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Key Words: Carbon,
Resin, Radioactive Waste,
Intermediate Level Vault

Retention: Permanent

Special Analysis:
**Atmospheric dose resulting from the release of ^{14}C from
Reactor Moderator Deionizers in a disposal environment (U)**

Robert A. Hiergesell
Robert F. Swingle

August 18, 2005

Westinghouse Savannah River Company
Savannah River Site
Aiken, SC 29808

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



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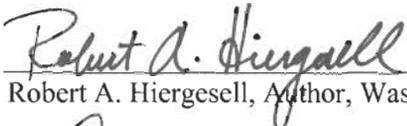
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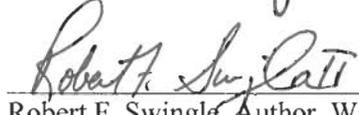
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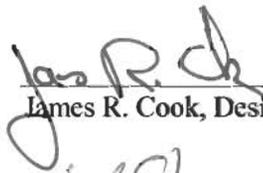
REVIEWS AND APPROVALS

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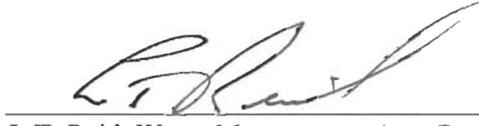

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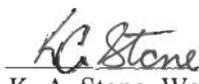

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LIST OF ACRONYMS

1-D	One dimensional
Ci	Curie
ILV	Intermediate Level Vault
LLWF	Low Level Waste Facility
m	Meter
mrem	millirem
MEI	Maximally Exposed Individual
PA	Performance Assessment
SA	Special Analysis
SRS	Savannah River Site
USDOE	U.S. Department of Energy

EXECUTIVE SUMMARY

The proposed action of disposing of 52 moderator deionizer vessels within the ILV was evaluated in this SA. In particular, a detailed analysis of the release of ^{14}C via the atmospheric pathway was conducted for these vessels since the major concern has been the nearly 20 Ci of ^{14}C that is associated with each vessel.

The more rigorous evaluation of the atmospheric pathway for ^{14}C included incorporation of new information about the chemical availability of ^{14}C when disposed in a grout/cement encapsulation environment, as will be the case in the ILV. This information was utilized to establish the source term for a 1-D numerical model to simulate the diffusion of $^{14}\text{CO}_2$ from the ILV Waste Zone to the land surface. The results indicate a peak surface emanation rate from the entire ILV of $1.42\text{E}-08$ Ci/yr with an associated dose of only $3.83\text{E}-05$ mrem/yr to the Maximally Exposed Individual (MEI) at 100m.

The fact that the atmospheric pathway exposure for ^{14}C is controlled by chemical solubility limits for ^{14}C between the solid waste, pore water and pore vapor within the disposal environment rather than the absolute inventory suggests that the establishment of specific facility limits is inappropriate. With the relaxation of the atmospheric pathway restriction, the groundwater pathway becomes the more restrictive in terms of disposing ^{14}C or $^{14}\text{C_KB}$ within the ILV. Since the resin-based ^{14}C of the 52 moderator deionizer vessels is highly similar to the $^{14}\text{C_KB}$ waste form, the inventory from the 52 deionizer vessels is compared against the groundwater limits for that waste form.

The small groundwater pathway fraction ($1.14\text{E}-05$) calculated for the proposed inventory of the 52 moderator deionizer vessels indicates that the proposed action will have an insignificant impact with respect to possible exposures via the groundwater pathway.

This investigation recommends that there be no ILV Atmospheric pathway limit for ^{14}C and $^{14}\text{C_KB}$. Further, in the absence of an Atmospheric pathway limit it was determined that there are no other applicable ILV limits (Groundwater or Intruder pathway) that would be impacted in any significant fashion should the waste package be disposed within the ILV. Thus, it is concluded that the disposal of 52 moderator deionizer vessels can easily be accommodated within the ILV.

INTRODUCTION

This Special Analysis evaluates the proposed permanent disposal of 52 Moderator Deionizers from use in the purification of the heavy water moderator of SRS reactors within the E-Area Intermediate Level Vaults (ILV). Each deionizer vessel contains resin which has been dedeuterized, dewatered and had absorbents added. Characterization of the waste material contained within each vessel is described in an SRS memorandum, Gibbs, 2005. A listing of the radionuclide inventory associated with an individual vessel is presented in the Appendix.

This report describes the analysis conducted to evaluate the potential magnitude of gaseous release of ^{14}C from the ILV over the 1,000-year performance assessment (PA) period of interest and the atmospheric pathway dose that this release might result in. The analysis considers the diffusion of ^{14}C upward from the ILV waste zone through the overlying closure cap material to determine emanation rates at the land surface. The peak emanation rate was then used to estimate the exposure of the Maximally Exposed Individual (MEI) at both the SRS boundary and at a location 100m downwind from the ILV. The exposure standard for the MEI is stated in USDOE Order 435.1 as 10 mrem/yr.

A 1-dimensional numerical model was employed to simulate a bounding (conservative) case to estimate the maximum plausible emanation rate of ^{14}C at the land surface. The ^{14}C source term for use in this model was developed in a separate investigation described in Kaplan, 2005, and the dose received by the MEI was determined using Dose Factors calculated for the ILV in Flach and Hiergesell, 2004.

ILV CLOSURE CONSIDERATIONS

The concepts for closure of the ILV are relevant to the determination of the ^{14}C flux at the land surface during the PA evaluation period (1,000 years). ILV construction specifics and closure concept are described in Phifer, 2004, *Preliminary E-Area Intermediate Level Vault Closure Cap Infiltration (U)* and are summarized below. The current IL Vault is a below grade, reinforced concrete vault. The bottom of the base slab is approximately 26 feet below existing grade and the top of exterior walls are approximately 5.5 feet above existing grade. The exterior vault wall footprint is approximately 279 feet long by 48 feet wide. It contains seven 25-foot by 44.5-foot by 28.5-foot deep bulk waste cells and two 25-foot by 44.5-foot by 26-foot deep cells (interior dimensions). One of the 26-foot deep cells contains 140 vertical silos. The silos are approximately 30 inches in diameter in the top 3 feet, 20 inches in diameter for the remainder, and 24.83 feet deep (interior dimensions). The other 26-foot deep cell is a bulk waste cell. There is currently one IL Vault in the E-Area LLWF.

Specific information with regard to the construction of the final closure cap is presented below. The final closure cap will exist far into the future and is the configuration that must be considered in evaluating the long-term ^{14}C release at the land surface.

Closure Configuration

During the operational period, the ILV will be filled with equipment and containerized waste placed in the cell in layers. Each layer of waste will be encapsulated in grout, which forms the surface for the placement of the next layer of waste. In this fashion, the full 27.3-foot (8.3 m)

depth of an ILV cell will be filled in, with the upper 17 inches (0.43 m) being filled in with grout (Phifer, 2004). It is anticipated that the 52 Moderator Deionizers would be placed within one or more layers within the ILV and individually surrounded with grout material.

As the individual cells of the ILV are filled they will be grouted shut with the upper 17-inch grout layer. A temporary roof will be maintained over each completed cell until the end of the 25-year operations period. At that point the concrete roof will be constructed and maintained over the 100-year institutional control period. At the end of institutional control, a permanent closure cap will be constructed to cover the entire ILV (Phifer, 2004).

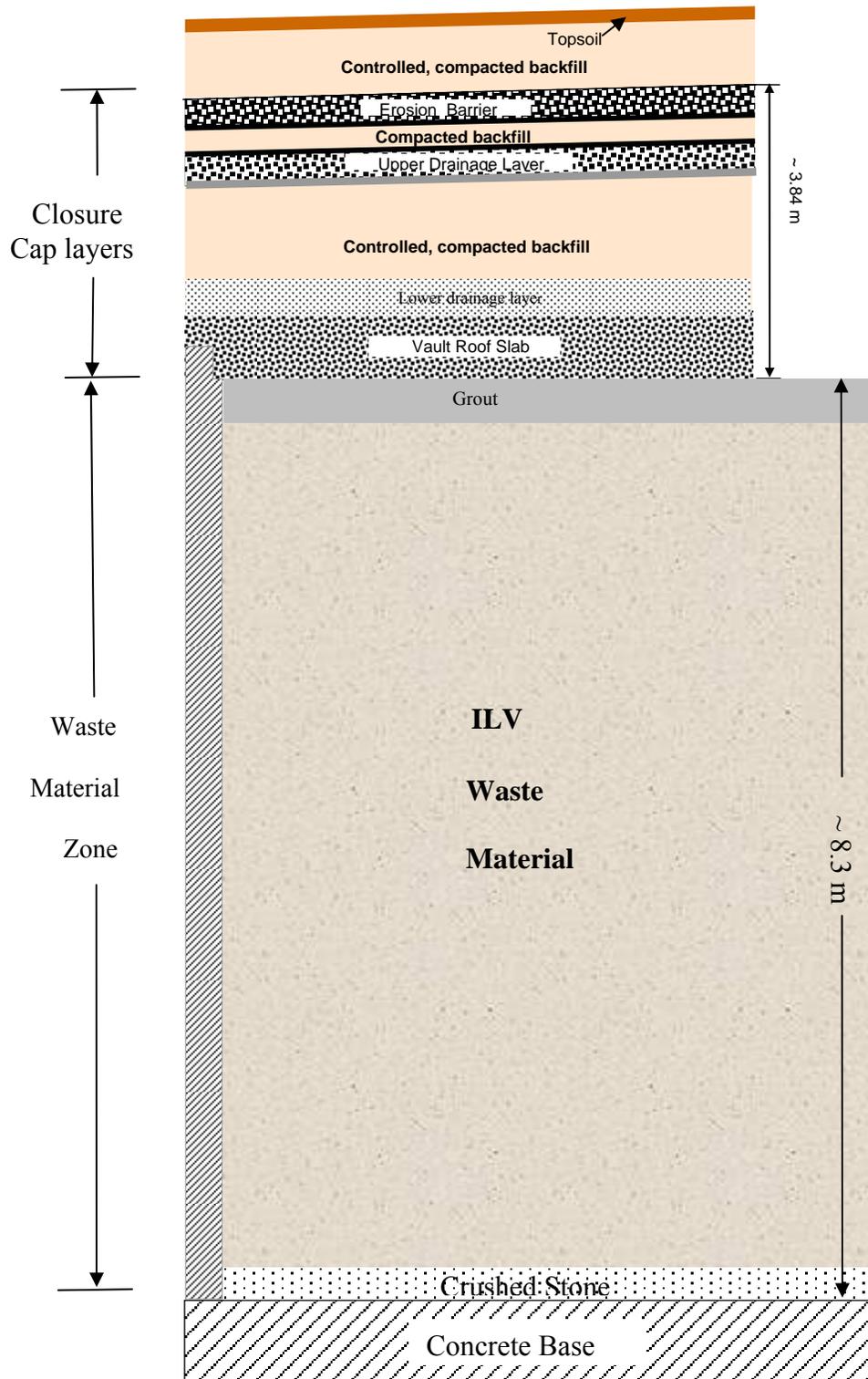
Table 1, below, describes the individual components of the closure cap. Materials are indicated with the associated thickness of each component, in inches, cm and m.

Table 1. Vertical layer sequence and associated thickness for ILV cover material.

(Adapted from Phifer, 2003 and Phifer, 2004)

Layer	Thickness (inches)	Thickness (cm)	Thickness (m)
Topsoil	6	15.2	0.15
Upper Backfill	30	76.2	0.76
Erosion Barrier	12	30.5	0.30
Middle Backfill	12	30.5	0.30
Upper Drainage Layer	12	30.5	0.30
Lower Backfill	58.65	149	1.49
Lower Drainage Layer	24	61	0.61
ILV Concrete Roof	33 (avg.)	83.8	0.84
Upper grout layer	17	43.2	0.43
Vault Waste Zone	~311	~790	~7.90

The closure cap components of concern for the long-term ¹⁴C performance calculation are those that will persist over the 1,000-year PA evaluation period. These components are situated below the top of the Erosion Barrier. A conceptual diagram of the ILV and its overlying closure cap are shown in Figure 1.



(diagram adapted from Phifer, 2004 and Phifer, 2003)

Figure 1. Conceptual illustration of the Intermediate Level Vault and its closure cap

MODEL DEVELOPMENT

Conceptual Model

The ^{14}C flux at the land surface above an ILV was evaluated for its specific closure configuration. The primary mechanism for release of ^{14}C at the land surface is diffusion within the vapor filled pore space of the ILV and overlying cover materials. While there may be other sources of ^{14}C within the ILV, the ^{14}C source evaluated in this investigation is that specifically generated from the placement of the 52 Moderator Deionizers within the ILV.

The release of ^{14}C from the deionizer resins is largely controlled by the chemical processes that drive the partitioning of C between the solid, liquid and gaseous phases at the pore scale under the elevated pH conditions of the ILV waste zone. In the vapor phase C would exist as CO_2 , however the concentration of CO_2 in the pore vapor phase is calculated to be very low under the elevated pH conditions within the ILV disposal environment (Kaplan, 2005). Using quite conservative assumptions regarding flow and concrete physical integrity, it was estimated in Kaplan and Myers (2001) that the vaults in SRS's E-Area would maintain a pH of >11 for more than 11,000 years. As reported in Kaplan, 2005, the concentration of $^{14}\text{CO}_2$ within vapor-filled pore space of the ILV is calculated to be $1.9 \times 10^{-7} \text{ Ci/m}^3$, which is expected to remain constant over the Performance Assessment (PA) evaluation period.

The chemical form of ^{14}C within the waste zone is important in evaluating its vapor diffusion transport to the land surface since the diffusion coefficients are calculated based on the molecular weight of the compound it exists as. In the vapor phase, $^{14}\text{CO}_2$ is the molecule that ^{14}C exists within and by which it diffuses outward from the waste emplacement zone.

While it is expected that the materials through which the ^{14}C will diffuse will largely maintain their character as porous media over the PA evaluation period, there is a possibility of large-scale cracks developing in the ILV due to differential settlement and seismic activity. Such cracks are likely to be vapor-filled, thus forming conduits that could enhance the upward diffusion of CO_2 toward the base of the closure cap.

Numerical model

The mathematical model utilized in this report was developed using the PORFLOW™ simulation package (ACRI, 2004). PC-based PORFLOW™ Version 5.97.0 was used to conduct a series of simulations. PORFLOW™ is developed and marketed by Analytic & Computational Research, Inc. to solve problems involving transient and steady-state fluid flow, heat and mass transport in multi-phase, variably saturated, porous or fractured media with dynamic phase change. PORFLOW™ has been widely used at the SRS and in the DOE complex to address major issues related to the ground water and nuclear waste management.

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

$$\frac{\partial C_k}{\partial t} + \frac{\partial}{\partial x_i} (V_i C_k) = \frac{\partial}{\partial x_i} (D_{ij} \frac{\partial C_k}{\partial x_j}) + \gamma_k \quad (1)$$

Where

C_k concentration of species k (M/L^3)

V_i	fluid velocity in the i^{th} direction (L/t)
D_{ij}	effective diffusion coefficient for the species (L^2/t)
γ_k	net decay of species k (M/t)
i, j	direction index (L)
t	time
x	distance coordinate (L)

This equation is solved within PORFLOW to evaluate transient radionuclide transport through the closure cap materials above ILV and to determine gaseous radionuclide flux at the land surface over time.

Model Assumptions

The numerical representation of the conceptual model is as a 1-dimensional vertical stack of elements configured to correspond to the vertical dimensions of the ILV and overlying cover material associated with final closure design. The conditions and assumptions of this model included the following:

The only radionuclide evaluated in this investigation is ^{14}C , which exists as part of the CO_2 molecule within the model domain. Since CO_2 exists as a gas in the vapor-filled pores and cracks, air was taken to be the fluid within which transport occurs. Air-diffusion was the only transport mechanism simulated in the model and advective air-transport was assumed to be negligible. A small percentage of CO_2 dissolves in residual pore water but since diffusion proceeds more slowly in that fluid, and because advective water movement carries dissolved CO_2 downward, air-diffusion is the only transport process by which CO_2 can reach the land surface from the ILV waste zone. Transport was allowed to proceed only through air-filled pore space and, therefore, residual pore water was treated as if it was part of the solid matrix material within the flow field.

The boundary conditions imposed on the model domain included:

- No-flux specified for $^{14}\text{CO}_2$ along sides and bottom
($dC/dX = 0$ at $x=0, x=1$ and $dC/dY = 0$ at $y=0$)
- $^{14}\text{CO}_2$ concentration set to 0 at land surface (top of erosion barrier)
($C = 0$ at $y=12.18$)

These boundary conditions force all of the gaseous $^{14}\text{CO}_2$ 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. Simulations were conducted in transient mode for diffusive transport in air, with results being obtained over 10,000 years.

A summary of the relevant information for ^{14}C and CO_2 are summarized in Table 2.

Table 2. Relevant information for ^{14}C and CO_2

Radionuclide	Half-life	Atomic wt.	Molecular form in gaseous state	Molecular Wt.
	(yrs)			
^{14}C	5.73E+03	14	CO_2	46.00

Grid Construction

The model grid was constructed as a node mesh 3 nodes wide by 36 nodes high. This mesh defines a vertical stack of model elements where each element is a block with a square base, 1m x 1m but having heights that varied to allow alignment of element faces exactly with the interfaces of material layers within the ILV and cover materials. The grid extends upward only as far as the closure cap erosion barrier, anticipating that this is the cover thickness that will prevail over the majority of the PA compliance period. A set of consistent units was employed in the simulations for length, mass and time, these being meters, grams and years, respectively.

Material Zones

The model domain was divided into two primary zones, the ILV waste zone occupying the lower 25.9 ft. (7.9 m) of the domain and the cover zone, extending ~14.1 ft. (~4.3 m) above the waste zone to the top of the domain. The cover zone includes the upper grout layer and vault concrete roof as well as the different backfill and drain layers of the closure cap.

Material Properties

Material properties utilized within the 1-D numerical model were specified for 7 material zones defined within the model domain. Each material zone was assigned values for total porosity, relative saturation, air-filled porosity, matrix density, air density, and an effective air-diffusion coefficient for each source element or compound. Tortuosity was assigned a unit value since its effect is incorporated with the use of an “effective” diffusion coefficient. The matrix density was selected based on the density of quartz, and is regarded to be representative of most SRS soils. The term “relative saturation” is used in this report to describe that fraction of the interconnected pore space that is filled with water.

Values for total porosity and long-term relative saturation for the ILV waste zone and concrete were obtained from vadose zone 2-D simulations conducted to evaluate the groundwater pathway as a part of a previous Special Analysis (Flach and Hiergesell, 2004). The ILV waste zone and concrete porosities were established at 0.42 and 0.18, respectively in that analysis. The steady-state relative saturations for the waste zone and concrete in this analysis ranged from greater than 0.95 for time periods less than 1075 years to ~0.63 for time periods greater than 1075 years.

For the purposes of evaluating the impact of a large-scale, through-penetrating, cracks in the ILV vault on the diffusion of $^{14}\text{CO}_2$ from the waste zone to the land surface, the porosity and relative saturation thought to be representative of a crack were assigned to the entire waste zone and cement roof., these being 0.9 and 0.1, respectively. These values were arbitrarily selected to represent a crack that has a variable aperture and which allows some moisture content to remain where crack walls are nearly touching one another. While assuming the entire waste zone has the air-filled porosity of a crack is not a physically valid representation, it is a conservative approach that sets a bounding case to evaluate the presence of air-filled cracks.

Values for total porosity and long-term relative saturation of the closure cap materials were selected based on a series of HELP model simulations conducted as part of the investigation summarized in Phifer and Nelson, 2003. HELP model analyses were conducted at different points in time, these being at 100, 300, 550, 1000, 1800, 3400, 5600 and 10,000 years in the future. Porosities of the different materials changed slightly through time as a result of leaching, and

relative saturations varied as porosities and expected recharge rates varied. Porosities changed less conspicuously than relative saturations, and by plotting relative saturation versus time representative values could then be selected. Air-filled porosity was estimated by subtracting the moisture content from the total porosity. While the HELP model results were used to guide the selection of long-term relative saturation values for all of the closure cap materials, even more conservative values were selected for the erosion barrier and lower backfill. Values representative of native soil were selected for these layers, these being 0.63 and 0.61, for the erosion barrier and lower backfill, respectively.

A summary of the values of porosity, long-term relative saturation and air-filled porosity used in the simulations is listed for each material type in Table 3. A value for the density of air was obtained for use in the numerical simulations from Bolz and Tuve, 1973.

Table 3. Porosity, relative saturation and air-filled porosity values

Layer Material	Representative Porosity	Long-term Relative Saturation	Air-Filled Porosity
Erosion Barrier	0.07	0.63	2.60E-02
Middle Backfill	0.38	0.63	1.39E-01
Upper Drainage	0.38	0.58	1.58E-01
Lower Backfill	0.37	0.61	1.44E-01
Lower Drainage	0.31	0.5	1.55E-01
Concrete Roof and Grout*	0.9	0.1	8.1E-01
ILV Waste Zone*	0.9	0.1	8.1E-01

* porosity and relative saturation of a crack were conservatively assigned for these materials.

The molecular diffusion coefficient for CO₂ was based on the effective (or “complete”) air diffusion coefficient of ²²²Rn, as reported in Rogers and Nielson, 1991. That study utilized the following relationship to determine effective ²²²Rn air-diffusion coefficients based on the material porosity and the degree of water saturation:

$$D_c = D_{op} p \exp(-6Sp - 6S14p) \tag{2}$$

Where
 D_c = the “complete”, or effective diffusion coefficient
 D_{op} = open air diffusion coefficient of ²²²Rn = 1.1 E-05 m²/sec
 S = relative saturation
 p = porosity

Using the effective radon air-diffusion coefficient calculated for each material zone in the model under the prevailing porosity and relative saturation conditions as the reference, the effective diffusion coefficients for CO₂ were then calculated by applying the following relationship, which is derived from Graham’s Law:

$$D = D' \sqrt{\frac{MWT'}{MWT}} \tag{3}$$

Where: D = the effective air diffusion coefficient for CO₂ (m²/yr)

D' = the effective air diffusion coefficient of ^{222}Rn (m^2/yr)

MWT' = the atomic weight of ^{222}Rn

MWT = the molecular weight of $^{14}\text{CO}_2$.

A summary of the $^{14}\text{CO}_2$ effective diffusion coefficients calculated for each material at the selected relative saturations are presented below in Table 4

Table 4. Effective CO_2 diffusion coefficients for specific material zones

Layer Material description	Layer Number	$D_c^{\text{Rn-222}}$ (m^2/yr)	$D_c^{\text{CO}_2}$ (m^2/yr)
Erosion barrier	7	4.11E-01	9.03E-01
Middle backfill	6	1.86E+01	4.08E+01
Upper drainage layer	5	2.50E+01	5.50E+01
Lower backfill	4	2.09E+01	4.58E+01
Lower drainage layer	3	3.16E+01	6.94E+01
Concrete Roof and Grout	2	1.82E+02	4.0E+02
ILV Waste Zone	1	1.82E+02	4.0E+02

Source Term

The inventory of ^{14}C introduced by a single moderator deionizer vessel is specified in Gibbs, 2005 and it is indicated that there will be "50+" such vessels to dispose within the ILV. It is therefore assumed that 52 moderator deionizer vessels provide a bounding inventory for evaluation in this SA. Furthermore, the inventory introduced by 52 deionizer vessels is within the bounding case evaluated in Kaplan, 2005.

The objective of the Kaplan, 2005 study was to estimate the concentration of gaseous ^{14}C in the waste pore space that is in contact with the resin leachate. That study indicates that as the resin ages, and equilibrates with slowly infiltrating water, it is expected that the ^{14}C will partition to the solid, liquid, and gaseous phases. The approach used to estimate this value was built largely around data generated from lysimeter studies that were conducted for 9 years. These lysimeters contained the same type of used resins (mixed-bed deionizer resins used in the purification of the heavy water moderator of SRS reactors) as are proposed for disposed in the ILV. During the 9-year period, pore water ^{14}C leaching concentrations were monitored to provide an excellent estimate of the long-term behavior of ^{14}C release rates from the resins.

Thermodynamic calculations were then performed to calculate $^{14}\text{CO}_2$ concentrations. These calculations included the ^{14}C pore water data from the lysimeter study, and data from a field study of a natural analogue to a long-term cementitious environment (Khoury et al. 1992). The calculations predicted an extremely low $^{14}\text{CO}_2$ concentration of $1.9\text{E-}07$ Ci/ m^3 in the air spaces above the resin leachate in the grout environment. This low concentration is not surprising in light of both laboratory and field observations that concrete acts as a strong sorbent of CO_2 . It is reported in Kaplan, 2005 that certain industrial entities dispose of large quantities of ^{14}C bearing resin (orders of magnitude greater than what is proposed in this SA) in conjunction with concrete slabs. These slabs are referred to as " ^{14}C getters".

Mathematically, the initial concentration, C_i , is expressed as $C_i = 1.9E-07 \text{ Ci/m}^3$ within model elements 1, 5 and 9. Within PORFLOW, the FIX keyword command was invoked to maintain air-filled pore concentration in these elements at $1.9E-07 \text{ Ci/m}^3$. Due to the boundary conditions imposed on this model and by maintaining $C_i = 1.9E-07 \text{ Ci/m}^3$, the simulation results are not sensitive to the exact placement location of the sources within the waste zone.

Measures Incorporated to Assure Conservative Results

In this analysis, several conditions incorporated that introduce a significant measure of conservatism into the calculations. These include:

- No re-equilibration of vapor phase CO_2 with pore water outside of source zone and thus not taking credit for the removal of $^{14}\text{CO}_2$ by its dissolving in pore water moving vertically downward through the flow field. This mechanism would likely carry off some dissolved $^{14}\text{CO}_2$, and therefore its omission causes an over-prediction of the instantaneous ^{14}C flux at the land surface in this investigation.
- The assignment of physical properties of an air-filled crack to the entire waste disposal zone to address the possibility that air-filled cracks might develop in concrete over time and act as conduits for $^{14}\text{CO}_2$ diffusion. This treatment provides more air-filled space in the waste zone that equilibrates to the equilibrium air concentration of ^{14}C , a larger cross-sectional area for diffusion to occur over and requires that the effective diffusion coefficient used in the model be very close to that for diffusion in open air. Incorporating these conditions leads to over-predicting the ^{14}C flux rate at the land surface.
- From the range of properties forecast to occur for each material type within the simulation domain over the period of interest, the combination of porosities and long-term relative saturations that yield the highest diffusive flux of ^{14}C over the PA period of compliance (higher porosity, lower relative saturation) was assumed to exist over the full period of simulation for each material.

RESULTS

Simulation Results

The results of the 1-D numerical simulation is in terms of an emanation rate of ¹⁴C activity at the land surface and is illustrated Figure 2.

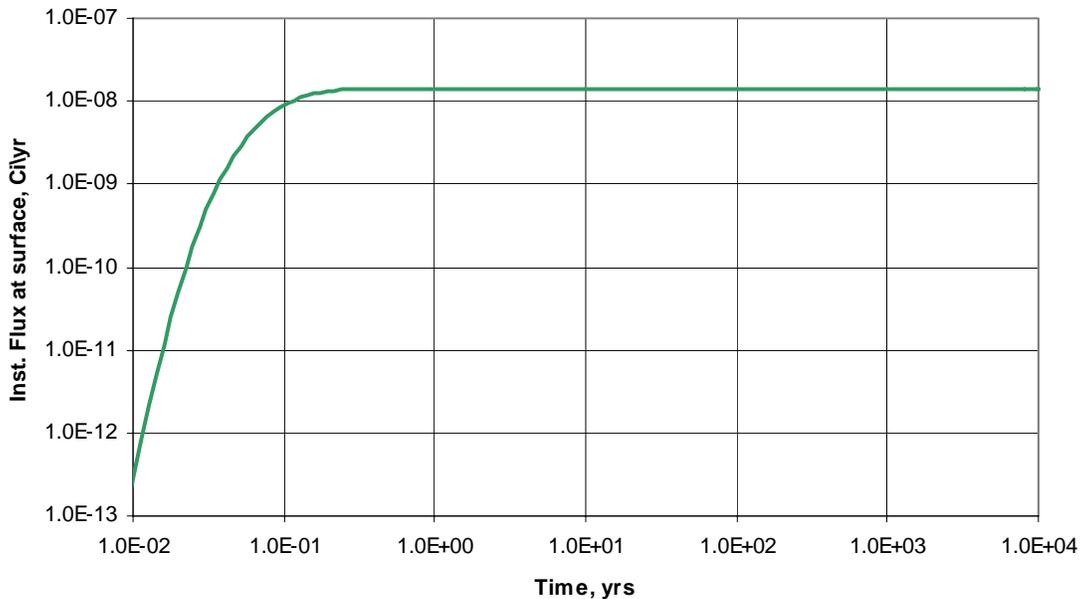


Figure 2. Simulated rate of ¹⁴C emanation at the land surface (Ci/yr).

The surface emanation rate is in Ci/year and is indicated for a 10,000-year period although the PA performance period is only 1,000 years. There is a rapid buildup to the maximum emanation rate of 1.42E-8 Ci/year within 0.4 years. This rate continues steadily for the remainder of the period of simulation. This feature is explained by the fact that the waste zone vapor phase (air-filled pore space) retains a constant activity level due to the solubility constraints of CO₂ in pore water and the equilibrium constant between the water and vapor phases. As ¹⁴CO₂ diffuses out of the waste zone, toward the land surface, ¹⁴C activity is replenished by the source of the ¹⁴C within the Moderator Vessels. Specific information on how the steady-state vapor phase concentration for ¹⁴CO₂ was calculated is documented in Kaplan, 2005.

Conversion of Model Results to Determine ILV Atmospheric Flux

The simulation results represent the ¹⁴C flux from 1 m² at the land surface above the ILV facility. If some of the Moderator Vessels are assumed to be disposed in each of the ILV cells, then diffusion of ¹⁴CO₂ will proceed over the entire footprint of the ILV facility. The dimensions of this footprint are 75m x 15m, or 1125 m².

The peak emanation rate from entire ILV is calculated to be:

$$1.42 \times 10^{-8} \text{ Ci/yr per m}^2 \times 1125 \text{ m}^2 = 1.6 \times 10^{-5} \text{ Ci/year} \quad (4)$$

To put this into perspective, the total ^{14}C flux at the land surface above the ILV ($1.6\text{E-}2$ Ci) over the full 1,000-year PA compliance period represents a fraction of only $1.54\text{E-}5$ of the proposed initial inventory of ^{14}C in the 52 moderator deionizer vessels.

Atmospheric Pathway Dose Calculations

Exposure of the Maximally Exposed Individual (MEI) in the PA process is calculated before and after the loss of institutional control, which is assumed to occur 100 years after closure of the ILV. Prior to the loss of institutional control, exposure is determined at the SRS boundary. Following the loss of institutional control the exposure point is at a location 100m from the ILV.

To accommodate the different exposure points, the maximum ^{14}C flux at the land surface is needed prior to, and after, 100 years. Dose calculations can then be made for the MEI. The peak flux in this investigation is the same for both time periods, 1.6×10^{-5} Ci/yr.

Dose conversion factors have been determined for each of the two exposure points for ^{14}C disposed within the ILV and are presented in Flach and Hiergesell, 2004. These values, along with the calculated dose the MEI at those points are presented in Table 5.

Table 5. Atmospheric pathway dose calculations

	SRS Boundary Dose Conversion Factor	Dose to MEI at SRS Boundary (from all vessels)	100-meter Dose Conversion Factor	Dose to MEI at 100-meters (from all vessels)
	(mrem/Ci)	(mrem/yr)	(mrem/Ci)	(mrem/yr)
^{14}C	1.1E-03	1.76E-08	2.4	3.83E-05

The dose to the MEI at the 100m exposure point is $3.83\text{E-}05$ mrem/yr compared to $1.76\text{E-}08$ mrem/yr at the SRS boundary. The higher value at the close-in location is not surprising considering that the surface emanation rate of ^{14}C is steady after 0.4 years and therefore the only difference in doses results from the degree of air-dispersion over different distances.

In this analysis, all ^{14}C is assumed to be associated with either anionic resins or an inorganic fraction that will eventually evolve to CO_2 gas. Because negligible amounts of cellulose degradation products (*e.g.*, paper, wood, cardboard) were placed in the ILV, no attempt to model their influence on ^{14}C presence was deemed necessary. Powell, et. al. (2002) reported extremely slow degradation rates of anionic resins, such that only 4% was estimated to be degraded in 10,000 years.

Kaplan (2005) indicates that irrespective of the amount of ^{14}C placed in the inventory, the same gaseous ^{14}C concentration occurs in adjacent air space. An explanation for this is depicted in Figure 3. In this schematic, it shows a sealed container with a ^{14}C -solid source material surrounded by water and an air space above the water. Equilibrium is established between the solid and the water, and between the water and air. The equilibrium between the solid and air in a cementitious environment is solubility controlled. In practical terms this means that the aqueous ^{14}C concentration slowly increases with increases in solid phase ^{14}C waste until the solubility

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limit is reached, beyond which the aqueous ^{14}C concentration does not increase any more. Kaplan (2005) provided the maximum aqueous ^{14}C value and associated maximum vapor phase ^{14}C values that were used in this SA. Therefore, the aqueous and gaseous concentration of ^{14}C will never exceed those used in this SA, irrespective of inventory disposed in the ILV because the limit is based on solubility constraints. It is therefore proposed that there be no atmospheric ^{14}C limit for the ILV.

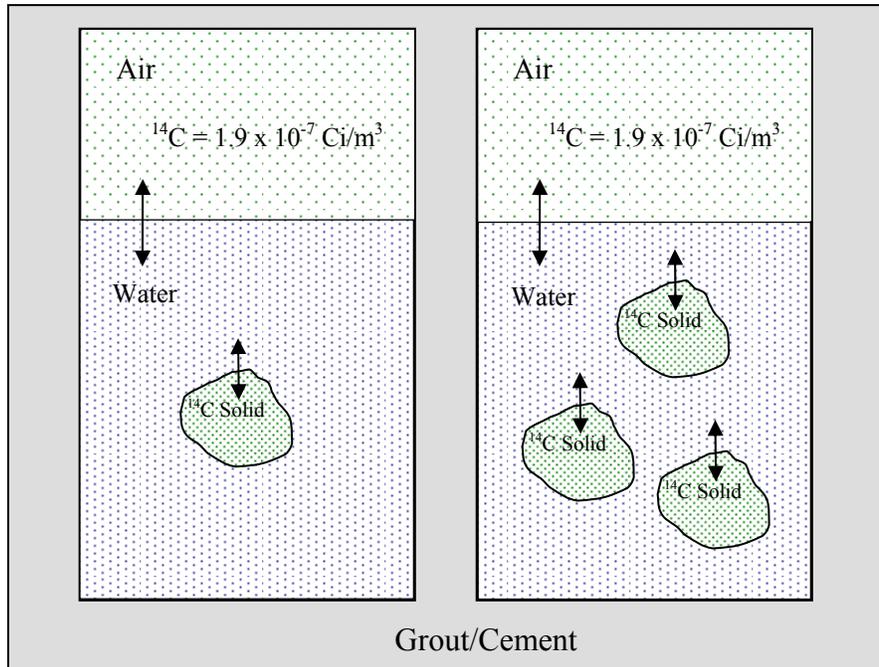


Figure 3. Relationship Between ^{14}C Solid Source and ^{14}C Air Concentration in a Grout/Cement Environment

Groundwater Pathway

The current ILV groundwater pathway limits for ^{14}C waste forms were established in a SA, documented in Flach and Hiergesell, 2004. The calculated groundwater pathway limits were of little consequence, however, since they were orders of magnitude higher than the atmospheric pathway limit presented in that same investigation. Due to the miniscule dose calculated to be received via the atmospheric pathway in this investigation, the groundwater pathway limits now become more restrictive.

There are currently two sets of groundwater pathway disposal limits for ^{14}C : one for generic ^{14}C and one for ^{14}C from K and L Basin Resin (which is similar to the resins contained in the deionizer vessels evaluated in this SA). As described in Flach and Hiergesell (2004), the basis for the distinction was in the use of different K_d values which were applied to the entire waste zone. The waste zone K_d used for derivation of the generic limit (5000 ml/g) was based on the behavior of ^{14}C in grout while the waste zone K_d used to determine the ^{14}C from K and L Basin resin (140 ml/g) was based on the behavior of ^{14}C in deionizer resin.

The GW2 limits for generic ^{14}C and $^{14}\text{C_KB}$ were determined to be $9.1\text{E}+07$ Ci and $2.6\text{E}+06$ Ci, respectively. The proposed inventory of resin-based ^{14}C from 52 moderator deionizer vessels ($1.04\text{E}+03$ Ci) is most appropriately compared to the $^{14}\text{C_KB}$ waste form limits due to the high degree of similarity between them. As such, the inventory represents a very small fraction of the ILV GW2 limit for $^{14}\text{C_KB}$, this being $1.14\text{E}-05$. Thus, for the 1,000-year PA period of compliance the inventory from 52 moderator deionizer vessels can easily be accommodated within the ILV.

Intruder Pathway

The radionuclide inventory for 52 deionizer vessels was compared against the current ILV Intruder Pathway limits, as determined in Flach and Hiergesell, 2004. That report indicates that a single form of ^{14}C was evaluated in the Intruder pathway screening and that the subsequent analysis calculated a limit $> 1.0\text{E}+20$. It is assumed that this applies equally to the $^{14}\text{C_KB}$ waste form, although Table E-1. in Flach and Hiergesell, 2004 indicates a blank entry for $^{14}\text{C_KB}$, suggesting the screening threshold for that waste form was not exceeded. No other radionuclide listed in the proposed inventory for 52 deionizer vessels contributes a fraction greater than $1.0\text{E}-4$, with virtually all of them many orders of magnitude less than that. It is concluded that the proposed disposal of 52 deionizer vessels has an insignificant impact with respect to the Intruder Pathway.

CONCLUSIONS

The proposed action of disposing of 52 moderator deionizer vessels within the ILV was evaluated in this SA. In particular, a detailed analysis of the release of ^{14}C via the atmospheric pathway was conducted for these vessels since the major concern has been the nearly 20 Ci of ^{14}C that is associated with each vessel.

The more rigorous evaluation of the atmospheric pathway for ^{14}C included incorporation of new information about the chemical availability of ^{14}C when disposed in a grout/cement encapsulation environment, as will be the case in the ILV. This information was utilized to establish the source term for a 1-D numerical model to simulate the diffusion of $^{14}\text{CO}_2$ from the ILV Waste Zone to the land surface. The results indicate a peak surface emanation rate from the entire ILV of $1.42\text{E}-08$ Ci/yr with an associated dose of only $3.83\text{E}-05$ mrem/yr to the MEI at 100m.

The fact that the atmospheric pathway exposure for ^{14}C is controlled by chemical solubility limits for ^{14}C between the solid waste, pore water and pore vapor within the disposal environment rather than the absolute inventory suggests that the establishment of specific facility limits is inappropriate. Since the atmospheric exposure to the MEI from ^{14}C will never exceed $3.83\text{E}-05$ mrem/yr as long as the high pH grout/cement environment persists, this study proposes that no atmospheric pathway limit be established for the ILV for either ^{14}C or $^{14}\text{C_KB}$. With the relaxation of the atmospheric pathway restriction, the groundwater pathway becomes the more restrictive in terms of disposing ^{14}C or $^{14}\text{C_KB}$ within the ILV. Since the resin-based ^{14}C of the 52 moderator deionizer vessels is highly similar to the $^{14}\text{C_KB}$ waste form, the inventory from the 52 deionizer vessels is compared against the groundwater limits for that waste form.

The status of ILV disposal limits for ^{14}C and $^{14}\text{C_KB}$ are presented below in Table 6 as is the GW2 fraction for $^{14}\text{C_KB}$.

Table 6. Limits for ¹⁴C in the ILV

Nuclide	52 Vessel Inventory (Ci)	Resident Limit (Ci)	Air Limit (Ci)	GW1 Limit (0-100 yrs) (Ci)	GW2 Limit (100-1350 yrs) (Ci)	GW2 Fraction
¹⁴ C	--	≥1E+20	--	≥1E+20	9.1E+07	
¹⁴ C_KB	1040	≥1E+20	--	≥1E+20	2.6E+06	1.14E-05

The small groundwater pathway fraction (1.14E-05) calculated for the proposed inventory of the 52 moderator deionizer vessels indicates that the proposed action will have an insignificant impact with respect to possible exposures via the groundwater pathway.

To provide a measure of screening, the inventory of all other radionuclides associated with 52 deionizer vessels was compared to the applicable ILV disposal limits. With respect to the intruder pathway, no radionuclide has a fraction greater than 1.E-04 hence there is no significant impact. With respect to the groundwater pathway limits, the largest fraction of any radionuclide in the inventory is that calculated for resin-based ¹⁴C, and its fraction has been shown to be insignificant.

In conclusion, this investigation recommends that there be no ILV Atmospheric pathway limit for ¹⁴C and ¹⁴C_KB. Further, in the absence of an Atmospheric pathway limit it was determined that there are no other applicable ILV limits (Groundwater or Intruder pathway) that would be impacted in any significant fashion should the waste package be disposed within the ILV. Thus, it is concluded that the disposal of 52 moderator deionizer vessels can easily be accommodated within the ILV.

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APPENDIX

Radionuclide Inventory
(from Gibbs, 2005)

Isotope	MK22, Ci/deionizer @ 20 yrs	MK31, Ci/resin, t=20 yr	MK16,Ci/moderator resin@t=20	Average MK1631	Aver MK1631 + MK22	Inventory X 52
C-14	20	2.00E+01	2.00E+01	20	2.00E+01	1.04E+03
H-3	7.85E-04	8.21E-05	3.97E-04	2.40E-04	5.12E-04	2.66E-02
Co-60	1.12E-03	1.52E-03	2.24E-04	8.72E-04	9.96E-04	5.18E-02
Se-79	8.39E-06	8.15E-06	1.01E-05	9.13E-06	8.76E-06	4.56E-04
Rb-87	5.42E-10	4.33E-10	6.82E-10	5.58E-10	5.50E-10	2.86E-08
Sr-90	1.26E+00	1.00E+00	1.06E+00	1.03E+00	1.15E+00	5.97E+01
Y-90	1.28E+00	1.02E+00	1.08E+00	1.05E+00	1.17E+00	6.06E+01
Zr-93	8.35E-05	3.64E-05	6.78E-05	5.21E-05	6.78E-05	3.53E-03
Tc-99	2.88E-04	2.87E-04	3.61E-04	3.24E-04	3.06E-04	1.59E-02
Ru-106	4.37E-03	1.79E-05	5.96E-11	8.96E-06	2.19E-03	1.14E-01
Pd-107	3.46E-09	1.68E-06	4.38E-07	1.06E-06	5.30E-07	2.76E-05
Sn-121m	9.30E-07	1.72E-06	9.24E-07	1.32E-06	1.13E-06	5.86E-05
Sn-126	7.74E-06	1.44E-05	9.53E-06	1.20E-05	9.87E-06	5.13E-04
Sb-125	9.02E-04	2.28E-03	1.63E-05	1.15E-03	1.02E-03	5.33E-02
I-129	4.70E-07	6.31E-07	5.84E-07	6.08E-07	5.39E-07	2.80E-05
Cs-134	1.89E-03	2.64E-04	7.57E-06	1.36E-04	1.01E-03	5.27E-02
Cs-135	3.75E-06	2.34E-06	4.08E-06	3.21E-06	3.48E-06	1.81E-04
Cs-137	1.33E+00	1.33E+00	1.13E+00	1.23E+00	1.28E+00	6.64E+01
Ce-144	1.05E-06	1.13E-06	4.23E-13	5.65E-07	8.09E-07	4.21E-05
Pm-147	2.36E-02	4.06E-02	3.59E-04	2.05E-02	2.20E-02	1.15E+00
Sm-147	1.23E-10	1.99E-10	1.60E-10	1.79E-10	1.51E-10	7.86E-09
Sm-151	7.51E-03	2.63E-02	7.52E-03	1.69E-02	1.22E-02	6.34E-01
Eu-152	7.47E-05	1.88E-04	3.87E-05	1.14E-04	9.41E-05	4.89E-03
Eu-154	1.47E-02	3.42E-03	4.53E-03	3.98E-03	9.34E-03	4.86E-01
Eu-155	2.71E-03	6.29E-03	2.65E-04	3.28E-03	2.99E-03	1.56E-01
Ra-224	3.74E-07	3.37E-08	8.40E-07	4.37E-07	4.05E-07	2.11E-05
Th-228	3.69E-07	3.34E-08	8.33E-07	4.33E-07	4.01E-07	2.09E-05
U-232	3.72E-07	3.25E-08	7.03E-07	3.68E-07	3.70E-07	1.92E-05
U-234	3.08E-07	3.84E-22	4.68E-07	2.34E-07	2.71E-07	1.41E-05
U-235	3.92E-09	1.09E-09	4.35E-09	2.72E-09	3.32E-09	1.73E-07
U-236	7.18E-08	1.13E-09	1.30E-07	6.53E-08	6.86E-08	3.56E-06
U-238	1.47E-10	1.02E-07	2.64E-10	5.11E-08	2.56E-08	1.33E-06
Np-237	6.56E-07	3.35E-08	9.75E-07	5.04E-07	5.80E-07	3.02E-05
Pu-236						
Pu-238	5.46E-02	6.54E-06	6.71E-02	3.35E-02	4.41E-02	2.29E+00
Pu-239	5.09E-04	2.21E-04	6.97E-04	4.59E-04	4.84E-04	2.52E-02
Pu-240	3.36E-04	4.93E-05	5.11E-02	2.56E-02	1.30E-02	6.74E-01
Pu-241						
Pu-242	5.04E-07	3.73E-09	7.12E-07	3.58E-07	4.31E-07	2.24E-05
Am-241	1.55E-03	9.99E-04	1.97E-03	1.48E-03	1.51E-03	7.87E-02
Am-242m	8.21E-07	1.17E-06	9.14E-07	1.04E-06	9.30E-07	4.84E-05
Am-243	9.12E-09	2.90E-09	1.19E-06	5.98E-07	3.04E-07	1.58E-05
Cm-242						
Cm-243	3.29E-07	5.04E-08	2.68E-07	1.59E-07	2.44E-07	1.27E-05
Cm-244						

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