

Radioactive Waste Contamination of Soil and Groundwater at the Hanford Site

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Abstract

The Hanford Site in southeastern Washington State is the location of the largest nuclear waste storage area in the United States. Since plutonium production facilities at Hanford were shut down in 1987, cleaning up the leftover waste at the Hanford Site has become one of the largest environmental cleanup projects in history. Legal, political, technical, and social aspects of the cleanup contribute to the overall complexity of the situation. There is, however, a sense of urgency surrounding the cleanup efforts. Of the 177 underground nuclear waste storage tanks on the Hanford Site 67 are known to be leaking currently and a large proportion of the tanks are at or approaching the end of their design lives. Cleanup efforts are presently focused on removing the waste from the storage tanks and immobilizing it for long-term storage. While it remains a primary concern, it is currently unknown what the long-term ecological consequences of the leaked nuclear wastes will be.

1. Introduction

1.1 The Hanford Site

In June 1942 the Manhattan Engineer District of the Army Corps of Engineers was instructed to locate a site where industrial facilities for a secret weapons project would be built. The Hanford Nuclear Reservation, which encompasses 560 square miles in southeastern Washington State, was established as the location for the plutonium production site in March 1943. The site was separated into several regions where different activities would take place (Figure 1). Within two years, nuclear reactors, processing facilities, support facilities, underground waste storage tanks, and nuclear fuel fabrication facilities had been constructed and were in operation to produce plutonium fuel for military defense (Gephart and Lundgren, 1998).

Between its establishment in 1943 and deactivation in 1987, approximately 110,000 tons of nuclear fuel were processed at the Hanford Site (Wald, 1998; Gephart and Lundgren, 1998). Hanford produced more than 73 tons of nuclear weapon- and reactor fuel-grade plutonium during its years of operation (Gephart and Lundgren, 1998). The production of plutonium fuel generated millions of gallons of highly radioactive waste, as well as non-radioactive hazardous waste and mixed waste. The U.S. Department of Energy (DOE) currently manages the Hanford Site, and is responsible for cleaning up the now-defunct plutonium processing facility.

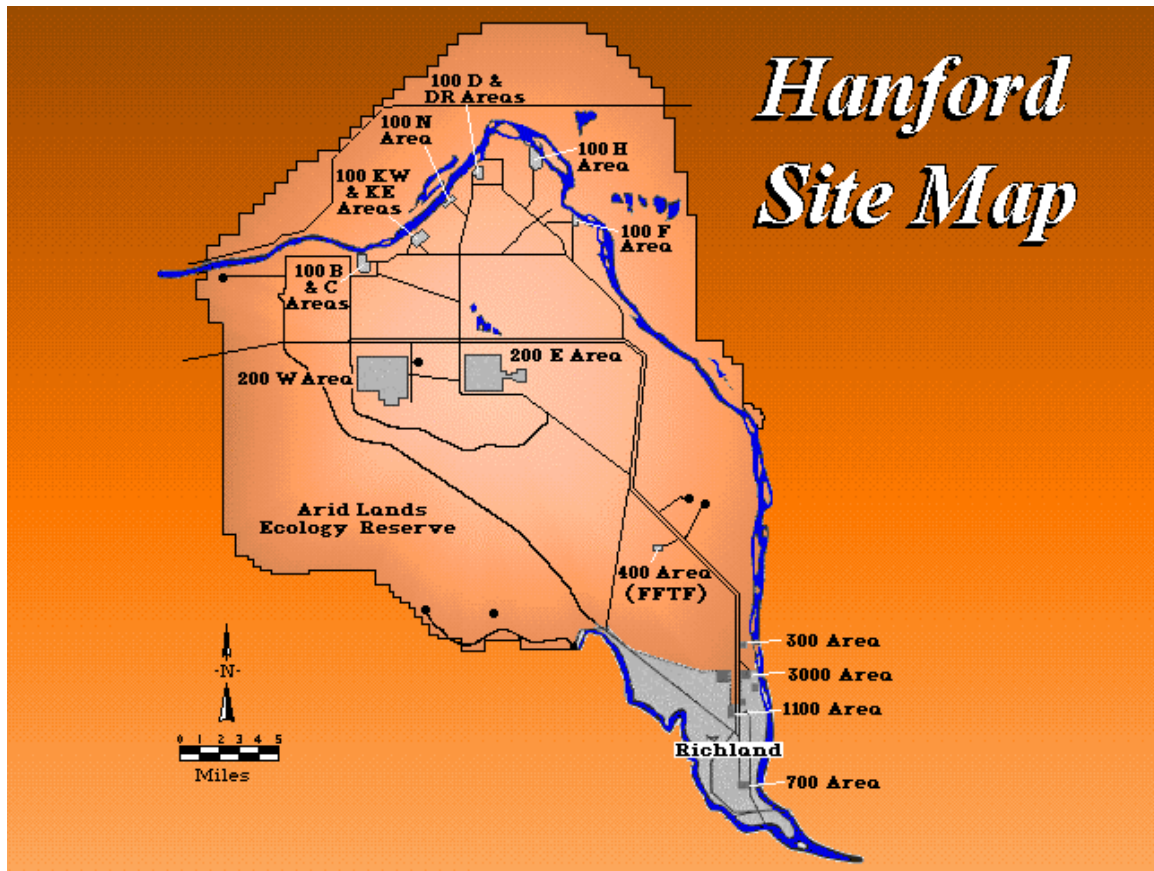


Figure 1. Map of the Hanford Site in southeastern Washington State (from USDOE: Hanford). The 100 Area of the Hanford Site housed nuclear reactors, the 200-East and –West Areas were home to fuel reprocessing facilities and waste storage areas, and the 300 Area was dedicated to nuclear fuel fabrication and research.

1.2 Occurrence of Waste in and around the Hanford Site

On a volume basis, Hanford contains over two-thirds of the DOE's highly radioactive waste (Gephart and Lundgren, 1998). About 50%, or 54 million gallons, of the radioactive waste at the Hanford Site is contained in 177 underground storage tanks, many of which are leaking. During operation, the primary consideration when making decisions regarding waste management was nuclear materials production. As a result, the methods by which Hanford's waste was managed are unacceptable by today's standards. For example, prior to 1970 solid low-level and transuranic waste

was frequently disposed of in cardboard boxes in unlined trenches (Wald, 1998). In 1989 the DOE began a comprehensive environmental cleanup effort at the Hanford Site at a cost of more than \$1 billion per year (Zorpette, 1996).

Cleaning up radioactive waste is a complex process because it cannot be degraded the way organic substances can. James D. Werner, director of strategic planning and analysis in the Office of Environmental Management of the DOE summarizes the complexity of cleaning up radioactive waste: "The stuff we're dealing with can't go away until it decays. You can containerize it, solidify it, immobilize it and move it, but you can't make it go away" (Zorpette, 1996). In 1996 the total radioactivity at the Hanford Site was estimated at approximately 437 million curies (Ci, see Appendix A), distributed as follows (Gephart and Lundgren, 1998):

- Underground waste tanks = approximately 215 million Ci
- Cesium and strontium capsules = 150 million Ci
- Stored irradiated fuel = 50 million Ci
- Facilities = 18 million Ci (in pipes, filters, etc.)
- Solid waste = 2.5 million Ci
- Soil and groundwater contamination = 1.5 million Ci

Several different types of waste were generated by plutonium processing at the Hanford Site, including high-level and low-level radioactive waste, transuranic waste, hazardous waste, and mixed waste. High-level waste (HLW) is generally defined as the waste generated during the chemical separation of uranium and plutonium from undesirable radioactive elements. Most of the radioactive elements discharged to the underground waste storage tanks were contained in HLW. Low-level waste (LLW) is any radioactive waste that is not spent fuel, high-level, or transuranic waste. The LLW is a general category and can include liquid waste, contaminated clothing, tools, and equipment. Transuranic waste is radioactive waste containing greater than 100 nanocuries per gram of alpha-emitting isotopes with atomic numbers higher than 92. Transuranic waste is generated primarily during nuclear fuel reprocessing and the manufacture of plutonium weapons. Hazardous wastes are non-radioactive wastes, such as metals or chemical compounds that are known to pose environmental or human health risks. Mixed waste is hazardous waste combined with radioactive material.

Approximately 99% of the total radioactivity contained in the underground waste storage tanks at the Hanford Site is due to cesium-137 (^{137}Cs) and strontium-90 (^{90}Sr). Cesium-137 is the most abundant of all radioactive isotopes present in the tank waste (Gephart and Lundgren 1998). The objective of this study is to examine the potential for widespread environmental contamination by the ^{137}Cs contained in the underground waste storage tanks at the Hanford Site.

2. Background Information

2.1 Radiation

Radiation is a form of energy that is transmitted by electromagnetic waves or by streams of subatomic particles. Variation in the frequency of electromagnetic waves produces different energy levels, ranging from low-frequency radio waves to high-frequency gamma rays. Low- to mid-frequency electromagnetic waves produce non-ionizing radiation. When electromagnetic waves at frequencies lower than approximately 10^{17} cycles per second are absorbed in biological material, energy is expended as heat. Electrons are excited but not removed. Non-ionizing radiation is not believed to pose a significant danger to humans, except at very high exposure (WA State Institute for Public Policy, 1985). Radio waves, radar waves, microwaves, infrared rays, visible light, and ultraviolet rays are all forms of non-ionizing radiation.

Ionizing radiation can penetrate and interact with a material by removing electrons surrounding nuclei, thus producing ions. The energy imported by ionizing radiation can damage living tissue. Tissues that continuously reproduce, such as intestinal lining, blood cells, hair cells, and reproductive cells, are more sensitive to ionizing radiation than those tissues that reproduce more slowly, such as muscle, nerve, and brain cells (WA State Institute for Public Policy, 1985). The impact ionizing radiation has on living tissue is governed by the amount and duration of exposure, the amount of energy absorbed, and the penetrating power of the radiation.

Gamma electromagnetic rays and alpha, beta, and neutron particles are types of ionizing radiation. Gamma radiation is the most energetic type of radiation. Nuclear

fission, radioactive decay of fission products, and the decay of some unstable natural isotopes produce gamma radiation. Gamma rays can penetrate biological material, and can pass through several inches of metal. Concrete and lead are used to provide shielding from gamma rays.

Alpha particles are positively charged and consist of two neutrons and two protons. The radioactive decay of heavy isotopes of atoms produces alpha radiation. Alpha particles impart a large amount of energy within a short distance, but they cannot penetrate skin, or even a sheet of paper. Alpha particles can only cause damage to cells if they are inhaled or ingested, in which case they come into intimate contact with internal tissue cells.

Beta radiation consists of high-speed electrons. Beta particles can penetrate up to approximately one-half inch of biological material, but are easily stopped by a thin sheet of aluminum. Neutron radiation is found in nuclear reactors, and is produced by nuclear fission and radioactive decay. Neutrons are highly penetrating, and can pass through several feet of metal. Water or concrete is used to provide shielding from neutron particles.

Cesium-137 emits both beta particles and gamma rays as it decays. Cesium-137 first releases beta particles as it decays to the metastable ^{137m}Ba , which has a half-life of 2.6 minutes (NCRP, 1977). Following the initial deterioration to the unstable daughter isotope, a high-energy gamma ray is emitted as the ^{137m}Ba decays to the stable element barium-137 (^{137}Ba).

Radioactive isotopes exhibit half-lives (t_H), which are defined as the amount of time required for half of any sample of a particular radionuclide to decay. The physical half-life of ^{137}Cs is 30 years. At any time, the fraction of radioactive particles remaining can be calculated by the equation $f = (1/2)^P$, where P = the ratio of time passed to the half-life, or t/t_H (Murray, 1989). Thus, we can calculate that the fraction of ^{137}Cs deposited at the Hanford Site in 1950 that is still remaining today is approximately 31.5%. A general rule of thumb is that 10 half-lives are required to eliminate a radioisotope (Murray, 1989). In 300 years all but approximately 0.1% of the ^{137}Cs present at the Hanford Site will have decayed to ^{137}Ba .

The rate of decay, or activity, of a radionuclide can be expressed as the number of disintegrations the radionuclide undergoes per second. Marie Curie was the first person to quantify radioactive decay during her study of radium-226 (^{226}Ra). A gram of ^{226}Ra undergoes 3.7×10^{10} disintegrations per second, a value known as the curie, abbreviated Ci. The modern unit of radionuclide activity is the becquerel (Bq), which is equal to one disintegration per second.

Several terms are used to describe radiation dose. The amount of ionizing radiation absorbed by inanimate and living matter is measured in terms of the rad, which is short for radiation absorbed dose. The modern unit used to describe absorbed dose or specific energy is the gray (Gy), which is equal to 100 rad. When a person is exposed to more than one type of radiation the rem, or rad equivalent man, is used to equate the damage produced from each type of radiation to gamma rays. The modern unit used to describe the dose equivalent is the sievert (Sv), which is equal to 100 rem.

The average person receives an annual dose of approximately 1-2 millisieverts (1-2 mSv, or 100-200 mrem) from the combination of natural background and man-made sources, such as medical tests, mining of radioactive materials, fallout from nuclear tests, nuclear energy production, consumer products, and defense activities (WA State Institute for Public Policy, 1985). Most information regarding the effects of ionizing radiation on humans is based on cases of accidental exposure to high doses. High doses of radiation are known to result in cancers, shortened life span, sterility, anemia, genetic effects, birth defects (in the case of the exposure of a developing fetus), and death. Radiation doses between 0.5 and 2 Sv have been shown to cause radiation-induced cancers, and a single dose of 4.0-5.0 Sv of radiation will lead to the death of half of the exposed individuals within 30 days (WA State Institute for Public Policy, 1985; Murray, 1989). The specific effects of ionizing radiation depend on the distribution of radiation exposure over time, as well as the absorbed dose.

While the effects of high doses of radiation on human health are clearly deleterious, the hazards posed by exposure to low doses of radiation (less than .5 Sv) are not well understood. Some experts contend that a linear relationship exists between radiation exposure and health effects, and that the effects of low-level radiation exposure can be predicted by extrapolating from effects of high-level exposures.

Extrapolation of high-level exposure effects is regarded by some as a conservative estimate of the dose-effect relationship between radiation exposure and human health (WA State Institute for Public Policy, 1985). Assuming a linear relationship between radiation exposure and health effects does not take into account the ability of some cells to repair radiation damage.

A contradictory theory suggests that low doses of radiation may actually be more dangerous per unit than high doses. Some scientists contend that there is a strong associative relationship between low doses of radiation and certain types of cancer, and that the types of cancer that are induced by low doses of radiation are different than those induced by high doses (WA State Institute for Public Policy, 1985). In 1980, the Committee on the Biological Effects of Ionizing Radiations suggested that there is a higher cancer risk at lower radiation doses than linear extrapolation of high-dose data would suggest (WA State Institute for Public Policy, 1985). However, determination of the health effects associated with low-dose radiation has proven difficult to determine conclusively.

2.2 Creation of Radioactive Waste

Nuclear fission is the splitting of a nucleus into two parts, and it is triggered when a neutron strikes nuclei having large atomic weights and the neutron is absorbed. Plutonium is produced by the fission of uranium metal (Figure 2). When a neutron strikes uranium-238 (^{238}U), it is absorbed and uranium-239 (^{239}U) is formed. ^{239}U emits a beta particle as it decays to neptunium-239 (^{239}Np), which also emits a beta particle as it decays. The final product is plutonium-239 (^{239}Pu), which is weapons-grade plutonium.

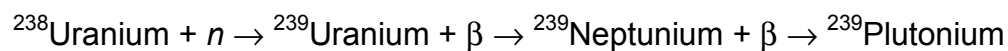


Figure 2. The chain reaction used to form weapons-grade plutonium at the Hanford Site.

The chemical processes used at Hanford to produce plutonium for nuclear weapons also resulted in the release of undesirable radioactive fission products including ^{137}Cs , ^{90}Sr , iodine-131 (^{131}I), krypton-85 (^{85}Kr), cesium-134 (^{134}Cs), cesium-135 (^{135}Cs), cerium-144 (^{144}Ce), iodine-129 (^{129}I), hydrogen-3 (tritium, ^3H), americium-241 (^{241}Am), and technetium-99 (^{99}Tc) (Murray, 1989; Gephart and Lundgren, 1998). Due to its relatively high yield of six atoms per hundred fissions, ^{137}Cs is one of the most significant fission byproducts of plutonium production (NCRP, 1977).

2.3 Reprocessing of Irradiated Uranium Fuel

A number of reprocessing techniques were employed to concentrate or reduce the volume of waste generated during plutonium production. The bismuth phosphate (BiPO_4) process, in operation at Hanford in 1944, was the first plutonium separation process used on an industrial scale at the Hanford Site (Gephart and Lundgren, 1998). The BiPO_4 process to separate plutonium from uranium and other radionuclides took place at T Plant in the 200-West Area and B Plant in the 200-East Area of the Hanford Site. While the BiPO_4 process successfully extracted plutonium from other fission products, it was unable to recover uranium for recycling into new nuclear fuel and large quantities of waste were generated.

The first step in BiPO_4 waste reprocessing involves dissolving the waste materials using sodium hydroxide (NaOH) and nitric acid (HNO_3). The resulting liquid was subjected to several precipitation processes to separate dissolved plutonium from other radioactive elements, namely chemical-induced precipitation using BiPO_4 and lanthanum fluoride (LaF_3). The BiPO_4 process yielded a plutonium nitrate (PuNO_3) paste and highly acidic waste containing uranium and other radionuclides. The acidic waste was neutralized and sent to single-shell underground storage tanks in the 200-West and 200-East Areas of the Hanford Site.

Between 1952 and 1958, wastes from the BiPO_4 process were diluted with water and pumped to the U Plant in the 200-West Area of the Hanford Site, where nitric acid was added to dissolve uranium remaining in the waste (Gephart and Lundgren, 1998). A solvent extraction process using tributyl phosphate and kerosene was used to recover uranium from the BiPO_4 waste. The remaining waste was pumped back into the single-

shell storage tanks then treated with ferrocyanide [$K_4Fe(CN)_6$] to precipitate cesium from the upper liquids in the tank. The addition of $K_4Fe(CN)_6$ to tank liquids largely removed the ^{137}Cs , making the liquids less radioactive. The uppermost liquids in the tanks were then discharged to the soil to make room for more HLW in the tanks.

Following World War II significant advances were made in solvent extraction chemical processes. Uranium and plutonium could be made soluble in some organic liquids, such as ethers, esters, and ketones, while undesirable fission products were insoluble in these liquids. The REDOX (for Reduction and Oxidation) process, which used methyl isobutyl ketone as the organic solvent with aluminum nitrate added to improve uranium and plutonium separation, was first used at the Hanford Site in 1952 (Gephart and Lundgren, 1998). The REDOX process offered some important advantages over the $BiPO_4$ process, including reduction in waste volume and the recovery of both uranium and plutonium. The REDOX process was in operation at the REDOX Plant located in the 200-West Area at the Hanford Site until 1967 (Gephart and Lundgren, 1998).

The REDOX process produced less waste volume than the $BiPO_4$ process because the REDOX plant had a concentrator that boiled the liquid to reduce waste volume before sending the waste to storage tanks. Waste from the REDOX process consisted principally of aluminum salts, which were made alkaline and sent to single-shell storage tanks in the 200-West and 200-East Areas.

In 1956, an advanced plutonium and uranium separations procedure known as the PUREX process (for Plutonium and Uranium Extraction) was employed at the Hanford Site (Gephart and Lundgren, 1998). The PUREX process differed from the REDOX process in that tributyl phosphate was used as the organic solvent and nitric acid, rather than aluminum nitrate, was added to improve uranium and plutonium separation from other radionuclides. Use of the PUREX process further reduced waste volume while providing greater flexibility in process conditions, posing less fire hazard, and decreasing operation costs. The PUREX Plant in the 200-East Area of the Hanford Site was in continuous operation from 1956 until 1972, and then was operated intermittently from 1983 to 1990 (Gephart and Lundgren, 1998).

Beginning in 1949, plutonium recovered as PuNO_3 through the BiPO_4 , REDOX, and PUREX processes was sent to Z Plant in the 200-West Area, a finishing plant where plutonium metal was created from PuNO_3 paste (Gephart and Lundgren, 1998). Waste from the finishing process contained low concentrations of fission products and high concentrations of metallic nitrates. Until 1973, waste from Z Plant was discharged to the soil. In 1973 a new regulation required that waste from the finishing plant be sent to the storage tanks rather than discharged into the ground (Gephart and Lundgren, 1998).

2.4 Storage of Waste

There were 149 single-shell waste storage tanks constructed at the Hanford Site between 1943 and 1964 to hold liquid radioactive waste from plutonium production. Four different sizes of single-shell tanks were constructed: 16 tanks with a 55,000 gallon capacity, 60 tanks with a 530,000 gallon capacity, 48 with a 758,000 gallon capacity, and 25 with a capacity of one million gallons. The single-shell tanks are cylindrical reinforced concrete with inner carbon steel liners. The domes of the tanks are comprised of concrete without a steel inner liner and covered with approximately ten feet of soil and gravel.

The 28 double-shell tanks at the Hanford Site were constructed between 1968 and 1986. Four of the double-shell tanks have a 1 million gallon capacity, while the remaining 24 tanks each hold 1.16 million gallons. The double-shell tanks are reinforced concrete structures with two carbon steel inner liners separated by a space. Tank leaks can be detected in the space, or annulus, between the steel liners. Unlike the single-shell tanks, both the cylinders and domes of the double-shell tanks at the Hanford Site are completely lined by steel. All of Hanford's waste storage tanks were built in groups called tank farms, which contain underground pipes so that waste can be moved between tanks. Seven tank farms are located in the 200-West Area, and eleven are located in the 200-East Area.

As early as 1956, waste leakage from storage tanks was suspected. The first storage tank leak was confirmed in 1961. Radioactive waste leaks have been detected from 67 of the 149 single-shell tanks at Hanford. This is not surprising considering that

the design life of the single-shell tanks was 10 to 20 years. All of the single-shell tanks at the Hanford Site have already exceeded their design life by more than 30 years. The double-shell waste storage tanks at Hanford had a design life of 25 to 50 years, and none of the double-shell tanks are known to have leaked. Waste from leaking single-shell tanks has been transferred to secure double-shell tanks. Waste removal from single- and double-tanks is expected to be completed in 2018 and 2028, respectively. By this time, the average single-shell tank will have exceeded its design life by approximately 50 years, and most double-shell tanks will also have exceeded their design lives.

2.5 Waste Mixtures

The radioactive and chemical contents of individual waste storage tanks are something of a mystery. Because different processes were used to remove plutonium and uranium from wastes, different chemicals and various metals have been added and there is insufficient record of waste composition and the volumes of waste transferred between tanks. Neutralization and radionuclide-scavenging processes have made wastes more chemically complex, and in some cases have created a safety concern. Waste contained in some storage tanks at the Hanford Site generates heat and flammable gases, which poses potential risks to workers, the public, and the environment (Gephart and Lundgren, 1998).

Since its deposition in storage tanks, the waste at the Hanford Site has separated into complex mixtures. Individual tanks may contain one or more different types of waste. Most of the waste storage tanks contain some amount of vapor at the top, which generally contains small amounts of hydrogen, nitrous oxide, ammonia, and water. The different types of waste found in the Hanford tanks are (Gephart and Lundgren, 1998):

- Supernatant liquid – a clear liquid that generally floats above a layer of settled solids.
- Interstitial liquid – the liquid found within pore spaces of saltcake and sludges.
- Sludge – a thick layer of water-insoluble chemicals precipitated to the bottom of a tank; tend to have small pore spaces which inhibit draining or pumping of liquids.

- Saltcake – a moist material created by the crystallization and precipitation of chemicals following the evaporation of the supernatant liquid; usually comprised of water-soluble chemicals.
- Slurry – a mixture of solids suspended in a liquid.

Materials such as organic ion exchange resin, cement, diatomaceous earth, plastic bottles, and metal parts are also present in some tanks. All these different wastes present in the storage tanks comprise a physically and chemically complex mixture that will be difficult to characterize, separate, process, and dispose of. The major problems with waste characterization are related to: the lack of sufficient record-keeping; use of multiple irradiated fuel reprocessing techniques; neutralization of acidic waste with NaOH, which caused precipitation and solidification of some waste; evaporation of some tank liquids leading to the formation of hard saltcakes and thick slurries, and the heterogeneous distribution of chemical compound and radionuclides; and, the addition of ferrocyanides (Gephart and Lundgren, 1998). Multiple samples will be required from each of Hanford's 177 waste storage tanks to determine key chemical and physical characteristics, making waste remediation more difficult and costly.

3. Soil and Groundwater Contamination

3.1 Source of Contamination

During the years of operation, there were several methods of disposal of wastes generated during plutonium production at the Hanford Site. Nine nuclear reactors were operated in the 100 Areas of the Site between 1944 and 1987. With the exception of the N reactor, the nuclear reactors operated at the Hanford Site discharged contaminated cooling water directly into the Columbia River (Delistraty and Yokel, 1998). Nuclear wastes were also directly disposed of in the ground by discharging liquids into cribs, or shallow buried tile fields. Liquid waste discharged into cribs eventually percolated to the groundwater (Gephart and Lundgren, 1998). Wastes generated from nuclear fuel fabrication and research in the 300 Area of the Hanford Site

were discharged into holding ponds near the Columbia River (Delistraty and Yokel, 1998).

During the 1950's, small-scale waste leaks from underground pipes and fittings in the tank farms were reported. However, it was 1959 before significant waste releases were confirmed. The first waste storage tank to leak was tank 104 in the U tank farm, a single-shell tank that leaked an estimated 55,000 gallons of waste before tank contents were transferred (Gephart and Lundgren 1998). Two additional tank leaks were confirmed in 1959: tank 101 in the U tank farm released approximately 30,000 gallons and tank 106 in the TY tank farm released an estimated 20,000 gallons (Gephart and Lundgren, 1998). The largest waste storage tank leak detected at the Hanford Site was a release of approximately 115,000 gallons from tank 106 in the T tank farm in 1973 (Gephart and Lundgren, 1998).

To date, 67 single-shell waste storage tanks at Hanford are known or suspected to have leaked significant amounts of radioactive waste, but no double-shell tanks are known to have leaked. Estimates on the total volume of waste that has leaked from the underground storage tanks at the Hanford Site range from 0.6 to 1.4 million gallons (Gephart and Lundgren, 1998). These estimates correspond to a total of 1 to 2 million Ci of radiation leaked from the storage tanks, primarily from ^{137}Cs .

While several methods are employed, leak detection is an imprecise activity in the case of single-shell tanks. Vertical monitoring wells referred to as drywells because they do not extend to the water table, were drilled around the single-shell tanks. Approximately 760 drywells around single-shell tanks are currently used to measure increases in radiation caused by waste release at the Hanford Site (Gephart and Lundgren, 1998). Horizontal drywells called laterals were also drilled underneath the single-shell tanks to measure radiation levels in the soil. A third leak detection method used for single-shell tanks at Hanford is to lower a radiation probes into the liquid observation well inside the storage tanks. The radiation probe method uses a measure of radioactivity to identify the level of liquid in an individual waste storage tank. Large leaks can be detected by comparing the current liquid level with a previously measured liquid level.

Detectors in the annuli between the two steel liners are the primary method used to monitor waste leaks in double-shell tanks. Some double-shell tanks are also equipped with a leak detection pit beneath the tank's outer liner should waste bypass both steel barriers in the tank. All of the double-shell tanks have a camera observation port through which a camera may be lowered to visually observe liquid levels.

Drainable liquid from the single-shell storage tanks at the Hanford Site has been pumped to the double-shell tanks in an effort to reduce the quantity of radioactive waste leaking from the single-shell tanks. An estimated 6 million gallons of liquid remain in the single-shell tanks within saltcake and sludge (Gephart and Lundgren, 1998). Some liquid waste is expected to leak from the single-shell tanks during tank cleanup because the removal of waste solids and non-pumpable sludges will require the addition of fluids to some waste storage tanks.

The nuclear reactors are no longer operating at Hanford, and wastes are no longer being generated. The current contamination of soils and groundwater at the Hanford Site is the result of activities occurring at the Site between 1944 and 1987. The only present source of radioactive waste is leaking waste storage tanks in the 200 Areas of the Hanford Site.

3.2 Waste Transport Pathway

The sediments underlying soils at the Hanford Site were formed over the last 8 million years in lacustrine and eluvial deposits from cataclysmic glacial floods (Boling et al., 1998). The uppermost sediments, which extend 200 to 300 feet below the surface, comprise the Hanford Formation. These soils consist of very permeable sands and gravels. The lower sediment layer, the Ringold Formation, varies from 200 to 600 feet in thickness. The Ringold Formation contains a mixture of clays, sands, silts, and gravels and is much less permeable than the Hanford Formation. Basalt bedrock underlies the soils and sediments at the Hanford Site.

The soils at the Hanford Site are predominantly dry and sandy, and were formed in sandy to silty eolian, or wind-blown, materials over glaciofluvial or glaciolacustrine deposits (Boling et al., 1998). The classification of soils at the Hanford Site as Xeric Torripsamments and Xeric Torriorthents indicates that these areas are dry, loamy sands

(Buol et al., 1997; Boling et al., 1998). Sandy soils, such as those at the Hanford Site have low water-holding capacity. Some of the contaminants directly discharged into the soil at the Hanford Site because absorbed to the soil solid phase, but others percolated through the soil matrix into the groundwater. Groundwater contamination is of particular concern in highly permeable soils, such as those in the Hanford Formation.

The upper surface of the water table lies 200 to 300 feet below ground level. In some areas at the Hanford Site, particularly the eastern half of the 200-East Area, groundwater exists within the Hanford Formation, which permits facile movement of contaminated groundwater outward from these areas. Depth to the water table decreases between the 200 Areas and the Columbia River, so groundwater from beneath the tank farms will eventually discharge to the surface of the river. The rate at which radioactive contaminants are discharged to the Columbia River depends on several factors, including sediment permeability, the slope of the water table, rate of groundwater flow, extent of chemical adsorption, and radionuclide decay. Radioactive wastes are no longer discharged into the Columbia River, thus the only active contaminant transport pathway is via groundwater.

An estimated 150 square mile plume of contaminated groundwater exists underneath the Hanford site (Gephart and Lundgren, 1998). This plume of contamination resulted from the release of an estimated 450 billion gallons of liquid radionuclide and hazardous waste since 1944, 346 billion gallons of which were released in the 200-East and 200-West Areas (Gephart and Lundgren, 1998). A portion of the wastes released to soil remains physically sequestered in or chemically sorbed to sediments overlying the groundwater. Contaminants presently contained in the soils and sediments are not necessarily irreversibly bound to the soil matrix, but the rate at which they may be released into the groundwater is presently unknown.

3.3 Receptors

Soil organisms and the aquatic biota are the primary receptors of contamination from the Hanford Site. Soil and aquatic organisms will be directly exposed to wastes, possibly for long periods of time. Contamination will be greatest in the soil directly underneath the leaking storage tanks and in areas historically used to dump wastes.

Contaminant concentrations will decrease as the wastes are diluted in groundwater or surface waters. Duration of exposure will determine the dose received. Humans in the area will also be exposed to some contaminants, primarily through hydrospheric pathways (e.g., drinking water, irrigation water, recreation in contaminated areas of the Columbia River, etc.), consumption of contaminated organisms, and through inhalation or ingestion of windblown dust.

In addition to radionuclides, wastes present in the soils underlying the Hanford Site contain toxic levels of many metal species. The waste is also extremely alkaline, and the high alkalinity and metal concentration make affected areas in the soils and sediments inhospitable to many indigenous microorganisms. Alteration of the subsurface chemical environment by waste materials will lead to altered community structure and reduced metabolic diversity (Stephen et al., 1999).

A study was undertaken by the Washington State Department of Ecology to evaluate the ecotoxicity of sediments along the Hanford Reach of the Columbia River. Sediments were collected from riverbank spring and riverbed sites along the 100 and 300 Areas of the Hanford Site. Analyses indicated varying levels of chromium, ^{137}Cs , and ^{90}Sr in the collected sediments. Both growth and survival of the freshwater amphipod *Hyalella azteca* were significantly lower in the presence of sediments collected in riverbank spring sites near the 100 Area of the Hanford Site as compared to an uncontaminated control (Delistraty and Yokel, 1998).

A corresponding study undertaken by the Washington State Department of Ecology sought to evaluate the toxicity of riverbank spring water collected along the Hanford Reach. The measurements of the springwater flowing from underneath Hanford into the Columbia River reflect the toxicity of the groundwater on the Hanford Site. Short-term chronic bioassays with fathead minnows (*Pimephales promelas*) and daphnids (*Ceriodaphnia dubia*) showed significant reduction in survival for both species in springwater from several test locations (Delistraty and Yokel, 1999). Daphnid reproduction was the measured variable most affected by the springwater, whereas fathead minnow reproduction was the least affected variable (Delistraty and Yokel, 1999). Delistraty and Yokel (1999) noted the importance of using several test species,

and concluded that further research is required for comprehensive ecological risk assessment of riverbank spring water.

In attempting to trace contaminant movement, effort should be made to understand the behavior of contaminants in a food web. In a food web, higher trophic-level species are expected to accumulate greater quantities of contaminants over time than species in lower trophic levels. Fish are frequently monitored to assess contaminant flow in aquatic food webs. Cesium-137 has a particular tendency to reconcentrate in aquatic ecosystems, primarily because cesium and potassium behave in a similar manner in biological systems (NCRP, 1977). Several studies have shown that water retention time and temperature are positively correlated with cesium accumulation in fish while a general negative correlation is seen between cesium activity in fish and the ionic strength of the water, particularly potassium concentration (Paller et al., 1999; Hessen et al., 2000).

A model designed to estimate the amount of time required for 50% reduction, or ecological half-life, of ^{137}Cs in brown trout (*Salmo trutta*) indicated that the ecological half-life of ^{137}Cs was considerably shorter than its radioactive half-life (Hessen et al., 2000). Ecological half-life of ^{137}Cs in Norwegian brown trout was approximately 2.5 years, a much shorter time period than the radioactive half-life of 30.2 years (Hessen et al., 2000).

Some evidence suggests that the decline of ^{137}Cs in fish may be more complex than initially believed. Since 1986, Jonsson et al. (2000) have measured ^{137}Cs levels in nearly 4,000 brown trout and Arctic char (*Salvelinus alpinus*). The researchers have concluded that cesium decline modeled by a simple one-component decay function underestimates the persistence of ^{137}Cs in fish (Jonsson et al., 2000). Jonsson et al. (2000) found that a two-component decay function provides a better model fit to the data collected, and suggests two contaminant pools. Cesium-137 contamination in Scandinavia is due to fallout from the accident at the Chernobyl nuclear reactor in 1986. Contamination was first introduced to water bodies by direct atmospheric deposition and runoff from catchment surfaces. Slow ^{137}Cs leaching from the lake catchment and cesium recycling within the lake may provide the second contaminant pool (Jonsson et al., 2000).

Cesium concentrations in fish are affected by the length of the food chain and the choice of prey organisms (Hessen et al., 2000). Organisms at the highest trophic levels are expected to exhibit the greatest degree of contaminant bioaccumulation. A study of ^{137}Cs biological half-life in fish occupying water bodies on the Savannah River Site, a nuclear weapons production facility in South Carolina, showed that piscivorous species generally contained the highest ^{137}Cs concentrations (Paller et al., 1999). Largemouth bass (*Micropterus salmoides*), which prey on other fish, had significantly higher ^{137}Cs body burdens than sunfish (*Lepomis* spp.) or bullheads (*Ameiurus* spp.), which occupy a lower trophic level (Paller et al., 1999). These results indicate that if restrictions are to be placed on consumption of fish in the Columbia River it is important to consider species in different trophic levels separately, as higher trophic-level species may bioaccumulate a significantly greater quantity of ^{137}Cs and other contaminants.

Consumption of fish from the Columbia River is only one pathway by which humans may be exposed to waste from the Hanford Site. Communities that extract drinking water from the river will have a higher rate of exposure than those exploiting groundwater resources. In addition, a substantial portion of the agricultural crops in the Columbia River basin is grown using irrigation water from the river. Soil-to-plant transfer of radionuclides depends on adsorption/desorption processes in the soil and plant root uptake processes (Nisbet and Woodman, 2000). Research indicates that soil-to-plant transfer of ^{137}Cs and ^{90}Sr is highest in sandy soils for cereal crops, green vegetables, brassicas, root vegetables, and legumes (Nisbet and Woodman, 2000). Soil-to-plant transfer values reported in a study by Nisbet and Woodman (2000) ranged from 0.0038 to 3.2 Bq kg⁻¹ dry plant mass per Bq kg⁻¹ dry soil mass. These results indicate that consumption of radionuclides in crops irrigated with water from the Columbia River as part of a normal diet is unlikely to cause significant human health problems due to the low radionuclide concentrations in the Columbia River.

Minor pathways through which humans may be exposed to nuclear wastes include recreation in the Columbia River downstream from the Hanford Site, inhalation of windblown dust, and consumption of terrestrial animals that have been consuming contaminated water and/or vegetation. The dose to which a human may be exposed by participating in any of these activities is likely to be extremely small.

3.4 Controls

A great deal of effort has been directed toward cleaning up the contamination at the Hanford Site since it was placed on the National Priorities List for environmental cleanup by the U.S. Environmental Protection Agency in 1988. Current cleanup priorities include the removal of nuclear waste from storage tanks to reduce risk of tank leakage. This is a daunting task, as wastes must first be safely dislodged and removed from the underground storage tanks. Because solid wastes remain in some of the tanks known to leak, subsurface barriers may have to be constructed prior to dissolution and removal of the solid wastes from these tanks. In addition, many of the pipes designed to transfer wastes between storage areas are clogged or leaking (Gephart and Lundgren, 1998).

Removing wastes from the storage tanks is only the first step in a long process. Wastes must be pretreated and separated to prevent hazardous chemical reactions between incompatible compounds. This step is complicated by the lack of characterization data available for the wastes contained in the storage tanks. Eventually, the goal is to separate radionuclides from other chemical compounds, destroy the organic compounds, and separately solidify LLW and HLW in a process known as vitrification. In vitrification, liquid tank wastes are turned into glass, thereby inhibiting radioactive materials from escaping into the environment. Vitrified wastes are suitable for long-term storage, but a location for the long-term storage of waste from the Hanford Site has not yet been determined.

Even after the nuclear wastes are removed from the underground storage tanks at the Hanford Site, wastes that have leaked into the soil and groundwater will remain. A number of studies are underway to characterize radionuclide sorption/desorption processes and mechanisms of radionuclide transport in Hanford sediments. Studies evaluating the ecological effects of the Hanford waste are ongoing. There is not enough information available at this point to make any decisions regarding remediation of contaminated sediments and groundwater at the Hanford Site.

4. Conclusion

What is the real risk posed to humans and wildlife occupying areas near the Hanford Site? When the Columbia River passes through the Hanford Site its radioactivity doubles (Gephart and Lundgren, 1998). Approximately 6,000 Ci of radioactivity flow down the Columbia River each year to Hanford, and approximately 12,000 Ci flow downstream from the site annually. The mean annual flow rate of the Columbia River is approximately 28 trillion gallons (Gephart and Lundgren, 1998). This equates to an average concentration of nearly 4.3×10^{-10} Ci, or 1.16×10^{-20} Bq, radioactivity per gallon of riverwater. This is a very small amount of radiation.

Further study is required to determine unequivocally whether or not the waste leaked from the underground storage tanks at the Hanford Site presents a significant human health risk. The main difficulty lies in determining the health effects of very small doses of radiation over time. Present indications are that the contaminated soil and water underlying the Hanford Site will present a low risk to the public (Gephart and Lundgren, 1998). More information regarding long-term, low-dose radiation exposure is required to accurately estimate the amount of risk associated with contamination from the Hanford wastes.

The Hanford Site cleanup will continue for at least another 30 years (Gephart and Lundgren, 1998). Between 1989 and 1996 the Department of Energy spent \$7.5 billion cleaning up the Hanford Site, and the DOE expects to spend at least \$1 billion per year for the duration of the project (Zorpette, 1996). The magnitude of contamination at the Hanford Site and the potential human health impacts of radionuclides in the environment make the environmental degradation at the Site one of the worst environmental disasters of the 20th century. Although there are few positives in all this, the lessons learned at the Hanford Site may prevent the same mistakes from being made in the future, and the technological advances and remedial technologies developed during the cleanup can be applied to other industries and environmental problems.

Appendix A: Measurements of Radioactivity

Measures of rate of decay

1 curie (Ci) = 3.7×10^{10} disintegrations per second (d/s)

1 becquerel (Bq) = 1 d/s

SI unit = Becquerel

Measures of Dose

1 radiation absorbed dose (rad) = 0.01 W-s/kg

1 rad = 0.01 grays (Gy)

SI unit - Gray

1 rad equivalent man (rem) = 0.01 sieverts (Sv)

SI unit = Sievert

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