

**DERIVATION OF RESIDUAL RADIONUCLIDE INVENTORY GUIDELINES FOR  
IMPLACE CLOSURE OF HIGH-LEVEL WASTE TANKS**

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**Abstract**

Residual radionuclide inventory guidelines were derived for the high-level waste tanks at a vitrification facility. The decommissioning scenario assumed for this derivation was that the tanks were to be stabilized at the present locations and the site is released for unrestricted use following a 100-year institutional control period. It was assumed that loss of institutional control would occur at 100-years following tank closure. The derivation of the residual radionuclide inventory guidelines was based on the requirement that the effective dose equivalent (EDE) to a hypothetical individual who lives in the vicinity of the site should not exceed a dose of 0.15 mSv/yr off-site and 5 mSv/yr on-site following closure of the tanks. The RESRAD computer code, modified for exposure scenarios specific for the site, was used for this evaluation. The results of the derivation indicate that the allowable off-site dose limit will not be exceeded. The estimated potential doses to individuals using water off-site from a creek are negligibly small fractions of the 0.15 mSv/yr allowable dose limit. With an assumed 3% heel remaining in the tanks, the estimated peak dose rate for the future offsite water user is about 0.00025 mSv/yr. The residual radionuclide inventory guidelines derived based on potential doses to the on-site resident farmer indicate that, with the exception of Tc-99 and C-14, a 3% heel remaining in the tanks would not result in doses exceeding the 5 mSv/yr allowable dose limit. For this on-site exposure scenario, the peak dose rates occur at about 2000 years after tank closure. The peak dose rate is calculated to be 25 mSv/yr, with greater than 99% produced by four radionuclides: C-14, Tc-99, Np-237, and Am-241. Ingestion of contaminated vegetation contributes most (90%) of the peak dose. Since the inventories used for the derivation are mostly estimated from fuel depletion calculations. There is a need to determine further the actual inventories of these radionuclides in tank heels after vitrification. If these relatively mobile radionuclides still remain in the tanks, specific methods to remove them should be fully investigated.

**Introduction**

The Federal and local government agencies are in the process of evaluating alternatives for the decontamination and decommissioning of high-level waste (HLW) tanks at a vitrification facility. One of the decommissioning alternatives being considered is to fill the tanks with concrete for in place closure at their present locations. A major consideration in developing a decommissioning plan based on this alternative is the amount of residual radionuclides that may be allowed to remain in the tanks. This paper presents the results of a pathway analysis to determine the residual radiological inventory that will be acceptable.

The guidelines to be derived are the radionuclide inventories (in curies) in the decontaminated tanks that must not be exceeded in order to meet a prescribed dose limit of 0.15 mSv/year off-site and 5 mSv/yr on-site to the reasonably maximally exposed individual (RMI). The closure scenario assumed was that: For the first 100-years after tank closure, the calculated rate of water infiltration (0.01 m/yr) is assumed to be limited by the clay cover

above the tank. After this 100-year period, an increasing time dependent infiltration rate representative of a gradually degraded clay cover is assumed for the remainder of the analysis.

The primary function of the closure scenario is to prevent long-term migration of contaminants into the environment. The primary path of migration is via rainwater that infiltrates from the surface into the tanks and then carries radionuclides from the waste to the environment. Therefore, reducing water infiltration into the waste tanks and the infiltration rate is one of the most important objectives of the closure system.

The RESRAD computer code (Gilbert et al. 1989; Yu et al. 1993), with enhancements for exposure scenarios specific for the site, was used for the pathway analyses and dose evaluations. The enhancements are necessary to model the exposure pathways to individuals residing off-site, and the surface contamination pathways associated with eventual tank overflow (bathtubbing).

The radionuclide inventory in the tanks at closure (the "heel") was assumed to be 3% of the total contents before the vitrification.

### Pathway Analysis Approach

The estimation of radiation doses is dependent on the physical and radiological characteristics of the HLW tanks and their contents as well as the scenarios of human exposure judged to be credible and to result in reasonable upper bound estimates. The magnitude of the potential doses to the RMIs is dependent on two factors: the projected uses of the site and the location of the RMIs.

It was assumed that use of the site will continue to be restricted or controlled for 100 years following site closure. During the 100 years of control, access to the site is limited by fences, markers, and intrusion barriers. Security surveillance continues and maintenance of fences and intrusion barriers is provided if required. During this period, the institutional controls are assumed to be effective. The only potential radiation exposure is from the off-site use of contaminated water.

Table I Summary of Pathways Included in the Exposure Scenarios

Pathway	(A) Off-Site Water User Scenario	(B) On-Site Resident Farmer Scenario
1. External Exposure	No	Yes
2. Inhalation (Resuspension)	No	Yes
3. Ingestion of Drinking Water	Yes	Yes
4. Ingestion of Vegetables	Yes	Yes
5. Ingestion of Meat	Yes	Yes
6. Ingestion of Milk	Yes	Yes
7. Ingestion of Aquatic Food	Yes	Yes

Unrestricted use of the site was assumed possible following the 100-year institutional control period. During unrestricted use of a radioactively contaminated site, the maximum exposed individual usually proves to be an individual exposed as a result of a residential/agricultural scenario. The two exposure scenarios assumed for assessing post-closure doses may be identified as: (A) the surface water user scenario; and (B) the on-site resident farmer scenario. The exposure pathways considered for each of these scenarios are summarized in Table I.

An enhanced version of the RESRAD code was used to calculate doses to both the on-site and off-site hypothetical resident farmers from the leaching and transport of radionuclides from the high-level waste tanks. RESRAD provides a comprehensive and reasonably conservative means to assess the long-term (greater than 1000 years) radiological impacts from a radioactively contaminated area. It includes major features necessary to model water infiltration through radioactive waste materials and the transport of radionuclides through the unsaturated (vadose) zone and saturated zone (aquifer) to a well or point of seepage into surface water. The code considers radionuclide decay and ingrowth and source leaching from the time of disposal to the time of human exposure. This feature of the code is especially important for the assessment of long half-lived americium-241, neptunium-237, and plutonium-239 and their decay series nuclides because all three series include a few progeny which are radiologically important.

### Derivation of Residual Radionuclide Inventory Guidelines

The residual radionuclide inventory guideline is defined as the radionuclide inventory (curies) of residual radioactive material that can remain in the tank farm without causing the allowable dose limit (ADL) of 0.15 mSv/yr off-site or 5 mSv/yr on-site to be exceeded

Table II - Derived Residual Radionuclide Inventories

	<b>Scenario A: Off-site Water User</b>	<b>Scenario B: On-site Resident Farmer</b>
<b>Radionuclide</b>	<b>Limiting Ci<sup>1)</sup></b>	<b>Limiting Ci<sup>2)</sup></b>
Am-241	1 E7	3 E3
C-14	3 E3	4
I-129	2 E4	10
Cs-137	4 E11 (SAL)	4 E11 (SAL)
Np-237	2 E3	8 E-1
Pu-238	2 E8	2 E3
Pu-239	1 E7	3 E2
Pu-241	4 E8	8 E4
Sr-90	7 E11 (SAL)	1 E8
Tc-99	1 E7	10
Total-Nuclide in Tank	2 E8	8 E4

1) Based on a maximum allowable off-site EDE of 0.15 mSv/year.

2) Based on a maximum allowable on-site EDE of 5 mSv/year.

SAL: Specific Activity Limit Maximum.

following in place closure of the tanks. The residual radionuclide inventory guideline,  $G_i$ , for individual radionuclide  $i$  at the tank farm is calculated as

$$G_i = ADL/DSR_i$$

where  $DSR_i$  is the calculated unit dose factor or the dose to source inventory ratio for individual radionuclide  $i$ . The calculated residual radionuclide inventory guidelines for individual radionuclides and the total-nuclides are presented in Table II. for both Scenario A (the off-site water user) and Scenario B (the on-site resident farmer). These inventory limits are linearly proportional to the dose limit used in the calculation. For an on-site ADL of 0.15 mSv/yr rather than 5 mSv/yr, the curie limits in Table II should be multiplied by 0.03.

When implementing the derived radionuclide inventory guidelines for decontamination of the HLW tanks, the sum-of-fraction rule would apply. That is, the summation of the fractions of radionuclide inventories,  $S_i$ , remaining in the tanks, divided by their guidelines,  $G_i$ , should not be greater than unity, or

$$\sum_i \frac{S_i}{G_i} \leq 1.$$

## Discussion of Results

The estimated potential doses to individuals using water off-site are negligibly small fractions of the 0.15 mSv/yr allowable dose limit. With the assumed 3% radionuclide heel remaining in the tanks, the estimated peak dose rate for the future water user (Scenario A) is about 0.00025 mSv/yr. This peak dose rate is primarily from the fast moving Np-237 and C-14 at about 400-500 years after tank closure.

The residual radionuclide inventory guidelines derived based on potential doses to the on-site resident farmer (Scenario B) indicate that, with the exception of Tc-99 and C-14, a 3% heel remaining in the tanks would not result in doses exceeding the 5 mSv/yr allowable dose limits (Table II.).

For this on-site exposure scenario, the peak dose rates occur at about 2,200 - 2,300 years after tank closure. Tc-99 produces the highest dose rate, about 66% of the total peak dose. The peak dose rate is calculated to be 25 mSv/yr, with greater than 99% produced by four radionuclides: C-14 (9%), Tc-99 (66%), Np-237 (15%), and Am-241 (10%). Ingestion of contaminated vegetation contributes most (90%) of the peak dose.

Since the peak doses are contributed from four radionuclides, Tc-99, Np-237, Am-241, and C-14, there is a need to determine more accurately the actual inventories of these radionuclides in HLW tanks. At present, the inventories available for use are mostly conservatively estimated values from fuel depletion calculations. If these relatively mobile radionuclides are found still remaining in the tanks following vitrification, specific methods to remove them should be fully investigated.

## References

- [1] Gilbert, T.L., Yu, C., Yuan, Y.C., Zielen, A.J., Jusko, M.J., and A. Wallo. June 1989. A Manual for Implementing Residual Radioactive Material Guidelines. Argonne National Laboratory. DOE/CH/8901; ANL/ES-160.
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