Key Words:
F-Area Tank Farm
Radon
Airborne Pathway
Performance Assessment

Retention: Permanent

AIR AND RADON PATHWAY MODELING FOR THE F-AREA TANK FARM

Kenneth Dixon
Mark Phifer

SEPTEMBER 2007

Savannah River National Laboratory
Washington Savannah River Company
Savannah River Site
Aiken, SC 29808

Prepared for the U.S. Department of Energy Under
Contract Number DE-AC09-96SR18500

SRNL

DISCLAIMER

Printed in the United States of America

Prepared For
U.S. Department of Energy

Key Words:
F-Area Tank Farm
Radon
Airborne Pathway
Performance
Assessment

Retention: Permanent

AIR AND RADON PATHWAY MODELING FOR THE F-AREA TANK FARM

SEPTEMBER 2007

Savannah River National Laboratory
Washington Savannah River Company
Savannah River Site
Aiken, SC 29808

Prepared for the U.S. Department of Energy Under
Contract Number DE-AC09-96SR18500

SRNL

REVIEWS AND APPROVALS

Authors

K. L. Dixon, Environmental Sciences and Biotechnology — 9/4/2007 Date

M. A. Phifer, Environmental Sciences and Biotechnology — 9/4/07 Date

Design Check

M. A. Phifer, Environmental Sciences and Biotechnology — 9/4/07 Date

T. Whiteside, Environmental Sciences and Biotechnology — 9/6/07 Date

Approvals

J. J. Mayer, Level 4 Manager, Environmental Sciences and Biotechnology — 9/5/07 Date

W. E. Stevens, Level 3 Manager, Environmental and Chemical Process Technology — 9/10/07 Date

Marcia Birk, SRIP Regulatory Documentation — 9/11/07 Date

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF ACRONYMS

Ci	Curie
DRF	Dose Release Factor
FTF	F-Area Tank Farm
K_s	Saturated Hydraulic Conductivity
MEI	Maximally Exposed Individual
NRC	Nuclear Regulatory Commission
PA	Performance Assessment
pCi	picoCurie
SRIP	Site Regulatory Integration and Planning
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
USDOE	United States Department of Energy
WMAP	Waste Management Area Projects
WSRC	Washington Savannah River Company

1.0 EXECUTIVE SUMMARY

The F-Area Tank Farm (FTF) is located within F-Area in the General Separations Area (GSA) of the Savannah River Site (SRS) as seen in Figure 1. The GSA contains the F and H-Area Separations Facilities, the S-Area Defense Waste Processing Facility, the Z-Area Saltstone Facility, and the E-Area Low-Level Waste Disposal Facilities. The FTF is a nearly rectangular shaped area and comprises approximately 20 acres, which is bounded by SRS coordinates N 76,604.5 to N 77,560.0 and E 52,435.0 to E 53,369.0.

SRS is in the process of preparing a Performance Assessment (PA) to support FTF closure. As part of the PA process, an analysis was conducted to evaluate the potential magnitude of gaseous release of radionuclides from the FTF over the 100-year institutional control period and 10,000-year post-closure compliance period. Specifically, an air and radon pathways analysis has been conducted to estimate the flux of volatile radionuclides and radon at the ground surface due to residual waste remaining in the tanks following closure. This analysis was used as the basis to estimate the dose to the maximally exposed individual (MEI) for the air pathway per Curie (Ci) of each radionuclide remaining in the combined FTF waste tanks.

For the air pathway analysis, several gaseous radionuclides were considered. These included carbon-14 (C-14), chlorine-36 (Cl-36), iodine-129 (I-129), selenium-79 (Se-79), antimony-125 (Sb-125), tin-126 (Sn-126), tritium (H-3), and technetium-99 (Tc-99). The dose to the MEI was estimated at the SRS Boundary during the 100 year institutional control period. For the 10,000 year post closure compliance period, the dose to the MEI was estimated at the 100 m compliance point.

For the radon pathway analysis, five parent radionuclides and their progeny were analyzed. These parent radionuclides included uranium-238 (U-238), plutonium-238 (Pu-238), uranium-234 (U-234), thorium-230 (Th-230), and radium-226 (Ra-226). The peak flux of radon-222 due to each parent radionuclide was estimated for the simulation period of 10,100 years.

2.0 INTRODUCTION

The F-Area Tank Farm (FTF) is located within F-Area in the General Separations Area (GSA) of the Savannah River Site (SRS) as seen in Figure 1. The GSA contains the F and H-Area Separations Facilities, the S-Area Defense Waste Processing Facility, the Z-Area Saltstone Facility, and the E-Area Low-Level Waste Disposal Facilities. The FTF is a nearly rectangular shaped area and comprises approximately 20 acres, which is bounded by SRS coordinates N 76,604.5 to N 77,560.0 and E 52,435.0 to E 53,369.0.

The FTF includes twenty-two waste tanks, which were emplaced between 1951 and 1976. Figure 2 provides an aerial view of the FTF looking southwest toward the 281-8F and 241-97F basins.

A detailed description of the construction of each tank group and the types of tanks used is given by Phifer et al. (2007a). In general, the FTF consists of four tank groups each with a different tank design. Tanks 1 through 8 (Group 1) were designated Type 1 Waste Tanks. Tanks 17 through 20 (Group 2) were designated Type IV Waste Tanks. Tanks 33 and 34 (Group 3) were designated as Type III Waste Tanks. Tanks 25 through 28 and 44 through 47 (Group 4) were designated as Type IIIA Waste Tanks.

SRS is in the process of preparing a Performance Assessment (PA) to support FTF closure. As part of the PA process, an analysis has been conducted to evaluate the potential magnitude of gaseous release of radionuclides from the FTF over the 100-year institutional control period and 10,000-year post-closure compliance period. Specifically, an air and radon pathways analysis has been conducted to estimate the flux of volatile radionuclides and radon at the ground surface due to waste stored in the tanks. The results from this analysis will be used to estimate the dose to the maximally exposed individual (MEI) at the SRS boundary (during the institutional control period) and at the 100 m boundary (during the post closure compliance period). The sections that follow discuss the conceptual model for the air and radon pathways analysis, the numerical implementation of the conceptual model, and the dose calculations for the MEI based on the results of the modeling.

3.0 FTF AIR AND RADON PATHWAY ANALYSIS

This section describes the details associated with computing the dose to the MEI due to the FTF for the air and radon pathways. The air and radon pathway analysis was divided into two time periods: 1) 100-year institutional control period and 2) 10,000 year post-closure compliance period. This results in a 10,100 year simulation period. During the operational period, wastes will be removed from the tanks and the tanks will be filled with grout. Therefore, the operational period was not considered in this analysis.

The method employed and the key aspects of the analysis performed are discussed in the sections that follow. For the radon pathway the peak flux at the ground surface of ^{222}Rn was calculated for five parent radionuclides for each time period. For the air pathway analysis, a list of eight radionuclides of interest was provided by SRIP Regulatory Documentation. The

dose to the MEI was also calculated for these radionuclides based on the gaseous flux of each at the land surface for each time period.

The method chosen is a hybrid approach where most parameters were set to their best estimate values (i.e., based on available site-specific measurements or engineering judgment), while other parameters were set to conservative/bounding values. The conceptual PORFLOW transport model used for the air and radon pathway analysis has imbedded within it biases that are intended to be conservative where possible. The conceptual model for both the air and radon pathway analysis is the same and the PORFLOW transport model used for both pathways utilizes the same input files. Section 3.1 and its associated subsections discuss the conceptual model for the air and radon pathway analysis. Sections 3.2 and 3.3 discuss the details specific to each analysis.

Four waste tank types were used in the FTF. Of these four waste tank types, the Type 1 tank was chosen for this analysis. This analysis does not consider any piping or ancillary equipment associated with the waste tanks. A schematic of the Type 1 tank is given in Figure 3. This tank type was selected because of the four tank types it will have the least grout and concrete thickness above the waste zone, which is located at the bottom of the tank. Additionally the minimum closure cap thickness over the tanks was assumed for conservatism. These assumptions should produce the maximum flux of gaseous radionuclides at the ground surface.

3.1 AIR AND RADON PATHWAY CONCEPTUAL MODEL

The approach taken focuses primarily on a baseline scenario where nominal settings for many of the input parameters have been conservatively chosen. The main analysis tool employed is the PORFLOW code which simulates the transport of radionuclide chains (i.e., parents and daughters) in porous media. The flux of radioactive gasses at the land surface above the FTF was evaluated for the closure configuration given by Phifer et al. (2007b). Gaseous radionuclides within the waste zone diffuse outward into the air-filled pore space of the overlying materials. Ultimately, some of the radionuclides emanate at the land surface. As such, air is the medium through which they diffuse. It is assumed that fluctuations in atmospheric pressure at the land surface that could induce small pulses of air movement into and out of the shallow soil profile over relatively short periods of time will have a zero net effect when averaged over longer time periods. Thus, advective transport of radionuclides in air-filled soil pores is not considered to be a significant process when compared to the rate of air diffusion.

The closure cap as described by Phifer et al. (2007b) consists of a top soil layer, an upper backfill layer, an erosion barrier layer, middle backfill layer, lateral drainage layer, a high density polyethylene (HDPE) geomembrane, a geosynthetic clay liner (GCL), an upper foundation layer, and a lower foundation layer. The HDPE geomembrane and the GCL are excluded from this analysis. By excluding these materials, the baseline analysis will be more conservative as these materials would be expected to significantly reduce gaseous flux at the land surface. The HDPE geomembrane would have very low gaseous diffusion coefficients and the GCL would have very little air-filled porosity, since it would be at or near saturation. The top soil layer and the upper backfill layer are also excluded from the baseline analysis,

since they are located above the erosion barrier and are therefore subject to erosion. For the purposes of this analysis, it is assumed that those components situated below the top of the erosion barrier remain intact for the duration of the simulation (10,100 years).

The Type 1 waste tank includes primary and secondary steel liners situated above a layer of base mat concrete as shown in Figure 3. The top of the tank is covered with a concrete roof. For the baseline analysis, the model domain begins at the top surface of the lower primary liner and extends through the waste material to the top of the erosion barrier. The baseline model excludes the upper primary steel liner. As with the exclusion of the geomembrane and GCL, this should make the model more conservative because including the steel liner would be expected to significantly reduce gaseous flux at the land surface.

The total thickness of the waste, tank, and cover materials (excluding the top soil, upper backfill, geomembrane, GCL, and steel liner) is 36.33 ft (10.77 m), with a waste layer thickness of 1.0 ft (0.30 m). The waste layer thickness was provided by SRIP. Table 1 lists the individual components of the Type 1 tank and closure cap included in the analysis. Materials are indicated with the associated thickness of each component, in inches, feet, and meters.

3.1.1 Air and Radon Pathway Diffusive Transport Model

A 1-dimensional PORFLOW based diffusive transport model was created for the FTF Type 1 tank baseline scenario. PC-based PORFLOW Version 5.97.0 was used to conduct the simulations (ACRI, 2004). PORFLOW has been widely used at the SRS and in the USDOE complex to address major issues related to the groundwater and nuclear waste management.

The governing equation for mass transport of species *k* in the fluid phase is given by

$$\frac{\partial C_k}{\partial t} + \frac{\partial}{\partial x_i}(V_i C_k) = \frac{\partial}{\partial x_i}(D_{ij}\frac{\partial C_k}{\partial x_j}) + \gamma_k$$

Where

C_k	concentration of species *k*, Ci/m^3
V_i	fluid velocity in the ith direction, m/yr
D_{ij}	effective diffusion coefficient for the species, m^2/yr
γ_k	net decay of species *k*, Ci/m^3 yr
i, j	direction index
t	time, yr
x	distance coordinate, m

This equation is solved within PORFLOW to evaluate transient radionuclide transport above the tank and to estimate gaseous radionuclide flux at the land surface over time. For this analysis, the advection term was disabled within PORFLOW and only the diffusive and net decay terms were evaluated.

The boundary conditions imposed on the entire model domain included:

- No-flux specified for all radionuclides along sides and bottom
  ($\partial C/\partial X = 0$ at x=0, x=1 and $\partial C/\partial Y = 0$ at y=0)
- Species concentration set to 0 at land surface (top of erosion barrier)
  ($C = 0$ at $y=y_{max}$)

These boundary conditions force all of the gaseous radionuclides to move upward from the waste disposal zone to the land surface. In reality, some lateral and downward diffusion occurs in the air-filled pores surrounding the waste zone; hence ignoring this lateral and downward movement has the effect of increasing the flux at the land surface. This should introduce some conservatism in the calculated results. Simulations were conducted in transient mode for diffusive transport in air, with results being obtained over 10,100 years which includes both the institutional control and post-closure compliance periods.

The initial condition imposed on the domain, except for the waste zone, included:

- Species concentration set to 0 at time = 0
  ($C=0$ for $0 \leq x \leq 1$ at t=0 and $C=0$ for $0 \leq y \leq y_{max}$ at t=0)

For the air pathway analysis, the initial conditions for the model assumed a 1 Ci inventory of each radionuclide uniformly spread over the waste zone. For the radon pathway analysis, an emanation factor of 0.25 was applied resulting in an initial inventory of 0.25 Ci for each parent radionuclide uniformly spread over the waste zone. This is discussed in more detail in Section 3.3.

3.1.1.1 Grid Construction

The model grid for the tank and overlying cover materials was constructed as a node mesh 3 nodes wide by 80 nodes high. This mesh creates a vertical stack of 78 model elements. Figure 4 shows a schematic of the PORFLOW model grid. The grid extends upward to the top of the erosion barrier, since this is the minimum possible cover thickness that could exist during the simulation period. A set of consistent units was employed in the simulations for length, mass and time, these being meters, grams and years, respectively.

3.1.1.2 Material Zone Properties and Other Input Parameters

Material properties utilized within the 1-D numerical model were specified for 8 material zones defined within the model domain. Each material zone was assigned values of particle density, total porosity, average saturation, air-filled porosity, air density, and an effective air-diffusion coefficient for each source element or compound. An effective air-diffusion coefficient was used for each radionuclide and material layer. Therefore, tortuosity was assigned a unit value in each material zone. An air fluid density of 1.24×10^3 g/m^3 at standard atmospheric conditions was used in the transport simulations (Bolz et al., 1973).

The waste layer was assumed to be 1 ft thick and confined to the bottom of the tank. The waste tank is to be filled with a reducing grout from the site concrete specification (OPDEXE-X-P-0-BS) and it was assumed that the waste layer would have similar properties.

The hydraulic and physical properties of this mix have been determined by Dixon and Phifer (2007). Based on the results of this testing, the waste layer and the reducing grout layer was assigned a particle density of 2.51 g/cm^3 and a total and air-filled porosity of 0.266. The concrete roof layer was assumed to be similar to the base mat surrogate tested by Dixon and Phifer (2007). This layer was assigned a particle density of 2.51 g/cm^3 and a total and air-filled porosity of 0.168. The waste layer, the reducing grout, and the concrete roof were conservatively assumed to be dry (i.e., total porosity = air-filled porosity).

The foundation layer is divided into the upper and lower foundation layers (Phifer et al. 2007b). It is anticipated that the lower foundation layer will need to promote drainage of infiltrating water away from and around the tanks, requiring a relatively high saturated conductivity such as 1.0E-03 cm/s. It is anticipated that the upper foundation layer will consist of soil with a moderately low permeability (i.e., ≤1.0E-06 cm/s) produced by blending typical SRS backfill with a small weight percent bentonite. The particle density of the lower and upper foundation layers was assigned that of control compacted backfill from Phifer et al., 2006 (i.e., 2.63 g/cm^3).

The particle density of the middle backfill layer was also assigned that of control compacted backfill from Phifer et al., 2006 (i.e., 2.63 g/cm^3). The lateral drainage layer and erosion barrier layer were assigned a particle density typical of quartz (i.e., 2.65 g/cm^3 (Hillel 1982)).

Phifer et al. (2007b) evaluated infiltration through the closure cap materials over time as the closure cap degraded using the HELP model. Values for total porosity and volumetric moisture content for the closure cap materials and foundation layers were taken from this analysis. These values were used to calculate the average saturation and the air-filled porosity for the closure cap materials (Table 2). The maximum air-filled porosity for each material layer over the 10,000-year simulation was utilized, since this represented the greatest air filled porosity in which a gas could diffuse.

Table 2 provides the values of particle density, total porosity, average saturation, and air-filled porosity utilized for all the layers used in the baseline scenario (i.e., waste layer to the erosion barrier) for the simulation period.

3.1.2 Summary of Key Air and Radon Pathway Assumptions

The following are the key air and radon pathway analysis assumptions associated with the FTF baseline scenario:

- The waste layer may be represented as a 1 ft thick layer of material located at the bottom of the tank.
- The waste layer is assumed to be dry and to have properties similar to reducing grout.
- Exclusion of the top soil, upper backfill, HDPE geomembrane, geosynthetic clay liner, and primary steel liner of the waste tank make the model more conservative.
- The final closure cap as outlined in Table 1 is assumed to remain intact for the duration of the simulation (10,100 years).

3.1.2.1 Measures Implemented to Ensure Conservative Results

In this analysis, several conditions introduce conservatism into the calculations. These include:

- The use of boundary conditions that force all of the gaseous radionuclides to move upward from the waste disposal zone to the land surface. In reality, some of the gaseous radionuclides diffuse sideways and downward in the air-filled pores surrounding the waste zone, hence ignoring this has the effect of increasing the flux at the land surface.
- Not taking credit for the removal of radionuclides by pore water moving vertically downward through the model domain. This mechanism would likely remove some dissolved radionuclides, and therefore its omission has the effect of increasing the estimate of instantaneous radionuclide flux at the land surface in simulations conducted as a part of this investigation.
- Exclusion of the HDPE geomembrane, the geosynthetic clay liner, and the primary steel liner of the waste tank. Inclusion of these materials in the model would significantly reduce the gaseous flux at the land surface due to their material properties (i.e., low air-filled porosity).
- Exclusion of the cover materials above the erosion barrier (i.e., top soil and upper backfill layers). Excluding these materials shortens the diffusion pathway and could increase the flux at the land surface.
- Assuming the waste layer, the reducing grout, and concrete roof are dry. This makes the air-filled porosity equal to the total porosity. This maximizes diffusive transport through these materials since gaseous flux is through the air-filled porosity.
- Use of the Type 1 tanks and minimum closure cap thickness to estimate dose to the MEI..

3.2 FTF AIR PATHWAY MODEL

For the air pathway analysis, a list of radionuclides of interest was provided by SRIP Regulatory Documentation. These radionuclides included carbon-14 (C-14), chlorine-36 (Cl-36), iodine-129 (I-129), selenium-79 (Se-79), antimony-125 (Sb-125), tin-126 (Sn-126), tritium (H-3), and technetium-99 (Tc-99). A summary of the radionuclides and compounds of interest is presented in Table 3.

The radionuclides of interest are assumed to be in the gas phase and uniformly distributed through the 1 ft thick waste layer at the bottom of the tank. Certain gaseous radionuclides will not likely remain in the monatomic elemental form. These radionuclides will likely combine with other gaseous elements or form diatomic molecules. The state of existence of each of these radionuclides in the gaseous phase is important in evaluating their transport to the land surface because the diffusion coefficient associated with each is related to its molecular weight.

In this investigation it is assumed that:

- C-14 exists as part of the CO_2 molecule
- Cl-36, H-3 and I-129 exist as diatomic gasses
- Sb-125, Se-79, Sn-126, and Tc-99 exist as monatomic gasses.

The effective air diffusion coefficient of each radionuclide or compound within each material zone was determined. Nielson et al. (1984) established a relationship between moisture saturation and the radon effective air-diffusion coefficient for various pore sizes of earthen materials. Using this method, a radon effective air-diffusion coefficient was determined for each material type based upon the average moisture saturation for the material. Subsequently, using Graham's Law, the effective air-diffusion coefficient of each radionuclide or compound evaluated was determined for each material type based on the radon effective air-diffusion coefficient using the following relationship:

$$D = D' \sqrt{\frac{MWT'}{MWT}}$$

Where:

D = the effective diffusion coefficient of the radionuclide of interest (m^2/yr) within the material zone of interest

D' = the effective diffusion coefficient of Rn-222 (m^2/yr) within the material zone of interest

MWT' = the molecular weight of the reference radionuclide (Rn-222)

MWT = the molecular weight of the element or compound of interest

A summary of the radon effective air-diffusion coefficients and the calculated effective air-diffusion coefficients for each radionuclide/compound by material zone are presented in Table 4.

3.2.1 Air Pathway Model Results

3.2.1.1 Air Pathway Flux to Ground Surface

Model simulations were conducted to evaluate the peak flux of each radionuclide emanating from the top of the model domain. A unit inventory of 1 Ci was assigned to the FTF Type 1 waste tank waste zone for each radionuclide considered in the analysis. Results were output in Ci/yr, consistent with the set of units employed in the model, and are presented for each radionuclide in Figure 5. The peak fluxes emanating at the land surface are presented for each time period in Table 5. The results are reported in this way to facilitate calculation of human exposure at the SRS boundary, the 100 m boundary, and the 1600 m boundary due to the FTF Type 1 waste tank.

3.2.2 Air Pathway Dose Calculations

An evaluation was conducted to assess the potential dose to a maximally exposed individual (MEI) located at the SRS boundary, the 1600 m location (seepline), and the 100 m location (Farfan, 2007). During the 100 year institutional control period, the SRS boundary is the compliance point for the dose calculations. Therefore, the peak flux during this time period was used to assess the dose to the MEI. For the remainder of the time period, the 100 m boundary is the compliance point. Thus, the peak flux between 100 and 10,100 years was used for these calculations. In addition, dose calculations were performed for the seepline location (i.e., 1600 m location) using the peak flux for the entire 10,100 year simulation period. Dose-release factors (DRF) were calculated for each radionuclide potentially released from the FTF using CAP88, the EPA model for National Emissions Standards for Hazardous Air Pollutants (NESHAP). DRFs represent the dose to the receptor exposed to 1 Ci of the specified radionuclide potentially released to the atmosphere. For the receptor located at the SRS boundary and at the seepline (1600 m), the distance from the FTF is sufficient for an assumption of a point source. However, the DRFs for the 100 m receptor requires evaluation of an area source because of the close proximity of the FTF to the 100 m receptor. For radionuclides not contained within the CAP88 library (Se-79, Cl-36) atmospheric transport was estimated by assigning surrogates with similar radiological properties (Farfan, 2007). Doses for these radionuclides were estimated by applying their dosimetric properties to the surrogate's relative air concentrations estimated by the model.

Specific SRS Boundary DRFs and the calculated exposure levels for the 0 to 100 year MEI at the SRS boundary are presented in Table 6. Specific SRS 100-meter DRFs and the calculated exposure levels for the 100 to 10,100 year MEI at 100-meters are presented in Table 7. Specific SRS 1600-meter DRFs and the calculated exposure levels for the 100 to 10,100 year MEI at 1600-meters are presented in Table 8. See Farfan (2007) for details on the estimation of all DRFs.

3.3 FTF RADON ANALYSIS

This section describes the investigation conducted to evaluate the potential magnitude of radon release from the FTF during the 10,100-year simulation period. This investigation addresses only Rn-222. It is assumed that the short half-life of Rn-220 (55.6 seconds) renders it unable to escape the FTF waste tanks and migrate to the land surface via air-diffusion before it is transformed by radioactive decay.

The permissible radon flux for USDOE facilities is addressed in DOE G 435.1-1 Appendix A. In this Appendix, Section IV. P.(c) states the radon flux limitations associated with the development of a disposal facility and maintenance of a performance assessment and the closure of the disposal facility. This requirement is that the release of radon shall be less than an average yearly flux of 20 $pCi/m^2/sec$ at the surface of the disposal facility. The requirements state that this standard was adopted from the uranium mill tailings requirements in 40 CFR Part 192 and 10 CFR Part 40. 10 CFR Part 40 discusses both Rn-222 from uranium and Rn-220 from thorium, therefore the performance objective refers only to radon, and the correct species must be analyzed depending on the characteristics of the waste stream. The instantaneous Rn-222 flux at the land surface was evaluated for the simulation period and the maximum flux was then compared to the USDOE performance objective.

The potential parent radionuclides that can contribute to the creation of Rn-222 are illustrated in Figure 6. The diagram indicates the specific decay chains that lead to the formation of Rn-222, as well as the half-lives for each radionuclide. The extremely long half-life of U-238 (4.468E+9 years) cause the other radionuclides higher up on the chain of parents to be of little concern with regard to their potential to contribute significantly to the Rn-222 flux at the land surface over the period of interest. In Figure 6, the parent radionuclides that were individually evaluated are indicated with the gray shaded area (i.e., beginning with Pu-238 and U-238). Rn-222 generated within the waste zone is in the gaseous phase and diffuses outward from this zone into the air-filled soil pores surrounding the FTF, eventually resulting in some of the radon emanating at the land surface. As such, air is the fluid through which Rn-222 diffuses, although some Rn-222 may dissolve in residual pore water.

The parent radionuclides are assumed to exist in the solid phase and therefore do not migrate upward through the air-filled pore space, although they could be leached and transported downward from the waste zone by pore water movement. This potential downward migration of the parent radionuclides was not considered in the radon analysis.

Decay chains evaluated were U-238→Th-234→Pa-234m→U-234→Th-230→ Ra-226→Rn-222 and Pu-238 →U-234→Th-230→Ra-226→Rn-222. Each parent in these chains, except Th-234 and Pa-234m, were simulated separately as the starting point of the decay chain. Th-234 and Pa-234m have extremely short half-lives compared to the other parent radionuclides in these chains. Only a fraction of the Rn-222 generated by the decay of each parent is available for migration away from its source and into open pore space. Since the Rn-222 parent radionuclides exist as oxides or in other crystalline forms, only a fraction of Rn-222 generated by decay of Ra-226 has sufficient energy to migrate away from its

original location into adjacent pore space before further decay occurs (3.82 day half-life for Rn-222).

The emanation coefficient is generally defined as the fraction of the total amount of Rn-222 produced by radium decay that escapes from soil particles and enters the pore space of the medium. This is the fraction of the Rn-222 that is available for transport. In the case of the FTF, the parent radionuclides are not embedded in soil but are contained within waste entombed in concrete/grout. Literature values for the Rn-222 emanation factor for these conditions are not available. Studies have shown the emanation factor to vary between 0.02 and 0.7 for various soil types depending primarily on moisture content. Generally, higher emanation factors are associated with higher moisture contents.

RESRAD is a model used to estimate radiation dose and risk from residual radioactive materials. This USDOE and Nuclear Regulatory Commission (NRC) approved code, assumes an emanation factor of 0.25 for Rn-222 which is representative of a silty loam soil with a low moisture content. For the FTF radon pathway analysis, the RESRAD default emanation factor of 0.25 was chosen recognizing that literature values for wastes similar to the FTF are not available. The use of 0.25 should be conservative since the waste is assumed to be dry and emanation factors reported in the literature for drier soils are much lower (Yu, et al. 2001). To account for the emanation factor in the model, an effective source term of 0.25 Ci of parent radionuclide was utilized for each Ci disposed within the facility.

Some radon dissolves in pore water but since diffusion proceeds more slowly in that fluid, air diffusion was the only transport process by which Rn-222 was allowed to reach the land surface of the FTF. This assertion is substantiated in Yu, et al. 2001. In that report the effective diffusion coefficient for soil is reported to range from the radon open air diffusion coefficient of 1.0×10^{-5} m^2/sec to that of fully saturated soil, 1.0×10^{-10} m^2/sec. This 5-order of magnitude difference is consistent with the comparison of water diffusion coefficients to air diffusion coefficients of other common molecular compounds and reported in many references. Thus, the larger volume of water-filled pore space compared to air-filled pore space (maximum of 1 order of magnitude difference) is inconsequential, in terms of the ability of water-dissolved radon to diffuse through water-filled pores as compared to the ability of the same compounds to diffuse as gas in the vapor-filled pore spaces.

The molecular diffusion coefficient of Rn-222 in open air is 347 m^2/yr (Nielson et al., 1984). Nielson et al. (1984) established a relationship between moisture saturation and the radon effective air-diffusion coefficient for various pore sizes of earthen materials. This method was used to calculate a radon effective air-diffusion coefficient for each material type based upon the average moisture saturation for the material. Tortuosity was assigned a unit value for each material type. A summary of the radon air-diffusion coefficients by material type are presented in Table 4.

3.3.1 Radon Pathway Model Results

Model simulations were conducted to evaluate the peak instantaneous Rn-222 flux at the land surface for the simulation period of 10,100 years. The simulation was divided into two time periods: 1) 100 year institutional control period and 2) 10,000 year post-closure compliance period. Model results were output in $Ci/m^2/yr$ per Ci of inventory, consistent with the set of units employed in the model. A graph of these results is shown in Figure 7, although the units are converted to $pCi/m^2/sec$ per Ci/m^2, which are the units used to define the regulatory flux limit in DOE G 435.1-1. The peak fluxes represent the peak Rn-222 flux per square meter at the land surface for the two time periods and are given in Table 9.

4.0 SUMMARY

SRS is in the process of preparing a Performance Assessment (PA) to support FTF closure. As part of the PA process, an analysis was conducted to evaluate the potential magnitude of gaseous release of radionuclides from the FTF over the 100-year institutional control period and 10,000-year post-closure compliance period. Specifically, an air and radon pathways analysis has been conducted to estimate the flux of volatile radionuclides and radon at the ground surface due to waste stored in the tanks. This analysis was used as the basis to estimate the dose to the maximally exposed individual (MEI) for the air pathway per Curie (Ci) of each radionuclide remaining in the combined FTF waste tanks.

For the air pathway analysis, several gaseous radionuclides were considered. These included carbon-14 (C-14), chlorine-36 (Cl-36), iodine-129 (I-129), selenium-79 (Se-79), antimony-125 (Sb-125), tin-126 (Sn-126), tritium (H-3), and technetium-99 (Tc-99). The dose to the MEI was estimated at the SRS Boundary during the 100 year institutional control period. For the 10,000 year post closure compliance period, the dose to the MEI was estimated at the 100 m compliance point.

For the radon pathway analysis, five parent radionuclides and their progeny were analyzed. These parent radionuclides included uranium-238 (U-238), plutonium-238 (Pu-238), uranium-234 (U-234), thorium-230 (Th-230), and radium-226 (Ra-226). The peak flux of radon-222 due to each parent radionuclide was estimated for the simulation period of 10,100 years.

5.0 REFERENCES

ACRI (Analytical & Computational Research, Inc.). 2004. *PORFLOW Version 5.0 User's Manual*, Revision 5, Analytical & Computational Research, Inc., Los Angeles, California.

Bolz, R.E. and G.L. Tuve, (Editors), 1973. *Handbook of tables for APPLIED ENGINEERING SCIENCE, 2'nd Edition*. CRC Press, 18901 Cranwood Parkway, Cleveland, OH.

Dixon, K. L. and M. A. Phifer. 2007. Hydraulic and Physical Properties of Grouts and Alternative Fill Materials for FTF Closure, WSRC-STI-2007-00369, Draft, Washington Savannah River Company, Aiken, SC 29808.

Farfan, E. B. 2007. *Air Pathway Dose Modeling for the F-Area Tank Farm,* WSRC-STI-2007-00343, Washington Savannah River Company, Aiken, SC 29808.

Glover, T. J. 2000. Pocket Ref, 2nd Edition. Sequoia Publishing, Inc. Littleton, CO.

Hillel, D. 1982. Introduction to Soil Physics, Academic Press, Inc., San Diego, California.

Nazaroff, W.W., and A.V. Nero (editors), 1998, *Radon and its Decay Products in Indoor Air*, John Wiley & Sons, New York, N.Y.

Nielson, K.K., V.C. Rogers and G.W. Gee, 1984. *Diffusion of Radon through Soils: A pore distribution Model*, Soil Science Society of America, J. 48:482-487.

Phifer, M. A., W. E. Jones, E. A. Nelson, M. E. Denham, and M. R. Lewis. 2007a. FTF Closure Cap Concept and Infiltration Estimates (U), WSRC-STI-2007-00184, Rev.1, Washington Savannah River Company, Aiken, SC 29808.

Phifer, M. A., W. E. Jones, E. A. Nelson, M. E. Denham, M. R. Lewis, and E. P. Shine. 2007b. FTF Closure Cap Concept and Infiltration Estimates (U), WSRC-STI-2007-00184, Rev.2, Draft, Washington Savannah River Company, Aiken, SC 29808.

Phifer, M.A., M. A. Millings, G. P. Flach, 2006. *Hydraulic Property Estimation for the E-Area and Z-Area Vadose Zone Soils, Cementitious Materials, and Waste Zones (U).* WSRC-STI-2006-00198. Westinghouse Savannah River Company, Aiken, South Carolina. September 2003.

Tuli, J.K. 2005. Nuclear Wallet Cards, sixth edition, Brookhaven National Laboratory, Upton, New York.

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, 2001. *Users Manual for RESRAD Version 6*, Environmental Assessment Division, Argonne National Laboratory. Chicago, Illinois.

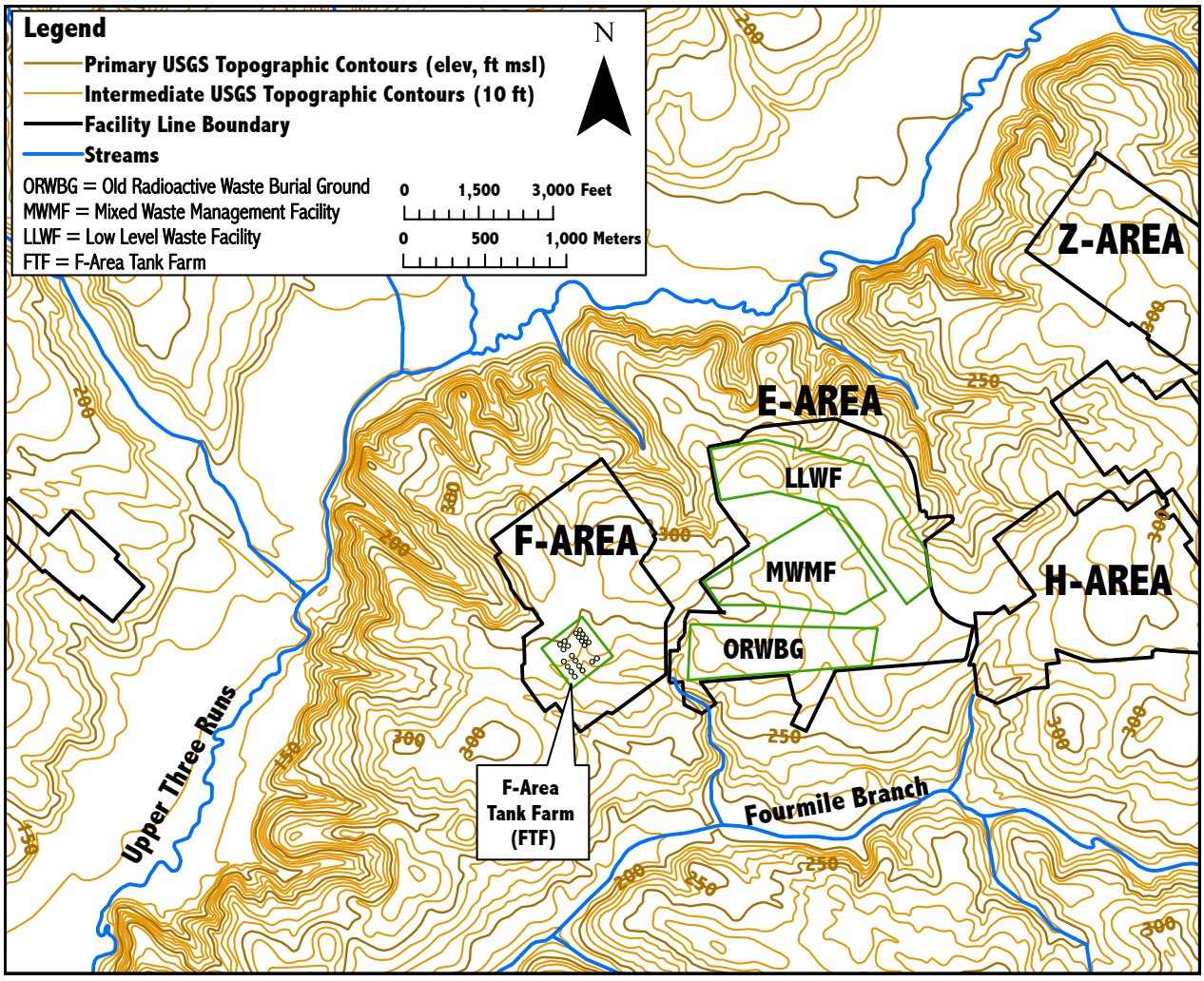


Figure 1. General Separations Area (GSA) Topography and FTF Location

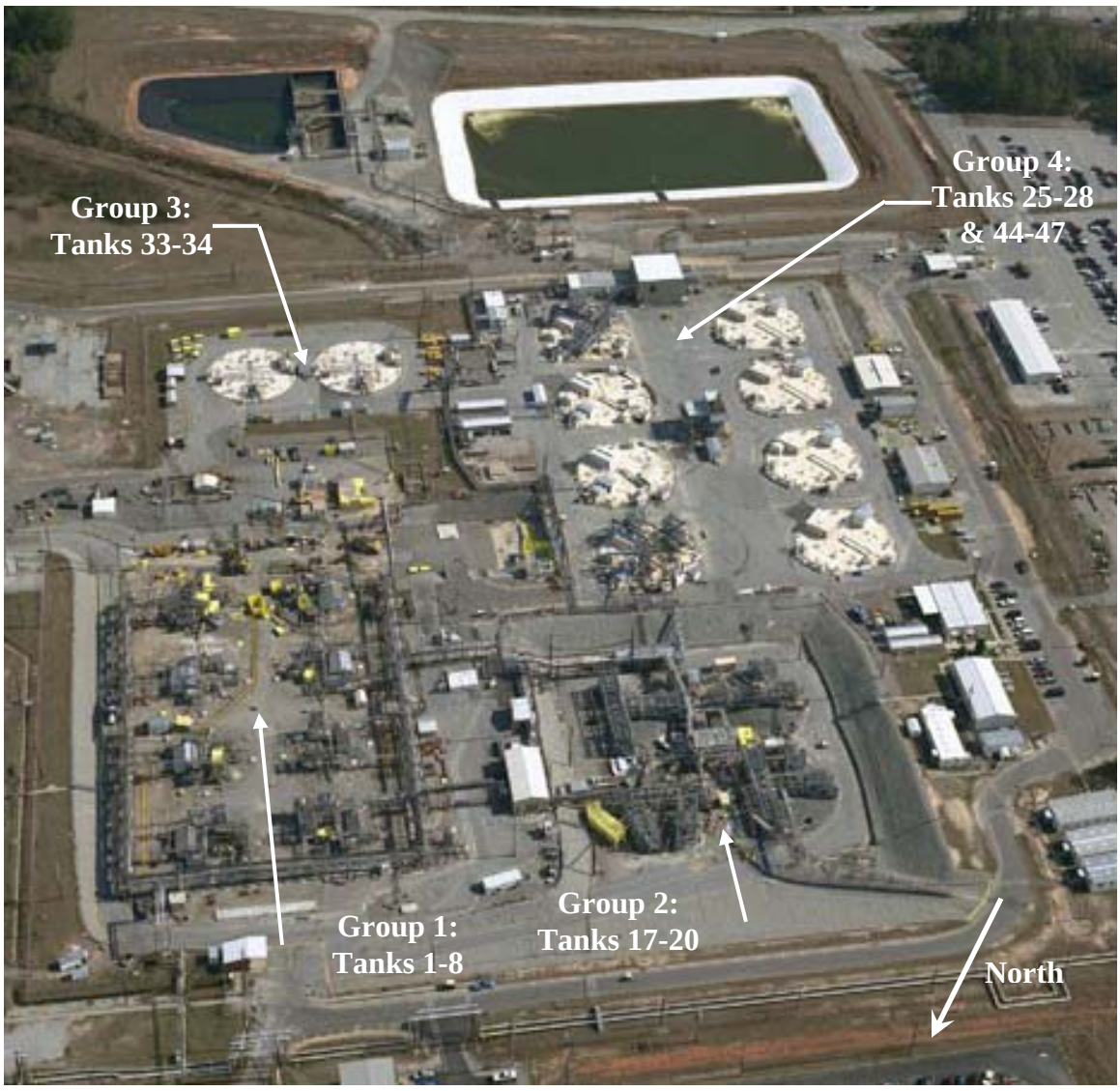
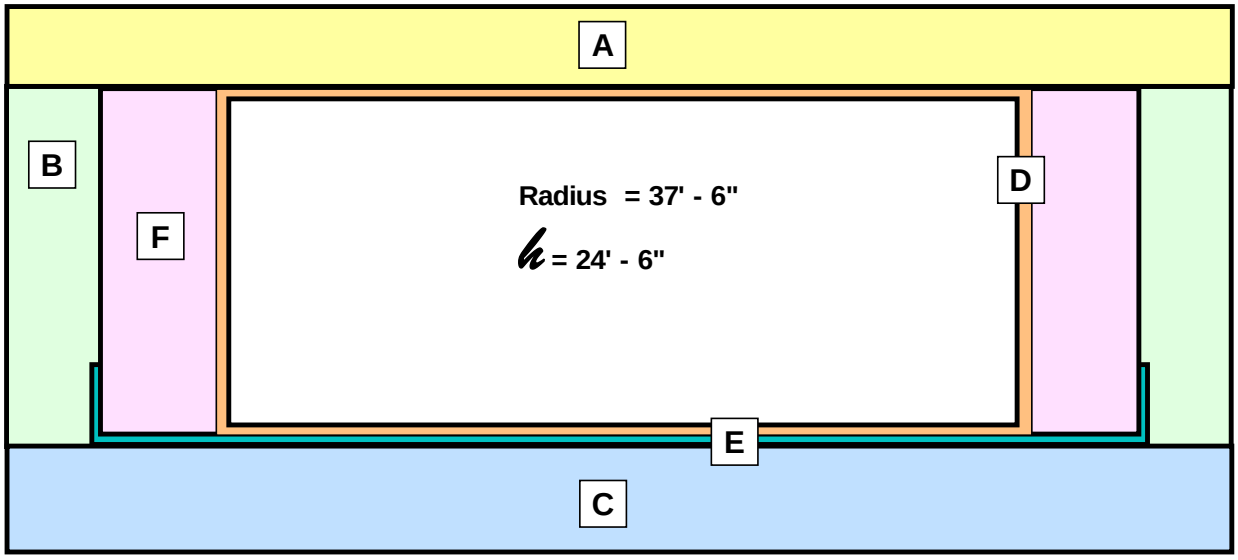


Figure 2. FTF Aerial View



[NOT TO SCALE]

LABEL	THICKNESS	MATERIAL
A Concrete Roof	22"	Concrete (Dupont Spec 3019, Sec. B)
B Concrete Wall	22"	Concrete (Dupont Spec 3019, Sec. B)
C Concrete Basemat	30"	Concrete (Dupont Spec 3019, Sec. B)
D Primary Liner	0.5"	Carbon Steel (ASTM A-285-50T)
E Secondary Liner	5' high and 0.5" thick	Carbon Steel (ASTM A-285-50T)
F Grouted Annulus	30"	Tank Fill Grout

Figure 3. Type 1 Waste Tank Modeling Dimensions

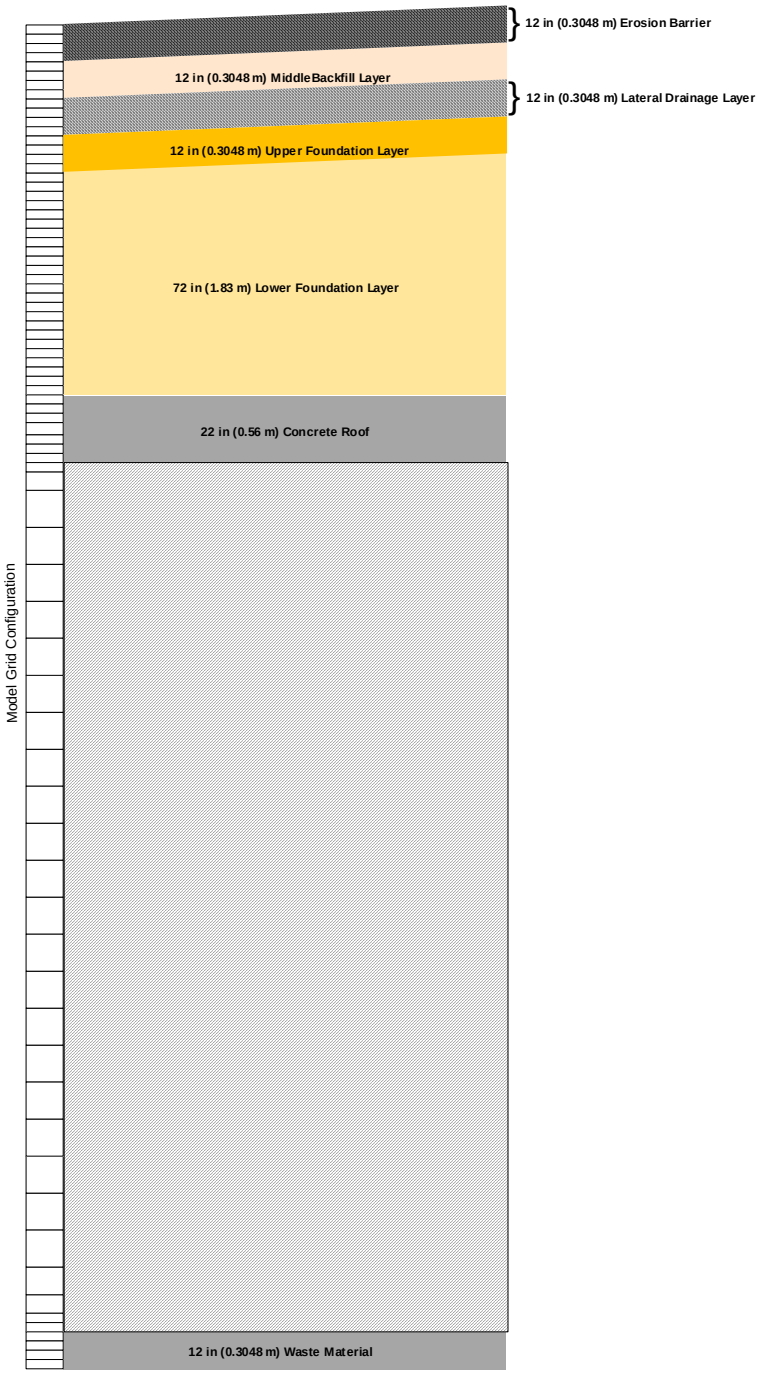


Figure 4. Schematic of PORFLOW Model Grid for Air and Radon Pathway Analysis

Note: For conservatism the model grid does not include the following layers: topsoil, upper backfill, HPDE geomembrane, and GCL.

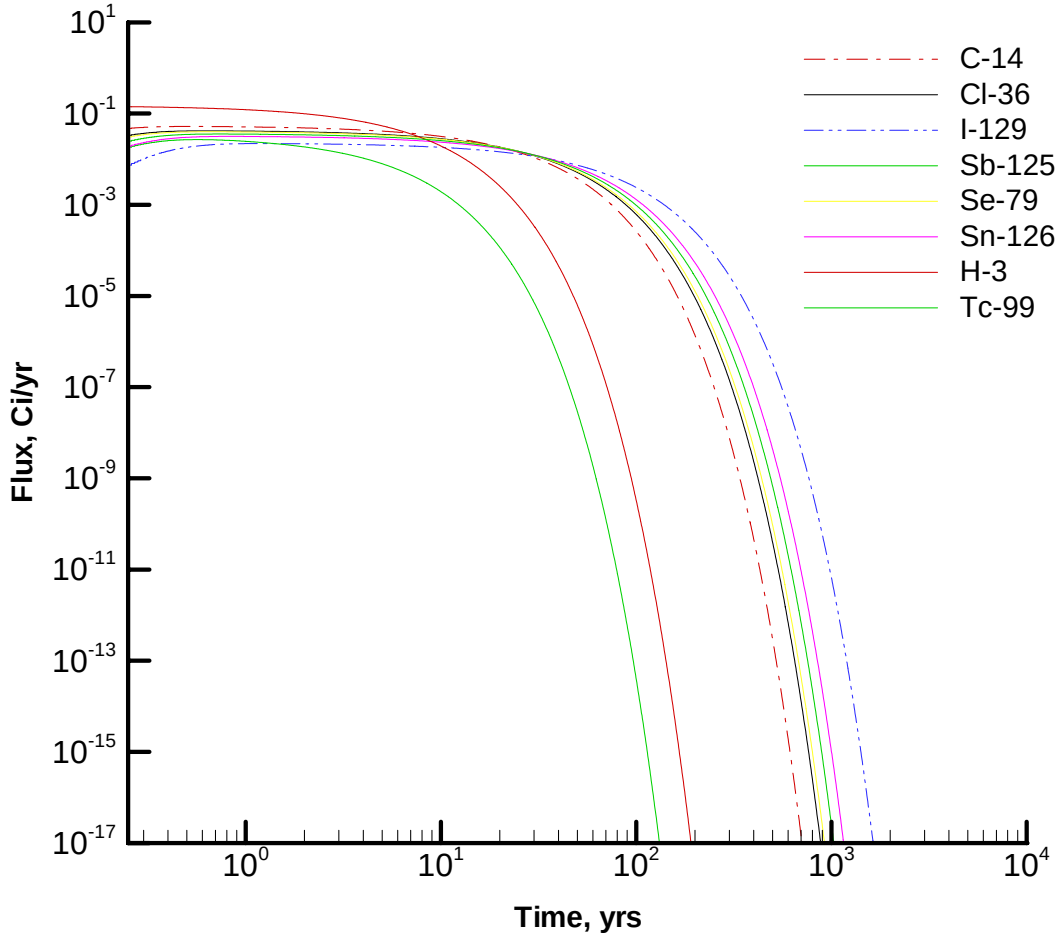


Figure 5. Flux at Land Surface for C-14, Cl-36, I-129, Sb-125, Se-79, Sn-126, H-3, and Tc-99 per Ci of Radionuclide Remaining in the Combined FTF Waste Tanks

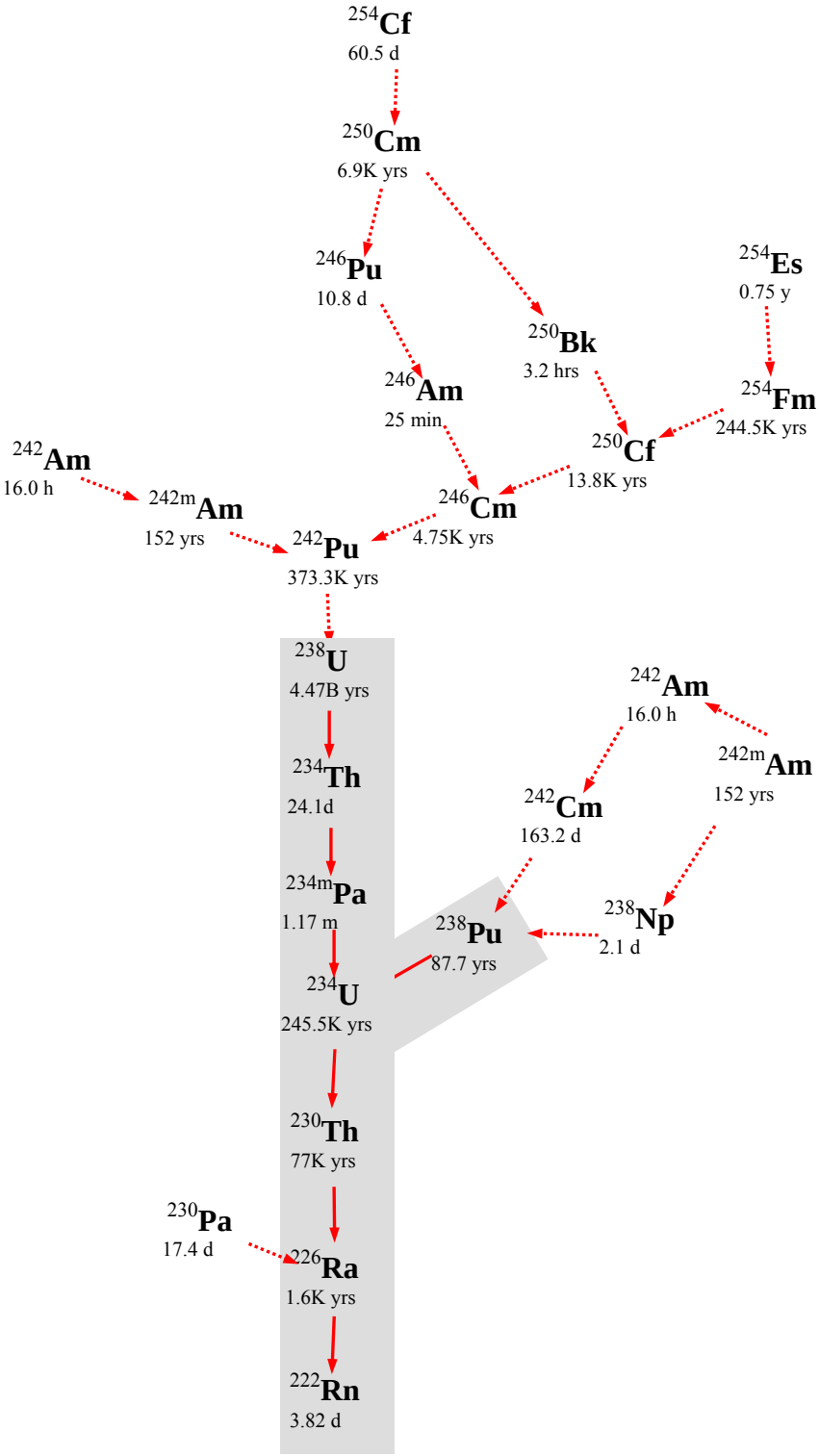


Figure 6. Radioactive Decay Chains Leading to Rn-222

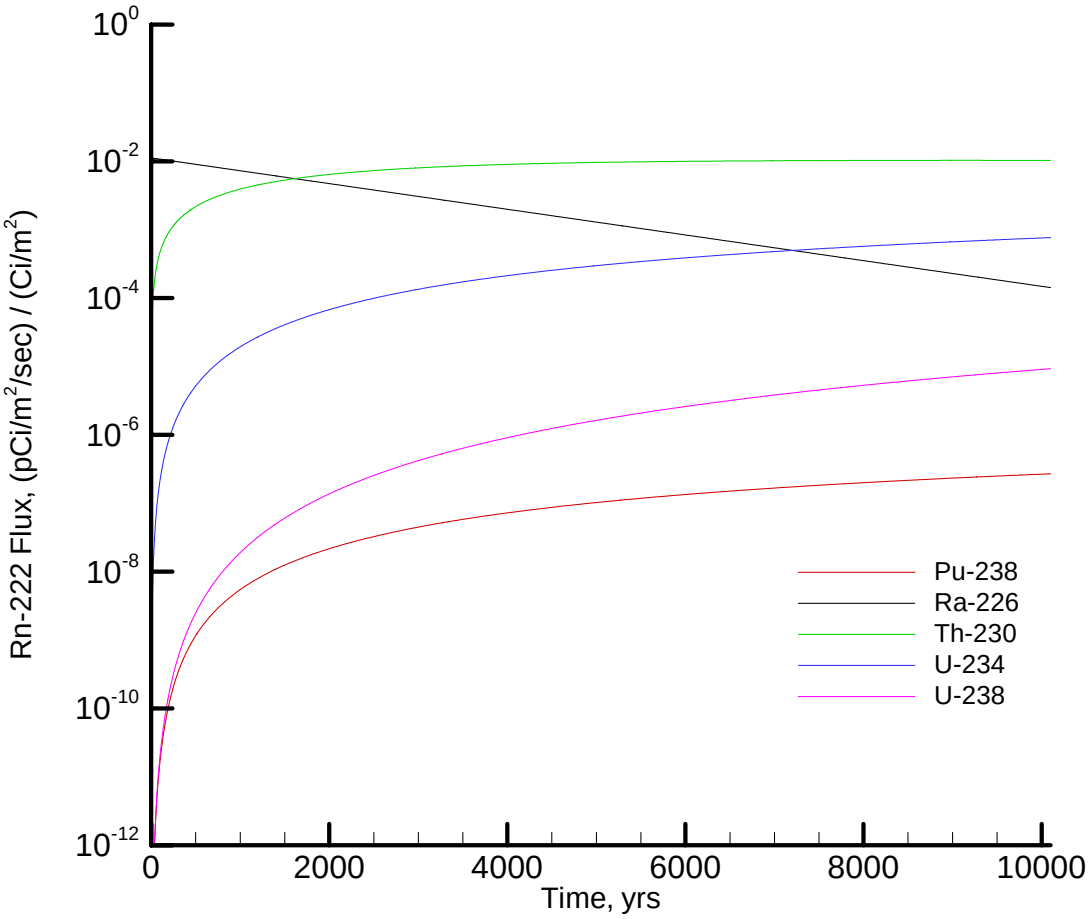


Figure 7. Rn-222 Flux at Land Surface Resulting from Unit Source Term

Table 1. Vertical Layer Sequence and Associated Thickness for FTF Type 1 Waste Tank and Cover Material

Layer	Thickness (inches)	Thickness (ft)	Thickness (m)
Erosion barrier	12	1.00	0.30
Middle backfill layer	12	1.00	0.30
Lateral drainage layer	12	1.00	0.30
Upper Foundation layer	12	1.00	0.30
Lower Foundation layer	72 (minimum)	6.00	1.83
Concrete Roof	22	1.83	0.56
Reducing Grout	282	23.5	7.16
Waste Layer	12	1.00	0.30

SOURCE: Adapted from Phifer, 2007b.

Table 2. Particle Density, Total Porosity, Average Saturation, and Air-Filled Porosity by Layer for the FTF Type 1 Tank Baseline Scenario.

Layer	Particle Density (g/cm^3)	Total Porosity (fraction)	Average Saturation (fraction)	Air-filled Porosity (fraction)
Erosion barrier layer [1, 3]	2.65	0.150	0.84	0.024
Middle backfill layer [2, 3]	2.63	0.371	0.82	0.067
Lateral drainage layer [1, 3]	2.65	0.417	0.61	0.162
Upper Foundation layer [2, 3]	2.63	0.35	0.72	0.098
Foundation Layer [2, 3]	2.63	0.457	0.28	0.328
Concrete Roof [4]	2.51	0.168	0.00	0.168
Reducing Grout [5]	2.51	0.266	0.00	0.266
Waste Layer [6]	2.51	0.266	0.00	0.266

[1] Particle density assumed to be that typical of quartz (Hillel 1982)
[2] Values for particle density taken as that of control compacted backfill from Phifer et al., 2006.
[3] Total porosity, average saturation, and air-filled porosity values derived from Phifer et al. (2007b).
[4] The concrete roof is assumed to be similar to the base mat surrogate as given by Dixon and Phifer, 2007. Particle density and porosity taken from Dixon and Phifer, 2007.
[5] Particle density and porosity of reducing grout taken from Dixon and Phifer, 2007.
[6] The waste is assumed to have the properties of reducing grout.
[7] The concrete roof, reducing grout, and waste layer are conservatively assumed to be dry; therefore the average saturation is taken as 0 and the air-filled porosity is taken as the total porosity.

Table 3. Radionuclides and Compounds of Interest for air and radon pathway analysis.

Radionuclide	Half-life[1] (yrs)	Approximate Atomic Wt.[2]	Molecular form in gaseous state	Molecular Wt.[2]
$^{14}CO_2$	5.700E+03	14	CO_2	45.99
2(^{36}Cl)	3.010E+05	36	Cl_2	72
2(^{129}I)	1.570E+07	129	I_2	258
^{125}Sb	2.759E+00	125	Sb	125
^{79}Se	2.950E+05	79	Se	79
^{126}Sn	2.300E+05	126	Sn	126
$^{3}H_2$	12.32E+00	3	H_2	6
^{99}Tc	2.111E+05	99	Tc	99
^{222}Rn	1.047E-02	222	Rn	222

[1]2005 Nuclear Wallet Cards (Tuli, 2005)
[2]Pocket Ref (Glover, 2000)

Table 4. Effective Air-Diffusion Coefficients for Each Radionuclide/Compound, by Material for FTF Type 1 Tank and Closure Cap.

Radionuclide	Tank Waste, Reducing Grout, and Concrete Roof Layer (m^2/yr)	Lower Foundation Layer (m^2/yr)	Upper Foundation Layer (m^2/yr)	Lateral Drainage Layer (m^2/yr)	Middle Backfill Layer (m^2/yr)	Erosion Barrier Layer (m^2/yr)
^{222}Rn[1]	3.470E+02	1.210E+01	2.618E+00	4.194E+00	1.455E+00	1.301E+00
^{14}C	7.623E+02	2.658E+01	5.752E+00	9.213E+00	3.196E+00	2.858E+00
^{36}Cl	6.093E+02	2.124E+01	4.597E+00	7.364E+00	2.555E+00	2.284E+00
^{129}I	3.219E+02	1.122E+01	2.429E+00	3.890E+00	1.350E+00	1.207E+00
^{125}Sb	4.624E+02	1.612E+01	3.489E+00	5.589E+00	1.939E+00	1.734E+00
^{79}Se	5.817E+02	2.028E+01	4.389E+00	7.030E+00	2.439E+00	2.181E+00
^{126}Sn	4.606E+02	1.606E+01	3.475E+00	5.567E+00	1.931E+00	1.727E+00
$^{3}H_2$	2.111E+03	7.359E+01	1.593E+01	2.551E+01	8.850E+00	7.912E+00
^{99}Tc	5.196E+02	1.812E+01	3.921E+00	6.280E+00	2.179E+00	1.948E+00

[1]The effective diffusion coefficient for ^{222}Rn was used to determine the effective air diffusion coefficient of each radionuclide/compound based on Graham's law.

Table 5. Summary of the Peak Fluxes for Each Radionuclide

		Peak Flux (Ci/yr/Ci)	
Radionuclide	**Activity in Waste (Ci)**	**0 - 100 Yrs**	**100 – 10,100 Yrs**
^{14}C	1.0	5.22E-02	2.59E-04
^{36}Cl	1.0	4.18E-02	6.07E-04
^{129}I	1.0	2.21E-02	2.38E-03
^{125}Sb	1.0	2.69E-02	3.71E-14
^{79}Se	1.0	3.99E-02	7.02E-04
^{126}Sn	1.0	3.16E-02	1.29E-03
$^{3}H_2$	1.0	1.43E-01	3.12E-10
^{99}Tc	1.0	3.56E-02	9.66E-04

Table 6. SRS Boundary Dose Release Factors Dose to the MEI for the 0-100 Year Time Period per Ci of Radionuclide Remaining in the Combined FTF Waste Tanks

Radionuclide	**Peak Flux (Ci/yr/Ci)**	**SRS Boundary Dose Release Factor[1] (mrem/Ci)**	**Dose to MEI at SRS Boundary[2] (mrem/yr/Ci)**
^{14}C	5.22E-02	1.4E-04	7.1E-06
^{36}Cl	4.18E-02	4.6E-04	1.9E-05
^{129}I	2.21E-02	7.4E-02	1.6E-03
^{125}Sb	2.69E-02	8.4E-03	2.2E-04
^{79}Se	3.99E-02	7.6E-04	3.0E-05
^{126}Sn	3.16E-02	3.8E-01	1.2E-02
$^{3}H_2$	1.43E-01	2.8E-06	4.0E-07
^{99}Tc	3.56E-02	2.3E-03	8.0E-05

[1]From (Farfan, 2007).
[2] Dose to MEI at SRS Boundary = Peak Flux × Dose Release Factor.

Table 7. 100-meter Dose Release Factors and Dose to the MEI for the 100 – 10,100 Year Time Period per Ci of Radionuclide Remaining in the Combined FTF Waste Tanks

Radionuclide	Peak Flux (Ci/yr/Ci)	SRS 100 m Dose Release Factor[1] (mrem/Ci)	Dose to MEI at 100 m Boundary[2] (mrem/yr/Ci)
^{14}C	2.59E-04	2.8E-04	7.2E-08
^{36}Cl	6.07E-04	2.9E-02	1.7E-05
^{129}I	2.38E-03	2.0E+01	4.8E-02
^{125}Sb	3.71E-14	3.9E-01	1.4E-14
^{79}Se	7.02E-04	3.8E-02	2.7E-05
^{126}Sn	1.29E-03	1.8E+01	2.3E-02
$^{3}H_2$	3.12E-10	1.3E-02	4.2E-12
^{99}Tc	9.66E-04	1.1E-01	1.0E-04

[1]From (Farfan, 2007).
[2] Dose to MEI at SRS Boundary = Peak Flux × Dose Release Factor.

Table 8. 1600-meter Dose Release Factors and Dose to the MEI for the 100 – 10,100 Year Time Period per Ci of Radionuclide Remaining in the Combined FTF Waste Tanks

Radionuclide	Peak Flux[1] (Ci/yr/Ci)	SRS 1600 m Dose Release Factor[2] (mrem/Ci)	Dose to MEI at 1600 m Boundary[3] (mrem/yr/Ci)
^{14}C	2.59E-04	2.4E-03	6.2E-07
^{36}Cl	6.07E-04	6.2E-03	3.7E-06
^{129}I	2.38E-03	2.3E+00	5.5E-03
^{125}Sb	3.71E-14	9.7E-02	3.6E-15
^{79}Se	7.02E-04	9.1E-03	6.4E-06
^{126}Sn	1.29E-03	4.4E+00	5.7E-03
$^{3}H_2$	3.12E-10	4.9E-05	1.5E-14
^{99}Tc	9.66E-04	2.6E-02	2.6E-05

[1]Peak flux from 100 to 10,100 yrs.
[2]From (Farfan, 2007).
[3]Dose to MEI at SRS Boundary = Peak Flux × Dose Release Factor.

Table 9. Simulated Peak Instantaneous Rn-222 Flux over 10,100-Years at the Land Surface

	Peak Instantaneous Rn-222 flux at Land Surface ($pCi/m^2/sec$) / (Ci/m^2)	
Parent Source ($1 Ci/m^2$)	**0-100 years**	**100-10,100 years**
Pu-238	1.75E-11	2.70E-07
U-238	2.10E-11	9.29E-06
U-234	2.22E-07	7.68E-04
Th-230	4.77E-04	1.03E-02
Ra-226	1.12E-02	1.08E-02

Distribution

J. J. Mayer, 773-42A
W. E. Stevens, 773-A
E. L. Wilhite, 773-43A
M. A. Phifer, 773-42A
M. B. Birk, 766-H
J. L. Newman, 766-H
M. H. Layton, 766-H
K. H. Rosenberger, 766-H
T. C. Robinson, 766-H
STI (4), 703-43A
E&CPT Files 773-43A, Rm. 213