Radionuclide Sources and Radioactive Decay Figures Pertinent to the Hanford Environmental Dose Reconstruction Project

C. M. Heeb

March 1991

Prepared for the Technical Steering Panel

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RADIONUCLIDE SOURCES AND RADIOACTIVE
DECAY FIGURES PERTINENT TO THE
HANFORD ENVIRONMENTAL DOSE
RECONSTRUCTION PROJECT

C. M. Heeb

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Prepared for
the Technical Steering Panel

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**RADIONUCLIDE SOURCES AND RADIOACTIVE DECAY
FIGURES PERTINENT TO THE HANFORD
ENVIRONMENTAL DOSE RECONSTRUCTION PROJECT**

March 1991

This document has been reviewed
and approved by the Technical Steering Panel.

John E. Till, Chairman
Technical Steering Panel
Hanford Environmental
Dose Reconstruction Project

March 9, 1991
Date

SUMMARY

The origin and radioactive decay schemes of radionuclides currently expected to be the major contributors to potential radiation doses that populations might have received as a result of nuclear operations at the Hanford Site since 1944 are identified in this report and illustrated in Figures 2.1 through 2.10. The reactions considered include actinide neutron capture and decay sequences, fission product decays, and neutron activation reactions.

It is important to note that the radioactive half-life of a given nuclide does not, by itself, fully determine the significance of a given radionuclide as a potential source term. This brief report does not address environmental transport mechanisms, behavior in the environment, or radiological dose impact of any of the radionuclides shown. These aspects will be addressed in other, separate documents supporting the goals of the Hanford Environmental Dose Reconstruction Project.

GLOSSARY

α	Alpha particle.
Alpha Decay	Radioactive decay of an atomic nucleus by emission of an alpha particle. Such a decay decreases the daughter nucleus' atomic number by 2 and decreases the atomic weight by 4.
Alpha Particle	A nucleus composed of two neutrons and two protons.
ANSI/ASME NQA-1	National Standard for Nuclear Grade Quality Assurance.
β^+	A positron, or positively-charged electron.
β^-	Beta particle.
Beta Decay	Radioactive decay of an atomic nucleus by emission of a beta particle. Such a decay increases the atomic number by 1 with essentially no change in the atomic weight.
Beta Particle	An electron ejected from an atom nucleus during the beta decay process.
d	Day.
EC	Electron capture.
Electron capture	A mode of radioactive decay in which an electron from outside the nucleus is captured. This process decreases the atomic number by 1 with essentially no change in the atomic weight.
Fission product	A fragment nucleus left after the parent nucleus undergoes nuclear fission.
γ	Gamma ray.
Gamma ray	High energy electromagnetic radiation with wavelengths shorter than X-rays.
h	Hour.
IT	Isomeric transition.

Isomeric transition	A mode of radioactive decay which involves the emission of a gamma ray by the nucleus with no change in atomic number or in atomic weight.
m	Minute.
ms	Millisecond, 1/1000 of a second.
Neutron activation	The process by which a previously non-radioactive nucleus becomes radioactive by absorbing a neutron.
s	Second.
y	Year.

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1.0 INTRODUCTION

Radionuclides were produced in the Hanford Site plutonium production reactors by neutrons acting upon 1) the uranium metal fuel and its cladding, 2) impurities in the coolant water itself, and 3) in coolant-water-deposited crud films in the reactor process tubes and effluent piping. Impurities in the graphite moderator material and in the cover gas present in the reactor graphite stack also resulted in radionuclide generation by neutron activation. The radionuclides contained in irradiated fuel were mainly bred-in actinides plus decay daughters and fission products. Except for in-reactor fuel failures, these radionuclides were entirely contained within the irradiated fuel element. Radionuclides produced by neutron activation in reactor process flow systems were released to the Columbia River, while radionuclides produced by neutron activation of impurities in the reactor's cover gas were released to the atmosphere.

A more detailed description of the Hanford Site plutonium production reactors and their operations and effluents is given by the report entitled A History of Major Hanford Operations Involving Radioactive Material (Ballinger and Hall 1989).

The sources of the radionuclides currently expected to be the major contributors to the radiation doses that populations could have received from nuclear operations at the Hanford Site since 1944 are identified in this report. In addition, the decay schemes for the most significant actinides found in irradiated fuel from the Hanford plutonium production reactors are identified even though they may not be significant contributors to any potential radiation dose to populations.

The work described in this report was conducted in accordance with the requirements of ANSI/ASME NQA-1 1986 Edition, Quality Assurance Program Requirements for Nuclear Facilities, as interpreted by the Pacific Northwest Laboratory (PNL) Quality Assurance (QA) program.

All staff performing work described here are adequately trained and qualified. All procedures used to support this report were appropriately written and controlled in accordance with PNL QA program requirements.

Records that support the data in this report were created and stored in accordance with applicable Hanford Environmental Dose Reconstruction (HEDR) Project record control requirements.

Drafts of this document underwent internal independent technical review. Review comments were satisfactorily resolved, and there were no controversial resolutions to the comments.

2.0 RADIONUCLIDES: SOURCES AND RADIOACTIVE DECAY

Source term radionuclides resulting from neutron capture and beta decay in the uranium metal fuel used for the Hanford Site plutonium production reactors are shown in Figure 2.1. The isotopes of uranium initially present in the unirradiated Hanford fuel are uranium-234, -235, -236, and -238. Naturally occurring uranium does not contain uranium-236; however, the uranium used in the Hanford Site reactors did contain uranium-236 because previously irradiated uranium, which had been recovered by chemical separation and recycled through the uranium gaseous diffusion cascades, was used at various times. Many of the actinides shown undergo radioactive decay by alpha particle emission; these alpha decay schemes are illustrated in subsequent figures.

The alpha and beta decay products of uranium-233 are given in Figure 2.2. This uranium isotope and its alpha and beta decay products are provided to complete the plutonium and neptunium radioactive decay schemes, which are subsequently illustrated in Figures 2.3 and 2.4, respectively.

The alpha and beta radioactive decay schemes for uranium-234, -235, -236, and -238 are illustrated in Figures 2.5 through 2.8, respectively. The very long radioactive half-lives of uranium-234 and its first radioactive decay product, thorium-230, drastically reduce any radioactive contributions from further decay products. Similarly, the very long radioactive half-life of uranium-235 reduces the radioactive contributions from its radioactive decay products. The alpha decay of uranium-236 and its first alpha decay product, thorium-232, is very slow. The longest-lived naturally occurring uranium isotope is uranium-238; the end product radioactive decay product of uranium-238 is uranium-234, another naturally occurring uranium isotope (see Figure 2.5). It is important to note that the radioactive half-lives of the uranium isotopes are long when compared with the radioactive half-lives of radionuclides resulting from neutron activation and fission.

The principal plutonium isotopes (and their radioactive decay daughters) that are of interest to the Hanford Environmental Dose Reconstruction Project are shown in Figure 2.3. The radioactive decay product for each plutonium

S8908008.1

FIGURE 2.1. Source Term Radionuclides from Neutron Capture and Beta Decay in Uranium Metal Fuel

S8908008.2

FIGURE 2.2. Source Term Radionuclides from Radioactive Decay of Uranium-233

isotope shown terminates with a uranium isotope whose radioactive decay scheme has been previously shown. The radioactive decay schemes of the plutonium-242 isotope is not shown in Figure 2.3 because it is present in insignificant amounts in the low irradiated fuel burnups of the Hanford Site plutonium production reactors. The production of plutonium-242 requires four neutron absorption steps and its radioactive decay results directly in the formation of the most stable uranium isotope, uranium-238.

Neptunium is formed in irradiated fuel as shown in Figure 2.1. The principal neptunium isotopes and their radioactive decay schemes are given in

S8908008.7

FIGURE 2.3. Source Term Radionuclides from Plutonium Radioactive Decay

Figure 2.4. Each radioactive decay scheme is terminated at a radioactive decay product of a uranium or plutonium isotope whose radioactive decay has previously been shown.

The neutron activation products of most significance as source terms for determining a potential radiation dose to populations are listed in Figure 2.9. As stated previously, neutron activation products are the result of impurities present in the water used to cool the reactors or impurities in the gas used to blanket the graphite of the reactor pile. The only neutron

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FIGURE 2.4. Source Term Radionuclides from Neptunium Radioactive Decay

activation products believed to be significant as possible source terms for a potential radiation dose to populations are those formed as a result of impurities in the coolant water. In Figure 2.9, the original isotopic impurities are shown first, followed by their neutron activation products, and then their radioactive decay products. The end product of each radioactive decay scheme is a stable (nonradioactive) nuclide.

Those fission products of most significance as source terms for determining a potential radiation dose to populations are indicated in Figure 2.10. Each fission product shown decays radioactively by beta emission except for the excited state nuclides that decay by isomeric transition and are so marked on the decay scheme. Again, the end product of each radioactive decay scheme is a nonradioactive nuclide.

S8908008.3

FIGURE 2.5. Source Term Radionuclides from Radioactive Decay of Uranium-234

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FIGURE 2.6. Source Term Radionuclides from Radioactive Decay of Uranium-235

FIGURE 2.7. Source Term Radionuclides from Radioactive Decay of Uranium-236

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FIGURE 2.8. Source Term Radionuclides from Radioactive Decay of Uranium-238

S8908008.9

FIGURE 2.9. Formation of Neutron Activation Products and Their Subsequent Radioactive Decay

S8908008.10

FIGURE 2.10. Fission Product Beta Decays

3.0 REFERENCE

Ballinger, M. Y., and R. B. Hall. 1989. A History of Major Hanford Operations Involving Radioactive Material. PNL-6964 HEDR, Pacific Northwest Laboratory, Richland, Washington.

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