Air Pathway Modeling of the E-Area Low-Level Waste Facility at the Savannah River Site - 12253

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ABSTRACT

This investigation was initiated to address a concern expressed by the Department of Energy's Low Level Waste Disposal Facility Federal Review Group (LFRG) Review Team during their review of the 2008 E-Area Performance Assessment (PA) [1]. The concern was the potential for overlapping of atmospheric plumes, emanating from the soil surface above SRS LLW disposal facilities within the E-Area, to contribute to the dose received by a member of the public during the Institutional Control (IC) period. The implication of this concern was that the dose to the maximally-exposed individual (MEI) located at the SRS boundary might be underestimated during this time interval. To address this concern a re-analysis of the atmospheric pathway releases from E-Area was required. A new atmospheric release model (ARM) capable of addressing the LFRG plume overlap concern was developed.

The conceptual approach to assessing the atmospheric dose to a member of the public from E-Area LLW disposal facilities is to perform sub-surface simulations of the release of volatile radionuclides from the waste zones through the overlying engineered barriers to determine a flux rate at the land surface. Then, atmospheric dispersion of the radionuclide flux is simulated in an atmospheric transport model [2] to compute air concentrations in the vicinity of the hypothetically exposed individual. A re-analysis of atmospheric dispersion portion of the atmospheric pathway releases was not undertaken, however the Dose Release Factors (DRF's) previously computed in a atmospheric transport study [3] were retained to convert the new surface emanation fluxes (computed with the ARM) into doses received by the MEI at the appropriate points of compliance (POC). In this way, the dose received by the MEI could be compared to the maximum permissible dose level, defined in DOE Order 435.1 [4] as being 100 uSv/yr (10 mrem/yr).

INTRODUCTION

The initial work conducted in this study was to develop a new Atmospheric Release Model (ARM) using the GoldSim® program [5]. This model simulates the subsurface vapor diffusion of volatile radionuclides as they release from E-Area disposal facility waste zones and migrate to the land surface. In the process of this work, many new features, including several new physical and chemical transport mechanisms, were incorporated into the model. Of all of the improvements, the most important one was to incorporate a mechanism to partition volatile contaminants across the water-air interface within the partially saturated pore spaces of the engineered and natural materials through which vapor-phase transport occurs. The ARM also combines the individual transport models constructed for each E-Area disposal unit into a single model, and was

ultimately was used to analyze the LFRG concern regarding the potential for atmospheric plume overlap at the SRS boundary during the IC period.

The first step in the re-analysis of the atmospheric pathway was to identify those radionuclides which might potentially become volatile and be available for diffusion through air-filled pores. A screening analysis [6] was conducted prior to the 2008 PA to identify the radionuclides to be more thoroughly analyzed to derive disposal limits for the E-Area disposal facilities based on the atmospheric pathway releases. The potentially volatile radionuclides were determined to be: C-14, Cl-36, H-3, I-129, S-35, Sb-124, Sb-125, Se-75, Se-79, Sn-113, Sn-119m, Sn-121, Sn-121m, Sn-123 and Sn-126. All of these radionuclides are built into the ARM, in their anticipated vapor-state molecular form, and are evaluated in every model realization.

The computational model utilized to simulate subsurface transport of volatile radionuclides was developed in the GoldSim® Version.10.5 [5] programming environment, which is an analytical contaminant transport code. When the flow field is specified, it has the ability to compute both advective and diffusive transport of contaminant species; however it lacks the ability to compute advective flow. The code is normally implemented using a 1D arrangement of computational elements to approximate a flow domain although 2D meshes can also be configured. Radioactive decay and chemical retardation within a flow field are easily implemented and multiple contaminants are simulated simultaneously.

MODEL STRUCTURE AND FEATURES

Description of Structural features

The conceptual model implemented in the ARM is a 1-D, vertical column of computational elements, having no-flow boundaries on sides and bottom, so as to divert all contaminant fluxes to the land surface. These boundary conditions are conservative in that, in reality, diffusion may also proceed laterally and downward in the subsurface. In this context an overestimation of the contaminant flux at the land surface is defined as “conservative”. Each of the separate types of disposal facilities within E-Area was incorporated within the GoldSim model so as to enable the simultaneous simulation of subsurface diffusive releases. There are three basic types of disposal facilities within E-Area for which a specific representation was built into the ARM. There are trench facilities, concrete vault facilities and surface pads. The materials contained in the model elements reflect the waste zones, engineered barriers, sand fill and closure cap of the individual disposal facilities. Model configurations of these zones conformed to the anticipated closure designs of each facility type (e.g., dimensions of waste zones and engineered barriers) and the general occurrence and timings of events encompassed in the loading and closure of the facilities in E-Area were emulated.

The ARM was constructed such that each simulation computed the vapor diffusive releases from each disposal facility simultaneously. The model structure and organization for an individual disposal facility is illustrated on the left side of Fig. 1. One such module was included for each disposal facility type. On the right hand side of that figure is an illustration of the vertical column of cells used to represent the waste zone of a disposal facility, and the portion of the ARM in which the source term was introduced.

The overall simulation length was 1,000 years, so as to enable an assessment over the performance period required by DOE Order 435.1 [4]. Material properties associated with each disposal facility, including porosity, particle density, residual water saturations, etc. identified in the earlier Performance Assessment [1] were largely adhered to in this investigation, except where better estimates could be obtained.

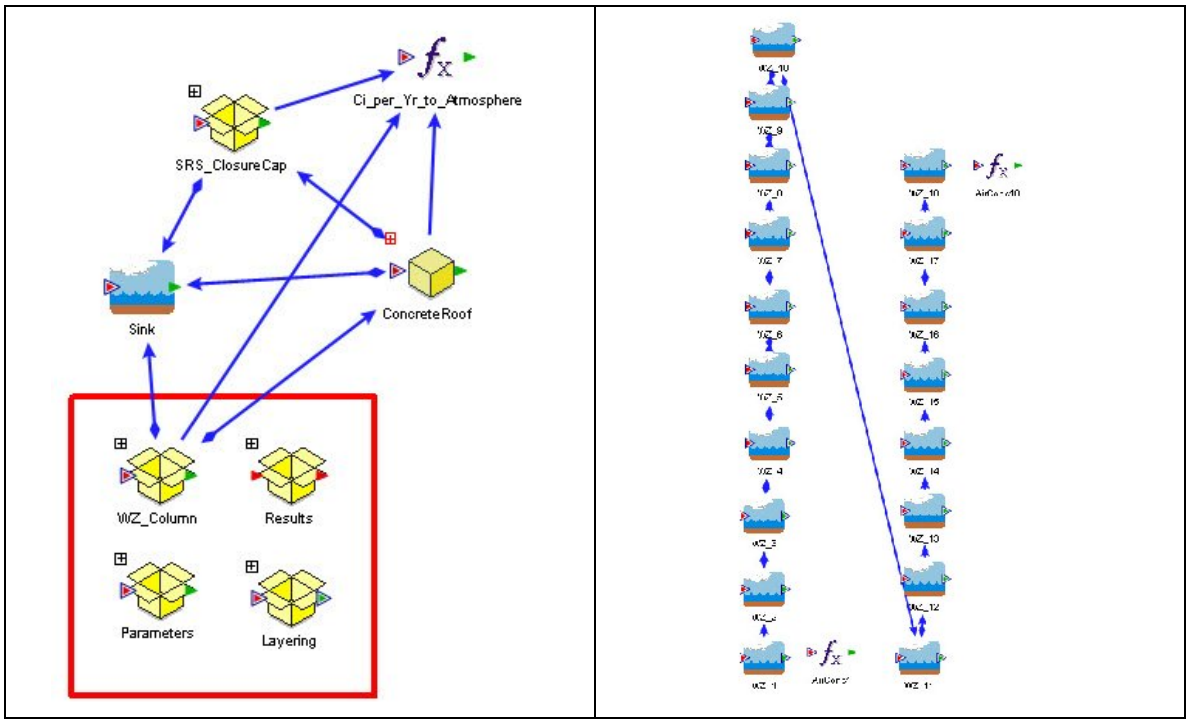

Fig. 1 Model organization for an individual disposal unit and waste zone cell structure

A standardized closure cap model, conforming to the engineering design of the anticipated final closure cap that will be placed over all of the E-Area facilities at the end of IC, was developed and implemented within the ARM. This closure cap was placed above each disposal facility at the end of IC. The specific features of the final closure cap are described in the Closure Plan for the E-Area Low-Level Waste Facility [7]. Fig. 2 illustrates the GoldSim® closure cap mixing cell arrangement within the model and is clearly labeled to indicate the material layers represented. The arrows in the figure indicate diffusive links that exist between adjacent cells. One unusual material contained in the closure cap is the Erosion Barrier (EB). This layer consists of granite cobbles with a fill material added to the spaces between the cobbles. A final decision has not been made as to what material will be used, however this investigation assumed a material with porosity of 0.328 and a long-term residual saturation of 0.825 will be used to fill the interstitial spaces between granite cobbles. The EB porosity, bulk density and residual saturation of the EB were calculated based on this assumption.

New Mechanisms

The ARM is a much more robust and flexible model than has been previously used for air dose modeling at the SRS. An effort was made to incorporate a more accurate representation of physical features and properties of each facility, honoring the configuration of the waste zones, engineered features and overlying closure cap layers.

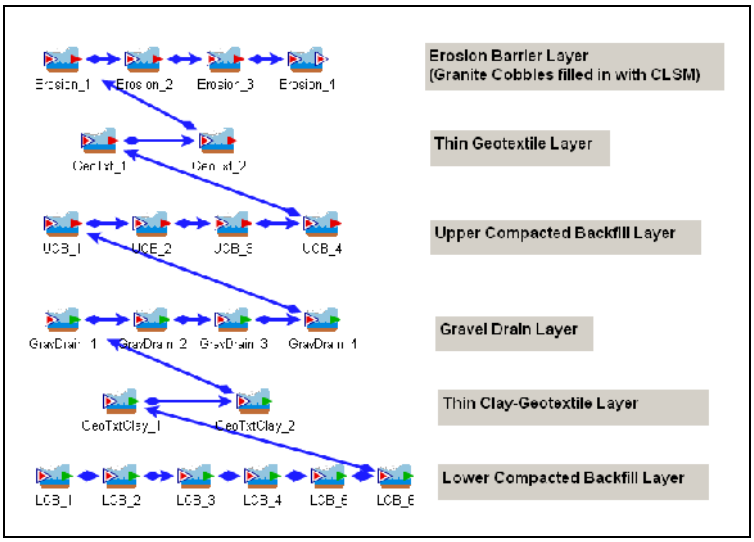

Fig. 2 GoldSim® Model Elements Representing the Standardized Closure Cap

Structurally, the combining of all disposal facility models into a single E-Area model provides the ability to evaluate the combined impact of multiple facilities on a single MEI, which is ultimately how the LFRG plume overlap concern is addressed. Related to this, certain parameters were constructed with "global" connections, making adjustments to these parameters much easier to simultaneously incorporate for all disposal facilities.

One of the most significant improvements in developing the ARM was the incorporation of the mechanism to equilibrate the concentration of contaminants across the air-water interface according to Henry's Law constants. This feature enables more accurate simulations of contaminant flux at the land surface to be realized. Another important new feature is the mechanism to automatically compute effective diffusion coefficients for each material, by radionuclide, during each simulation. This introduces a flexibility that enables the user to easily investigate the impact of varying selected parameter values (e.g., porosity and residual saturation) on the result. Expanded discussions of the new features are provided below.

Henry's Law describes the partitioning of species between the aqueous phase and the gas phase. A recent study [8] gives the Henry's Law constants for relevant radionuclides under a variety of conditions. Conditions representing soil (pH 5.4, Eh 0.37) and aged concrete (pH 8.23 Eh 0.73) were incorporated as new models were constructed for each E-Area disposal facility. The analysis used to derive the Henry's Law constants also gave the chemical form of each radionuclide that would be most stable in the vapor phase under each condition. This information is shown in Table I.

Table I. Henry's Law Constants (mole/kg-atm) for Chemical Species [8]

	C	Cl	H	I	Sb	Se	Sn
Soil	3.8E-2	5.2E11	2.1E3	6.3E14	6.9E32	2.8E25	9.6E53
Aged Concrete	2.8E0	3.6E14	2.1E3	1.3E29	4.9E38	3.8E87	6.1E61

Note: C as CO_2, Cl as HCL, H as H_2O, I as HI, Sb as $SbCl_3$, Se as H_2Se, and Sn as $SnCl_4$

The ARM requires the Henry's Law constants to be expressed as the dimensionless ratio:

$$\frac{\text{quantity in vapor phase}}{\text{quantity in aqueous phase}}$$

The conversion factor for this is 4.1E-02 (kg-atm)/mole [9]**.** The values presented in this table were implemented within the new ARM model as partitioning coefficients for individual radionuclides within the different material zones.

The ability to compute the effective diffusion coefficients (D_e) for volatile radionuclides and molecules within various porous media was built directly into the ARM. The advantage of performing the calculation internally within the transport model is that it allows evaluations of the sensitivity of the model results to the values assumed for porosity and residual water saturation to easily be performed. Furthermore, transient water saturations of materials based on computations external to the ARM can easily be incorporated by simply adjusting that property for the selected material at the appropriate times.

The initial step in this computation is to evaluate the D_c for Rn-222 as a function of the porosity and moisture saturation as a reference case for the other volatile radionuclides. The equations to perform this computation are defined in [10].

$$D_c = D_0 p \times \exp\left[-6Sp - 6S^{14p}\right] \qquad \text{(Eq. 1)}$$

where:

D_o = diffusion coefficient of Rn-222 in open air
p = total porosity
S = volume fraction of water saturation

While implementing these equations in GoldSim®, a minor issue was discovered in that all isotopes of an element are assumed to have the same diffusion coefficient. While this is not the case for an air diffusion model, the affect in the ARM is quite small, and is proportional to the difference in atomic weights of the isotopes. Therefore, this effect could be ignored.

Once the D_e of Rn-222 is computed for a particular material, with unique porosity and a specified long-term residual saturation, the D_e of the other volatile radionuclides or molecular compounds evaluated in this SA were computed using the relationship described by Graham's Law:

$$D_e = D'_e \sqrt{\frac{MWT'}{MWT}} \qquad \text{(Eq. 2)}$$

where:

D_e = the diffusion coefficient of the radionuclide of interest (m^2/yr)

D'_e = the diffusion coefficient of the reference radionuclide (Rn-222) (m^2/yr)

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

MWT = the molecular weight of the vapor form of the radionuclide of interest

One last mechanism implemented in the ARM was to invoke the imposition of a maximum concentration of $^{14}CO_2$ in the vapor-filled pore spaces of cementitious materials. The basis for this is documented in [11] where the thermodynamic equations describing the equilibration of a C-14 waste source with high pH water were evaluated. The key processes evaluated in that study included: (1) the equilibration of $^{12}CO_3^{2-}{}_{(aq)}$ and $^{14}CO_3^{2-}{}_{(aq)}$ with cementitious pore water, and (2) volatilization of $^{12}CO_{2(g)}$ and $^{14}CO_{2(g)}$ from the cement-equilibrated $^{12}CO_3^{2-}{}_{(aq)}$ and $^{14}CO_3^{2-}{}_{(aq)}$. The study concluded that C-14 concentrations in the gaseous state would not exceed 7.0E+03 Bq/ m^3 (1.9E-07 Ci/m^3).

Conceptually, the cementitious material provides a very strong sorbent for the $^{14}CO^2{}_{(g)}$. In fact, industries that generate 1000's of times greater amounts of C-14 than anticipated for the ILV, such as the Ontario Hydro's Reactors [12, 13], dispose C-14 bearing resins along with concrete slabs; the concrete slabs are referred to as "C-14 getters." Consequently, little $^{14}CO_{2(g)}$ is expected to be released from E-Area facilities containing cementitious material. The use of this concept to simulate releases of $^{14}CO_2$ from E-Area waste disposal facilities is also reinforced by widespread attention currently being directed to the use of concrete as a means to sequester atmospheric CO_2.

ANALYSIS AND RESULTS

The ARM simulation computed the rate of diffusive flux at the land surface above each facility over the IC and post-Closure time periods. One of the purposes of the simulation was to establish the dose to the MEI with respect to time that results from placing a single Ci source term in each E-Area disposal facility. This dose is computed within the ARM model by first computing the diffusive flux of each radionuclide at the land surface and then multiplying that flux rate by the appropriate DRF for each facility with respect to time. Two DRFs are used for each facility, depending upon the location of the POC for the MEI. Initially the MEI is located at the SRS boundary and the E-Area is considered to be a point source, thus the same DRF is used for each disposal facility. During the post-Closure period (after 100 years of IC) a disposal facility specific DRF is available for each facility. The appropriate DRF is automatically invoked within the ARM depending on the elapsed time of the simulation.

The total simulation time was for 1000 years. This includes a 25-yr operations period, a 100-yr IC period and an additional 875-year post-Closure period. Although simulation of the operations period was not necessary in the analysis, it was retained in this model because it was built into the original ARM which was then easily adapted simply by changing the time in which the source term was inserted into it. The total 1000-yr simulation length was sufficient to observe the peak land surface fluxes for each E-Area

disposal facility, although the simulation did not extend to the full 1000 years beyond the Final Closure (e.g., t_{max} = 1125). The atmospheric releases from all facilities except the NRCDA's peak relatively early in this time period. The waste material loaded into the NRCDA's is contained within stainless steel vessels that are welded shut. The welds and walls of the vessel are assumed to retain their integrity for 750 years and only begin to release any remaining volatile inventory at that time.

While the ARM evaluated the full suite of potentially volatile radionuclides identified in the INTRODUCTION, it only computes a flux at the land surface for C-14 and H-3. The model computes a zero flux for all other radionuclides. The main factors in this phenomenon are 1) the tendency of the other radionuclides to partition into the available pore water of the disposal facility layers and overlying layers during the simulation and 2) radioactive decay.

Table II presents the peak contaminant fluxes for C-14 and H-3 above each E-Area disposal facility that results from an initial source term of 3.7E+10 Bq (1 Ci) and the elapsed time at which the peak occurred. Note that time = 25 yr is the end of operations, time = 125 yr is the end of IC, and times > 125 yr represent the post-Closure period. Similarly, Table III presents the computed peak dose to the MEI that is associated with each E-Area disposal facility and the time of occurrence of that dose. Simulation of a unit source term is performed at SRS to establish disposal limits for individual E-Area disposal facilities.

Table II. Peak Contaminant Fluxes and the Time of Peak

	Peak Flux H-3 (Bq/yr)	**Time of Peak (Yr)**	**Peak Flux C-14 (Bq/yr)**	**Time of Peak (Yr)**
ST/ET	6.1E-02	25	1.8E+10	25
CIG	5.2E-04	125	5.2E+03	25
LAWV	4.4E+03	102	5.1E+03	26
ILV	2.6E-02	125	2.9E+03	25
NRCDA 643-26E	NA	NA	7.4E+08	842
NRCDA 643-7E	NA	NA	7.4E+08	837

Table III. Peak Doses to the MEI and the Time of Peak

	Peak Dose H-3 (uSv/yr)	**Time of Peak (Yr)**	**Peak Dose C-14 (uSv/yr)**	**Time of Peak (Yr)**
ST/ET	3.6E-17	125	5.4E-04	25
CIG	3.1E-19	125	2.9E-08	145
LAWV	2.6E-12	102	2.8E-08	138
ILV	1.6E-17	125	1.4E-07	138
NRCDA 643-26E	NA	NA	9.9E-03	842
NRCDA 643-7E	NA	NA	2.0E-02	837

Plume Overlap Analysis

As was described earlier, the primary motivation in updating the E-Area Low Level Waste facility atmospheric pathway models was to be able to address the concern of the LFRG PA review team regarding the need to assess the potential for overlapping atmospheric plumes during the IC period. During this period the POC (and therefore the location of potential impact) is located at the SRS boundary, at which distance the plumes emanating from separate facilities within E-Area may indeed co-mingle. Should atmospheric plumes co-mingle a dose to the MEI that is higher than expected dose from an individual facility could occur. This is the essence of the LFRG review team concern.

To evaluate this, a conservative approach was adopted in whereby the MEI at the SRS boundary is exposed simultaneously to the releases from all E-Area disposal facilities. This is equivalent to evaluating a 100% overlap of all atmospheric plumes emanating from E-Area. Should the dose received from this level of atmospheric plume overlap still fall below the permissible exposure level of 100 uSv/yr (10 mrem/yr), then the LFRG issue would be demonstrated to no longer be a concern. The structuring of the ARM allows this evaluation to be easily performed.

The strategy in this analysis was to introduce the full radionuclide inventory that is anticipated for each E-Area disposal facility, as indicated in the 2008 PA [1] (Appendix C, Closure Inventory Estimate), into their waste zones. Since it was earlier demonstrated that of all the potentially volatile radionuclides only C-14 and H-3 emanate from the land surface, those were the only radionuclides considered in the plume overlap analysis. Also, since both C-14 and H-3 have certain special waste forms listed in the inventory (e.g., resin based C-14) they were combined with the inventory of the generic isotope to produce the total inventory. This strategy is thought to be conservative since any special waste form would have a slower vapor release rate of the isotope than the generic form, which is assumed to be immediately available for vapor diffusion. This, in effect, causes the peak release rates to be higher than they otherwise would be if the special waste form inventory released more slowly. The total anticipated radionuclide inventory for C-14 and H-3 utilized in the plume overlap evaluation are shown in Table IV.

Table IV. ELLWF Estimated Total Inventory at Closure, by disposal facility.

E-Area Disposal Facility	**C-14** (Bq)	**H-3** (Bq)
Slit Trenches	1.11E+09	4.44E+10
Engineered Trenches	1.30E+10	3.11E+11
Component In Grout Trenches	1.26E+10	4.07E+14
Low Activity Waste Vaults	5.55E+10	7.77E+17
Intermediate Level Vault	3.70E+13	1.55E+17
NRCDA Pad 663-26E	1.26E+13	--
NRCDA Pad 663-7E	5.18E+12	--

Note: Combines generic isotope inventory and special waste form inventory

Within the ARM the total dose delivered to the MEI was computed. The total dose comprises contributions from both C-14 and H-3 and from the contribution from all E-Area disposal facilities, simultaneously. Since individual facilities don't receive their total inventory emplacement simultaneously, there is a degree of temporal overlap as well as the spatial overlap imbedded within the analysis. Combined, these conditions are thought to create a "worst-case" scenario for the MEI. The results of this analysis, including the total plume overlap dose and the dose contribution from each individual E-Area disposal facility, are displayed in Fig. 3. Total plume overlap dose is the sum of the individual facility doses contributed to the MEI and hence is essentially the trace of the uppermost individual facility dose contributions shown with the dark blue line. Doses contributed by individual facilities are also indicated.

During the IC period, the peak of the "total plume overlap dose" was computed to be 1.95E-04 uSv/yr (1.9E-05 mrem/yr), which occurs at time = 25 years. Later, after the NRCDA begin to release C-14, the overall peak of the "total plume overlap dose" occurs.

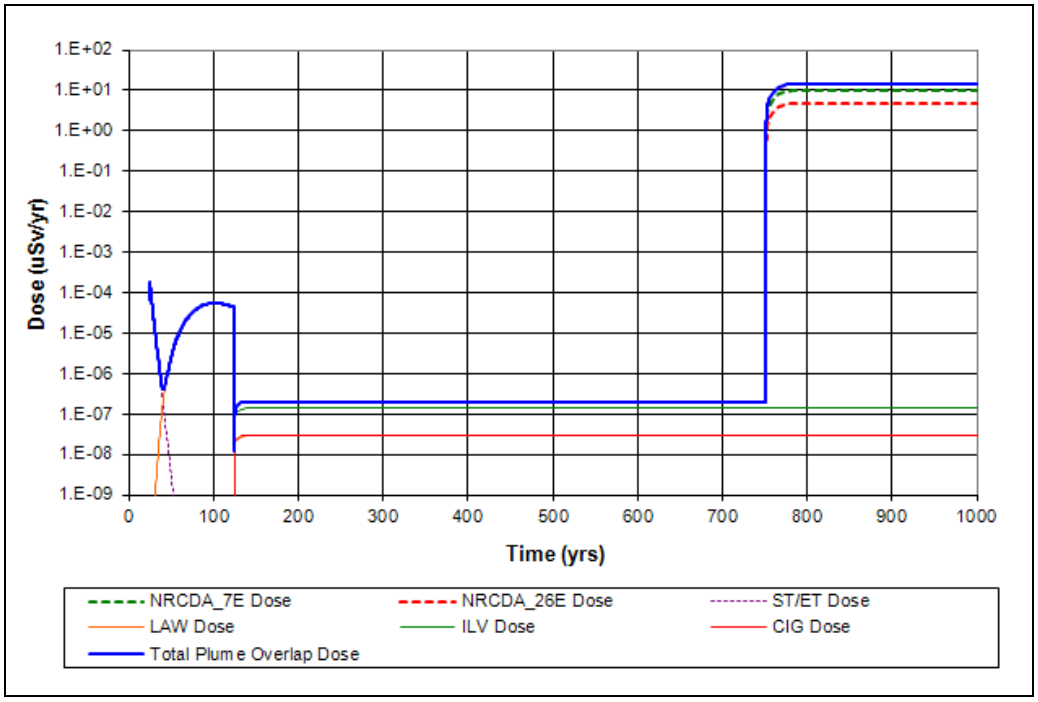

Fig. 3. Plume Overlap Dose and Dose from Individual Disposal Facilities

The total plume overlap dose during the IC period (0-125 years) is driven C-14 and H-3 releases. After the IC, total plume overlap dose illustrated in the figure is driven by C-14 releases. The large jump that occurs at 750 years is from the C-14 release from NRCDA, as the steel vessels containing the C-14 waste are assumed to corrode all the way through, simultaneously.

The LFRG PA review team concern was for plume overlap occurring in the IC period and the worst case plume overlap dose is shown to be 1.95E-04 uSv/yr (1.9E-05 mrem/yr) during this time-period, which is five orders of magnitude less than the PA performance

measure of 100 uSv/yr (10 mrem/yr). It can safely be said that overlap of atmospheric plumes emanating from E-Area disposal facilities is not a concern during the IC period.

Additionally, the potential for plume overlap was assessed in the post-Closure period. Atmospheric plume overlap is less likely to occur during this period but conceivably could occur if the prevailing wind direction shifted so as to pass directly over all E-Area disposal facilities and transport airborne radionuclides to the MEI at the 100 m POC. This was also demonstrated to be of little concern, as the maximum plume overlap dose was found to be 1.45E+01 uSv/yr (1.45E+00 mrem/yr), or ~15% of the performance measure) during this period and under these “worst-case” conditions.

DISCUSSION

This investigation was initiated to address a concern, expressed by DOE’s LFRG review team. The concern was that overlapping of atmospheric plumes, emanating from the soil surface above SRS LLW disposal facilities, would lead to the underestimating of the dose received by a member of the public during the IC period. An entirely new atmospheric release model was developed and, using this improved model, it was discovered that, even though the full suite of potentially volatile radionuclides identified in [6] were introduced into the disposal facility waste zones, it computed a zero flux at the land surface for all of the radionuclides except C-14 and H-3. The main factors in this phenomenon is the tendency of those radionuclides to partition into the available pore water of the disposal facility and within the overlying engineered barriers during the simulation and the radioactive decay process. Thus, these two radionuclides are the only ones for which new atmospheric pathway disposal limits were computed for E-Area disposal facilities and for which a plume overlap assessment was made.

The main conclusion of this study is that for atmospheric releases from the E-Area disposal facilities, plume overlap does not cause the total dose to the MEI at the SRS boundary during IC to exceed the PA performance objective. This conclusion directly addresses the LFRG PA review team issue. Furthermore, even though the LFRG comment did not express concern for such overlap to occur during the post-Closure period, applying the 100% plume overlap strategy to atmospheric releases during this period also demonstrates that the PA performance objective for the MEI will also not be exceeded during that time-frame either.

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