

OVERVIEW OF HANFORD OPERATIONS

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PREFACE

Following this preface this report is in two parts. "Part I. Introduction" and "Part II. Overview of Hanford Operations" are meant to provide the reader with information to understand what type of activities took place at Hanford, how these activities led to significant releases of radioactive materials, how these radioactive materials exposed members of the public to excessive and harmful levels of radiation, and how scientist have attempted to reconstruct the radiation exposure to these groups of people and estimate the health effects caused by this exposure.

I. INTRODUCTION

A. Terminology.

In this report ``**radiation**," refers to energetic x-rays, gamma-rays, alpha particles, beta particles and neutrons. These different forms of radiation are emitted when radionuclides undergo radioactive decay. They are referred to as ``**ionizing radiation**" because as they pass through, or are absorbed within matter, including human tissue, they excite or ionize atoms and molecules. This in turn can cause chemical and physical changes in the matter. When this radiation is absorbed in the human body these changes can result in deleterious health effects, including cancer and genetic effects.

Chemical compounds are made up of **elements**, of which there are just over 100, e.g., hydrogen, helium, oxygen, lead, zinc, uranium, plutonium, etc. A hydrogen atom consisting of a nucleus containing a single proton and an electron orbiting around it. The nucleus of a helium atom has two protons in the nucleus and two orbiting electrons. Heavier elements have an even greater number of protons. Elements are distinguished by the number of protons in the nucleus of the atom. Uranium has 92 protons in the nucleus and plutonium has 94. With one exception the nuclei of atoms also contains neutrons, which are similar to protons but without an electrical charge. Atoms of the same element must have the same number of protons, but can have a different number of neutrons. We distinguish them by calling them ``**isotopes**" of the element. Thus, an element can have several isotopes. We call the collection of isotopes of the various elements ``**nuclides**." We distinguish and identify the nuclides by the name of the element and a number that identifies the total number of protons and neutrons in the nucleus. For example, uranium-235 has 92 protons and 143 neutrons ($92+143=235$) in the nucleus, and uranium-238 has 92 protons and 146 neutrons ($92+146=238$). For our purpose we can use the terms ``nuclides" and ``isotopes" interchangeably.

Many isotopes (or nuclides) are stable, meaning they stick around essentially forever. The element lead, for example, has four stable isotopes: lead-204, lead-206, lead-207 and lead-208. Most isotopes (or nuclides) are not stable, but undergo spontaneous emissions of radiation by a process that we call ``**radioactive decay**." We call these ``**radioisotopes**" (shorthand for ``radioactive isotope"), or ``**radionuclides**" (shorthand for ``radioactive nuclide"). As before, for our purposes the terms ``radionuclide" and ``radioisotopes" are interchangeable.

The process of radioactive decay, or the property of the radionuclides (or radioisotopes) to emit radiation, is called ``**radioactivity**." Natural radioactivity has been known since 1896 when Becquerel discovered that uranium compounds emitted a penetrating radiation similar to x-rays, which we now call gamma-rays. Some radionuclides emit particles when they undergo radioactive decay. These include alpha particles, beta particles and neutrons. **Alpha particles** are two protons and two neutrons bound together, which makes them the same as the nucleus of a helium atom. **Beta particles** are electrons emitted from the nucleus.

When a radionuclide emits an x-ray or a gamma-ray, the number of protons and neutrons in the nucleus remains unchanged, and therefore it is left as the same nuclide but in a different "energy state." But if a radionuclide emits an alpha particle, beta particle, or neutron, it changes into a new isotope that may be stable or itself radioactive. The original nuclide literally "disintegrates" or "decays." As noted above we call the process "radioactive decay," and we refer to the new nuclide as a "radioactive decay product," or "daughter product." Thus, "radioisotopes" emit "radiation" in a process we call "radioactivity" or "radioactive decay."

We also refer to the amount of "radioactivity," meaning the number of spontaneous disintegrations per unit time. There are two commonly used units of measure for this. The older unit, and the one most often used by the Hanford Environmental Dose Reconstruction (HEDR) project, is called the "curie" (abbreviated "Ci"), defined as 37,000,000,000 disintegrations per second. This unit was chosen in 1930 because it is the number of disintegrations per second taking place in one gram of naturally occurring radium. A newer unit, called the "becquerel" (abbreviated "Bq"), is defined as one disintegration per second. Thus, $1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$, where 3.7×10^{10} is a short hand way of writing 37,000,000,000.

B. Radioactive Decay.

Each radionuclide decays (or disintegrates) at its own characteristic rate. It is convenient to measure the rate of radioactive disintegration of a particular radionuclide in terms of its "half-life," defined as the time it takes for the concentration to be reduced to one-half of the amount that was there at time zero, that is, at the time when one starts counting. For example, the radioactive half-life of iodine-131 is known accurately to be 8.040 days (8 days, 57 minutes and 36 seconds). This means that regardless of when you started counting, after 8.040 days only one-half of the iodine-131 that was there when you started counting is left. Stated another way, the amount of iodine-131 is reduced by another factor of 2 every 8.04 days. The half-lives of some of the other radionuclides of interest to us are:

<u>Nuclide</u>	<u>Half-life</u>
strontium-90	28.5 years
ruthenium-103	39.258 days
ruthenium-106	1.020 years
iodine-131	8.040 days
cesium-137	30.0 years
cerium-144	284.9 days
plutonium-239	24,110 years
plutonium-240	6,563 years

Because the number of disintegrations of a particular isotope occurring per unit time is dependent on the amount present, it turns out that so long as the nuclide is not being replenished, then

at the end of 2 half-lives the concentration is reduced by a factor of $2 \times 2 = 4$
 at the end of 3 half-lives the concentration is reduced by a factor of $2 \times 2 \times 2 = 8$
 at the end of 4 half-lives the concentration is reduced by a factor of $2 \times 2 \times 2 \times 2 = 16$
 ...
 ...
 ...
 at the end of 10 half-lives the concentration is reduced by a factor of $2^{10} = 1024$.
 and so forth.

Students of calculus have a shorthand way of expressing the same thing:

$$I = I_0 e^{-\lambda t},$$

where I is the concentration at time t , I_0 is the concentration at time $t = 0$, $e \approx 2.71828$ (called the Naperian constant), and $\lambda = \ln(2)/T_{1/2}$, where $T_{1/2}$ is the half-life, and $\ln(2) \approx 0.69315$. Since the times of interest are rarely exact multiples of one half-life, the calculations are performed using this more complicated formula. Otherwise, the results are the same.

C. Assessing Health Effects.

In assessing the health effects due to exposure to radioactive materials, it is sometimes convenient to divide the calculations into several logical components. The first of these, called the "**source term**," identifies the types and quantities of radionuclides released as a function of time and the locations of the release points. Once the source terms have been identified, one calculates the transport of the radionuclides from the points of release to where the radionuclides can be either directly inhaled or ingested, or to where people can be exposed to external radiation from radionuclides concentrated in the surrounding air, water or as surface deposits. This is called the "**transport**" or "**pathway**" analysis.

Some radionuclides are in the form of particulate, or attach themselves to particulate, such as dust or rain drops. Others remain a gaseous form, for example, the noble gases krypton-85, or xenon-133. Radionuclides that are in the form of, or that attach themselves to, particulates can fall out of the atmosphere along the transport pathway and concentrate on the ground, in water, or on vegetation. Ground deposits can be re-suspended and blown to other locations, or can be leached into the soil locally. Once leached into the soil or groundwater some fraction of the radioactivity is taken up by plants. Since humans eat vegetables, meat and dairy products and drink water, the transport analysis must also consider a variety of food chains. Measured concentrations of radionuclides in/on the ground, water, vegetation and foods provide additional sources of data for estimating the releases and transport of selected radionuclides and to check

estimates made by other means. These environmental measurements often provide a better starting point for estimating exposure to humans. Alternatively, they can be used to "renormalize" calculations based on source term and transport analyses.¹

Unlike a chemical poison, the damage caused by radioactive materials is not caused by the radionuclides, but by the radiation they emit. The radiation can come from radionuclides undergoing radioactive decay while inside or outside of the body, so long as the radiation is absorbed within. Thus, the next step is to calculate the **radiation dose** to different parts of the body as inhaled or ingested radionuclides pass through, or concentrate in, various organs, other tissue, or the skeleton; or as the various body parts are irradiated by external gamma-rays and other types of radiation from the nearby concentrations in the air, in the water, or on surfaces.

For many years the basic unit of **absorbed dose** was the **rad**, defined as the absorption of 100 ergs of radiation in one gram of matter. Subsequently all radiation units have been converted to the metric system. [NCRP Report No. 82] The metric unit of absorbed dose is the **gray**, defined as the absorption of one joule of energy in one kilogram of matter and abbreviated (Gy). Since the biological damage caused by a given absorbed dose may differ depending upon the type of radiation -- x-radiation, beta-radiation, gamma-radiation, alpha radiation, or neutrons -- there is another quantity called the **dose equivalent** such that a unit of dose equivalent causes the same biological damage regardless of the type of radiation. The old unit of dose equivalent is the **rem**, and the new metric system unit is the **seivert**, abbreviated (**Sv**). To convert from the old system to the new metric system of units:

$$1 \text{ Gy} = 100 \text{ rad}$$

$$1 \text{ Sv} = 100 \text{ rem}$$

These quantities, as well as others such as the curie (Ci), are also expressed with a prefix that is used to indicate a decimal multiple or submultiple of the basic unit, e.g.,

$$1 \text{ millirem} = 1 \text{ mrem} = 0.001 \text{ rem}$$

$$1 \text{ microCi} = 1 \text{ } \mu\text{Ci} = 0.000001 \text{ Ci}$$

$$1 \text{ nanoCi} = 1 \text{ nCi} = 0.000000001 \text{ Ci}$$

$$1 \text{ picoCi} = 1 \text{ pCi} = 0.000000000001 \text{ Ci}$$

To convert from rad to rem (or Gy to Sv, mGy to mSv, etc.), the rad (or Gy) is multiplied by a quality factor and other modifying factors where appropriate. For x-radiation, beta-radiation and gamma-radiation the quality factor and the other modifying factors are all usually assumed to equal to one. In this case, 1 rad = 1 rem, and 1 Gy = 1 Sv. Many of the isotopes of interest in this proceeding, including iodine-131, emit beta- and/or gamma-radiation. For radiation emitted from such isotopes:

¹ "Renormalization" is the process of adjusting the parameters of the model, so that the model predictions represent an improved fit to available data.

$$1 \text{ rem} = 1 \text{ rad} = 0.01 \text{ Gy} = 0.01 \text{ Sv.}$$

or

$$1 \text{ Gy} = 1 \text{ Sv} = 100 \text{ rad} = 100 \text{ rem.}$$

Also, in such situations it is convenient to use the abbreviated term "dose" to refer to absorbed dose or dose equivalent.

Finally, one must estimate the **health effects** caused by the radiation absorbed in the various body parts.

All of these calculations are complicated by the fact that the releases, transport and exposures are all time dependent, and more often than not much of the desired data is missing and must be approximated if it is not to be ignored. Measurements are often sparse, uncertain, and sometimes erroneous. Scientists deal with these problems using a variety of formal and informal approaches, including by making "best estimates," giving ranges to estimates, and by attaching "uncertainty limits" to estimates.

II. OVERVIEW OF HANFORD OPERATIONS

A. Early History.

In early 1943, the Hanford site in the state of Washington was selected by the U.S. Corp of Engineers' Manhattan Engineering District (the co-called Manhattan Project) to build several nuclear reactors and related facilities to produce plutonium for atomic bombs. A 200 square-mile tract was acquired by the U.S. Government, and construction of the Hanford Engineering Works was begun in March 1943.

B. The Reactors.

Construction on the first plutonium production reactor, the B Reactor, was begun on 7 June 1943 and it began operating in September 1944. Subsequently, eight additional plutonium production reactors--called ``piles" in the early years--were built and operated at Hanford. The operating periods of the nine plutonium production reactors were [Ballinger, PNL-6964, March 1991, p. 3.15]:

<u>Reactor</u>	<u>Initial Start Up Date</u>		<u>Final Shut Down Date</u>	
B	September	1944	February	1968
D	December	1944	June	1967
F	February	1945	June	1965
H	October	1949	April	1965
DR	October	1950	December	1964
C	November	1952	April	1969
KW	January	1955	February	1970
KE	April	1955	January	1971
N	December	1963	January	1987

The last reactor, the N Reactor, was initially called the ``New Production Reactor." It differed from the original eight reactors in that it was designed not only to produce plutonium, but also to byproduce steam which was sold for commercial electricity generation. As such it is called a dual purpose reactor.

All nine reactors were graphite-moderated light-water cooled reactors located at the north sector of the site along the Columbia River in what was called the 100 Area. ``Graphite-moderated" means the neutrons in the nuclear reactor are purposely slowed down by carbon in the form of graphite blocks. ``Light-water" means ordinary water, as opposed to ``heavy water" where the nuclei of the hydrogen atoms (which are chemically combined with oxygen atoms to form water molecules) have an extra neutron in them. The original eight reactors had single-pass (once-through) cooling systems, meaning that water from the Columbia River was pumped directly through reactor core to cool the reactor. The N Reactor had a recirculating primary

coolant system meaning the water that flowed through the reactor to cool it was continually recycled through the reactor. In this case the river water never flowed through the reactor, but instead flowed through a separate heat exchanger where it cooled the primary coolant.

Reactor Operations. The core of each Hanford production reactor consisted of a lattice of horizontal tubes, or channels, embedded in large stack of graphite blocks, called a moderator. Most of the tubes contained reactor fuel elements in the form of cylindrical slugs of uranium metal clad with a thin layer of aluminum (or zirconium in the case of the N Reactor). Some tubes contained reactor control rods, instrumentation, and occasionally targets to make materials other than plutonium. Coolant water also passes through the channels to cool the fuel. The number of channels and the amount of fuel depended on the reactor design and the desired reactor power. Typically, there were more than one thousand channels and several hundred tons of uranium fuel in each reactor. The number of uranium fuel slugs in each channel varied, but during the early period typically was on the order of 32-35 slugs per channel.

In the early years the reactor fuel was made from natural uranium which consists of 0.711% uranium-235 (U-235) and 99.289% uranium-238 (U-238). Beginning in the early-1950s, a ring of channels near the outside (comprising only a few percent of the total fuel) were filled with high-enriched uranium (HEU)--that is, uranium with a high concentration of U-235--to flatten the power profile across the reactor core.² The fuel in the N Reactor was low-enriched uranium (LEU) in two zones--a zone of 0.95% U-235 and a zone of 1.25% U-235 fuel.

When the control rods, which are made of neutron absorbing material, are partially removed from the reactor core, a nuclear chain reaction begins in the uranium fuel. The nuclei of the U-235 atoms fission, releasing thermal energy (heat) and on average about 2.5 neutrons each. The graphite moderator serves to slow the neutrons down to very slow speeds where they are preferentially absorbed by the U-235.

On average at least one of the neutrons from each fission must be reabsorbed and fission another nucleus in order to maintain the chain reaction. A fraction of the excess neutrons are absorbed by the more plentiful U-238, producing plutonium-239 (Pu-239), the principal isotope in the plutonium used in nuclear weapons. The Pu-239 is also a fissile material and serves as added fuel, just like the U-235. When Pu-239 absorbs neutrons it usually fissions, but sometimes it captures neutrons producing Pu-240. Heavier isotopes of plutonium are produced by the capture of neutrons by Pu-240. The longer the fuel is left in the reactor the more Pu-239 and Pu-240 is produced.

For the production of atomic bombs and other nuclear weapons, Pu-240 can be troublesome because its high rate of spontaneous fissions can initiate the nuclear chain reaction in the weapon prior to the optimum time for achieving the maximum yield of the weapon. Consequently, the

² The power profile is the spacial distribution of the rate at which energy is produced in the reactor. The rate of energy production, which in turn is proportional to the rate of fissions. The peak (or highest) power level in the reactor is safety limited to prevent fuel melting. Thus, by flattening the power profile the average power level throughout the reactor can be increased thereby increasing the rate of plutonium production.

Hanford operators were required to keep the Pu-240 concentration low.

Plutonium is graded according to its Pu-240 concentration:

<u>Plutonium Category</u>	<u>Percent Pu-240</u>
Supergrade (high purity)	2 to 3
Weapon-grade	less than 7
Fuel-Grade	7 to less than 19
Reactor-Grade	19 or greater

To prevent the buildup of Pu-240 beyond weapon-grade, the uranium fuel must be removed from the reactor early and often, typically after residing in the reactor for only a couple of months, or so. Thus, reactors primarily for weapon-grade plutonium production are designed for rapid refueling and require a large amount of fuel to be "pushed" through the reactor. The N-Reactor was somewhat different in this regard. During most of its operating life the N-Reactor was optimized for electricity production, and produced fuel-grade plutonium for use in civil research and development reactors.

During the nuclear chain reaction, when the U-235 (and Pu-239) nuclei fission, they almost always split into two unequal masses but these two masses are not always the same. The U-235 atom, for example, splits up in more than forty different ways and into nuclides with more than 90 mass numbers. Consequently, the fission process produces a wide range of radionuclides, collectively called fission products.

The composition of uranium and plutonium isotopes and radioactive fission products is a function of the type of reactor, its power level history, the initial fuel composition, and the length of time the fuel remains in the reactor. For a given reactor type, and initial fuel composition, the subsequent fuel composition is conveniently expressed as a function of the "fuel burnup," a measure of the amount of fuel that is fissioned before it is removed. Fuel burnup can be measured in terms of the percent of the U-235 that is fissioned, but since the average amount of heat energy produced per ton of fuel can be calculated, a more convenient unit of fuel burnup used at Hanford was the number of megawatt-days of thermal energy produced per ton of heavy metal (MWd/ton). (Heavy metal includes the initial mass of the uranium and/or plutonium in the fuel, but not the mass of the cladding material.)

In addition to fission products, other radioactive isotopes are produced when neutrons are absorbed by uranium and plutonium isotopes that do not fission. These products are called transuranium isotopes (or elements) because the elements are heavier than uranium. Finally, a small fraction of the neutrons are absorbed in the reactor structural materials and impurities in the coolant water producing radioactive isotopes that are referred to collectively as neutron activation products.

Most of the fission products and transuranics are retained in the uranium fuel, which is

sent to the chemical separation plants where they are separated from the recovered plutonium and uranium. A fraction of the radioactivity, from leaking fuel slugs and corrosion of reactor structural materials leaks into the cooling water and passes into the Columbia River. Similarly, a fraction of the volatile fission products leak into the reactor building atmosphere and are released to the environment.

The uranium fuel must be left in the reactor long enough to build up the desired Pu-239, but it must be removed before the percentage of Pu-240 concentration gets too high. Because the neutron flux (and power level) is not uniform throughout the reactor core, the fuel in the channels nearest the center of the reactor reach the desired average burnup before the fuel in the outer channels. Therefore, the plutonium production reactor operators would discharge (or "push") the fuel from the center channels first. These channels were then reloaded. Batches of fuel from other channels are pushed in turn as the average Pu-240 concentration, defined by the fuel burnup of each batch, approaches the desired value. Except for the dual purpose N Reactor, during normal operations batches of fuel were typically discharged at intervals between one and three weeks. In order to stay below the desired average burnup, some mixing and matching of fuel from different channels occurred.

"The procedure for charging and discharging fuel tubes was simple. The pile [i.e., reactor] was shut down by inserting the control rods and the water supply was reduced but still had a flow capable of carrying off the radioactive heat of the fuel elements [i.e., fuel slugs]. The end caps were removed from the front and rear face ends of the tubes to be charged, and a charging machine was coupled to the front face of the tube. The charge of the fuel elements was placed in the charging machine one at a time, starting with the rear face dummy charge, followed with the fuel charge and finishing with the front-face dummy charge. As the new fuel elements were pushed in, the old ones fell out of the rear end of the tube into [the water of a discharge chute where they slid into] a basin of water. When the end caps were replaced, the pile was restarted. The irradiated fuel elements removed from the pile were kept under 20 ft of water to shield personnel from radiation exposure. Using long tongs to handle the fuel elements from above, workers picked up the fuel elements, sorted them, and placed them in buckets [that held approximately 105 fuel slugs, weighing about 824 lb] at the bottom of the basin." [Ballinger and Hall, 1991, pp. 3.5-3.6]

At the time of discharge the fuel was very hot, both thermally and in terms of its radioactivity content. The slugs were stored in the buckets under water, for about 30 to 60 days in the early years and for more than 100 days in later years, to permit the radionuclides--particularly those with short radioactive half-lives--to decay, thereby reducing the radioactive emissions when the fuel was dissolved. "Initially, spent fuel from the reactors was stored [in buckets under water] in the reactor buildings and in three identical buildings in the 200-N area, just north of the 200 East Area [one of the two chemical separation plant areas]. These buildings, designated 212-N, 212-P, and 212-R, contained storage basins and transfer facilities for moving buckets of irradiated fuel elements in and out of well cars." [Ballinger and Hall, 1991, p. 3.13] The 200-N storage area was in use prior to 2 July 1945. ["200 Area Daily Logs," HAN-45761, 1946]

The buckets of fuel elements were transported from the 100 Area and ultimately to one of the chemical separation plants in lead casks weighing about 17 tons. The casks were covered by water in railroad well cars that could carry two casks at a time. [Ballinger and Hall, 1991, p. 3.13]

C. Chemical Separation Plants.

In addition to the reactors, a series of large chemical separation plants were constructed and operated in order to extract the plutonium from the irradiated nuclear reactor fuel. The operating periods of the chemical separation plants were [Ballinger, PNL-6964, March 1991, pp. 4.19 and 4.21]:

<u>Chemical Plant</u>	<u>Start Up Date</u>		<u>Shut Down Date</u>	
T	December	1944	August	1956
B	April	1945	October	1952
REDOX (S Plant)	January	1952	December	1966
PUREX	January	1956	June	1972
	November	1983	?	1985

All Hanford chemical separation plants were based on dissolving the irradiated uranium metal in nitric acid and conducting multiple purification steps. [Ballinger and Hall, 1991, p. 4.1] The **T and B Plants** employed the bismuth phosphate process to separate plutonium from production reactor irradiated uranium fuel slugs. The operations included: (1) dissolution of the fuel slugs with nitric acid; (2) coprecipitation of the plutonium with bismuth phosphate (BiPO_4); (3) decontamination of the plutonium (i.e., removal of radioactive fission products) by a series of oxidation and reduction steps involving successive dissolution and participation operations. [Hogerton, 1963, p. 71.]

The bismuth phosphate process recovered plutonium but it had serious disadvantages, namely, its inability to separate and recover uranium, the large amount of process chemicals used, the large volume of wastes and its batch operation. [Benedict, Pigford and Levi, 1981, p. 459.] As a consequence B Plant was shut down in 1952 after the REDOX Plant (S Plant) came on line, and T Plant was shut down in 1956 after the PUREX Plant came on line. Since 1957 T Plant has been used primarily to decontaminate highly radioactive equipment. [Blush and Heitman, March 1955, p. A-101] B Plant was converted in 1968 to a waste fractionation plant designed to separate strontium and cesium from the high-level radioactive wastes. [ERDA, FEIS, Waste Management Operations, Hanford, 1975, Vol. 1, p. II.1-10.] The U Plant was a third chemical separation plant designed to use the bismuth phosphate process, but it was never used for that purpose. Following startup of the REDOX Plant, the U Plant was converted to recover uranium from stored radioactive waste from 1952 to 1958. [ERDA, FEIS, Waste Management Operations, Hanford, 1975, Vol. 1, p. II.1-12.]

The **REDOX** [REDuction-Oxidation] Plant (also called the S Plant) used a countercurrent solvent extraction process that was developed at Argonne National Laboratory and used only at the REDOX Plant at Hanford between 1952 and 1967. (The REDOX and bismuth phosphate processes are important historically, but are no longer employed.) The uranium fuel slugs were first dissolved in nitric acid, producing an aqueous solution of uranyl nitrate, plutonyl nitrate, and fission product nitrates. This was followed by the introduction of an organic solvent, methyl isobutyl ketone called "hexone," in which the uranyl and plutonyl nitrates concentrated. The fission product nitrates were left in the aqueous phase. In three subsequent steps, the fission products were first removed from the aqueous phase, the plutonium was then chemically reduced and removed in the aqueous phase as plutonium nitrate, and finally uranyl nitrate was transferred back to the aqueous phase. The plutonium, uranium and fission products were discharged in three separate streams. The aqueous fission product stream was neutralized, concentrated by evaporation, and piped to the high-level radioactive waste tanks. Unlike the bismuth phosphate process, the REDOX process could operate continuously rather than in batches. Between January 1952 and June 1967, when the process was discontinued, some 12,000 tons of irradiated fuel were reportedly processed at the REDOX Plant. [Heeb, May 1994, Table A.3].

In the **PUREX** [Plutonium-Uranium-Extraction] process the irradiated reactor fuel is first chopped up and dissolved in nitric acid. The dissolver solution is then fed into an extraction tank fed with tributyl phosphate (TBP), an organic solvent, in kerosene. Most of the fission products are separated here with the plutonium and uranium leaving the tank as organic compounds. The uranium and plutonium are then separated from one another in a second extraction stage. While the Hanford PUREX Plant is now shut down, the PUREX process is still employed at other chemical separation plants elsewhere in the world, primarily to separate plutonium and unused uranium from radioactive waste in spent power reactor fuel. In this case the separated uranium and plutonium are then chemically converted to uranium-dioxide (UO_2) and plutonium-dioxide (PuO_2).

D. Which Radionuclides Are Important?

As described above, for more than 40 years commercial contractors, working for the U.S. Government, made plutonium for nuclear weapons at the Hanford site. As a result of these operations, huge quantities of radioactivity were released to the environment to both the air and to the Columbia River, [HEDR Technical Steering Panel, April 21, 1994, p. 1.] and Hanford soils and groundwater are severely contaminated and thousands of metric tons of radioactive materials are currently stored on site. [Blush and Heitman, March 1955, p. 1-9] Most of the radioactivity released to the atmosphere came from the chemical separation plants following the dissolution of the irradiated fuel slugs. Most of the radioactivity released to the Columbia came from neutron activation products and fission products from leaking fuel slugs that contaminated the cooling water of the eight production reactors that used a once-through cooling system.

As noted above there are many radioactive isotopes released to the environment from these operations. Not all of them are of equal importance. Since we are ultimately interested in their impact on human health, we can narrow the list of radionuclides to those that have a significant impact on humans. There are a variety of considerations that are important in determining which nuclides are significant. Some of the more important considerations include:

- a) the rate of production,
- b) the rate of radioactive decay,
- c) the fraction that is released which depends on the environmental control technologies used,
- d) pathway effects, such whether the material concentrates in the food chain, and
- e) how the material moves and concentrates in the human body.

Following years of study scientists have learned that for particular operations there are a few isotopes that are worthy of careful analysis. For example, for atmospheric releases from reactors and chemical separation plants iodine-131 is likely to be important because it is produced in quantity, it is volatile and therefore readily released, and because it concentrates in the grass-cow-milk food chain of children. Isotopes HEDR has singled out as potential major contributors to radiation dose as a consequence of atmospheric releases are: iodine-131, ruthenium-103, ruthenium-106, curium-144, strontium-90, plutonium-239, and neptunium-239. In analyzing the Columbia River pathway HEDR considered the potential doses from 71 radionuclides and evaluated 19 radionuclides as major contributors to radiation dose. [Farris, et al., PNWD-2227, p. 2.1] According to HEDR, five of these radionuclides contributed over 94 percent of the dose via this pathway, namely, phosphorus-32, zinc-65, arsenic-76, neptunium-239, and sodium-24. [Ibid., pp. 2.1 and 4.2.]

In estimating the radiation exposure from Hanford operations, one of the important issues that will be addresses in a subsequent phase of this proceeding is whether all of the significant isotopes have been identified.

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