyl phosphate solvent extraction processes relied upon in the PUREX Plant generated 5 cubic meters (1300 gallons) to less than 1 cubic meter (264 gallons) of waste per metric ton of spent fuel.

Hanford waste and nuclear materials can be informally categorized as contained waste and nuclear materials, buried and stored solid waste, and released waste¹³. Additional radionuclides and chemicals from non-plutonium production activities also exist onsite.

Contained Waste and Nuclear Materials

Contained waste is held inside structures such as underground tanks, buildings, or concrete basins. There are more than 500 waste facilities at Hanford. Some are contaminated; others are not. Largest are the reactors and reprocessing plants. Hanford facilities may contain 1 million curies. These numbers include the radioactivity present in the stack filters of plants.

From 1944 through the late 1980s, Hanford generated nearly 2 million cubic meters ¹⁴ (525 million gallons) of high-level tank waste (Agnew 1997). Liquid evaporation, discharge to the ground, chemical treatment, and tank leakage reduced this volume by 90%—to 204,000 cubic meters (54 million gallons) (Hanlon 2003). This is about 60% of the tank waste existing in the U.S. Department of Energy (DOE) complex. Today, this waste contains about 195 million curies of radioactivity and 220,000 metric tons (240,000 short tons) of chemicals.

Of the 177 tanks onsite, 149 are single-shell carbon-steel tanks built from 1943 to 1964. They range in volume from 210 cubic meters (55,000 gallons) to 3800 cubic meters

¹³ All numbers given in this report are approximations based upon reports, conversations, assumptions, and calculations. Most estimates of curies present are decayed to the year 2000. Many numbers are rounded. ¹⁴ If placed inside standard 20,000-gallon railroad tanker cars, this high-level waste would fill enough tanker cars to make a train 260 miles long.

(1 million gallons) and had a life expectancy of 20 years (DOE 1998a). Sixty-seven have leaked or are suspected to have leaked 3800 cubic meters (1 million gallons) or more of liquids into the underlying sediment (Hanlon 2003; Agnew 1997). The first waste leak was suspected in 1956 and confirmed in 1959 (Gephart and Lundgren 1998). To provide better containment, 28 double-shell carbon-steel tanks were built starting in 1968. Their volumes are 3800 to 4400 cubic meters (1.0 to 1.16 million gallons) each, with design lives as much as 50 years. All 177 tanks are grouped into 18 tank farms.

Over the years, most liquids in single-shell tanks have been pumped into the newer double-shell tanks. Pumping continues to remove the remaining 9,000 cubic meters (2.4 million gallons) of drainable liquid (Hanlon 2003). Thick waste sludge and salt cake are left behind. To date, no double-shell tank has leaked, though the design life of the oldest ones has been reached.

Nuclear materials stored at Hanford include 1936 stainless-steel capsules (cylinders 6.6 centimeters wide by 52 centimeters long—2.6 inches by 20 inches) containing 130 million curies of radioactive cesium and strontium, plus their decay products. These are kept in water-filled pools in the Waste Encapsulation and Storage Facility adjoining B Plant in the 200 Area. From 1967 to 1985, these radionuclides were recovered from tank waste to reduce its curie load so more tank liquids could be released to the soil to make room for new tank waste.

Eighty percent of DOE's remaining irradiated uranium fuel was stored in two aging water filled concrete basins (called the K Basins) at the KE and KW reactors. The fuel consists of about 105,000 assemblies (2100 metric tons or 2300 short tons) containing about 55 million curies of radioactivity¹⁵. The largest of these elements are 66 centimeters (26 inches) long and 6 centimeters (2.4 inches) wide. Each fuel assembly is composed of two hollow uranium tubes, one inside the other. Each weights 25 kilograms (55 pounds). The fuel was irradiated inside N Reactor, moved to the K Basins for storage, and never reprocessed. Some fuel has corroded. As a result, radionuclides are found in the basins' water, bottom sludge, and concrete walls. Past water leaks released radionuclides into the local soil and groundwater.

Hanford has about 11 metric tons (12 short tons) of plutonium onsite, in various forms, compared to the DOE and Department of Defense inventory of 99.5 metric tons (109 short tons) (DOE 1996)¹⁶. Most of the Hanford plutonium rests at three locations—in the spent fuel at K Basins, spent fuel at the Fast Flux Test Facility, and Plutonium Finishing Plant.

¹⁶ Two-thirds (66 metric tons) of this plutonium inventory is with the Department of Defense at Pantex, Texas.

¹⁵ The number of curies stored in the Basins decreases as spent fuel is prepared for interim storage in the Canister Storage Facility in the 200 East Area. As of late 2003, 60% of the spent fuel has been removed, dried, repackaged and placed in the Canister Storage Facility.

Buried and Stored Solid Waste

Solid waste consists of boxes, crates, and drums holding materials such as clothing, rags, and tools contaminated with chemicals and low-levels of radioactivity. Some radioactivity is from long-lived transuranic elements such as plutonium, neptunium, and americium. Seventy-five solid waste burial grounds exist at Hanford (DOE 1997b).

Hanford contains 710,000 cubic meters¹⁷ (25 million cubic feet) of solid waste buried or stored in above-ground facilities. About 97% is buried in landfills. This waste contains 5 to 6 million curies of radioactivity (decayed as of 1999) and 65,000 metric tons (70,000 short tons) of chemicals. An estimated 590 metric tons (650 short tons) of uranium and 360 kilograms (800 pounds) of plutonium exist in solid waste buried in the 200 Area (Wodrich 1991). Some large machines, including contaminated railroad cars and locomotives, are stored in underground tunnels such as adjoining the PUREX Plant.

Before 1970, it was accepted practice to bury solid waste that was not well characterized or sorted. Little effort was taken to separate transuranic and non-transuranic contaminated materials or to enable later waste retrieval, repackaging, and offsite shipment. Solid waste buried since 1970 is better characterized. Today, Hanford solid waste burial takes place in the 200 West Area.

In 1999, the 9-acre 618-11 burial ground, located 13 kilometers (8 miles) north of the 300 Area and adjacent to the Energy Northwest commercial reactor complex, started receiving considerable attention. Groundwater samples revealed elevated levels of tritium—as much as 400 times above drinking water standards (Hartman et al. 2001). This is the highest tritium concentration found onsite in recent years.

From 1962 to 1967, this burial ground received low- and high-activity waste from nuclear fuel experiments and other research conducted in the 300 Area. It may contain 10,000 cubic meters (360,000 cubic feet) of transuranic-contaminated materials having 680 curies of radioactivity—decayed as of 1995 (DOE 2000). Records show waste buried inside trenches as well as in dozens of vertical pipes (steel barrels welded together) and galvanized metal caissons. Some waste contains slivers of irradiated nuclear fuel including 5 to 10 kilograms (11 and 22 pounds) of plutonium and other potentially dispersible radionuclides (Petersen et al. 2001).

Released Waste

Perhaps two million curies of radioactivity and between 90,000 and 270,000 metric tons (100,000 to 300,000 short tons) of chemicals are in the soil and groundwater beneath Hanford. Most of this is in or near the 200 Area.

During the first months of waste generation and release, mildly contaminated liquids were simply discharged to a low spot on the ground (Brown and Ruppert 1950). Most liquids seeped into the sandy soil, adding contaminants to the soil, deep sediment, and

¹⁷ 710,000 cubic meters of waste would form a cube 90 meters (300 feet) per side

¹⁸ 10,000 cubic meters of waste would fill a cube 21 meters (70 feet) per side.

groundwater. Some liquids evaporated, leaving surface residues for plant and animal uptake as well as being dispersed by the wind. This disposal practice created contaminated wetlands.

These practices quickly became unacceptable. Liquids were then pumped down wells. While this got contaminants underground, it also injected them closer to, and sometimes into the underlying aquifer. Within months, well use was discontinued except for the disposal of small waste quantities (Du Pont 1945). Liquids were then pumped to underground box-like structures called cribs, gravel-filled tile fields, French drains (vertical buried concrete pipes), and open trenches later backfilled with gravel. Surface ponds and unlined ditches received about 90% of non-contaminated to slightly contaminated liquids discharged onsite. The rest was released underground via cribs, etc.

Over the last 50 years, 1.5 to 1.7 billion cubic meters (400 billion to 450 billion gallons) of liquids ¹⁹ were discharged to the soil and groundwater—most in the 200 Area (Hartmann and Dresel 1998). This provided a hydraulic driving force that moved some contamination deeper and faster through the subsurface than would have otherwise taken place.

Today, groundwater plumes covering 400 square kilometers (150 square miles) contain a variety of contaminants such as metals (e.g., chromium), chemicals (e.g., nitrates, trichloroethene, carbon tetrachloride), and radionuclides (e.g., tritium, technetium-99, strontium-90). Portions of these plumes have contamination at concentrations above safe drinking-water standards or other recommended guidelines (Poston et al. 2001).

Some of the fastest-moving contaminants are tritium, technetium-99, iodine-129, chromium, and nitrate (DOE 1998a). These migrate close to groundwater speed. Other contaminants, such as cobalt-60, plutonium-239, cesium-137, and strontium-90, take longer to move because they are more tightly bound to the sediment. Certain radionuclides, as well as some non-radioactive metals, undergo strong chemical sorption on the surface of or into the crystalline structure of the sediment. The movement and behavior of all contaminants are strongly influenced by their chemical form, chemical characteristics of the released waste, and the chemistry of the subsurface environment. This is particularly evident in waste leaked from single-shell tanks.

Groundwater travel time along the 16-kilometer (10-mile) stretch from the 200 East Area to the Columbia River is a few decades for the most mobile contaminants. For example, tritium first reached the river 20 years after disposal. Groundwater travel time to the river from the 200 West Area may approach a century or more. This is because the sediment in the underlying aquifer has a lower permeability than that beneath the 200 East Area. For the same reason, contaminated groundwater from the 200 East Area has spread across some 250 square kilometers (100 square miles), while contaminated groundwater from the 200 West Area covers a much smaller area (Hartman et al. 2001).

¹⁹ 1.5 to 1.7 billion cubic meters of liquids equals the average amount of water flowing down the Columbia River every 5 days.

As noted, 67 single-shell tanks have leaked or are suspected to have leaked perhaps 5700 cubic meters (1.5 million gallons) of mostly sodium nitrate- and cesium-contaminated liquids into the underlying sediment. The underground spread of these contaminants is receiving considerable attention because the chemistry of the leaked waste and the properties of the subsurface sediment allowed contaminants to migrate deeper than originally acknowledged (GAO 1998).

In addition to leaked waste, as much as 450,000 to 490,000 cubic meters (120 to 130 million gallons) of tank waste were intentionally discharged to the ground to provide more tank space for newly generated waste coming from the reprocessing plants (Agnew 1997). This occurred between 1946 and 1958. Some waste was treated to remove selected radionuclides. Other waste went untreated. This waste contained more than 275,000 metric tons (300,000 short tons) of chemicals and 60,000 curies of radioactivity (Waite 1991).

Liquids released into the ground have contaminated portions of the underlying aquifer. Examples of contaminants that are of concern in the groundwater beneath portions of the 100 Area include tritium, nitrate, strontium-90, technetium-99, uranium, nickel, chromium, trichloroethene, and plutonium (Hartman et al. 2001). Beneath portions of the 200 Areas, examples of groundwater contaminants of concern include carbon tetrachloride, nitrate, tritium, iodine-129, strontium-90, technetium-99, cobalt-60, cesium-137, uranium, and plutonium (Hartman et al. 2001).

Uranium fuel preparation in the 300 Area also generated contamination. This included trichloroethene and released acids (nitric, sulfuric, and hydrofluoric) containing uranium, zirconium, copper, and beryllium (DOE 1997a). Contaminants were discharged to surface ponds and trenches located along the banks of the Columbia River. About 25% of Hanford's solid waste lies in or near the 300 Area.

Past Radionuclide Releases to the Atmosphere and Columbia River

The following measurements on thyroid activity of sheep in regions adjacent to the Plant [Hanford] were obtained under conditions which avoided the excitement of public curiosity.

--K. E. Herde (1946)

Today, small amounts of radionuclides are released from Hanford into the air and the Columbia River. During 2000, the average member of the local public received a radiation dose of about 0.0008 millirem²⁰ from Hanford releases (Poston et al. 2001). A maximally exposed individual is estimated to have received 0.014 millirem during the same year.

²⁰ Rem is an acronym for *roentgen equivalent man*; a unit of <u>ionizing</u> radiation dose that indicates the potential for damage to human cells. A millirem is one thousandth of a rem. The average radiation dose a member of the local public receives from the natural environment each year is 300 millirem. This is the same dose received by an average U.S citizen. A radiation dose of 0.0008 millirem per year equals what an average resident receives in about 1 minute from the natural environment.

Radionuclide releases and dose were much larger in the past. Over 140 million curies of radionuclide were released into the atmosphere and Columbia River between the mid-1940s and the mid-1960s.

Radionuclide Releases to the Atmosphere

Thirty-two million curies were released into the atmosphere from stack gases venting from operating reactors and reprocessing plants (HEDR 1994).

Twelve million curies came from the reactors. Heeb (1994) reported that between 1944 and 1970, 10 million curies were discharged into the atmosphere from the operation of the first eight reactors. Later, an additional 2 million curies came from N Reactor. More than 99% of these releases were argon-41 with small amounts of carbon-14 and tritium. HEDR (1994) and Heeb (1994) noted these releases resulted in an increased radiation dose of between 2 and 4 millirem per year. This was about a 1% increase from natural background radiation levels for people living near Hanford.

Between 1944 and 1972, some 20 million curies were released into the atmosphere from reprocessing plants (Napier 1992; HEDR 1994; Heeb 1994). Most came from krypton-85 (18.5 million curies).

Napier (1992) reported that six radionuclides released into the atmosphere during reprocessing operations contributed 99% of the potential radiation dose to humans from air pathways. These radionuclides were iodine-131, ruthenium-103, ruthenium-106, strontium-90, plutonium-239, and cerium-144.

However, the 739,000 curies of iodine-131 dominated the dose to humans. Most iodine releases occurred between 1944 and 1947 before stack filters were installed on T and B Plants. People living adjacent to and downwind of Hanford received the highest dose. The range in dose to the thyroid of an adult at a maximally exposed location over a few-year period was 10 to 150 rad²¹ (equal to a radiation dose of 10,000 to 150,000 millirem) (HEDR 1994). The range in dose to the thyroid for a child at a maximally exposed location was higher—54 to 870 rad (equal to a radiation dose of 54,000 to 870,000 millirem). Final thyroid disease study results were released in 2002 (CDC and Fred Hutchinson Cancer Research Center 2002).

A special case of airborne contamination took place in 1949. In September of that year, U.S. officials were startled by the Soviet Union's first nuclear bomb explosion. It was unannounced but first detected in air samples collected by U.S. planes near the Kamchatka Peninsula in the eastern Soviet Union (Rhodes 1995). Officials thought the Soviets were reprocessing young or "green" spent fuel—spent fuel reprocessed less than 3 weeks after being pulled from a reactor.

²¹ Rad is an acronym for *radiation absorbed dose*; a unit that measures the amount of <u>ionizing</u> radiation (energy) absorbed by any material, such as human tissue.

This initiated a U.S. experiment to estimate Soviet plutonium production. The experiment, called the Green Run, took place in early December when 1 ton of spent fuel was reprocessed in T Plant just 16 days after irradiation. Stack filters were circumvented and the reprocessed fuel released radioactive gases, including 11,000 curies of iodine-131 and xenon-133. Radionuclides were tracked as they drifted across the landscape. Analyses permitted a comparison of air samples, and therefore plutonium production, between Hanford and the Soviet site.

Radionuclide Releases to the Columbia River

An estimated 110 million curies of radiation were released to the Columbia River from 1944 to 1971 during the operation of Hanford's first eight reactors (Heeb and Bates 1994)²². These were single-pass reactors where river water was used to cool the reactor's core and then returned to the river. The ninth reactor recirculated much of its cooling water.

Most radionuclides were in small amounts or had short-half-lives. For example, manganese-56, with a half-life of 2.6 hours, contributed nearly two-thirds of the curies released. The five radionuclides contributing most (94%) of the estimated radiation dose to people living downstream were:

Radionuclide	<u>Curies</u>
sodium-24	12,600,000
phosphorus-32	230,000
neptunium-239	6,300,000
zinc-65	490,000
arsenic-76	2,500,000

Many of these radionuclides were activation products. Some additional radiation releases resulted from the failure of about 2000 fuel slugs between 1951 and 1965.

Longer-lived radionuclides were also released into the river and are now buried in the sediment, especially in slack waters near islands or downstream dams. Examples include cobalt-60, strontium-90, cesium-137, uranium-238, and plutonium-238, -239, and -240 (Patton 1998). According to the Washington State Department of Health (Wells 1994), if these sediments were dredged and placed on top of the ground, the maximum average yearly radiation dose to a person over a lifetime would be 1.6 millirem.

The largest radionuclide releases to the Columbia River occurred between 1956 and 1965 when most reactors were operating. An average of 10,000 to 12,000 curies per day was discharged (HEDR 1994). In 1963, an average of 14,500 curies were released each day (Gerber 1997). Larger peak releases took place during any single day.

-

²² When discharged, reactor water was 67°C (120°F) higher than river temperature (Foster et al. 1954). When river flows were low and all reactors were operating, the ambient river temperature for the first 90 kilometers (56 miles) downstream was increased by about 3°C (5°F) (Becker 1990).

During these times, a typical nearby resident may have received a yearly radiation dose increase of 1 to 5 millirem per year (HEDR 1994). On the other hand, a person who was a significant user of the river and who ate large quantities of fish may have received a 50-to 130-millirem increase in their annual dose—a 15 to 45% increase compared to local background levels. Some Native Americans may have received ten times this radiation dose because they ate more fish from the river than the general population (Till et al. 2002).

Today, the amount of radioactivity flowing down the Columbia River is significantly lower. Low but detectable amounts of radionuclides, chemicals such as nitrate, and various metals still enter the river. For example, water analyses reported by Patton and Poston (2000) detected 10 curies of radioactivity dissolved in Columbia River water as it entered Hanford. About 98% of this radioactivity comes from tritium found in rainfall, and therefore in the river water²³. Most of the remaining radioactivity is from naturally occurring uranium and potassium.

As the river passes Hanford, some contaminated groundwater enters the river. Each day downstream of Hanford, about 20 curies of natural and human-made radioactivity are detected. Compared to up-river water samples, greater than 99% of this 10-curie-per-day increase comes from tritiated Hanford groundwater. Though the amount is small, iodine-129, and sometimes strontium-90, concentrations are slightly higher downstream.

Hanford Today

The greatest immediate need is to recognize the importance of factoring the future into present decision making.

--National Academy of Public Administration (1997)

Hanford is perhaps the largest and most complex environmental cleanup site in the United States. It's a macrocosm of the environmental and nuclear material management problems facing the federal government at the 134 sites once used for uranium mining, nuclear weapons research, material production, and bomb testing (DOE 1998b). Of the 1 billion curies of human-made radioactivity that exist across the nuclear weapons complex, 40% of it is at Hanford.

Today, about 390 million curies²⁴ (mostly cesium and strontium) and about 375,000 to 555,000 metric tons (410,000 to 610,000 short tons) of chemicals remain onsite from the plutonium production mission. In addition, about 40 million curies is onsite from other government and commercial activities. Thus, Hanford contains about 430 million curies of human-made radioactivity.

The number of curies onsite decreases by about 10 million each year due to radioactivity decay. In the future, this amount of decrease will become smaller.

²³ Most of the tritium in rain comes from the atmospheric testing of nuclear weapons during the 20th century.

Locations containing waste and nuclear material associated with the plutonium mission are:

Waste or Material	Volume ²⁵	<u>Curies</u>	Chemicals ²⁶
Tank Waste	204,000 m ³	195 million	220,000 metric tons
Solid Waste	710,000 m ³	6 million	65,000 metric tons
Soil/Groundwater	1.0 billion m ³	2 million	90,000 to 270,000 metric tons
Facilities	5 million m ³	1 million	
Nuclear Material ²⁷	700 m^3	185 million	

The 39 to 44 million curies in facilities and burial grounds not associated with Hanford's original plutonium-production mission include:

- 2 million curies in spent fuel stored inside T Plant from the decommissioned Shippingport, Pennsylvania, commercial nuclear reactor
- 5 million curies of activation products inside the Navy reactors stored in a solid waste burial ground in the 200 East Area
- 4 million curies of commercial low-level waste in the U.S. Ecology landfill located between the 200 East and 200 West Areas
- 15 to 20 million curies (as of the year 2000) of irradiated fuel inside the Fast Flux Test Facility
- 13 million curies of special waste (e.g., 7 to 8 million curies contained in the "German vitrified logs" removed from a 300 Area building and stored in the 200 West Area).

_

²⁵ Multiply cubic meters (m³) by 35.3 to calculate cubic feet or 264 to calculate gallons.

²⁶ Multiply metric tons by 1.1 to estimate short (english) tons.

²⁷ These 185 million curies are contained in the spent fuel (55 million curies) found in the K Basins and Canister Storage Facility plus the cesium and strontium capsules (130 million curies) stored in the Waste Encapsulation and Storage Facility.

Hanford Cleanup

I'm often asked why it has taken so long to clean up and stabilize Hanford's radioactive and hazardous wastes. These queries generally come from individuals who have never seen the site, cannot envision the size and complexity of its buildings and their contents, and have little understanding about the short and long-term risks to workers, the public, or the environment.

---Past chair of HAB²⁸ Merilyn Reeves (Hanford Advisory Board 1999)

Hanford was subject to a flurry of public concerns in the 1980s spearheaded by increased public awareness of waste management practices, declassification of formally secret documents, study of the site as a possible geologic repository for radioactive waste, and the application of environmental protection and cleanup legislation to federal sites.

In May 1989, DOE, the U.S. Environmental Protection Agency, and the Washington State Department of Ecology entered into an agreement to cleanup Hanford and better manage the treatment, storage, and disposal of hazardous materials. This agreement, known as the Tri-Party Agreement, was the first cleanup agreement signed by DOE.

The plutonium production era officially ended in 1990 when the PUREX Plant shut down. Along with it went the goals of plutonium production and the uses of Hanford land and facilities for that purpose. The fundamentals of chemical and nuclear-based processes that once framed decision-making were now steeped in a new set of technical, environmental, health, and social issues. Independent oversight grew and public input to decision-making became commonplace. The Hanford mission and culture had changed dramatically.

References

Agnew SF. 1997. *Hanford tank chemical and radionuclide inventories: HDW model Rev. 4.* LA-UR-96-3860, Los Alamos National Laboratory, Los Alamos, New Mexico.

Becker CD. 1990. Aquatic bioenvironmental studies: the Hanford experience 1944-84. Elsevier, New York.

Brown RE and HG Ruppert. 1950. *The underground disposal of liquid wastes at the Hanford Works Washington*. HW-17088, General Electric Company, Hanford Works, Richland, Washington.

Centers for Disease Control and Prevention (CDC) and Fred Hutchinson Cancer Research Center. June 21, 2002. *A Guide to the Hanford Thyroid Disease Study—Final Report.* Atlanta, Georgia.

_

²⁸ Hanford Advisory Board

- Cooper CM. December 31, 1943. *Proposed program for study of stack dispersion at Hanford*. Letter to RS Stone. DUH-47, U.S. Department of Energy Public Reading Room, Richland, Washington.
- DOE (U.S Department of Energy). 2000. Buried transuranic-contaminated waste information for U.S. Department of Energy facilities. U.S. Department of Energy-Headquarters, Washington D.C.
- DOE (U.S. Department of Energy). January 26, 1999. *DOE resumes plutonium stabilization at Hanford plutonium finishing plant*. Press release, U.S. Department of Energy-Richland Operations Office, Richland, Washington.
- DOE (U.S. Department of Energy). 1998a. *Groundwater/vadose integration project specification*. DOE/RL-98-48, Draft C, U.S. Department of Energy-Richland Operations Office, Richland, Washington.
- DOE (U.S. Department of Energy). 1998b. *Accelerating cleanup: paths to closure*. DOE-EM-0362, U.S. Department of Energy, Office of Environmental Management, Washington, D.C.
- DOE (U.S. Department of Energy). 1997a. *Linking legacies: connecting the Cold War nuclear weapons production processes to their environmental consequences*. DOE/EM-0319, U.S. Department of Energy, Washington, D.C.
- DOE (U.S. Department of Energy), 1997b. *Hanford site environmental management specifications: Section 4.2.2 waste management project.* DOE/RL-97-55, Rev. 1, U.S. Department of Energy-Richland Operations Office, Richland, Washington.
- DOE (U.S. Department of Energy). 1996. *Plutonium: the first 50 years, United States plutonium production, acquisition, and utilization from 1944 through 1994.* U.S. Department of Energy, Washington, DC.
- Du Pont (E.I. Du Pont De Nemours & Co), 1946. *Operations of Hanford Engineer Works, History of the Project, book 11*. HAN-73214. Du Pont, Hanford Engineer Works, Richland, Washington.
- Du Pont (E.I. Du Pont De Nemours & Co), 1945. *Memoranda for the file, book 12*. Du Pont, Hanford Engineer Works, Richland, Washington.
- Foster RF, JJ David, and PA Olson, October 11, 1954. *Studies on the effect of the Hanford reactors on aquatic life in the Columbia River*. HW-33366, General Electric, Hanford Atomic Products Operation, Richland, Washington.
- GAO (General Accounting Office). 1998. *Nuclear waste: understanding of waste migration at Hanford is inadequate for key decisions*. GAO/RCED-98-80, General Accounting Office, Washington, D.C.

Gephart RE and RE Lundgren. 1998. *Hanford tank cleanup: a guide to understanding the technical issues*. Battelle Press, Columbus, Ohio.

Gerber MS. 1997. On the home front: the Cold War legacy of the Hanford nuclear site. University of Nebraska Press, Lincoln, Nebraska.

Gerber MS. 1993. *Brief history of the PUREX and UO₃ facilities*. WHC-MR-0437, Westinghouse Hanford Company, Richland, Washington.

Goldberg S. 1998. *General Groves and the atomic west: the making and meaning of Hanford.* in Hevly B and JM Findlay. *The Atomic West.* Center for the Study of the Pacific Northwest in association with the University of Washington Press, Seattle, Washington.

Groves LR. 1962. *Now it can be told: the story of the Manhattan Project*. Harper and Brothers, New York.

Hanford Advisory Board. 1999. *Hanford Advisory Board progress report fiscal year 1999: managing the focus on cleanup in a time of transition*. Available online at http://www.hanford.gov/boards/hab/advice/adviceindex.htm

Hanlon BM. 2003. Waste tank summary report for month ending July 31, 2003. HNF-EP-0182-184, CH2MHILL Hanford Group, Inc., Richland, Washington.

Hartman MJ and PE Dresel (editors). 1998. *Hanford site groundwater monitoring for fiscal year 1997*. PNNL-11793, Pacific Northwest National Laboratory, Richland, Washington.

Hartmann MJ, LF Morasch, and WD Webber (editors). 2001. *Hanford site groundwater monitoring for fiscal year 2000*. PNNL-13404, Pacific Northwest National Laboratory, Richland, Washington.

HEDR (Hanford Environmental Dose Reconstruction Project). April 21, 1994. *Summary:* radiation dose estimates from Hanford radioactive material releases to the air and the Columbia River. Issued by the Technical Steering Panel of the Hanford Environmental Dose Reconstruction Project.

Heeb CM. 1994. *Radionuclide releases to the atmosphere from Hanford operations,* 1944-1972. PNWD-2222 HEDR, Pacific Northwest Laboratory, Richland, Washington.

Heeb CM and DJ Bates. 1994. *Radionuclide releases to the Columbia River from Hanford operations*, 1944-1971. PNWD-2223 HEDR, Pacific Northwest Laboratory, Richland, Washington.

Herde KE. March 1, 1946. I^{131} accumulation in the thyroid of sheep grazing near H.E.W. Letter to JW Healy, HW 3-3455, General Electric Company, Hanford Atomic Products Operation, Richland, Washington.

Lide DR, ed. 1995. CRC handbook of chemistry and physics: 76th edition for 1995-1996. CRC Press, New York.

May K. 1944. *Time interval between discharge of metal from the pile and processing in canyon*. Letter to DF Babcock. DUH-160, U.S. Department of Energy Public Reading Room, Richland, Washington.

Miller NR. March 4, 1976. *Directory of ERDA-owned nuclear reactor*. Letter to O.J. Bennett (U.S. Energy Research and Development Administration-Richland Operations Office), United Nuclear Industries, Richland, Washington.

Miller RL and JM Steffes. 1987. *Radionuclide inventory and source terms for the surplus production reactors at Hanford*. UNI-3714, Rev. 1, UNC Nuclear Industries, Richland, Washington.

Napier BA. 1992. *Determination of radionuclides and pathways contributing to cumulative dose*. BN-SA-3673 HEDR, Pacific Northwest Laboratory, Richland, Washington.

National Academy of Public Administration. 1997. *Deciding for the future: balancing risks, costs, and benefits fairly across generations.* National Academy of Public Administration, Washington, D.C.

Patton GW. 1998. "Surface water and sediment surveillance." Contained in *Hanford site* environmental report for calendar year 1997. PNNL-11795, Pacific Northwest National Laboratory, Richland, Washington.

Patton GW and TM Poston. 2000. "Appendix A: additional monitoring results for 1999." In *Hanford site environmental report for calendar year 1999*. PNNL-13230, Pacific Northwest National Laboratory, Richland, Washington.

Peterson, SW, RJ Cameron, MD Johnson, and MJ Truex. March 2001. *Technology Alternatives Baseline Report for the 618-10 and 618-11 Burial Grounds, 300-FF-2 Operable Unit.* BHI-01484, Rev. 0, Bechtel Hanford Company, Richland, Washington.

Poston TM, RW Hanf, RL Dirkes, and LF Morasch. 2001. *Hanford site environmental report for calendar year 2000*. PNNL-13487, Pacific Northwest National Laboratory, Richland, Washington.

Rhodes R. 1995. *Dark Sun: The making of the hydrogen bomb*. Touchstone, New York, New York.

Till, JE, et al. 2002. A Risk-based Screening Analysis for Radionuclides Released to the Columbia River From Past Activities at the U.S. Department of Energy Nuclear Weapons Site in Hanford, Washington. Risk Assessment Corporation, Neeses, South Carolina.

Usdin, S. February 9, 1996. "DOE lifts the plutonium veil," *Nuclear Remediation Week*. King Publishing Group, Washington, D.C.

Waite JL. 1991. *Tank wastes discharged directly to the soil at the Hanford Site*. WHC-MR-0227, Westinghouse Hanford Company, Richland, Washington.

Wells D. 1994. Special report: radioactivity in the Columbia River sediments and their health effects. Washington State Department of Health, Olympia, Washington.

Wodrich D. 1991. Historical perspective of radioactivity contaminated liquid and solid wastes discharged or buried in the ground at Hanford. TRAC-0151-VA, Westinghouse Hanford Company, Richland, Washington.

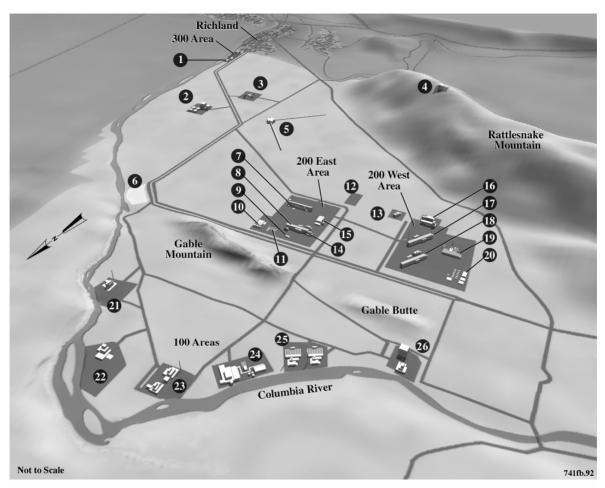
The Half-Lives of Radionuclides

This is a list of the natural and human-made radionuclides referred to in this report and their half-life (Lide 1995).

Radionuclide	Half-Life	
Argon-41	1.8 hours	
Carbon-14	5715 years	
Cerium-144	285 days	
Cesium-137	30 years	
Cobalt-60	5.3 years	
Iodine-129	17 million years	
Iodine-131	8.0 days	
Krypton-85	10.7 years	
Manganese-56	2.6 hours	
Neptunium-239	2.4 days	
Phosphorus-32	14 days	
Plutonium-238	87.7 years	
Plutonium-239	24,000 years	
Plutonium-240	6537 years	
Potassium-40	1.26 billion years	
Ruthenium-103	39 days	
Ruthenium-106	1.0 year	
Sodium-24	15 hours	
Strontium-90	29 years	
Technetium-99	213,000 years	
Tritium	12.3 years	
Uranium-235	704 million years	
Uranium-238	4.46 billion years	
Xenon-133	5.2 days	

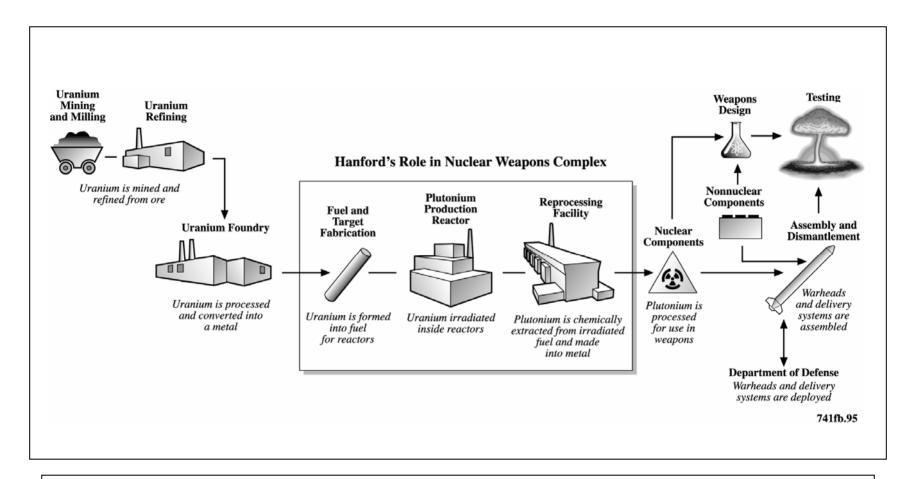


Hanford Site Location. The Hanford Site covers 1517 square kilometers (586 square miles), an area about half the size of Rhode Island. It's located along the banks of the Columbia River in the desert of southeastern Washington State.

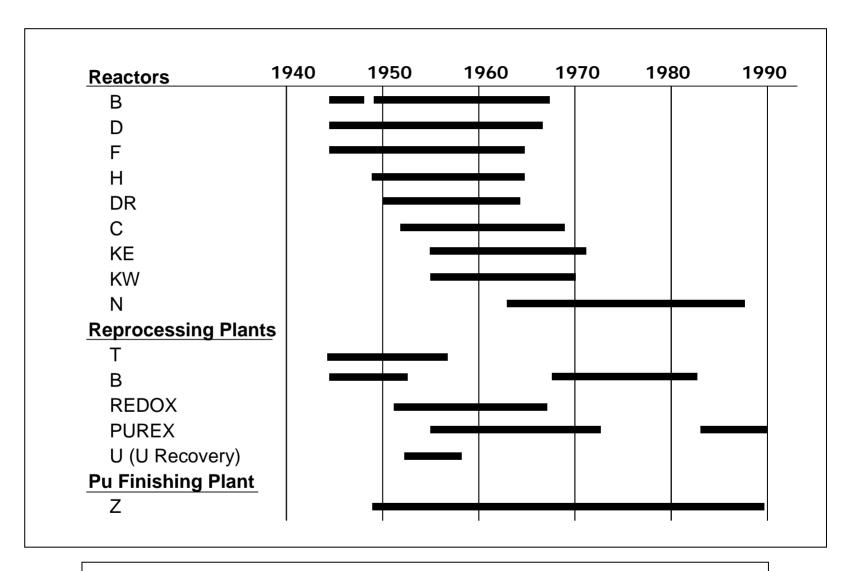


- 1. 300 Area Liquid Effluent Treatment Facility
- 2. Commercial Operating Nuclear Power Plant
- 3. Fast Flux Test Facility
- 4. Observatory
- 5. Laser Interferometer Gravitational Wave Observatory (LIGO)
- 6. Old Hanford Townsite
- 7. Plutonium-Uranium Extraction (PUREX) Plant
- 8. B Plant
- 9. Prototype Surface Engineered Barrier
- 10. 200 Area Liquid Effluent Treatment Facility
- 11. Submarine Burial
- 12. U.S. Ecology Commercial Solid Waste Site
- 13. Environmental Restoration Disposal Facility (ERDF)

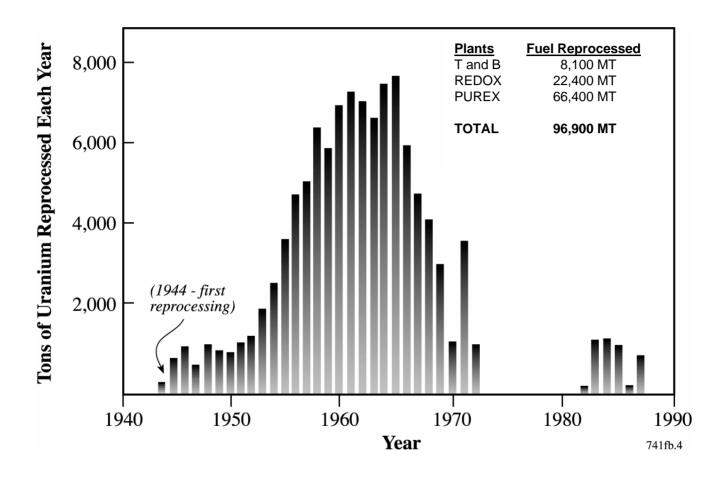
- 14. Waste Encapsulation and Storage Facility (WESF)
- 15. Canister Storage Facility
- 16. Reduction-Oxidation (REDOX) Plant
- 17. U Plant
- 18. T Plant
- 19. Plutonium Finishing Plant
- 20. Waste Receiving and Processing (WRAP) Facility
- 21. F Reactor
- 22. H Reactor
- 23. D and DR Reactors
- 24. N Reactor
- 25. KE and KW Reactors; Cold Vacuum Drying Facility
- 26. B and C Reactors



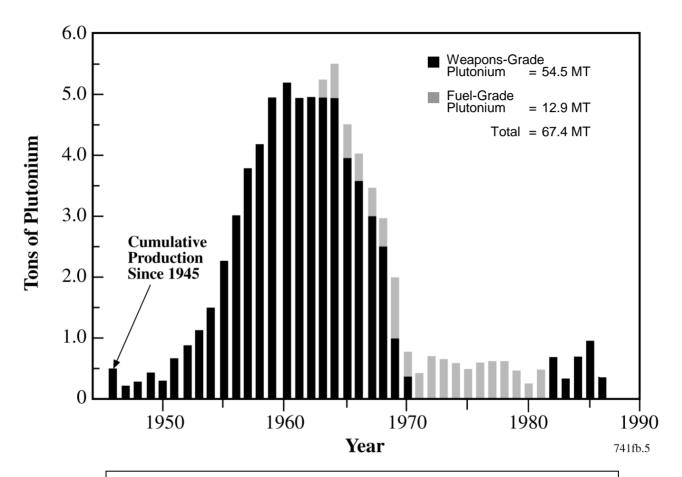
Hanford's Past Mission. The role of Hanford in the nuclear weapons complex was to create plutonium and separate it chemically from the irradiated uranium fuel. More than 130 other sites across the United States were used for uranium mining through weapon testing to support the development of nuclear materials and weapons.



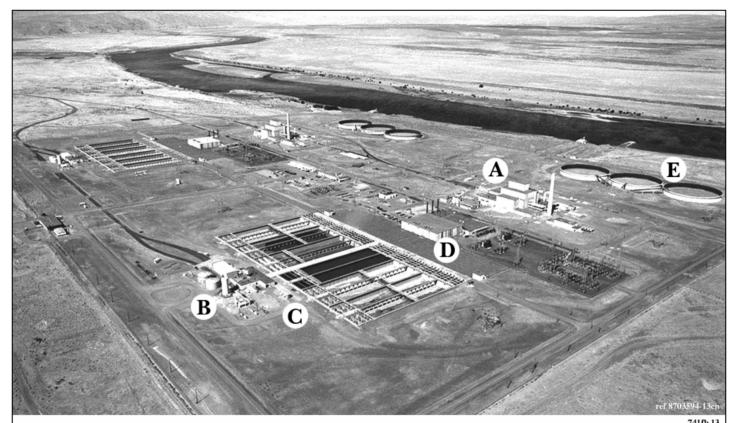
Operating History. Nine nuclear reactors, five reprocessing plants, and other support facilities once operated at Hanford to produce, separate, and purify plutonium.



Uranium Reprocessed. Most of the 96,900 metric tons (107,000 short tons) of uranium fuel at Hanford were reprocessed in the 1950s and 1960s. The PUREX Plant reprocessed 70% of this fuel.



Plutonium Produced. Hanford produced 67 metric tons (74 short tons) of plutonium. This was 65% of the plutonium produced in government-owned reactors in the United States.



A. Reactor Building

B. Water Treatment Plant

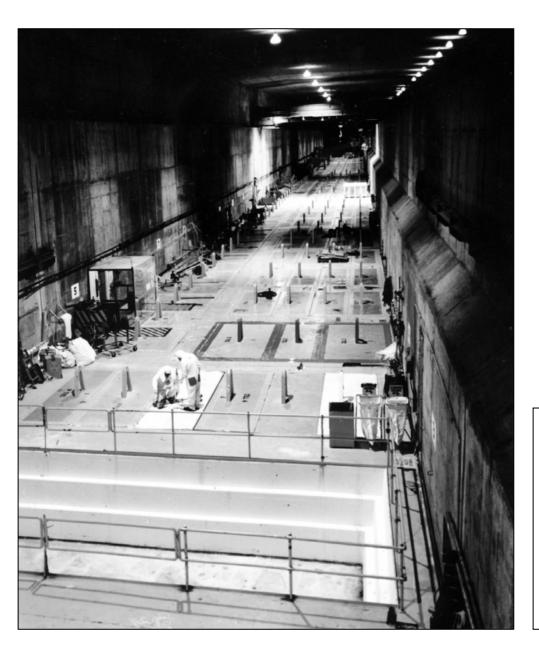
C. Treated Water Storage Basins

- D. Pump House Supplying Cooling Water to Reactor
- E. Water Retention Tanks for Radiation and Heat Decay

Two Reactors. This is an aerial view of the KE and KW Reactors and their support structures along the Columbia River. Today, two basins at these reactors store 80% of the Department of Energy's spent fuel.

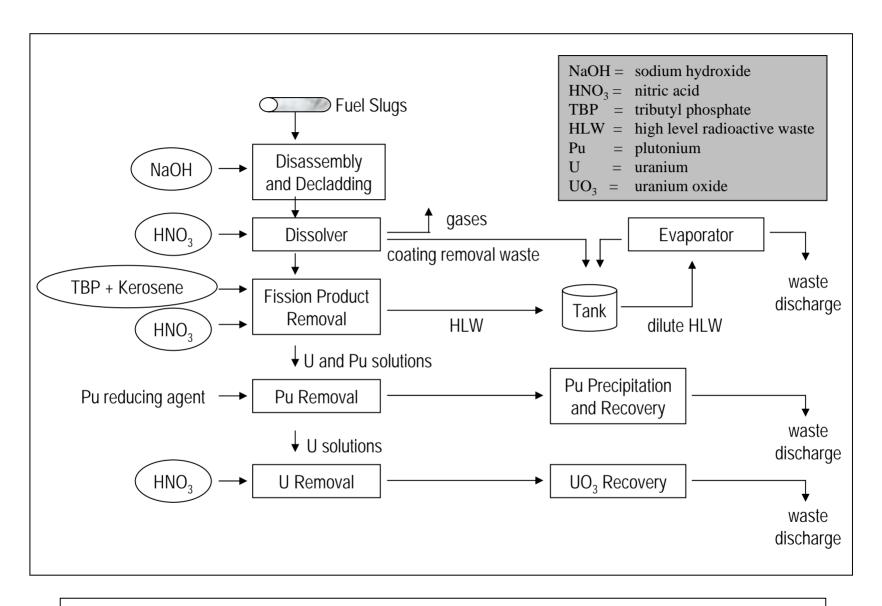


Reprocessing Plant. Five plants were built for chemically reprocessing irradiated spent fuel and recovering plutonium. Four were used for that purpose. The PUREX Plant was the largest. It's about 304 meters (1000 feet) long, 18 meters (60 feet) wide, and 30 meters (100 feet) tall. Forty percent of its height is underground.



Inside a Reprocessing Plant.

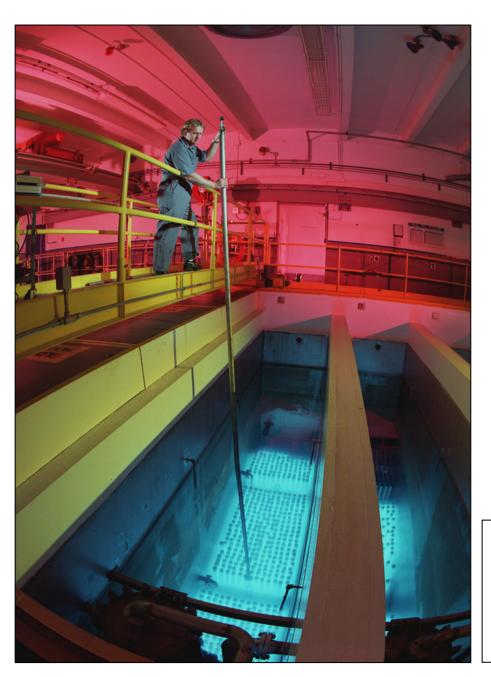
The insides of reprocessing plants resemble long, narrow canyons with concrete and steel walls. Beneath the floors are the heavily shielded and remotely operated chemical reprocessing laboratories (cells) where irradiated uranium fuel was dissolved and plutonium extracted.



Reprocessing of Uranium Fuel. This illustration summarizes the reprocessing approach used in the PUREX Plant. It outlines where waste was generated and disposed.



Tank Construction. Workers are installing the steel roof on a double-shell tank built in 1984. Steel-reinforced concrete was later placed around the tank before it was buried under 3 meters (10 feet) of sediment. Metal pipes, called risers, provide access into the top of each tank.

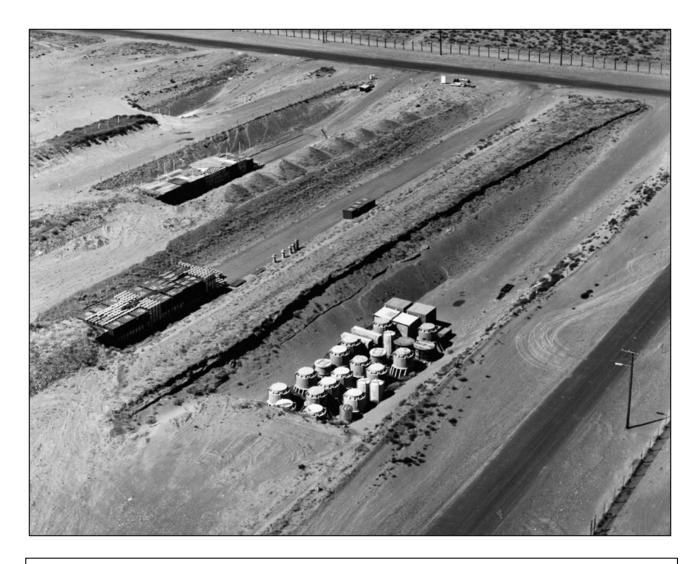


Cesium and Strontium Capsules.

Water-filled pools inside the Waste Encapsulation and Storage Facility, adjoining B Plant, contain 1936 stainless-steel capsules holding intensely radioactive cesium and strontium.

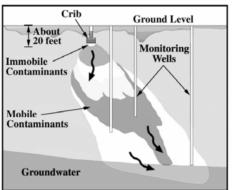


Spent Fuel in K Basins. This photograph shows a portion of the 2100 metric tons (2300 short tons) of spent fuel stored in two water-filled concrete basins at the KE and KW Reactors. The picture is looking through about 4 meters (14 feet) of water at the 6-centimeter (2.4-inch) fuel elements grouped inside individual canisters. As of late 2003, 60% of this fuel has been retrieved, dried, repackaged and placed into the Canister Storage Facility.

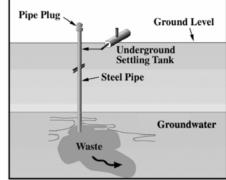


Solid Waste Burial. This photograph shows an open trench receiving solid waste in the 200 West Area burial grounds. Afterwards, burial grounds are covered with soil.

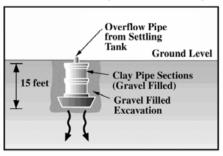
Cribs (used 1944-1990s)



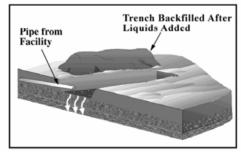
Injection Wells (used 1945-1955; one to 1980)



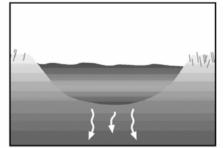
French Drains (used 1944-1980s)



Specific Retention Trenches (used 1944-1973)

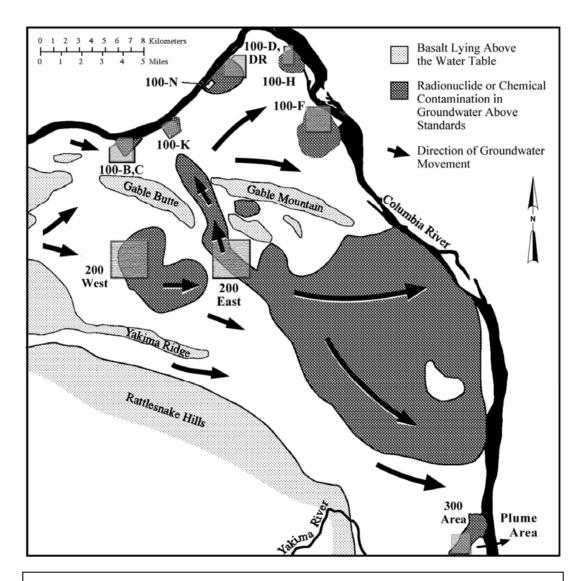


Ponds and Ditches (used 1944-1990s)

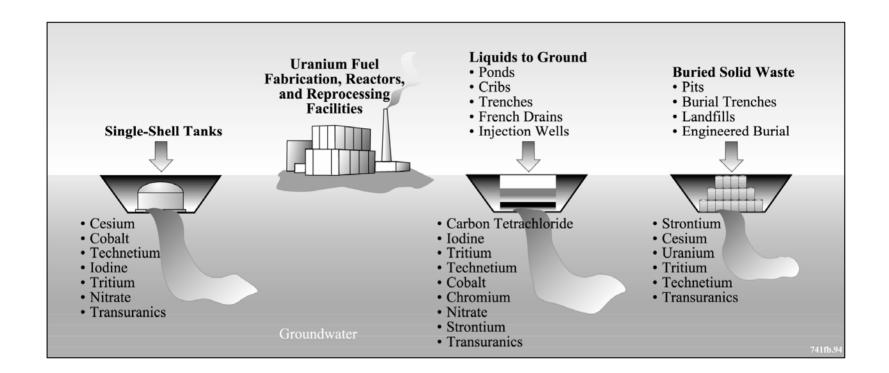


741fb.10

Liquid Releases to Ground. Both clean and contaminated liquids were released to the ground using different approaches. Most liquids were discharged into unlined ponds and ditches.



Contaminated Groundwater Plumes. Small to large groundwater plumes containing chemical and radionuclide contamination above drinking water or other standards exist beneath Hanford. Generally, groundwater moves east toward the Columbia River.



Contaminants Released into the Ground. A variety of radioactive and chemical contaminants were released into the soil and groundwater from various sources. Contaminant mobility depends upon the chemical and physical characteristics of the contaminant, waste release, and natural subsurface setting.