

**An Exposure Assessment of Radionuclide Emissions Associated with  
Potential Mixed-Low Level Waste Disposal Facilities at Fifteen DOE Sites**

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## INTRODUCTION

For more than 50 years, the United States has produced materials for nuclear weapons and has conducted research with nuclear materials. These activities generated mixed wastes (i.e., those that are both radioactive and chemically hazardous). The U.S. Department of Energy (DOE) is faced with the challenge of managing these wastes. The DOE currently generates, stores, or is expected to generate over the next five years about 458,000 m<sup>3</sup> of mixed low-level waste (MLLW).<sup>1</sup>

Mixed waste has a hazardous component and must be treated to comply with Land Disposal Restrictions (LDRs) of the Resource Conservation and Recovery Act (RCRA). However, there is insufficient capacity, and in some cases a lack of available technologies, to treat these wastes. The Federal Facility Compliance Act (FFCAct) of 1992 requires the Secretary of Energy to develop and submit site treatment plans for the development of treatment capacity and of technologies for treating mixed waste for each facility at which the DOE stores or generates these wastes. These plans identify how the DOE will provide necessary mixed waste treatment capacity, including schedules for bringing new treatment facilities into operation. In collaboration with the National Governors' Association, representing the States, the DOE has been evaluating candidate treatment options and developing the required treatment plans.

Although the FFCAct does not specifically require the DOE to address disposal of treated mixed waste, both the DOE and the States realize that the method of treatment for a specific waste is an integral component of considering for its eventual disposal. As a result, the DOE established the FFCAct Disposal Workgroup (DWG) in June 1993 to work with the States to define and develop a process for evaluating disposal options. The focus of the DWG was to identify sites, from among those currently storing or expected to generate MLLW, that were suitable for further evaluation regarding their disposal capability. Some sites that have been determined to have marginal or no potential for disposal activities were removed or postponed from further evaluation under this process. Fifteen sites, identified in Figure 1, were evaluated using a performance evaluation (PE) process that served as a preliminary screening analysis. Ultimately, a number of sites are expected to be technically acceptable for disposal activities.

The goal of the PE was to quantify and compare the limitations of 15 DOE sites for disposal of MLLW. The objective was, therefore, to use a set of modeling assumptions of sufficient detail to capture major site-specific characteristics and yet generic enough for consistent application at all sites. In addition, the analyses were developed to ensure that no systematic biases were introduced, that sites were analyzed consistently, and that all major assumptions were clearly stated.

The PE adopted an analysis simplified from that used in many low-level radioactive waste performance assessments. The PE was based solely on radiological assessment for disposal even though the wastes under consideration also contain hazardous components that are subject to RCRA requirements. The PE analysis assumed that the chemical components of the wastes would be treated to land LDRs according to RCRA's treatment processes and that a MLLW disposal facility would comply with all RCRA design criteria.

Because the PE is a radiological assessment for disposal, it follows DOE Order 5820.2A as the basis for the analysis. The DOE Order lists *performance objectives* that are used to demonstrate whether a proposed facility will be in compliance. These same performance objectives were used in the PE as *performance*

*measures*, which are used to back calculate maximum concentrations of each radionuclide in the disposed waste. These waste concentrations are calculated for potential releases of radionuclides from a disposal facility via the air, water, and inadvertent intruder pathways (generally referred to as transport pathways). The atmospheric transport pathway was one of the three transport pathways analyzed in the PE, and is the focus of this paper.

## ANALYSIS METHOD

The DOE Order 5820.2A performance objective applicable to atmospheric releases is to meet the requirements of 40 CFR 61, National Emissions Standards for Hazardous Air Pollutants (NESHAPs). That is, radiological doses through all intake routes from releases to the atmosphere shall not exceed 0.01 rems per year. This includes inhalation of radionuclides and ingestion of foodstuffs contaminated by atmospheric releases. The downwind receptor site was set at 100 m from the edge of the disposal facility and covered a period of 10,000 years from the time the disposal facility was closed, based on recommendations of the DOE's Performance Assessment Task Team.<sup>2</sup> Two generic types of disposal facilities were assessed at each site: a RCRA-compliant, below-ground trench and a RCRA-compliant, above-ground concrete vault (also called a tumulus). To provide consistency in the evaluation of the 15 sites, it was assumed that the size and shape of each generic facility was the same.

The attenuation of radionuclides between the waste and the disposal facility and the performance boundary was labeled as a "concentration reduction factor" (CRF). More generally, CRFs are the ratio of concentrations between selected points along the pathway (e.g., the ratio of the radionuclide concentration in the disposed waste to the radionuclide concentration in the soil surface above the disposal facility). These were used so that intermediate results could be displayed in a transparent fashion that allowed comparisons of the effects of site location on overall performance. This approach allowed comparisons of results from different sites. The CRFs pertain to transport effects only, ignoring radioactive decay. When applicable, the effects of radioactive decay were applied separately in the calculations of permissible waste concentrations. Radionuclides were assumed to be retained in the facility for 100 years corresponding to the period of institutional control.<sup>1</sup>

For the atmospheric pathway, the CRFs were broken into two components: (1) diffusion of radionuclides upward through the soil cover above the disposal facility ( $CRF_{diff}$ ) and (2) emission of radionuclides and subsequent atmospheric dispersion ( $CRF_{disp}$ ). The PE analyzed 58 radionuclides for all sites for the water and inadvertent intruder pathways. Only two of those radionuclides,  $^3\text{H}$  (tritium) and  $^{14}\text{C}$ , were analyzed for the atmospheric pathway because these radionuclides were considered the only ones sufficiently volatile to migrate upward through the disposal facility cover and into the atmosphere in amounts large enough for assessment. A schematic of radionuclide transport is shown in Figure 2.

Dose conversion factors (DCFs), modified to include inhalation and ingestion exposure parameters (e.g., inhalation rate, ingestion rate of milk, plant uptake of radionuclides), are used to convert predicted atmospheric concentrations to annual doses. These modified DCFs are expressed as scenario dose conversion factors (SDCFs). The SDCFs and CRFs are applied to the performance measure (0.01 rem per year) to calculate a maximum permissible concentration in the disposal facilities for  $^3\text{H}$  and  $^{14}\text{C}$ . The following subsections describe the atmospheric pathway analysis for calculating permissible waste concentrations.

**CRF<sub>diff</sub> Calculation**

The definition of  $CRF_{diff}$  is as follows:

$$CRF_{diff} = \frac{C_w}{C_{ss}} \quad (1)$$

where

$C_w$  is the radionuclide concentration in the disposed waste ( $\mu\text{Ci}/\text{m}^3$ ), and

$C_{ss}$  is the radionuclide concentration in the soil surface above the waste disposal unit resulting from the concentration in the waste ( $\mu\text{Ci}/\text{m}^3$ ).

Volatile radionuclides are assumed to be transported to the soil surface by diffusion in the vapor phase.<sup>3</sup> The vapor flux is assumed to be a first-order, linear process, and flux is assumed to occur from the top of the waste disposal unit to the soil surface. Therefore, the volatile radionuclide vapor concentration,  $C_v$  ( $\mu\text{Ci}/\text{m}^3$ ), at the top of the waste disposal unit is approximated by:

$$C_v = J \left( \frac{x}{D} \right) \quad (2)$$

where

$J$  is the radionuclide flux density through the soil above the waste disposal facility ( $\mu\text{Ci}/\text{m}^2\text{-s}$ ),

$x$  is the cover thickness above the waste disposal facility (m), and

$D$  is the  $^3\text{H}$  diffusion coefficient in air,  $2.39 \times 10^{-5} \text{ m}^2/\text{s}$ , or the  $^{14}\text{CO}_2$  diffusion coefficient in air,  $1.40 \times 10^{-5} \text{ m}^2/\text{s}$ .

For  $^3\text{H}$ , the relation between its concentration in the water vapor and in the liquid water bound in the waste must be determined before  $C_w$  can be calculated. This can be accomplished by determining the ratio of water density in air to that in the liquid phase. If the air is saturated with water at  $10^\circ\text{C}$ , then the density of water vapor in dry air, or absolute humidity, is 9.2 grams of water vapor per cubic meter of dry air.<sup>3</sup> Assuming the density of liquid water is  $1 \times 10^6$  grams per cubic meter, the ratio of water vapor present in dry air to that in the liquid phase,  $r$ , is:

$$r = \frac{9.2 \text{ g}/\text{m}^3 \text{ (water vapor)}}{1 \times 10^6 \text{ g}/\text{m}^3 \text{ (liquid water)}} \quad (3)$$

The relation between the concentration of  $^{14}\text{C}$  in the air and in the liquid water bound in the waste can be calculated using Henry's Law. This law is a linear, first-order relationship derived by approximating equilibrium conditions between dissolved gas with a particular concentration in liquid water and the same gas with a particular concentration in the air adjacent to the liquid water. All of the  $^{14}\text{C}$  in the disposal facility is conservatively assumed to be  $^{14}\text{CO}_2$  (i.e., some of the carbon would be in the form of  $\text{H}_2\text{CO}_3$ ,  $\text{HCO}^-$ , and  $\text{CO}_3^{2-}$ ). For use in Henry's Law, the unit mole fraction,  $\chi$ , of  $^{14}\text{CO}_2$  in the liquid water is determined as:

$$\chi = \frac{N_{CO_2} \text{ per g of } CO_2}{N_{H_2O} \text{ per g of } H_2O} \quad (4)$$

where

$N_{CO_2}$  is the number of moles in 1 gram of  $CO_2$ , and  
 $N_{H_2O}$  is the number of moles in 1 cubic meter of liquid water.

Assuming the density of liquid water as  $1.0 \times 10^6$  grams per cubic meter,  $\chi$  is equal to  $4.1 \times 10^{-7}$ . The partial pressure of  $CO_2$  in the air,  $p$  (atm), is calculated using Henry's Law:

$$p = k\chi \quad (5)$$

where

$k$  is the Henry's Law constant for  $CO_2$ . At  $10^\circ C$ ,  $k$  is equal to 1040 atm/mole fraction.<sup>4,5</sup>

Substituting the values for the unit mole fraction and Henry's Law constant for  $CO_2$  at  $10^\circ C$ ,  $p$  is calculated to be  $4.3 \times 10^{-4}$  atm. If the air is at atmospheric pressure, the concentration of  $CO_2$  in the air is approximately equal to  $4.3 \times 10^{-4}$  g/m<sup>3</sup>, and therefore, the ratio,  $r$ , of  $CO_2$  in the air to that dissolved in the water is:

$$r = \frac{4.3 \times 10^{-4} \text{ g/m}^3 \text{ (CO}_2 \text{ vapor)}}{1 \text{ g/m}^3 \text{ (CO}_2 \text{ dissolved water)}} \quad (6)$$

Because  $^3H$  is assumed to be completely bound in the pore water and  $^{14}C$  is assumed to be dissolved as  $^{14}CO_2$ , the volatile radionuclide concentration in the pore water,  $C_{pw}$  ( $\mu Ci/m^3$ ) is:

$$C_{pw} = \frac{C_v}{r} \quad (7)$$

The radionuclide bulk concentration in the waste,  $C_w$ , is related to the  $C_{pw}$  as:

$$C_w = C_{pw} ns \quad (8)$$

where

$n$  is the soil porosity, and  
 $s$  is the fraction saturation in the soil void space.

Substituting Equation 7 into Equation 8 yields:

$$C_w = \frac{C_v ns}{r} \quad (9)$$

and substituting Equation 2 into Equation 9 gives:

$$C_w = \frac{Jxns}{Dr} \quad (10)$$

The flux density out of the soil surface is assumed to be equal to the flux density through the soil,  $J$ . Therefore,  $C_{ss}$  is calculated as:

$$C_{ss} = \frac{Jd}{D} \quad (11)$$

where,  $d$  is the depth of the surface soil. The value for  $d$  is assumed to be 0.01 m for all sites.<sup>6</sup>

Substituting Equation 10 and 11 into Equation 1 gives:

$$CRF_{diff} = \frac{xns}{rd} \quad (12)$$

#### **$CRF_{disp}$ Calculation**

The definition of  $CRF_{disp}$  is as follows:

$$CRF_{disp} = \frac{C_{ss}}{C_{Atm}} \quad (13)$$

where

$C_{ss}$  is the radionuclide concentration in the soil surface above the waste disposal unit ( $\mu\text{Ci}/\text{m}^3$ ), and  $C_{Atm}$  is the radionuclide concentration in the ambient air at the performance boundary ( $\mu\text{Ci}/\text{m}^3$ ).

The radionuclide flux density out of the surface soil is assumed to be emitted directly into the atmosphere where it is mixed with the ambient air flowing above the facility. Therefore,  $C_{Atm}$  is determined by:

$$C_{Atm} = JA_D \quad (14)$$

where,  $A_D$  is the atmospheric dispersion term specifying the concentration in ambient air at the performance boundary produced by unit flux density leaving the soil at the disposal facility ( $\mu\text{Ci}/\text{m}^3$  per  $\mu\text{Ci}/\text{m}^2\text{-s}$ ).  $A_D$  is the maximum annual average value among a set of receptors located at the performance boundary, it can be obtained using a Gaussian air dispersion model as described below.

Equation 11 and Equation 14 are substituted into Equation 13, yielding:

$$CRF_{disp} = \frac{d}{A_D D} \quad (15)$$

### Transport Time To Receptors

Assuming that the diffusive velocity,  $v_d$ , is uniform and that it is approximated as one-dimensional, first-order, and linear, the diffusion transport time,  $t$ , for volatile radionuclides, is given by:<sup>7</sup>

$$t = \frac{x^2}{D} \quad (16)$$

Once the radionuclide is airborne, the transport time to receptors located at the performance boundary downwind would take less than 100 seconds. Therefore, there would not be sufficient time for appreciable radionuclide decay during both the mixing dispersion phase of transport.

### Atmospheric Dispersion Term Determination

The atmospheric dispersion term,  $A_D$  ( $\mu\text{Ci}/\text{m}^3$  per  $\mu\text{Ci}/\text{m}^2\text{-s}$ ), was estimated using the Industrial Source Complex-Version 2 Long Term Air Dispersion Model (ISCLT2).<sup>8</sup>  $A_D$  is defined as the maximum annual average value calculated for a set of receptors located 100 m from the waste disposal facility. ISCLT2 is the EPA's refined air dispersion model for calculating long-term (annual average) atmospheric concentrations in simple terrain. GENII and AIRDOS-PC are two Gaussian dispersion models that are specifically approved for use in modeling effects of radionuclide emissions; however, both models have features that go well beyond the need of the PE analysis and would be considerably more cumbersome to use than ISCLT2. Because these three models incorporate the same basic dispersion equations, differences in the concentration estimates generated by the models would not be significant (less than an order of magnitude, which is less than the resolution of this screening analysis).

The area of the waste disposal facility was an important input into the model. For the general tumulus and shallow trench designs, flux areas are the same for all 15 sites. For those sites that have existing plans for mixed waste disposal, site-specific waste disposal facility areas were used. In order to improve the accuracy of the model with receptors at close distances, the surface area of the disposal facility was divided into 256 equal squares based on several simulations using different numbers of areas. Note the length of the smaller squares is 1/16th the length of the side of the facility area. Site-specific meteorological data were also used. Because the area flux is assumed to occur at ground-level, terrain was modeled as flat. This is an upper-bound assumption that resulted in the maximum ground-level receptor concentrations.

### Exposure Analysis and Scenario Dose Conversion Factors

The performance objective for atmospheric releases (0.01 rem per year) includes doses from all potential exposure pathways and intake routes associated with such releases. The annual doses for the two volatile radionuclides of interest ( $^3\text{H}$  and  $^{14}\text{C}$ ) were calculated from four exposure pathways: (1) inhalation of airborne radionuclides, (2) ingestion of vegetation exposed to airborne radionuclides (i.e., airborne-contaminated



vegetation), (3) ingestion of beef from cattle consuming airborne-contaminated vegetation, and (4) ingestion of milk from cows consuming airborne-contaminated vegetation.

No external doses are expected since the beta particles emitted by these two radionuclides have very low energies; in fact, these two radionuclides have external dose conversion factors equal to zero.<sup>9</sup> The total dose from the atmospheric transport pathway is the sum of the doses from each of the four exposure pathways listed above. The equations used to calculate each exposure pathway dose are described below and the parameter values used in these equations are listed in Table 1. This methodology is based on a conservative specific activity model presented in NRC Regulatory Guide 1.109.<sup>10</sup> Note that because this is a pathway analysis, radioactive decay is not included in the calculations. To determine dose or, conversely, permissible waste limits, radioactive decay at the time of exposure must be taken into account.

The dose from direct inhalation of volatiles is calculated using the following equation:

$$H_{inh} = C_{Atm} \times IR_a \times DCF_{inh} \quad (17)$$

where

$H_{inh}$  is the annual dose from inhalation in air (rem/year),  
 $IR_a$  is the intake rate of air (adult inhalation rate) ( $m^3$ /year), and  
 $DCF_{inh}$  is the internal dose conversion factor from inhalation (rem/ $\mu$ Ci).

To calculate the doses due to  $^3H$  contamination in food, it is assumed that the source of the contamination is the air surrounding the vegetation. The concentration in the vegetation is based on the amount of water in the vegetation and the amount of  $^3H$  that would be in the plant water. The equation for calculating the concentration of  $^3H$  in vegetation is as follows:

$$C_{v,H-3} = (C_{a,H-3} / AH) \times R_{v/w/a} \times f_{vw} \quad (18)$$

where

$C_{v,H-3}$  is the concentration of  $^3H$  in vegetation ( $\mu$ Ci/kg),  
 $C_{a,H-3}$  is the concentration of  $^3H$  in air ( $\mu$ Ci/ $m^3$ ),  
 $AH$  is the absolute humidity of the atmosphere (kg/ $m^3$ ),  
 $R_{v/w/a}$  is the concentration ratio of  $^3H$  in vegetation water to  $^3H$  in atmospheric water (dimensionless),  
 and  
 $f_{vw}$  is the fraction of vegetation that is water (dimensionless).

The concentration of  $^3H$  in air,  $C_{a,H-3}$ , divided by the absolute humidity,  $AH$ , is equivalent to the concentration in water. This, when multiplied by the fraction of water in the vegetation,  $f_{vw}$  and the concentration ratio of  $^3H$  in vegetation water to atmospheric water,  $R_{v/w/a}$  equals the concentration of  $^3H$  in the vegetation.

To estimate the doses from the ingestion of  $^{14}C$ -contaminated food, it is again assumed that the source of the contamination is the air surrounding the vegetation. It is also assumed that the ratio of  $^{14}C$  to the natural carbon in vegetation is the same as the ratio of  $^{14}C$  to natural carbon in the atmosphere surrounding the

vegetation. For airborne releases, it is also assumed that plants obtain all their carbon from airborne CO<sub>2</sub> and that animals obtain all their carbon through ingestion of plants.<sup>10,11</sup>

The equation for calculating the concentration of <sup>14</sup>C in vegetation from contaminated air is:

$$C_{v,C-14} = (C_{air,C-14} / C_{air,C}) \times f_{Cv} \quad (19)$$

where

$C_{v,C-14}$  is the concentration of <sup>14</sup>C in vegetation ( $\mu\text{Ci}/\text{kg}$ ),  
 $C_{air,C-14}$  is the concentration of <sup>14</sup>C in air ( $\mu\text{Ci}/\text{m}^3$ ),  
 $C_{air,C}$  is the concentration of natural carbon in air ( $\text{kg}/\text{m}^3$ ), and  
 $f_{Cv}$  is the fraction of natural carbon in vegetation (dimensionless).

The concentration in vegetation is then used to calculate the dose from ingestion of contaminated vegetation. As shown in Table 1, approximately 50% of the exposed person's vegetable intake is assumed to involve contaminated vegetation.<sup>3</sup> The resulting dose can be estimated as follows:

$$H_v = C_v \times IR_v \times DCF_{ing} \quad (20)$$

where

$H_v$  is the annual dose in vegetation (rem/year),  
 $C_v$  is the concentration in vegetation ( $\mu\text{Ci}/\text{kg}$ ),  
 $IR_v$  is the adult intake rate of vegetables (kg/year), and  
 $DCF_{ing}$  is the internal dose conversion factor for ingestion (rem/ $\mu\text{Ci}$ ).

The concentration in vegetation is also used to determine the concentration in beef and milk that will be consumed by humans. It is conservatively assumed that 100% of the cow's vegetation consumption is from grazing on fresh pasture grass contaminated with airborne radionuclides and that the animal grazes 365 days of the year. The concentrations in beef and in cow's milk are estimated as follows:

$$C_b = C_v \times F_b \times IR_{v(cow)} / (365 \text{ days/year}) \quad (21)$$

$$C_m = C_v \times F_m \times IR_{v(cow)} / (365 \text{ days/year}) \quad (22)$$

where

$C_b$  is the concentration in beef from cattle that consumed contaminated vegetation ( $\mu\text{Ci}/\text{kg}$ ),  
 $C_m$  is the concentration in milk from dairy cows that consumed contaminated vegetation ( $\mu\text{Ci}/\text{L}$ ),  
 $C_v$  is the concentration in vegetation consumed by beef cattle or dairy cows,  
 $F_b$  is the ratio of equilibrium concentration in meat to daily intake by beef cattle ( $\mu\text{Ci}/\text{kg}$  in meat per  $\mu\text{Ci}/\text{d}$  intake),

$F_m$  is the ratio of equilibrium concentration in milk to daily intake by dairy cows ( $\mu\text{Ci/L}$  in milk per  $\mu\text{Ci/d}$  intake), and

$IR_{(cow)}$  is the consumption rate of vegetation by beef cattle or dairy cows (kg/year).

The annual radiation doses from the ingestion of beef and milk are calculated by multiplying the concentration in each medium by the human intake rate and the radionuclide-specific internal dose conversion factor:

$$H_b = C_b \times IR_b \times DCF_{ing} \quad (23)$$

$$H_m = C_m \times IR_m \times DCF_{ing} \quad (24)$$

where

$H_{(b,m)}$  is the annual dose in beef (b) or milk (m) (rem/year),

$C_{(b,m)}$  is the concentration in beef (b) ( $\mu\text{Ci/kg}$ ) or milk (m) ( $\mu\text{Ci/L}$ ),

$IR_{(b,m)}$  is the adult intake rate of beef (b) (kg/year) or milk (m) (kg/L), and

$DCF_{ing}$  is the internal dose conversion factor for ingestion (rem/ $\mu\text{Ci}$ ).

The annual dose from atmospheric releases,  $H_T$ , is the sum of the annual doses from the four exposure pathways described above (i.e., inhalation of contaminated air and ingestion of contaminated vegetables, beef, and milk):

$$H_T = H_{inh} + H_v + H_b + H_m \quad (25)$$

Each of the terms in Equation 25 was normalized using a unit air concentration  $1 \mu\text{Ci/m}^3$  to yield scenario dose conversion factors,  $SDCF_{Atm}$ , of  $8.33 \times 10^2$  mrem/year per  $\mu\text{Ci/m}^3$  for  $^3\text{H}$  and  $2.20 \times 10^5$  mrem/year per  $\mu\text{Ci/m}^3$  for  $^{14}\text{C}$ . Table 2 illustrates the contribution of each pathway to the overall dose. The  $SDCF_{Atm}$  for  $^3\text{H}$  is dominated by inhalation while the  $SDCF_{Atm}$  for  $^{14}\text{C}$  is dominated by vegetable consumption (see Table 2).

#### Calculating Permissible Waste Concentrations via the Atmospheric Pathway

The back-calculation to determine the permissible waste concentration,  $C_{W-Atm}$  ( $\mu\text{Ci/m}^3$ ), is:

$$C_{W-Atm} = C_{Atm} \times CRF_{diff} \times CRF_{disp} \times r_{decay} \quad (26)$$

where,  $r_{decay}$  accounts for the radioactive decay of  $^3\text{H}$  and  $^{14}\text{C}$  during detention time in the waste plus the travel time,  $t$ , via environmental transport to the receptor and is given by:

$$r_{decay} = \exp\left[\frac{\ln(2)t_a}{t_{1/2}}\right] \quad (27)$$

where

$t_a$  is the radionuclide detention time (100 years) in the RCRA-covered disposal facility plus  $t$ , and because  $t$  is very small, is approximately equal to 100 years.  
 $t_{1/2}$  is the half-life of  $^3\text{H}$  (12.3 years) or  $^{14}\text{C}$  (5730 years).

The radionuclide concentration in the atmosphere can be expressed as:

$$C_{Atm} = \frac{H_{Atm}}{SDCF} \quad (28)$$

where,  $H_{Atm}$  is the performance measure of 0.01 rem per year for all pathways resulting from atmospheric releases. Substituting Equations 27 and 28 into the Equation 29 yield the overall equation used to determining the permissible waste concentration,  $C_{W-Atm}$ :

$$C_{W-Atm} = H_{Atm} \times \frac{1}{SDCF_{Atm}} \times CRF_{diff} \times CRF_{disp} \times \exp\left[\frac{\ln(2)t_a}{t_{1/2}}\right] \quad (29)$$

## RESULTS

Figure 3 shows  $C_{W-Atm}$  calculated for the 15 DOE sites. Even though site-specific data were used in the calculations, results between sites did not appreciably differ. For  $^3\text{H}$ ,  $C_{W-Atm}$  values ranged from a low of  $2 \times 10^9 \mu\text{Ci}/\text{m}^3$  for the Hanford Site to a high of  $3 \times 10^{10} \mu\text{Ci}/\text{m}^3$  for the Savannah River Site. Generally, arid sites (i.e., sites in the western United States) had lower permissible waste concentrations because the fraction saturation in the soil pore space,  $s$ , was lower than the humid site (i.e., sites located in the eastern United States). A lower value of  $s$  leads to higher radionuclide concentrations in the water trapped in the soil, and therefore, a greater amount of the radionuclide would be released into the atmosphere. The same effect occurs for  $^{14}\text{C}$ , where the  $C_{W-Atm}$  ranges from a low of  $1 \times 10^3 \mu\text{Ci}/\text{m}^3$  at the Hanford and Los Alamos sites to a high of  $2 \times 10^4 \mu\text{Ci}/\text{m}^3$  at the Savannah River Site. For all sites, the greatest concentration reduction occurred as the radionuclides diffused upward through the soil compared to the concentration reduction that occurred with atmospheric dispersion.

Site-specific meteorology resulted in only minor differences in  $C_{W-Atm}$  among all of the sites. Meteorological data (i.e., wind speed and direction, and atmospheric stability class) were used to develop the atmospheric dispersion term,  $A_D$ , which varied over a relatively small range among the sites (minimum was 0.17 at Argonne National Laboratory and maximum was 0.75 at Oak Ridge Reservation). Lower values of  $A_D$  were associated with sites that exhibited relatively even distributions of wind directions and higher values of  $A_D$  were associated with sites where distributions favored specific wind directions. In addition, the effect of varying meteorological parameters among sites was minimized because the performance boundary of 100 m was so close to the disposal facility that significant dispersion effects were not developed.

Calculated values of  $C_{W-Atm}$  for  $^3\text{H}$  were about six orders of magnitude higher for  $^3\text{H}$  than for  $^{14}\text{C}$ . A little over two orders of magnitude (about a factor of 270) are accounted for in  $SDCF_{Atm}$  where the  $^{14}\text{C}$  contributes a much higher dose per curie. Another two orders of magnitude are accounted for in the radioactive decay term.  $^3\text{H}$  decays much more quickly than  $^{14}\text{C}$  and much of the amount of  $^3\text{H}$  is substantially reduced at 100 years. The last two orders of magnitude are accounted for in the ratio term,  $r$ .

These results were compared to similar analyses performed for the water and inadvertent intrusion pathways. The atmospheric pathway was limiting (i.e., resulted in the lowest  $C_{W-Atm}$ ) for  $^{14}\text{C}$  at Portsmouth Gaseous Diffusion Plant and at all arid sites except Rocky Flats Environmental Technology Site. The water pathway was limiting for  $^{14}\text{C}$  at all remaining sites.  $^3\text{H}$  was not limited by the atmospheric pathway at any of the sites. The intruder pathway was limiting at most sites, with the water pathway limiting at the remaining sites<sup>1</sup>.

## SUMMARY AND CONCLUSIONS

A screening method was developed to compare the doses received via the atmospheric pathway at 15 potential DOE MLLW sites. Permissible waste concentrations in the potential disposal facilities were back-calculated using a performance objective applicable to atmospheric releases (i.e., the radioactivity NESHAP, which specifies that doses from atmospheric emissions shall not exceed 0.01 rem per year through all intake exposure routes). Site-specific soil and meteorological data were used to determine permissible waste concentrations.

When looking at a particular radionuclide, results show that permissible waste concentrations for each site do not vary by more than about one order of magnitude. Permissible waste concentrations of  $^{14}\text{C}$  are about six orders of magnitude more restrictive than permissible waste concentrations of  $^3\text{H}$  because of differences in liquid to vapor partitioning, radioactive decay, and exposure dose. When comparing results from the atmospheric pathway to the water and intruder pathways,  $^{14}\text{C}$  disposal concentrations were limited by the atmospheric pathway for most arid sites. For  $^3\text{H}$ , the atmospheric pathway was not limiting at any of the sites compared to the water and intruder pathways.

The results of the PE analysis are to be used by decision-makers to begin planning for siting of disposal facilities. This screening analysis is to be followed by more in-depth performance assessments at selected sites to determine disposal facility design and waste inventory.

## REFERENCES

1. R. D. Waters, M. M. Gruebel, M. B. Hospelhorn, A. M. Parsons, B. M. Thomson, M. S. Y. Chu, G. P. Zimmerman, J. D. Tauxe, D. A. Lombardi, M. L. Socolof, J. Wang, D. Kocher, and D. W. Lee, *Performance Evaluation of the Capabilities of DOE Sites for Disposal of Mixed Low-Level Waste, Volume 2: Technical Basis and Discussion of Results*. U.S. Department of Energy, Idaho Falls, Idaho, 1996.
2. D. E. Wood, R. U. Curl, D. R. Armstrong, J. R. Cook, M. R. Dolenc, D. C. Kocher, K. W. Owens, E. P. Regnier, G. W. Roles, R. R. Seitz, and M. I. Wood, *Performance Assessment Task Team Progress Report*. DOE/LLW-157, Rev. 1., Idaho National Engineering Laboratory, Idaho Falls, Idaho, 1994.

3. Martin Marietta Energy Systems, Inc., EG&G Idaho, Inc., and Westinghouse Savannah River Company, 1994. *Radiological Performance Assessment for the E-Area Vaults Facility (U)*, WSRC-RP-94-218. Aiken, SC: Westinghouse Savannah River Company, 1994.
4. Faust *et al.*, *Principles of Unit Operations*. New York, NY, John Wiley & Sons, Inc., 1960.
5. C. D. Cooper and F. C. Alley, *Air Pollution Control: A Design Approach*. Prospect Heights, Illinois, Waveland Press, Inc., 1986.
6. S. J. Maheras *et al.*, *Radioactive Waste Management Complex Low-Level Waste Radiological Performance Assessment*. EGG-WM-8773. Idaho Falls, ID: Idaho National Engineering Laboratory, 1994.
7. H. B. Fisher, E. J. List, R. C. Koh, J. Imberger, and N. H. Brooks, *Mixing in Inland and Coastal Waters*. New York, NY, Academic Press, 1979.
8. U.S. Environmental Protection Agency, *User's Guide for the Industrial Source Complex (ISC2) Dispersion Models, Volume I: User Instructions*, EPA Publication No. EPA-450/4-92-008a. Research Triangle, NC, 1992.
9. U. S. Department of Energy, *Internal Dose Conversion Factors for Calculation of Dose to the Public*, DOE/EH-0071, July, 1988.
10. U. S. Nuclear Regulatory Commission, *Regulatory Guide 1.109. Calculation of Annual doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I*, 1977.
11. Napier *et al.*, *GENII - The Hanford Environmental Radiation Dosimetry Software System, Volume 1: Conceptual Representation*, Battelle, Pacific Northwest Laboratory, Hanford, Washington, December, 1988.
12. International Commission on Radiological Protection, *International Commission on Radiological Protection: Task Group Report on Reference Man*, ICRP Publication No. 23, Pergamon Press, New York, 1975.
13. U. S. Environmental Protection Agency, *Limiting Values of Radionuclides Intake and Air Concentration and Dose Conversion Factors for Inhalation, Submersion, and Ingestion*, Federal Guidance Report No. 11, Office of Radiation Programs, EPA 520/1-88-020, 1988.
14. U. S. Environmental Protection Agency, *User's Guide for CAP88-PC Version 1.0, Air and Radiation*, 402-B-92-001, Las Vegas, Nevada, 1992.

Table 1. Parameter values used to determine doses from all potential exposure pathways associated with atmospheric emissions from waste disposal facilities.

Parameter	Value	Nuclide-specific value		Reference number
		<sup>3</sup> H	<sup>14</sup> C	
$IR_a$ (adult average) (m <sup>3</sup> /year)	8000			12
$DCF_{inh}$ (rem/ $\mu$ Ci)		6.4E-05	2.20E-05 <sup>b</sup>	13
$AH$ (kg/m <sup>3</sup> )	0.0092			3
$R_{ww/wa}$ (dimensionless)	0.5			10
$f_{ww}$ (dimensionless)	0.75			10
$C_{air,C}$ (kg/m <sup>3</sup> )	0.00016			10, 11
$f_{cv}$ (dimensionless)	0.11			10
$IR_v$ (kg/year)	90 <sup>a</sup>			3
$DCF_{ing}$ (rem/ $\mu$ Ci)		6.4E-05	2.1E-03	13
$F_b$ (day/kg)		1.2E-02	3.1E-02	10
$IR_{v(cow)}$ (kg/year)	5694			14
$F_m$ (day/L)		1.0E-02	1.2E-02	10
$IR_b$ (kg/year)	85			14
$IR_m$ (L/year)	112			14

<sup>a</sup>Assumes approximately half of an individual's vegetable intake is from locally-grown, contaminated vegetation.

<sup>b</sup>Carbon as CO<sub>2</sub>.

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Table 2. All-pathways dose resulting from exposure to air with an ambient concentration of  $1\mu\text{Ci}/\text{m}^3$  of  $^3\text{H}$  or  $^{14}\text{C}$ .

Pathway	Radionuclide			
	$^3\text{H}$		$^{14}\text{C}$	
	Dose (mrem/year)	Percent of total dose	Dose (mrem/year)	Percent of total dose
Direct inhalation	512	61	190	<1
Consumption of contaminated vegetables	234	28	129,900	59
Consumption of contaminated beef	42	5	59,380	27
Consumption of contaminated milk	45	6	30,390	14
<b>TOTAL</b>	<b>833</b>	<b>100</b>	<b>219,860</b>	<b>100</b>



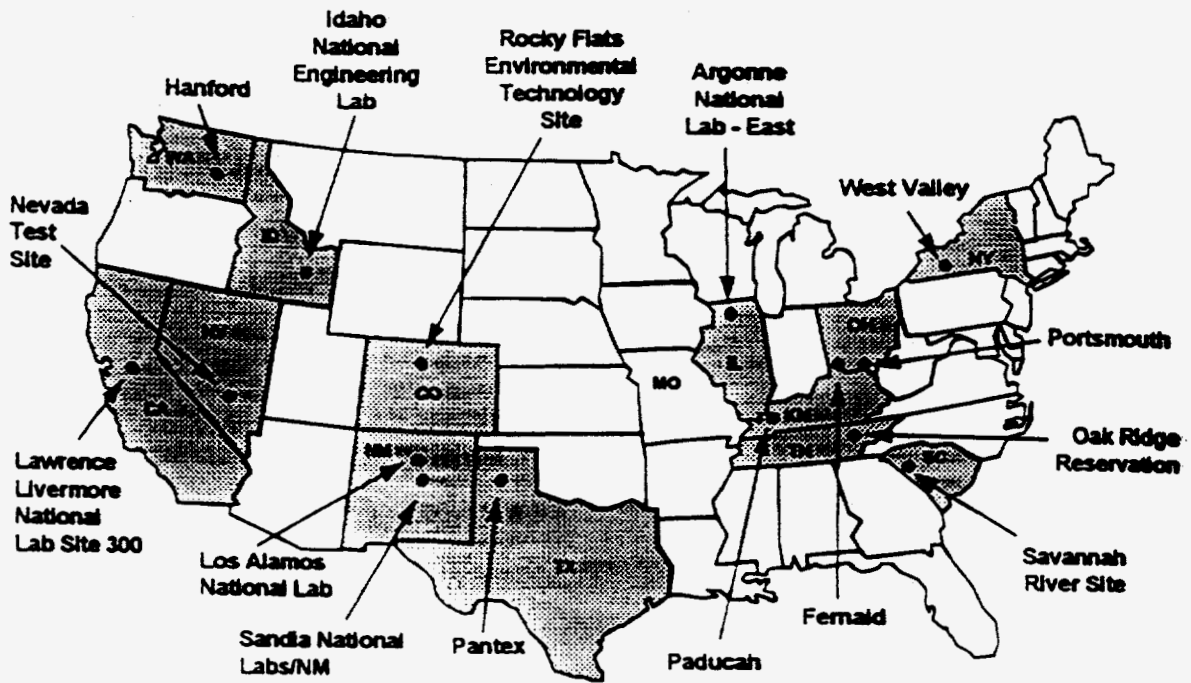


Figure 1. Sites considered in the Performance Evaluation.

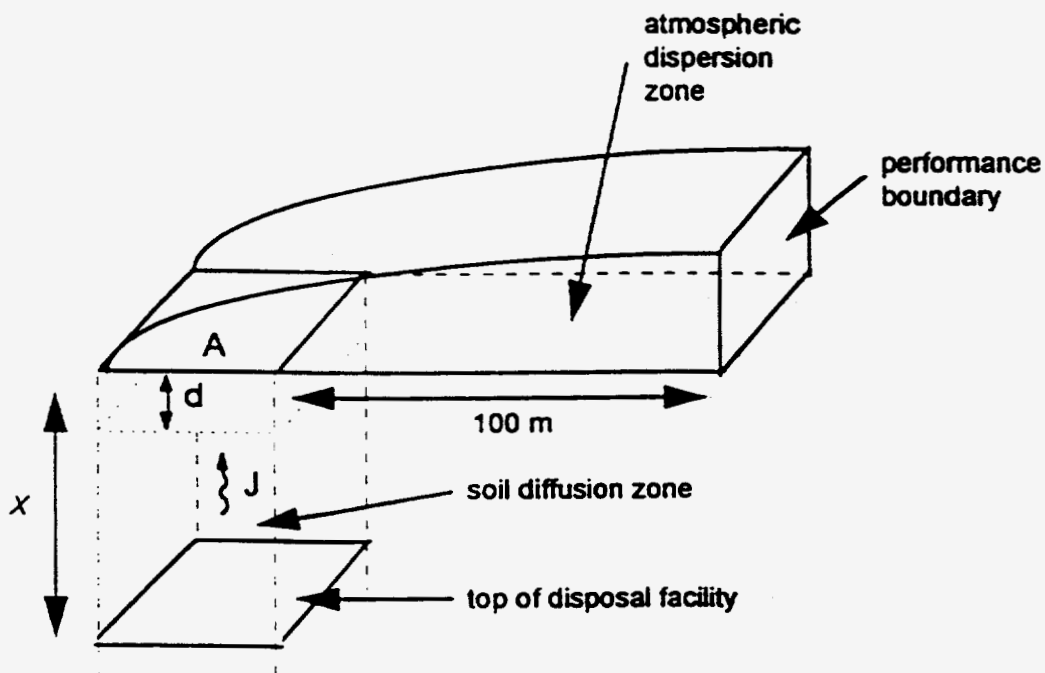


Figure 2. Conceptual model for the atmospheric pathway.

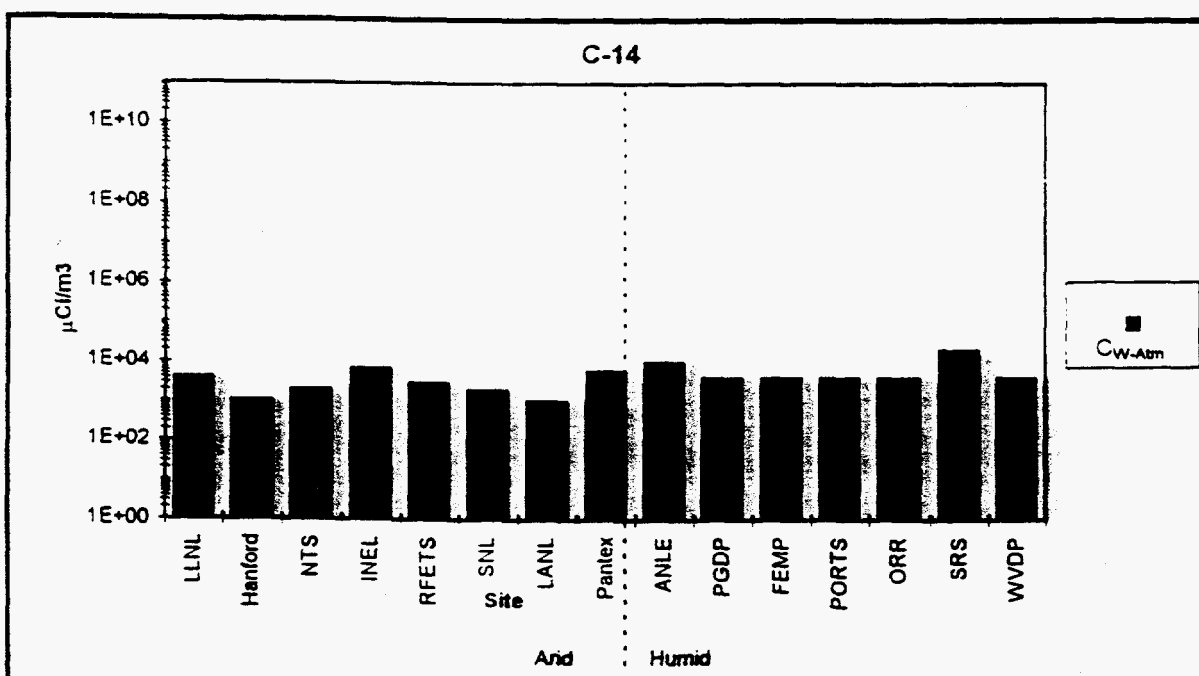
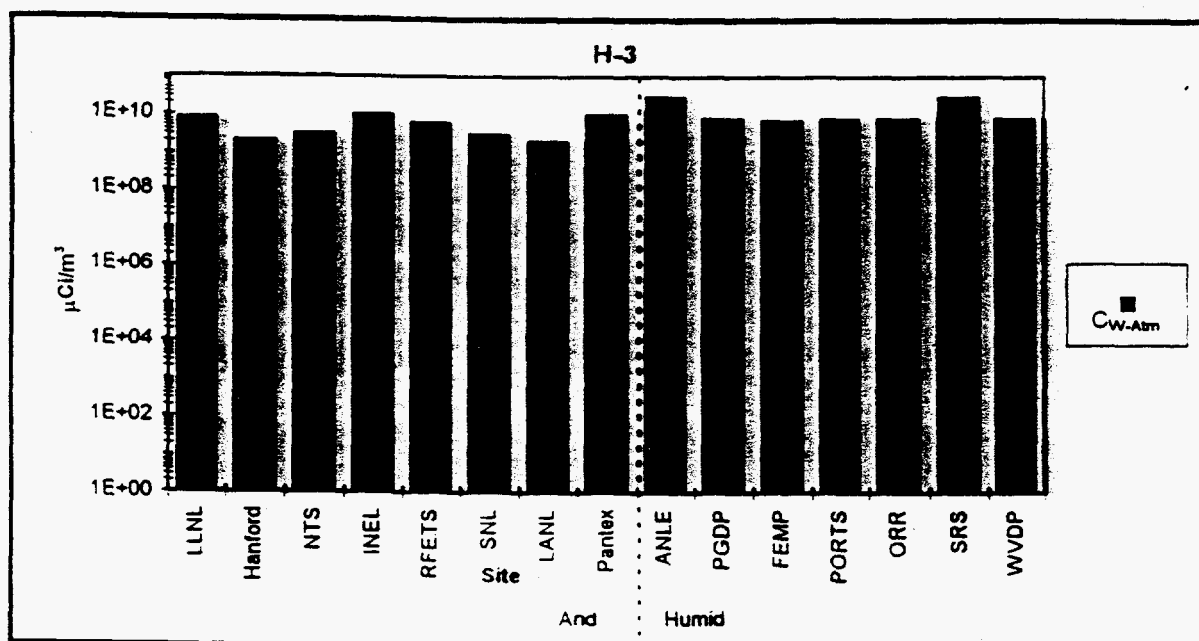


Figure 3. Permissible waste concentrations ( $\mu\text{Ci}/\text{m}^3$ ) for  $^3\text{H}$  and  $^{14}\text{C}$  for the atmospheric pathway.