Physics and Chemistry of the Earth 35 (2010) 298–306

Contents lists available at ScienceDirect

Physics and Chemistry of the Earth

journal homepage: www.elsevier.com/locate/pce

A short history of waste management at the Hanford Site

Roy E. Gephart *

Pacific Northwest National Laboratory, P.O. Box 999, MSIN K6-84, Richland, WA 99352, United States

ARTICLE INFO

Article history:
Received 20 October 2009
Received in revised form 3 December 2009
Accepted 10 March 2010
Available online 15 March 2010

Keywords:
Radioactive waste
Hanford Site
Manhattan Project
Cleanup

ABSTRACT

The world's first full-scale nuclear reactors and chemical reprocessing plants built at the Hanford Site in the desert of southeastern Washington State produced two-thirds of the plutonium generated in the United States for nuclear weapons. Operating these facilities also created large volumes of radioactive and chemical waste, some of which was released into the environment exposing people who lived downwind and downstream. Hanford now contains the largest accumulation of nuclear waste in the Western Hemisphere.

Hanford's last reactor shut down in 1987 followed by closure of the last reprocessing plant in 1990. Today, Hanford's only mission is cleanup. Most onsite radioactive waste and nuclear material lingers inside underground tanks or storage facilities. About half of the chemical waste remains in tanks while the rest persists in the soil, groundwater, and burial grounds. Six million dollars each day, or nearly two billion dollars each year, are spent on waste management and cleanup activities. There is significant uncertainty in how long cleanup will take, how much it will cost, and what risks will remain for future generations.

This paper summarizes portions of the waste management history of the Hanford Site published in the book "Hanford: A Conversation about Nuclear Waste and Cleanup." (Gephart, 2003).

1. Site selection

"I thought the Hanford Site was perfect the first time I saw it... we found the only place in the country that could match the requirements for a desirable site." Colonel Franklin Matthias, Sanger (1995).

In the early 1940s, scientists and government officials expressed concern about the hazardous nature of new types of radioactive materials to be generated from the industrial-scale production of plutonium. This was a public and worker safety problem facing the Manhattan Project, which was responsible for building the world's first nuclear weapon industrial complex in the shortest possible time. For expediency, officials circumvented typical pilot-scale engineering steps normal to constructing prototype chemical factories.

For security and safety, General Leslie Groves, army commander of the Manhattan Project, and his staff wanted to avoid any accident taking place in the eastern United States that might release large quantities of radiation—causing a "Congressional investigation to end all Congressional investigations" (Groves, 1962). Such publicity would destroy project secrecy and likely halt work on plutonium production facilities. Therefore, site selection shifted from the more populated areas near Chicago, Illinois and Knoxville, Tennessee to the western United States in the states of Montana, Washington, Oregon, and California.

General Groves' team selected a sparsely settled desert site, now known as Hanford, located along the banks of the Columbia River in southeastern Washington State. Hanford offered various advantages including electricity, abundant river water for cooling and contaminant disposal, a railroad line, sand deposits, nearby hills for encamping security, and a mild, dry climate to expedite construction.

In March 1943, the federal government acquired 1740 square kilometers of land in Washington State[1] (Fig. 1).

The 1300 residents living there were given 2 weeks to 3 months to leave. Government officials paid former residents for the land—cents to a few tens of dollars per acre (Sanger, 1995). The cost for all land acquired was $5.1 million. Lawsuits delayed some payments until higher fair market land values were negotiated.

The government then hired nearly 50,000 workers for Hanford. Site construction began in earnest and in relative secrecy.

The first nuclear reactor (B Reactor) and reprocessing plant (T-Plant) began operating in late 1944—less than 2 years after Enrico Fermi and his team of physicists at the University of Chicago proved a controlled nuclear chain reaction was possible.

Informally, Du Pont officials gave the B Reactor just a 60% chance of working (Gephart, 2003). Candidly, General Leslie Groves

* Tel.: +1 509 371 6142.
E-mail address: roy.gephart.@pnl.gov

[1] Today, the Hanford Site covers 1518 square kilometers.

1474-7065/$ - see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.pce.2010.03.032

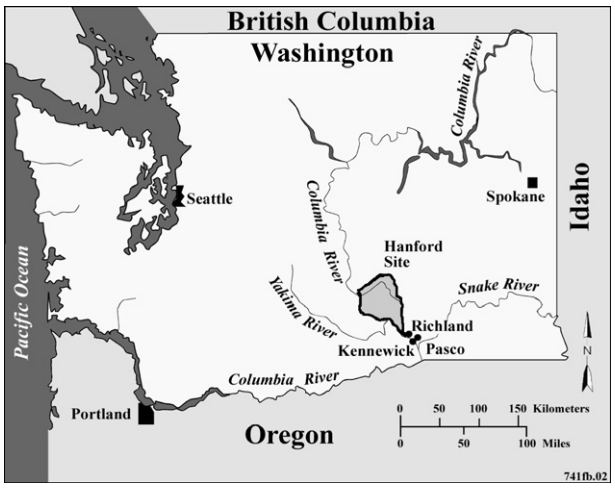

Fig. 1. The Hanford Site covers an area over half the size of the state of Rhode Island.

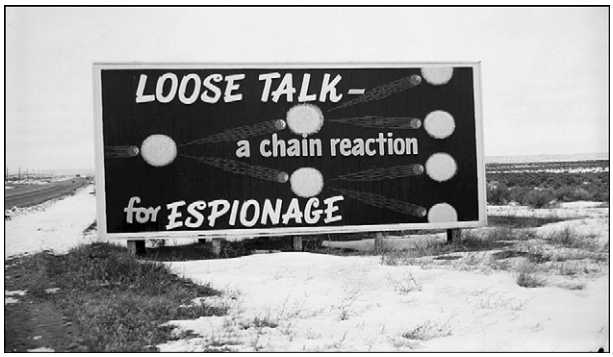

Fig. 2. This 1954 billboard underscored the importance of Hanford workers not talking about their jobs with anyone who did not have a need to know (US Department of Energy).

told his onsite Hanford manager, Colonel Matthias, "If the reactor blows up, jump in the middle of it, and save yourself a lot of trouble." (Sanger, 1995).

Hanford produced the plutonium used in the world's first nuclear explosion at the Trinity Site, New Mexico, in July 1945 and in the atomic bomb dropped on Nagasaki, Japan, the following month.

Facilities located in Fernald, Ohio, and Weldon Spring, Missouri, prepared most of the uranium metal shipped to Hanford. Further milling, metal cladding, and fuel preparation took place in the southern tip of Hanford called the 300 Area. Nearly 20 million uranium fuel slugs were prepared there.[2] These slugs resembled short metal cylinders measuring about 23 cm long and 3.5 cm wide. The last reactor constructed onsite, known as N Reactor, used larger uranium slugs.

The secrecy of site operations, spent fuel reprocessing, plutonium production, and waste management prevailed for years after the Atomic Energy Commission took control of Hanford management from the militarily run Manhattan Project in 1947 (Fig. 2).

[2] Most uranium used inside Hanford reactors was unenriched or only slightly enriched. Unenriched uranium contained 99.3% (by mass) uranium-238 and 0.7% uranium-235. Enriched uranium held slightly more uranium-235. Fuel containing enriched uranium was inserted along the outer edges of a reactor core to generate extra neutrons for creating additional plutonium where the neutron flux was lower compared to the core's center.

2. The first 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).

Between 1943 and 1963, nine block-shaped reactors rose along the shoreline of the Columbia River in what is known as the 100 Area (Fig. 3). Tens of thousands of finely machined and tightly stacked graphite blocks formed the core or "pile" inside each reactor. The graphite slowed neutrons for easier capture by uranium-235 atoms, enabling those atoms to fission.

Inside each reactor, the fragmented uranium-235 atoms released fast-moving neutrons then captured by more abundant uranium-238 atoms also contained in the fuel slugs. In less than 3 days, a portion of the uranium-238 changed into plutonium-239—the metal of choice for nuclear bombs. One uranium atom out of about every 4000 uranium atoms converted to plutionum-239.

Hanford reactors came in different sizes and power levels. The size of each reactor's core ranged from 7 to 11 m per side. Between 200 and 390 metric tons of uranium fuel were inserted into 1000 to more than 3000 aluminum tubes running through each reactor's core. (Miller, 1976; Miller and Steffes, 1987). B Reactor had an initial power level of 250 MW (Du Pont, 1946). Power levels increased to 4400 MW when Hanford's largest reactors, named KE and KW, were built in the mid-1950s in response to the escalating Cold War (Fig. 4).

Chemically treated river water flowed through sealed pipes inside each reactor to cool them. These chemicals adjusted the water's pH, prohibited algal growth, removed dissolved solids, and lessened metal corrosion (Foster et al., 1954). The fission of the uranium-235 atoms and the resulting radiation releases heated reactor water to near the boiling point. Pumped water remained inside the reactor for a few seconds before discharge. Helium gas circulated through the air-sealed pile to remove moisture and reactor-generated gases that absorbed neutrons. Later, carbon dioxide was added to the helium flow, reducing radiation-induced swelling of the reactor's graphite core.

In the first eight reactors built, surface basins temporarily stored the heated, yet now irradiated, cooling water before its discharge or leakage into the Columbia River.

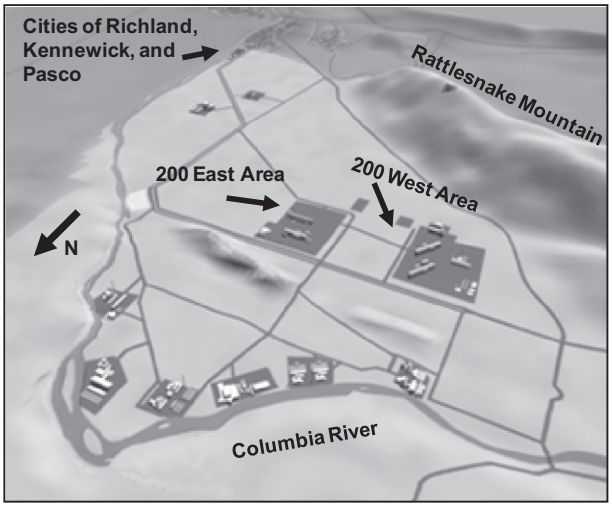

Fig. 3. Illustration shows Hanford's nine reactors built along the southward flowing Columbia River and five reprocessing plants and other support facilities in the 200 East and West Areas. The reprocessing plants are located 8–16 km south of the reactors. Image not to scale.

Fig. 4. This 1987 aerial photograph shows the two K Reactors (adjacent to stacks) plus support facilities built along the shoreline of the Columbia River. Similar layouts typified other reactors (US Department of Energy).

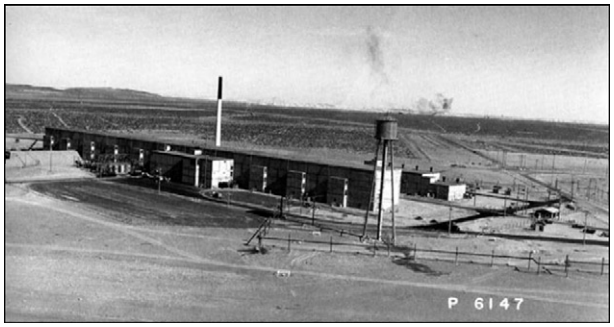

Fig. 5. This photograph, taken in 1944, shows the world's first reprocessing facility (T-Plant). The facility is 245 m long (US Department of Energy).

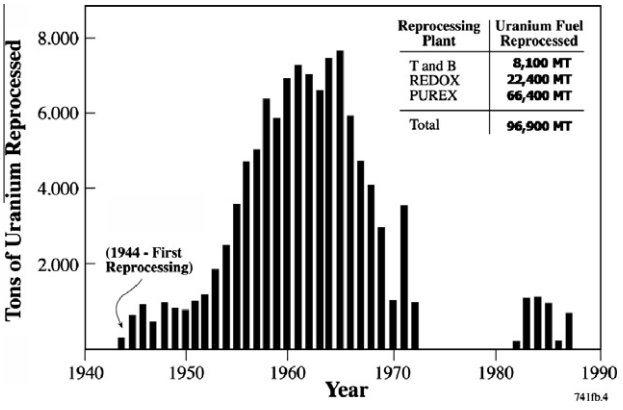

Fig. 6. Hanford facilities processed 96,900 metric tons of uranium fuel.

Radioactive products from Hanford rather than natural radioactivity now dominated radiation levels into the river. These radionuclides were known as activation products, created when elements dissolved in the water captured neutrons released during the fission of uranium, transforming stable, non-radioactive elements into unstable, radioactive elements.

When the metal cladding enclosing uranium fuel ruptured, some fission products escaped into the Columbia River before reactor operators diverted the contaminated water to a nearby trench or liquid disposal site. Nearly 2000 fuel elements ruptured during the 43 years Hanford reactors ran.

Table 1
Tonnage of uranium fuel chemically dissolved and treated inside Hanford reprocessing plants.

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

Once irradiated, uranium fuel was dangerously radioactive and thermally hot. Originally, operators considered 60–65 days adequate for spent fuel to cool before chemically stripping out the plutonium, an activity known as reprocessing (Cooper, 1943; May, 1944). This permitted large amounts of short-lived radionuclides time to decay into non-radioactive elements, or at least reach lower concentrations. Reprocessing was done near the center of the Hanford Site in the 200 East and West Areas.

When pressure mounted for increased plutonium production, fuel was sometimes reprocessed 30–60 days after irradiation (Goldberg, 1998). Inferential evidence suggests that in 1945 some fuel may have been reprocessed less than 3 weeks after irradiation. By the late 1950s to early 1960s, the average storage time increased to between 200 and 250 days (US Department of Energy, 1997).

Various chemical precipitation and solvent extraction techniques separated plutonium from unwanted radionuclides and chemicals. This took place inside large rectangular concrete buildings called reprocessing plants or "canyons."

Workers built five reprocessing plants in the 200 East and West Areas. Starting in late 1944 and 1945, T and B Plants used a bismuth phosphate batch processing technology (Fig. 5).

Higher efficiency and safer solvent extraction technologies functioned in both the REDOX (Reduction-Oxidation) and PUREX (Plutonium Uranium Extraction) Plants beginning in the 1950s. The fifth plant, U Plant, which operated from 1952 to 1958 to recover uranium from tank waste, did not reprocess spent fuel.

The largest reprocessing plant called PUREX, stretched 305 m long, 50 m wide, and 30 m tall. As much as 40% of the height of all reprocessing plants rested underground. The lower level contained remotely operated chemical cells where hot solutions of sodium hydroxide and nitric acid dissolved spent fuel and their metal jackets, and recovery of such desired radionuclides as plutonium and uranium took place.

Hanford reprocessed 96,900 metric tons of uranium (Fig. 6; Table 1). During the early years, T and B Plants reprocessed 2 metric tons of fuel each day. This increased to as high as 30 metric tons per day after the PUREX Plant came online in 1956 (Anderson, 1990).

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

Onsite reprocessing recovered 67.4 metric tons of plutonium (Fig. 7) (Usdin, 1996). This comprised 65% of the nation's supply of 104 metric tons of plutonium generated inside government reactors.[3]

Hanford produced 54.5 metric tons of weapons-grade plutonium and 12.9 metric tons of fuel-grade plutonium.[4] Most of the

[3] Plutonium production in the United States occurred inside 14 government-owned reactors (9 at Hanford and 5 at Savannah River) plus 6 reprocessing plants (4 at Hanford and 2 at Savannah River).

[4] Weapons-grade plutonium consists 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 82–94% plutonium-239.

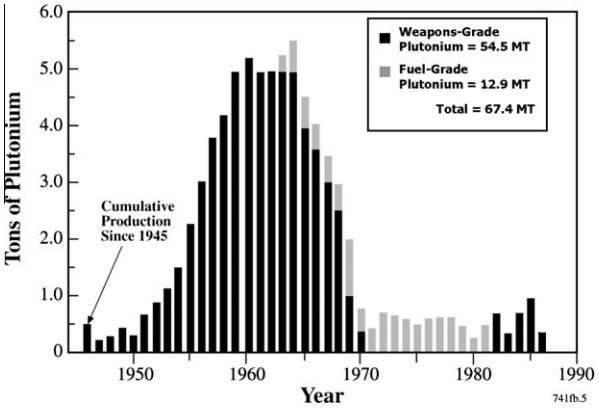

Fig. 7. Hanford produced 67 metric tons of weapons- and fuel-grade plutonium.

remaining 35% of the nation's plutonium supply came from the Savannah River Site, South Carolina.

3. Waste and nuclear materials

"The disposal of contaminated waste in present quantities and by present methods... if continued for decades, presents the gravest of problems." Williams (1948).

Underground tanks received the most radioactive waste discharged from reprocessing plants (Fig. 8).

Cribs, trenches, French drains, reverse wells, and ponds acquired uncontaminated to slightly contaminated liquids plus cooling water. Radioactively contaminated reactor water flowed into the Columbia River. Shallow trenches and storage facilities bore solid waste, and concrete stacks rising above reactors and reprocessing plants released gasses into the air. More than 1600 waste sites once existed at Hanford (US Department of Energy, 1999).

Fig. 8. Workers install the steel roof on a double-shell tank built in 1984 to store highly radioactive waste. Following construction, all tanks were buried underground with their tops about 3 m below the surface (US Department of Energy).

Reprocessing spent fuel in the 200 East and West Areas generated the largest volume of site waste—more than 85% of the liquid releases generated during plutonium production and all tank waste (US Department of Energy, 1997).

Hanford waste and nuclear materials can be informally categorized as contained waste and nuclear materials, buried and stored solid waste, and released waste.[5]

4. Contained waste and nuclear materials

Underground tanks, buildings, and concrete basins hold Hanford's contained waste and nuclear materials. Some structures are contaminated; others are not. The largest are the reactors and reprocessing plants. Filters, pipes, reactor cores, and other portions of Hanford facilities may still contain 1 million curies of radioactivity.

From 1944 through 1988, Hanford generated nearly 2 million cubic meters of tank waste (Agnew, 1997). Liquid evaporation, discharge to the ground, chemical treatment, and tank leakage reduced this volume by 90%—to 200,000 cubic meters[6] (Hanlon, 2003). This is about 60% of the tank waste existing across the nuclear weapons complex. Today, Hanford tanks hold about 190 million curies of radioactivity and 170,000 metric tons of chemicals. Each cubic meter of tank waste contains nearly 1000 curies of radioactivity.

Adding large volumes of sodium hydroxide to the acid waste stream, generated from reprocessing spent fuel, transformed the stream into a caustic solution before being pumped into underground storage tanks. The pH was adjusted because acidic waste would dissolve the carbon-steel tanks; wartime shortages made it impossible to construct tanks from more acid-resistent stainless steel. The resultant tank liquids contain high concentrations of alkali, salt, aluminate, and cesium-137.

The 177 underground tanks built in the 200 East and West Areas are grouped into 18 tank farms. Eighty-five percent or 149 of these are single-shell tanks constructed between 1943 and 1964. These range in volume from 210 cubic meters to 3800 cubic meters. Their life expectancy for containing liquid waste was 20 years (US Department of Energy, 1998).

Sixty-seven single-shell tanks have leaked or are suspected to have leaked as much as 5700 cubic meters of sodium nitrate- and cesium-contaminated liquids into the underlying sediment. The first waste leak was confirmed in 1959 (Gephart, 2003).

To improve containment, 28 double-shell carbon–steel tanks were constructed between 1968 and 1986. Volumes ranged from 3800 to 4200 cubic meters, with design lives as long as 50 years.

Over the years, drainable liquids inside single-shell tanks were pumped into the newer double-shell tanks. Thick waste sludge and semi-dried salt cake remained behind. To date, no double-shell tank has leaked, though the oldest ones have reached the end of their design life.

Nuclear materials stored onsite include 1936 stainless-steel capsules containing 110 million curies of radioactive cesium and strontium, plus their decay products. These are kept in water-filled pools inside the Waste Encapsulation and Storage Facility adjoining B Plant in the 200 East Area. From 1967 to 1985, these radionuclides had been recovered from tank waste to reduce the heat

[5] All waste and nuclear material inventories in this article are approximations based upon reports, conversations, assumptions, and calculations. Most estimates of curies present are decayed to the year 2009. Numbers are rounded and will change over time due to radioactive decay, improved knowledge, and site cleanup.

[6] If placed inside standard-sized railroad tanker cars, this volume of tank waste would fill enough tanker cars to assemble a train 42 km long.

and radiation load inside tanks so they could receive newly generated waste.

For years, 80% of the US Department of Energy's (DOE) remaining irradiated uranium fuel remained stored inside two aging water-filled concrete basins at the KE and KW Reactors. Spent fuel removal began in 2000 and was completed four years later with dried and repackaged fuel rods stored in the Canister Storage Building located in the 200 East Area. The fuel weighed 2100 metric tons and contained less than 40 million curies of radioactivity as of 2009.

This uranium fuel had been irradiated inside N Reactor and moved to the K Basins between 1975 and 1989 for storage. Because the PUREX Plant was not restarted in the late 1980s, this fuel remained unprocessed. Some fuel corroded. Water leaks released radionuclides into the soil and groundwater near the K Reactors.

An additional 30 metric tons of irradiated fuel from non-Hanford production reactors are also stored onsite.

5. 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/or low levels of radioactivity. Some radioactivity comes from such long-lived transuranic elements as plutonium, neptunium, and americium.

As of the late 1990s, 75 solid waste burial grounds existed at Hanford (US Department of Energy, 1997) containing 700,000 cubic meters of solid waste buried or stored in facilities. About 97% was buried in landfills, and more than 95% is classified as low-level waste (Duncan et al., 1995). This waste contained 6 million curies of radioactivity and 65,000 metric tons of chemicals. An estimated 590 metric tons of uranium and 360 kilograms of plutonium may exist in solid waste buried within the 200 Area (Wodrich, 1991).

Before 1970, solid waste was not well characterized or sorted (Fig. 9). For example, transuranic and non-transuranic contaminated materials were packed together along with a variety of chemicals and combustibles. The waste was not buried to enable later retrieval, repackaging, and offsite shipment. Solid waste buried since 1970 is better characterized. Today, any new solid waste burial takes place in the 200 West Area. Shipments of containerized transuranic waste are now trucked to the Waste Isolation Pilot Plant in New Mexico for storage in an underground salt formation.

Beginning in the late 1990s, the six-acre 618-10 and nine-acre 618-11 burial grounds, located in southern Hanford, received considerable attention because of their content and contribution to subsurface contamination. From 1953 to 1967, these sites received low- and high-activity waste from nuclear fuel experiments and other research conducted in the 300 Area. Groundwater samples revealed elevated levels of tritium—as much as 400 times above drinking-water standards near 618-11 (Hartmann et al., 2001). As much as 11 kg of dispersible plutonium, as well as slivers of irradiated uranium fuel and other waste materials, might be buried at these two sites (Peterson et al., 2001).

6. Released waste

Perhaps 1 million curies of radioactivity and 100,000–300,000 metric tons of chemicals remain in the soil and groundwater beneath Hanford. Most exist in or near the center of Hanford where reprocessing plants once operated and waste tanks are buried.

During the first months of spent fuel reprocessing in late 1944, mildly contaminated liquids were simply dumped into depressions on the ground (Brown and Ruppert, 1950). There liquids seeped into the sandy soil, adding contaminants to the sediment and eventually the groundwater. Some liquids evaporated, leaving radioactive salt residue for plant and animal uptake. Contaminants also blew downwind, enlarging marked radiation zones, and exposing workers and the public. Such uncontrolled waste disposal and dispersal became unacceptable.

Fig. 9. Poorly characterized radioactive and non-radioactive solid waste was buried onsite during the early years of Hanford (US Department of Energy).

Liquids were then pumped down "reverse" wells. While this removed contaminants from the near surface, it injected them closer to and sometimes directly into, the underlying aquifer, bypassing the chemical sorptive benefit of overlying sediment.

Within months, most well use also discontinued except for disposal of small waste quantities (Du Pont, 1945). Liquids were then pumped into shallow buried box-like structures called cribs, gravel-filled tile fields, French drains (vertical buried concrete pipes), and open trenches later backfilled with gravel. These approaches to liquid waste management continued into the late 20th century.

As much as 1.7 billion cubic meters of uncontaminated to slightly contaminated liquids[7] were discharged into 30 ponds and unlined ditches—most in or near the 200 East and West Areas (Hartman and Dresel, 1998). This provided a hydraulic driving force moving contamination deeper and faster into the subsurface than otherwise possible.

Hanford may contain as much as 28,300 cubic meters of soil contaminated from radionuclides contained in liquid waste streams released near reprocessing plants (Gee et al., 2007). According to Kincaid et al. (2006), and Corbin et al. (2005), and conversations with Hanford staff responsible for tracking onsite waste inventories, some 225,000 curies of cesium-137; 180,000 curies of tritium; 52,000 curies of strontium-90; 50,000 curies of plutonium; and 700 curies of technetium-99 may exist in the Hanford soil and groundwater from past liquid discharges and tank leaks.

In 2008, groundwater plumes covering 185 square kilometers or 12% of the Hanford Site contain contaminants such as metals (e.g., chromium), chemicals (e.g., nitrates, trichloroethene, and carbon tetrachloride), and radionuclides (e.g., tritium, iodine-129, and technetium-99) at concentrations above safe drinking-water standards or other guidelines (Poston et al., 2009). Smaller pockets of cobalt-60, cesium-137, uranium, and plutonium contamination are also found.

Geochemical reactions with Hanford sediments retain some contaminants, such as cesium-137, strontium-90, and cobalt-60, effectively immobilizing them except under conditions of extreme saline or acidic conditions (Gee et al., 2007). However, tritium,

[7] A total of 1.7 billion cubic meters equals the average volume of water flowing down the Columbia River every 5 days.

technetium-99, and iodine-129 are mobile, enabling those radionuclides to potentially move deep in the vadose zone and into the unconfined aquifer. Certain radionuclides, such as uranium and transuranic elements, can undergo chemical sorption on the surface of or into the crystalline structure of sedimentary minerals, effectively holding them in place.

Groundwater travel time along the 16-km stretch separating the 200 East Area from 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 longer because sediment comprising the underlying aquifer has a lower permeability than beneath the 200 East Area.

For the same reason, broad plumes of contaminated groundwater emanate from the 200 East Area while contaminated groundwater beneath the 200 West Area covers a far smaller region.

In addition to leaked tank waste entering the soil, nearly 500,000 cubic meters of treated and untreated tank waste were intentionally discharged to the ground (Agnew, 1997). This occurred between 1946 and 1958. This waste contained more than 275,000 metric tons of chemicals and 60,000 curies of radioactivity (Waite, 1991).

Laboratory studies of contaminated sediments collected from beneath tanks demonstrate that ion exchange, precipitation and dissolution, plus surface complexation reactions can retard the migration of select radionuclides (Zachara et al., 2007). The use of such information, coupled with subsurface geohydrologic studies, is irreplaceable in supporting regulatory decisions influencing contaminant removal, stabilization, and/or monitoring.

7. Past radionuclide releases into 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." (Herde, 1946).

Today, small amounts of radionuclides are released from Hanford into the air and Columbia River. During 2008, the average member of the public received a radiation dose of 0.0009 mrem from Hanford releases (Poston et al., 2009). A maximally exposed individual received 0.04 mrem during the same year.

The average radiation dose a member of the public receives from the natural environment each year is about 300 mrem. Therefore, a radiation dose of 0.0009 mrem per year received from living near Hanford equals what an average person gains in 2 min of exposure to the natural environment.

Radionuclide releases and doses were higher in the past. Over 140 million curies of radionuclides were discharged into the atmosphere and Columbia River between the mid-1940s and the mid-1960s (HEDR, 1994).

8. Radionuclide releases into the atmosphere

The atmosphere received 32 million curies of radioactivity released in stack gases venting reactors and reprocessing plants.

Twelve million curies originated 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, N Reactor emitted an additional 2 million curies. More than 99% of these releases were argon-41 with small amounts of carbon-14 and tritium. The Hanford Dose Reconstruction Report (HEDR, 1994) and Heeb (1994) reported these releases resulted in an increased radiation dose to the public of 2–4 mrem per year. This was about a 1% increase from natural background radiation levels for people living near Hanford.

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

However, neither argon-41or krypton-85 contributed much to the public's radiation exposure because these elements are noble gases and do not accumulate inside the body; also, their energy levels are thousands to millions of times less than other radionuclides of concern such as iodine-131.

Napier reported that six radionuclides discharged from reprocessing plants contributed most to public radiation doses (Napier, 1992). These radionuclides were iodine-131, ruthenium-103, ruthenium-106, strontium-90, plutonium-239, and cerium-144. However, 99% of this dose came from just one radioisotope—iodine-131.

Most iodine releases (697,000 out of 740,000 curies) occurred between 1944 and 1949 before the first two reprocessing plants (T and B) had stack filters installed.

People living adjacent to and downwind of Hanford received the highest dose. The range in dose to the thyroid of an adult living at a maximally exposed location was 10–150 rad (HEDR, 1994). The range in dose to the thyroid for a child at a maximally exposed site was higher: 54–870 rad.

Early in Hanford operations, onsite officials recognized they had an iodine-131 problem. However, "revelation of a regional iodine-131 problem would have had a tremendous public relations impact," including employees hesitant to work onsite (Stannard, 1988). Secrecy surrounded the monitoring of iodine uptake in live animals. Therefore, Hanford staff, masquerading as US Department of Agriculture specialists trained in testing the "health and vigor" of farm animals, visited local ranchers where they serendipitously passed a radiation detector across the necks of cattle to detect radionuclide uptake.

The Center for Disease Control and Prevention and the Fred Hutchinson Cancer Research Center published thyroid disease study results in 2002 (CDC, 2002). These centers could not find an association between Hanford's past iodine releases and thyroid disease. However, the report stated "the findings do not prove that Hanford radiation had no effect... if there is an increased risk of thyroid disease... it is probably too small to observe using the best epidemiologic methods available."

9. Radionuclide releases into the Columbia River

Between 1944 and 1971, the Columbia River received an estimated 110 million curies of radioactivity from activation products released by the operation of Hanford's first eight reactors (Heeb and Bates, 1994).[8]

River water cooled these single-pass reactors. The ninth reactor re-circulated cooling water.

Most radionuclides existed in small amounts or had short half-lives. For example, manganese-56, with a half-life of 2.6 h, contributed nearly two-thirds of the curie load released. Most of this radioisotope decayed away before reaching down-river populations. The five radionuclides contributing most (94%) of the estimated radiation dose people living downstream received were as follows:

[8] When discharged, the temperature of reactor water was 67°C higher than river temperature (Foster et al., 1954). When river water levels remained low and all reactors operated, the ambient river temperature for the first 90 km downstream increased by 3°C (Becker, 1990).

Radionuclide	Curies
Sodium-24	12,600,000
Phosphorus-32	230,000
Neptunium-239	6,300,000
Zinc-65	490,000
Arsenic-76	2,500,000

Additional radiation releases resulted from the periodic failure of uranium fuel slugs. These longer-lived fission products remain buried in the river sediment, especially in slack waters downstream of islands or in sediment behind 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 atop the ground, the maximum average yearly radiation dose to a person over their lifetime would be 1.6 mrem.

The largest radionuclide releases to the Columbia River occurred between 1956 and 1965 when most reactors operated simultaneously. An average of 10,000–12,000 curies per day was discharged (HEDR, 1994).

During these times, a typical nearby resident may have received a yearly radiation dose increase of 1–5 mrem per year (HEDR, 1994). However, a person who significantly used the river and ate large quantities of fish might have received a 50- to 130-mrem increase in their annual dose—a 15–45% increase compared to background levels. Some Native Americans may have accumulated ten times this dose because they consumed more river fish than the general population (Till et al., 2002).

To detect radionuclide uptake in the public, especially children, schools along the path of the Columbia River hosted visits of whole-body radiation counters operated by Hanford personnel (Fig. 10).

Today, radioactivity flowing down the Columbia River is significantly reduced. Low but detectable amounts of radionuclides, chemicals such as nitrates, and various metals still enter the river from Hanford plus upstream ore mines and agricultural runoff.

For example, water analyses reported by Poston et al. (2009) detected an average of 7 curies of radioactivity dissolved in river water before it entered Hanford. About 90% of this radioactivity comes from tritium—both naturally occurring tritium and that contained in fallout from past atmospheric testing of nuclear weapons.

Fig. 10. This 1965 photograph records the inside of a truck-mounted whole-body radiation counter hauled to schools to monitor radiation levels in children (US Department of Energy).

Most of the remaining radioactivity originates from naturally occurring uranium and potassium.

As river water passes along the northern and eastern boundary of Hanford, groundwater from beneath the site enters through shoreline springs and direct discharge. Downstream of Hanford, an average of 11 curies of natural and human-made radioactivity flow in the river each day. Compared to upriver water samples, this 4 curie-per-day increase mostly originates from Hanford tritium-contaminated groundwater. Sometimes iodine-129, technetium-99, and strontium-90 levels are higher downstream of Hanford (Poston et al., 2009).

10. 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 is a macrocosm of the environmental and nuclear material management problems facing the federal government at sites once used for uranium mining, nuclear weapons research, material production, and bomb testing.

Today, nearly 350 million curies of mostly cesium-137 and strontium-90 plus 350,000–550,000 metric tons of chemicals remain onsite from the plutonium production mission (Table 2). Of the human-made radioactivity existing across the nuclear weapons complex, 35% of it lingers at Hanford.

Hanford's plutonium production era ended when N Reactor closed in 1987 and the PUREX Plant shut down in 1990. Now all reactors and reprocessing plants are permanently closed.

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

The fundamentals of chemical and nuclear-based processing that once framed decision-making were now steeped in a new environmental, health, and social language never before applied to Hanford. Independent oversight grew, and public input to decision-making became commonplace.

Initial cleanup schedules and costs were overly optimistic. Two decades later, the once anticipated 10- to 30-year cleanup program has stretched well into the mid-21st century as harder problems are faced. For the first time, Hanford cleanup costs reaching $100 billion—just covering tank waste—are being reported (Government Accountability Office, 2009).

Hanford's present remediation strategy centers upon (1) cleanup of lands bordering the Columbia River; (2) shrinking the site's active waste management area to less than 50-square-kilometers inside the Central Plateau where the 200 Areas are located; (3)

Table 2
General waste and nuclear material inventories at the Hanford Site. Numbers are approximate and rounded updated after Gephart (2003).

Waste or material	Volume	Curies (million)	Chemicals (metric tons)
Tank waste	200,000 m^3	190	170,000
Solid waste	700,000 m^3	6	65,000
Soil/groundwater	1.0 billion m^3	1	100,000–300,000
Facilities	5 million m^3	1	–
Nuclear material[a]	700 m^3	150	–

[a] These 150 million curies are contained in 2100 metric tons of spent fuel (<40 million curies) now in dry storage inside the Canister Storage Building plus 1936 cesium and strontium capsules (110 million curies) held in the Waste Encapsulation and Storage Facility.

remediating tank farms plus the remaining waste sites and facilities and (4) implementing institutional controls and long-term stewardship over waste forms and contaminants remaining onsite.

Though considerable attention has focused on the challenge of tank waste remediation, there is growing awareness of the lack of knowledge and capability hindering the effective remediation of contaminants released into the vadose zone and groundwater aquifer.

The National Academies has reported upon these gaps (National Research Council, 2000, 2009). Examples include the following:

- Locating and characterizing the concentrations, speciations, release rates, and movement of contaminants distributed within a heterogeneous sedimentary environment.
- Characterizing the coupled physical, chemical, and biological properties of the subsurface controlling contaminant fate and transport.
- Creating validated conceptual and predictive models to depict subsurface dynamics and contaminant behavior spanning the molecular to field scale.
- Developing less costly and more effective contaminant treatment, recovery, containment, and stabilization techniques through integrated laboratory and field test programs.
- Advancing subsurface monitoring technologies including novel sensors, detectors, and data transmission techniques.

Without this advanced knowledge and capabilities, it will be difficult to perform reliable performance assessments supporting regulatory decisions or planning/executing remedial actions where projected outcomes match field results.

How long Hanford cleanup will take and how much it will cost remains uncertain. What we do know is that cleanup activities must be framed upon informed debate, social consensus, defensible standards, achievable goals, and high-quality science and technology.

References

Readers interested in accessing historical Hanford documentation, such as those referenced in this article, can do so by linking onto the Hanford declassified document retrieval system at: http://www2.hanford.gov/ddrs/.

Agnew, S.F., 1997. Hanford Tank Chemical and Radionuclide Inventories, HDW Model. LA-UR-96-3860, Rev. 4, Los Alamos National Laboratory, Los Alamos, New Mexico.

Anderson, J.D., 1990. A History of the 200 Area Tank Farms. WHC-MR-0132, Westinghouse Hanford Company, Richland, Washington.

Becker, C.D., 1990. Aquatic Bioenvironmental Studies. The Hanford Experience 1944–1984. Elsevier, New York.

Brown, R.E., Ruppert, H.G., 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, 2002. A Guide to the Hanford Thyroid Disease Study—Final Report, Atlanta, Georgia.

Cooper, C.M., 1943. Proposed Program for Study of Stack Dispersion at Hanford. Letter to RS Stone, DUH-47, US Department of Energy Public Reading Room, Richland, Washington (December 31, 1943).

Corbin, R.A., Simpson, B.C., Anderson, M.J., Field, F.G., Jones, T.E., Kincaid, C.T., 2005. Hanford Soil Inventory Model. RPP-26744, Rev. 1, CH2MHILL, Richland, Washington.

US Department of Energy (DOE), 1997. Hanford Site Environmental Management Specifications: Section 4.2.2, "Waste Management Project." DOE/RL-97-55, Rev. 1, US Department of Energy-Richland Operations Office, Richland, Washington.

US Department of Energy (DOE), 1998. Groundwater/Vadose Integration Project Specification. DOE/RL-98-48, Draft C, US Department of Energy-Richland Operations Office, Richland, Washington.

US Department of Energy (DOE), 1999. DOE Resumes Plutonium Stabilization at Hanford Plutonium Finishing Plant. Press Release, US Department of Energy-Richland Operations Office, Richland, Washington (January 26, 1999).

Du Pont (E.I. Du Pont De Nemours & Co), 1945. Memoranda for the File, Book 12, Du Pont. Hanford Engineer Works, Richland, Washington.

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.

Duncan, D.I., Weyns-Rollosson, D.I., Pottmeyer, J.A., Stratton, T.J., 1995. A History of Solid Waste Packaging at the Hanford Site. WHC-SA-2772-FP, Westinghouse Hanford Company, Richland, Washington.

Foster, R.F., David, J.J., Olson, P.A., 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.

Gee, F.W., Oostrom, M., Freshley, M.D., Rockhold, M.L., Zachara, J.M., 2007. Hanford Site vadoze zone studies: an overview. Vadose Zone Journal 6, 899–905.

Gephart, R.E., 2003. Hanford: A Conversation about Nuclear Waste and Cleanup. Battelle Press, Columbus, Ohio.

Goldberg, S., 1998. General groves and the atomic west: the making and meaning of Hanford. In: Hevly, B., Findlay, J.M. (Eds.), The Atomic West. Center for the Study of the Pacific Northwest in Association with the University of Washington Press, Washington.

Government Accountability Office (GAO), 2009. Uncertainties and Questions about Costs and Risks Persist with DOE's Tank Waste Cleanup Strategy at Hanford. GAO-09-913, US Government Accountability Office, Washington, DC.

Groves, L.R., 1962. Now It Can Be Told: The Story of the Manhattan Project. Harper and Brothers, New York.

Hanlon, B.M., 2003. Waste Tank Summary Report for Month Ending July 31, 2003. HNF-EP-0182-184, CH2MHILL Hanford Group Inc., Richland, Washington.

Hartman, M.J., Dresel, P.E. (Eds.), 1998. Hanford Site Groundwater Monitoring for Fiscal Year 1997. PNNL-11793, Pacific Northwest National Laboratory, Richland, Washington.

Hartmann, M.J., Morasch, L.F., Webber, W.D. (Eds.), 2001. Hanford Site Groundwater Monitoring for Fiscal Year 2000. PNNL-13404, Pacific Northwest National Laboratory, Richland, Washington.

Hanford Environmental Dose Reconstruction Project (HEDR), 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 (April 21, 1994).

Heeb, C.M., 1994. Radionuclide Releases to the Atmosphere from Hanford Operations, 1944–1972. PNWD-2222 HEDR, Pacific Northwest Laboratory, Richland, Washington.

Heeb, C.M., Bates, D.J., 1994. Radionuclide Releases to the Columbia River from Hanford Operations, 1944–1971. PNWD-2223 HEDR, Pacific Northwest Laboratory, Richland, Washington.

Herde, K.E., 1946. I^{131} Accumulation in the Thyroid of Sheep Grazing Near H.E.W. Letter to J.W. Healy. HW 3-3455, General Electric Company, Hanford Atomic Products Operation, Richland, Washington (March 1, 1946).

Kincaid, C.T., Eslinger, P.W., Aaberg, R.L., Miley, T.B., Nelson, I.C., Strenge, D.L., Jr. Evans, J.C., 2006. Inventory Data Packages for Hanford Assessments. PNNL-15829, Rev. 0, Pacific Northwest National Laboratory, Richland, Washington.

May, K., 1944. Time Interval Between Discharge of Metal from the Pile and Processing in Canyon. Letter to DF Babcock. DUH-160, US Department of Energy Public Reading Room, Richland, Washington.

Miller, N.R., 1976. Directory of ERDA-owned Nuclear Reactor, letter to O.J. Bennett. US Energy Research and Development Administration-Richland Operations Office, United Nuclear Industries, Richland, Washington (March 4, 1976).

Miller, R.L., Steffes, J.M., 1987. Radionuclide Inventory and Source Terms for the Surplus Production Reactors at Hanford. UNI-3714, Rev. 1,UNC Nuclear Industries, Richland, Washington.

Napier, B.A., 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, DC.

National Reseach Council, 2000. Research Needs in Subsurface Science. National Academies Press, Washington, DC.

National Research Council, 2009. Advice on the Department of Energy's Cleanup Technology Roadmap. National Academies Press, Washington, DC.

Patton, G.W., 1998. Surface water and sediment surveillance. In: Hanford Site Environmental Report for Calendar Year 1997. PNNL-11795, Pacific Northwest National Laboratory, Richland, Washington.

Peterson, S.W., Cameron, R.J., Johnson, M.D., Truex, M.J., 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, T.M., Duncan, J.P., Dirkes, R.L. (Eds.), 2009. Hanford Site Environmental Report for Calendar Year 2008. PNNL-18427, Pacific Northwest National Laboratory, Richland, Washington.

Sanger, S.L., 1995. Working on the Bomb: An Oral History of WWII Hanford. Portland State University, Portland, Oregon.

Stannard, J.N., 1988. Radioactivity and Health: A History. DOE/RL/01830-T59, Pacific Northwest Laboratory, Richland, Washington.

Till, J.E., Grogan, H.A., Rood, A.S., Aanenson, J.W., Liebow, E.B., 2002. A Risk-based Screening Analysis for Radionuclides Released to the Columbia River from Past Activities at the US Department of Energy Nuclear Weapons Site in Hanford, Washington. Risk Assessment Corporation, Neeses, South Carolina.

US Department of Energy (DOE), 1997. Linking Legacies: Connecting the Cold War Nuclear Weapons Production Processes to Their Environmental Consequences. DOE/EM-0319, US Department of Energy, Washington, DC.

Usdin, S., 1996. DOE Lifts the Plutonium Veil. Nuclear Remediation Week (February 9, 1996).

Waite, J.L., 1991. Tank Wastes Discharged Directly to the Soil at the Hanford Site. WHC-MR-0227, Westinghouse Hanford Company, Richland, Washington.

Washington State Department of Ecology, 1989. US Environmental Protection Agency, and US Department of Energy. Hanford Federal Facility Agreement and Consent Order. Document No. 89-10, Rev. 5 (Tri-Party Agreement), Olympia, Washington.

Wells, D., 1994. Special Report: Radioactivity in the Columbia River Sediments and Their Health Effects. Washington State Department of Health, Olympia, Washington.

Williams, S.J., 1948. Report of the Safety and Industrial Health Advisory Board. US Atomic Energy Commission, Washington, DC.

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.

Zachara, J.M., Serne, J., Freshley, M., Mann, F., Anderson, F., Wood, M., Jones, T., Myers, D., 2007. Geochemical processes controlling migration of tank wastes in Hanford's Vadose Zone. Vadose Zone Journal 6, 985–1003.