BNL-74698-2005-IR

Assessment of Radionuclide Release from Intact Structures Backfilled with Contaminated Concrete at the Yankee Nuclear Power Station

Terry Sullivan

March 2004

Environmental Sciences Department
Environmental Research & Technology Division

Brookhaven National Laboratory
P.O. Box 5000
Upton, NY 11973-5000
www.bnl.gov

Managed by
Brookhaven Science Associates, LLC
for the United States Department of Energy under
Contract No. DE-AC02-98CH10886

DISCLAIMER

Calculation Title Page

Assessment of Radionuclide Release from Intact Structures Backfilled with Contaminated Concrete at the Yankee Nuclear Power Station

Title

YA-CALC-00-003-04

Calculation Number

Approvals (Print & Sign Name)

Preparer: Terry Sullivan Date:

Reviewer: Eric Darois Date:

Reviewer: Alice Carson Date:

Approval(Cognizant Manager): Date:

Table of Contents

List of Tables

Assessment of Radionuclide Release from Intact Structures Backfilled with Contaminated Concrete at the Yankee Rowe Nuclear Power Station

Purpose

This calculation determines the release of residual radioactivity (including H-3, C-14, Co-60, Ni-63, Sr-90, and Cs-137), from subsurface structures filled with concrete debris at the Yankee Nuclear Power Station. Analyses were performed to assess the rate of release from the source of contamination and the resulting dose in the groundwater pathway.

Summary of Results

Two mechanisms were considered, diffusive release from the concrete structures (walls and floors) that remain intact and sorption onto concrete backfill placed within these structures.

RESRAD was used to calculate the predicted maximum dose assuming a unit loading of 1 pCi/g on the intact structures. To the extent possible, the same assumptions in the soil DCGL calculations performed for Yankee Atomic were used in the calculation. However, modifications to some input parameter values were needed to represent the geometry of the subsurface facilities, flow through these facilities, and releases from the backfill and intact structures. Input parameters specific to these calculations included the leach rate, disposal geometry, pumping rate, porosity and bulk density. The dose results for a unit loading of 1 pCi/g on intact structures showed that Sr-90 had the highest dose (3.67E-02 mrem/yr).

References

[Carson, 2004]
A. Carson, “Basis for Selection of Concrete Kd Values,” YA-REPT-01-003-03, August 2004.

[Darman, 2004],
YAEC Internal Communication, J. Darman to AP-0831 File, “IX-Pit Sample Plan Close-Out,” January 7, 2004 (also filed as RP 04-008., Darman to Heath, dated March 4, 2004).

[Sullivan, 2004]
Sullivan, T.M., “Assessment of Radionuclide Release from Contaminated Concrete at the Yankee Nuclear Power Station,” YA-CALC-00-001-04, March 2004.

[YA 2004]
YA-CALC-01-002-03, “Derived Concentration Guideline Levels for Soil at the Yankee Rowe Site,” January 2004.

[Yu, 2001]
Yu, C, A.J. Zielen, J.-J. Cheng, D.J. LePoire, E. Gnanapragasam, S. Kamboj, J. Arnish, A. Wallo III,* W.A. Williams, and H. Peterson, Users Manual for RESRAD Version 6, ANL/EAD-4, Argonne National Laboratory, 2001.

Method of Calculation

Yankee Nuclear Power Station (YNPS) is undergoing decontamination and decommissioning (D&D) and, eventually, license termination. As a part of the process, the YAEC is required to demonstrate that any radioactivity that remains at the time of license termination will not cause exposure to exceed the limits stated in Subpart E to 10CFR20.

Yankee Rowe has identified four structures that may remain, in part, and be backfilled with concrete debris resulting from D&D activities or other fill, at the time of license termination:

a. Primary Auxiliary Building (PAB) drain collection tank (PDCT) cubicle
b. PAB gravity drain tank (GDT) cubicle
c. Spent Fuel Pit (SFP)
d. Waste Disposal Building (WDB) cubicle

Because these partial structures and the backfill may contain residual radioactivity, the possible dose attributable to these must be evaluated. A separate calculation addresses the dose from the concrete debris backfill, and, thus, the focus of this study is the dose from the subsurface structures. The resulting dose depends upon the distribution of radionuclides in the remaining material and the mechanisms by which radionuclides in the material migrate into groundwater. This calculation addresses the potential release of radionuclide contamination from subsurface concrete structures and the associated dose.

Body of Calculation

1.0 Introduction

The YNPS is undergoing the process of decontamination and decommissioning (D&D) and license termination. In doing so, a number of concrete structures will be demolished, with the debris potentially used as backfill onsite. Other structures will be partially demolished, with their subsurface foundations left intact. The demolition debris may be used as backfill in these partially intact subsurface structures. Part of the license termination process requires analyses that demonstrate that any radioactivity that remains will not cause exposure to radioactive contaminants to exceed acceptable limits. This requires knowledge of the distribution of radionuclides in the remaining material and their potential release mechanisms from the material to the contacting groundwater.

This study concerns the potential release of radionuclide contamination from the partially intact subsurface structures. A separate calculation assesses the dose from the concrete debris.

1.1 Objective

To determine the potential dose that may result from subsurface contaminated concrete as a function of initial contamination levels in the remaining partial structures for selected radionuclides.

1.2 Approach

YNPS has four subsurface partial structures that may remain and may be backfilled with concrete debris. These partial structures are:

1. Primary Auxiliary Building (PAB) drain collection tank room
2. PAB gravity drain tank room
3. Spent Fuel Pit (SFP)
4. Waste Vault

Release of the contaminants from the facilities will occur through diffusion out of the concrete into the groundwater. Diffusion-controlled release from each of the four facilities was modeled independently using DUST-MS (Sullivan, 2004). The total release from the walls of the subsurface structures will be combined to form a single source to be used by RESRAD. RESRAD will calculate exposure from the various pathways using the resident farmer scenario. The details of this approach are provided in the following sections.

2.0 Subsurface Structures Geometry

To calculate the magnitude of the source, the maximum potential volume of contaminated concrete is needed. Dimensions of the walls for facilities that may be left in situ are in Table 1. To estimate the mass of concrete, the density of concrete is needed.

This value is not known, but a density of 2.5 g/cm^3 is assumed. The actual value is expected to be less than the assumed value based on other concretes. Therefore, mass and total contamination will be overestimated.

Table 1 Dimensions of Subsurface Structures After Removal of Above Grade Sections.

		Below-Grade Dimensions (ft)					
Structure	Component	Component Height	Component Width	Component Thickness	Area (ft^2)	Volume (ft^3)	Mass (g)
PAB	Floor			2.50	193.8	484.4	3.4E+07
(drain collection tank room)	wall 1	18.50	12.50	1.00	231.25	231.25	1.6E+07
	wall 2	18.50	12.50	1.00	231.25	231.25	1.6E+07
	wall 3	18.50	15.50	1.00	286.75	286.75	2.0E+07
	wall 4	18.50	15.50	1.75	286.75	501.81	3.6E+07
PAB	Floor			2.50	157.6	394.1	2.8E+07
(gravity drain tank room)	wall 1	18.50	10.17	1.00	188.15	188.15	1.3E+07
	wall 2	18.50	10.17	1.00	188.15	188.15	1.3E+07
	wall 3	18.50	15.50	1.00	286.75	286.75	2.0E+07
	wall 4	18.50	15.50	1.00	286.75	286.75	2.0E+07
SFP	Floor			3.00	555.5	1666.7	1.2E+08
	wall 1	14.67	16.50	6.00	242.06	1452.33	1.0E+08
	wall 2	14.67	16.50	6.00	242.06	1452.33	1.0E+08
	wall 3	14.67	33.67	6.00	493.94	2963.63	2.1E+08
	wall 4	14.67	33.67	6.00	493.94	2963.63	2.1E+08
Waste Vault	Floor			1.17	126	147.4	1.0E+07
	wall 1	9.83	9.00	1.00	88.47	88.47	6.3E+06
	wall 2	9.83	9.00	1.00	88.47	88.47	6.3E+06
	wall 3	9.83	14.00	1.00	137.62	137.62	9.7E+06
	wall 4	9.83	14.00	1.00	137.62	137.62	9.7E+06

2.1 Contaminant Initial Concentrations

The total release from the concrete walls to the backfill region is assumed to be independent of the contamination levels in the backfill. It is assumed that the initial contamination level is 1 pCi/g for all radionuclides in the walls and floors of the subsurface structures. Thus the results can be scaled up or down to determine the dose for the actual levels of residual radioactivity in the structures.

3.0 Modeling Release from Contaminated Concrete

The radionuclides in the concrete will eventually be released to the surrounding backfill. The contaminants will move through the backfill undergoing sorption and eventually reach a receptor well. Therefore, the contaminant concentration in the well is a function of the release rate from the concrete and the transport to the well. For conservatism, all four subsurface areas will be combined, assuming that one well withdraws all the water from each area.

An important outcome of modeling diffusion controlled releases is the ability to obtain a diffusion profile that can be used to help develop characterization data needs. An analysis of these profiles is presented in this section.

3.1 Key Radionuclides

Measurable levels of tritium were found throughout the entire core of the wall between the IX Pit and the SFP. Average concentrations in the concrete for tritium were 25 pCi/g (Darman, 2004). Several radionuclides including Cs-137, Co-60, Ni-63, Sr-90, and C-14 were found within the first inch of the interface of the wall and the SFP. Contamination levels ranged from 1075 pCi/g for Cs-137 down to 0.91pCi/g for Sr-90 (Darman, 2004). All of these radionuclides will be simulated.

3.2 Diffusion- Controlled Release from Intact Structures

Calculations of diffusion controlled release from the subsurface structures has been previously reported (Sullivan, 2004). In these calculations, conservative values for the diffusion coefficients were selected based on literature values. The initial distribution of contaminants for the release calculation is a uniform concentration of 1 pCi/g. The maximum yearly release rate occurs in the first year and continually declines afterwards. This analysis uses the first year release rates for conservatism. Table 2 presents the diffusion coefficient, fractional release of the inventory and the peak release rate for the six radionuclides under consideration.

Table 2 Average Fractional Release and Total Release from All Subsurface Facilities for Initial Uniform Contamination of 1 pCi/g in All Walls and Floors.

Radionuclide	Diffusion Coefficient (cm^2/s)	Average fractional release	Peak Release Rate (pCi/yr)
H-3	5.5 E-07	6.29E-02	6.45E+07
C-14	1.0E-12	3.02E-04	3.10E+05
Co-60	4.0E-11	1.02E-03	1.05E+06
Ni-63	1.1E-09	4.88E-03	5.00E+06
Sr-90	5.2E-10	3.41E-03	3.49E+06
Cs-137	3.0E-09	7.82E-03	8.02E+06

Using the assumption that the groundwater concentration is controlled by sorption onto the backfill with the total mass defined from the peak release rate in Table 2, the maximum activity released within a year and the maximum contaminated zone concentration for each radionuclide from the subsurface structures are presented in Table 3. The maximum concentration is calculated as follows:

$$C_{cz} = M_r/(V*\beta*\eta*R_d) \text{ (in pCi/L)} \qquad (1)$$

Where M_r = mass released
V= Volume of the fill (m^3)
β =1000 conversion factor for m^3 to liters.
η = saturated zone porosity = 0.31
R_d = retardation coefficient = $1 + \rho K_d/\eta$

This approach does not account for dilution that will occur due to groundwater flow.

Table 3 Activity and contaminated zone concentrations through diffusion release from intact subsurface structures.

Radionuclide	Activity Released through Diffusion (pCi)	Maximum Contaminated Zone Concentration (pCi/L)
H-3	6.45E+07	4.36E+02
C-14	3.10E+05	5.95E-03
Co-60	1.05E+06	5.05E-03
Ni-63	5.00E+06	1.91E-01
Sr-90	3.49E+06	4.44E-01
Cs-137	8.02E+06	7.96E-02

4.0 Modelling the Source Term for RESRAD

The modeling performed above was used as the basis for assessing dose using RESRAD. RESRAD assumes that the source is controlled by the sorption properties of the contaminated zone. RESRAD also assumes that the contaminated zone is above the water table, whereas in this case, the source is below the water table. These conceptual model differences lead to the need to make assumptions about parameter values to make the calculations in RESRAD match the desired release rates, geometry, and dilution effects.

4.1 Matching release rates

RESRAD assumes that release from contaminated material is controlled by the following equation (Yu, 2001).

$R(t) = L(t)*\rho_b*A*T*S(t)$ (2)

Where R(t) is the time-dependent release rate (pCi/yr)
L(t) is the leach rate (1/yr) for the contaminant
ρ_b is the bulk density of the source zone (kg/m^3)
A is the area of the contaminated zone (m^2)
T is the thickness of the contaminated zone (m)
S(t) is the average concentration of the radionuclide available for leaching (pCi/kg).

The RESRAD Leach Rate, L (t), is calculated from the following expression (Yu, 2001),

$L(t) = I/(\theta*T*R_{dcz})$ (3)

Where I = infiltration rate (m/y)
θ = moisture content in the contaminated zone
R_{dcz} = retardation coefficient in the contaminated zone (dimensionless)

$R_{dcz} = 1 + \rho_b K_d/\theta$ (3)

Where ρ_b = bulk density of the concrete backfill = 1.54 g/cm^3
K_d = radionuclide distribution coefficient (g/cm^3)
θ = moisture content (porosity in the saturated zone) = 0.31

4.2 Selection of Contaminated Zone Geometry for RESRAD

RESRAD treats the source of contamination as a single region. Therefore, for modeling purposes, the different building structures are combined to form a single source. RESRAD requires three parameters to define the geometry of the contaminated zone: area, thickness, and length parallel to aquifer flow. The most conservative assumption is that all of the facilities are lined up with the longest dimension in the direction of flow. Using only the interior dimensions of the intact structures (Table 1), the distance parallel to flow is 24 m. The average height of the structures is 4.7 m. The volume of the backfill region of the contaminated structures is 450 m^3. Therefore, the effective width is 4 m. The area perpendicular to groundwater flow is the product of the average height and effective width and is 18.8 m^2.

Table 4 Contaminated zone geometry factors

Area (m^2)	Height (m)	Length parallel to flow (m) (thickness)	Volume (m^3)
18.8	4.7	24	450

4.3 Leach Rates

RESRAD assumes that the contaminated zone is above the water table and uses unsaturated flow parameters (infiltration rate and moisture content) to calculate leach rates. In this simulation, the source of contamination is below the water table and, therefore, the flow parameters of the aquifer and the geometry relative to flow in the aquifer should be used to calculate the leach rates. Equation (2) and (3) were used to calculate the "effective" leach rates for use in RESRAD using the K_d values in Table 2, the geometry values in Table 4, and a moisture content of 0.31 and bulk density of 1.54 g/cm^3. Table 5 presents the effective leach rate for each radionuclide.

Table 5 Effective leach rates for RESRAD.

Radionuclide	Effective Leach Rate (yr^{-1})
H-3	3.368E+01
C-14	9.56E-02
Co-60	2.40E-02
Ni-63	1.90E-01
Sr-90	6.34E-01
Cs-137	4.94E-02

The effective leach rate for H-3 is much greater than 1. This reflects the lack of sorption and high groundwater flow velocity. This high effective leach rate suggests that all of the contamination is released within much less than 1 year. The effective leach rates in Table 5 were used as the contaminated zone leach rate in RESRAD. Use of the leach rate for the contaminated zone overrides the use of a distribution coefficient in the contaminated zone. Therefore, the input for this parameter in RESRAD does not effect the calculations

4.4 Source Inventory

In RESRAD, all concentrations are directly proportional to the initial concentration in the contaminated zone. The source inventory is presented in Table 6 for the intact structures with an initial concentration of 1 pCi/g.

Table 6 Activity released from subsurface structures contaminated to 1 pCi/g and the effective concentration for RESRAD calculations.

Radionuclide	Activity Released from subsurface structures (pCi)	Effective Contaminated Zone Soil Concentration (pCi/g)
H-3	6.45E+07	8.78E-02

C-14	3.10E+05	4.22E-04
Co-60	1.05E+06	1.42E-03
Ni-63	5.00E+06	6.81E-03
Sr-90	3.49E+06	4.75E-03
Cs-137	8.02E+06	1.09E-02

5.0 RESRAD Dose Modelling

Using the effective leach rates in Table 5 and the effective initial concentrations in Table 6 RESRAD was used to calculate peak concentration and dose.

A few parameter values used in the analysis differ from those used by Yankee Rowe in calculating soil DCGLs (YA, 2003) These were changed to fit the conditions being simulated for release controlled by diffusion from intact concrete and sorption on concrete debris used as backfill in the saturated zone.

In calculating soil DCGLs, the cover thickness is zero. However, in this analysis, the depth to the water table will be at least 2 feet and, because of the assumption that the source is in the aquifer, a cover thickness of 2 feet (0.6 m) is used. In the soil DCGLs, the depth for root penetration was 1.17 m and the unsaturated zone thickness was 1.82 m. Therefore, the roots did not penetrate the saturated zone. Retaining the root depth value used in the soil DCGL analysis implies the roots penetrate the saturated zone. The root depth value of 1.17 m will be changed to 0.5 m for this analysis. This prevents the non-water pathways from being non-zero, which is appropriate for the conditions being modeled. The non-water pathways assume that the entire inventory in the contaminated zone is available for transfer to plants modified by the thickness of the cover divided by the thickness of the root zone. However, this is not the situation being modeled as most of the contamination will be bound in the cement and unavailable for release to the plants even if the roots were near the concrete.

A second difference between the assumption used in the analyses of the soil DCGLs and the concrete release model is that the water table drop rate is set to 0. In the soil DCGLs analysis (YA, 2003), a RESRAD default value of 0.001 m/y was used. That analysis had a 1.8 m unsaturated zone, and the results were not sensitive to the water table drop rate. In the current analysis, the unsaturated zone has zero thickness and a non-zero water table drop rate causes a non-zero unsaturated zone thickness to form. This leads to RESRAD calculating transport through the unsaturated zone to reach the aquifer. This effect is not desired because the source is assumed to reside in the saturated zone.

The saturated zone K_ds were set to zero. This assumption was used to provide an upper bound on well concentrations.

The contaminated zone geometry was changed to fit the conditions for release from the partial subsurface structures as defined in Table 4. A bulk density of 1.54 g/cm^3 was used as being representative of the expected average for backfilled concrete and an effective porosity of 0.31 was used.

In the following analysis, the deterministic model in RESRAD was used. The deterministic model was selected because the calculated well concentrations are conservative. This is because the entire source was considered to be in the aquifer, the release rate calculations were biased high due to the use of the largest value of diffusion coefficient found in the literature, a K_d of zero is used in the saturated zone, and the mass balance model which takes all of the radionuclides released and places them in the well was used in place of the non-dispersion model.

Table 7 summarizes the results for the peak well concentrations and dose using the release rates calculated for diffusion controlled release, Table 2, the leach rate parameters in Table 5 and the parameters used in the analysis for soil DCGLs, with the exceptions noted above. The results suggest that Sr-90 is the radionuclide of highest dose. The RESRAD prediction for H-3 appeared to be low considering it released an order of magnitude more mass than any other radionuclide. As a basis of comparison, the concentration that would occur if all of the mass released was put into the volume of water that flows through the contaminated zone in one year was calculated. This value is almost a factor of 3 higher than predicted by RESRAD. The cause for this discrepancy is that physically the H-3 is modeled to be released instantly. The contaminant flow velocity is the volumetric low rate per unit area (250.6 m/yr) divided by the porosity and retardation factor. For H-3, the retardation factor is 1 ($K_d = 0$) and its transport velocity is 808 m/yr. Therefore, the residence time of H-3 in the 24 meters of contamination is 0.03 years. RESRAD does not allow time steps smaller than 1 year and this limitation leads to large inaccuracy when the residence time is on the order of 1 year or less. Several attempts were made to change input parameters to RESRAD (leach rate, geometry, precipitation rate, etc) to obtain concentrations that were closer to those obtained using a simple mass balance. However, due to the interrelationships between parameters and other restrictions in RESRAD (e.g., maximum precipitation rate is 10 m/yr) this was not possible.

Table 7 Peak Dose for initial partial structure concentrations of 1 pCi/g

Radionuclide	Peak Dose (mrem/yr)
H-3	1.42E-03
H-3*	3.67E-03
C-14	2.14E-04
Co-60	1.45E-04
Ni-63	2.05E-04
Sr-90	3.67E-02
Cs-137	8.73E-03

* H-3 concentration determined by mass balance of the total inventory divided by the annual flow rate through the contaminated region.

6.0 Discussion and Conclusions

Release of residual radioactive contaminants, including H-3, C-14, Co-60, Ni-63, Sr-90, and Cs-137, from subsurface concrete structures at the Yankee Nuclear Power Station is of concern. Consideration is being given to the effect of using demolition debris containing residual radioactivity as backfill in four subsurface structures (PAB drain collection tank room, PAB gravity drain tank room. Spent Fuel Pit, and the Waste Vault). These facilities may have residual contamination in their walls and floors that may contribute to release to the groundwater. As the dose from the backfill is being calculated separately, this analysis was performed to assess the peak dose and well water concentration that could occur as a result of the residual radioactivity in the partially intact subsurface structures. Two mechanisms were considered, diffusive release from the intact concrete and equilibrium sorption onto the concrete backfill that fills the facilities. The majority of the volumes of the concrete walls and floors that may remain are below the water table. Thus, for these analyses it is assumed the entire facility is below the water table. Using these assumptions and appropriate parameters, the calculations showed that Sr-90 had the highest predicted dose rate (3.67E-02 mrem/yr).