DEVELOPMENT OF DUST: A COMPUTER CODE THAT CALCULATES RELEASE RATES FROM A LLW DISPOSAL UNIT

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ABSTRACT

Performance assessment of a Low-Level Waste (LLW) disposal facility begins with an estimation of the rate at which radionuclides migrate out of the facility (i.e., the disposal unit source term). The major physical processes that influence the source term are water flow, container degradation, waste form leaching, and radionuclide transport. A computer code, DUST (Disposal Unit Source Term) has been developed which incorporates these processes in a unified manner. The DUST code improves upon existing codes as it has the capability to model multiple container failure times, multiple waste form release properties, and radionuclide specific transport properties. Verification studies performed on the code are discussed.

INTRODUCTION

Evaluation of the radiological impacts of LLW disposal is accomplished through a performance assessment which includes estimation of the following processes for each radionuclide: (a) the rate of release from the disposal unit (i.e., the source term); (b) the transport from the disposal unit to the accessible environment; and (c) the conversion of the radionuclide concentration at the receptor site into an equivalent dose.

In general, the source term is influenced by the radionuclide inventory and its origin (i.e., waste stream), the waste forms and containers used to dispose of the inventory, and the physical processes that influence release from the facility (fluid flow, container degradation, waste form leaching, and radionuclide transport).

The inventory data during the period of 1987 - 1989 from the three commercial LLW sites have been reviewed to determine which of the various radionuclides, waste streams, and waste forms are most widely used. This is discussed in a companion paper (1).

This paper will discuss the development of the computer code DUST (Disposal Unit Source Term) which calculates release rates from the disposal facility. Currently, computer codes that predict release from the disposal facility either are very restrictive in the modeling of releases or require extensive input and computer execution times. The objective behind the development of the DUST code is to improve upon existing models by adding the flexibility to model multiple waste forms and containers each with their own performance parameters while still retaining a code that is relatively easy to use and can be executed quickly on desk top computer systems. The output of the DUST code will be compatible with the PAGAN performance assessment methodology developed for the Nuclear Regulatory Commission (NRC) by Sandia National Laboratories (2).

To verify that the DUST computer code works as intended a number of test problems were performed and the results of these simulations are presented.

MODEL SELECTION

A LLW disposal unit is a complex, heterogeneous collection of wastes/waste forms/containers, soils, and engineered structures (clay caps, concrete vaults, drains, etc.). Release of most radionuclides from such a system is controlled by access of water to the waste form, release of the radionuclide from the waste form, and transport to the disposal unit boundary. These processes are influenced by the design of the disposal unit, precipitation, hydrology, geochemistry, and waste form/container characteristics. To model the complete disposal unit, including every waste container individually would require a three dimensional model that considered all of these processes simultaneously. Even if such a model did exist, its use would require extensive computing times and the accuracy of the predictions would be questionable due to limitations in the data.

Therefore, simplifications from a fully descriptive three-dimensional model are justified. These "simplified" models should account for the most important physical processes and parameters influencing release while retaining as much accuracy as possible. Further, the models should be flexible enough to simulate a wide range of conditions and not be overly conservative. For example, one could require that all of the containers fail instantly upon emplacement. Such a model would be conservative, but not realistic and inflexible. A better model would be one that allowed a range of container failures times based on the container properties. Such a model would have the flexibility to permit simulation of extremely low probability worst case scenarios, such as instantaneous failure of all containers, as well as more likely scenarios, such as time-distributed container failures.

The DUST computer code has been developed in a general manner which allows the simulation of the majority of situations expected to occur. However, to account for the possibility of special cases and to allow easy modifications of the models to reflect new and better information, the code structure is modular. The following sections present the models selected to simulate the four physical processes believed to be most important in controlling release, e.g. water flow, container degradation, waste form leaching, and radionuclide transport.

Water Flow

Disposal facilities will most probably be located above the water table in the unsaturated zone. Infiltration of water into a facility will involve many processes including precipitation, evapotranspiration, and surface run-off. This is further complicated by the engineered barriers (trench cap, concrete

structures, etc.) any disposal facility will have to minimize water flow into the waste containing region. Finally, the flow rate will vary in time due to short term events (precipitation) and long term events (barrier degradation). Even if the change in material properties and precipitation rates are known, water flow in the unsaturated zone is difficult to model due to the non-linearity of the flow properties.

A simple model for infiltration into a disposal unit does not exist. Further, there is no acceptable model for predicting infiltration into *soils* in arid sites under all conditions (3). The complexities involved with modeling the evolution of unsaturated flow through a disposal unit requires extensive computing expense. This is inconsistent with the objective for having a simplified model that executes quickly on desktop systems.

For the above reason, in the DUST computer code water flow is modeled through tabular input of the volumetric flow velocity (Darcy velocity) versus time. This velocity could be determined through more detailed calculations or chosen conservatively. If a computer simulation is not performed an upper bound for the flow rate is the annual precipitation rate. If the evapotranspiration rate is accurately known this could be subtracted from the precipitation rate at humid sites. At arid sites this may lead to large errors in predicted recharge (3). Alternatively, if the recharge rate through the disposal facility is known due to measurement at the site, this value could be used.

In the actual situation, infiltration may be very low until significant degradation of the trench cap occurs. If one accounts for degradation of the trench cap, this will require additional modeling. At this time, there is no widely accepted model for the degradation of earthen materials or engineered (i.e., concrete) trench caps. However, work is being performed to determine the degradation mechanisms of underground concrete structures. Models based on these studies are under development (4, 5, 6).

Container Degradation

Based on a review of the commercial disposal inventory data it appears that carbon steel drums and boxes are most widely used to dispose of Class A wastes, the largest volumes of wastes. Most class B and C wastes are disposed of in High Integrity Containers (HIC's) but a small fraction have been stabilized in cement and placed in carbon steel 55 gallon drums. Recently, there has been a trend to rely more and more exclusively on HIC's for Class B and C wastes due to their ease of use, lack of need for processing equipment, reduced worker exposure, problems encountered with solidification of some waste streams, and their approval by NRC as a means of demonstrating structural stability.

Mechanistic modeling of the degradation of the container materials requires knowledge of the local chemistry and is quite complicated. Further, the data for mechanistic modeling of corrosion of container materials is not well known in a disposal environment and subject to large uncertainties. These uncertainties would lead to large uncertainties in the predicted corrosion rates. Therefore, the work required to perform such a calculation is probably not justified within the framework of the DUST computer code. Rather, container degradation is modeled for each container through two processes:

a. time to failure after which the container no longer protects the waste form from contact with water;
b. partial failure due to localized effects (e.g. pitting) which permits earlier, limited access of water to the waste form.

Selection of the time to failure should be based on site specific data. When this is not possible, for carbon and stainless steels, the data base generated by the National Bureau of Standards (currently, National Institute of Standards and Technology) could be used to estimate container life (7,8). A summary of this data can be found in Ref. 9.

An analysis of the data necessary to use the localized failure model has been presented elsewhere (9,10). This model allows the fractional area of the container that has failed to increase with time until the entire container is breached, or until the time of failure as specified by a) above is reached. This model provides a means of estimating releases due to localized failure that occur earlier in time than general failure. A detailed discussion of the model, its input parameters, and selection of appropriate values for the parameters has been reported previously (9,11). The data collected applies only to carbon steel systems. For other materials, the code user must justify the choice of model parameters.

Waste Form Release

Radionuclide release from the waste form begins immediately after container failure. In a LLW facility, there will be several different waste forms, a partial list of which includes: activated metals; wastes solidified by one of several processes (cement, VES, bitumen); compacted lab trash, dewatered resins; and filter media (12). Each of these may have its own release mechanism. To cover a wide range of situations, four nuclide specific waste form release models are incorporated into the DUST computer code. They are:

a. Surface rinse limited by partitioning;
b. Diffusion through solidified waste forms;
c. Dissolution at a constant rate; and
d. Solubility limited release.

In general, a waste form may release radionuclides by more than one mechanism. This will be modeled in the DUST code through user-supplied input. For example, the user could specify that 10% of the mass is released through a rinse process with partitioning and 90% is controlled by diffusion. This flexibility may be useful when homogenizing the number of waste streams/waste forms or in modeling large boxes containing several waste forms.

The surface rinse model assumes that the radionuclides with this release characteristic are available as soon as water contact occurs. Prior to water accessing the wastes, the radionuclides may be held on the surface by adsorption, chemisorption, adhesion, and ion exchange processes. To account for these factors a partition factor, which is an equilibrium ratio relating the amount on the waste to that in solution, can be used. This partition factor depends on the properties of the waste form and the local chemistry. Obtaining a reliable estimate of this factor may be difficulty because it is a lumped parameter that represents many physical processes.

Experimental leaching data from solidified wastes often indicate that diffusion is the rate controlling process. In fact, the ANS 16.1 standard leach test (13) and the NRC technical position on waste forms (14) both interpret leach tests data in terms of diffusion. Diffusion controlled release is

characterized by relatively high leach rates at early times that continually decrease in time. In fact, analytically, the release rate, although it is integrable, approaches infinity as time approaches zero. For this reason, a release model based on a constant release rate may prove to be difficult to justify for diffusion controlled release. Choosing a constant release rate based on short term releases may be overly conservative while choosing the rate based on some type of average value may underpredict early releases. Therefore, one of the release models in the DUST code is based on diffusion.

The diffusion model considers two geometries most widely used in LLW disposal: cylindrical (drums) and rectangular (boxes). To simplify the situation it is assumed that the concentration in the contacting solution is zero. This leads to the highest predicted release rates and permits an analytical solution to be obtained. These analytical solutions account for radioactive decay and have been presented elsewhere (11,15).

The dissolution model assumes that radionuclides are released congruently. The release rate is assumed to be constant in time and limited by solubility constraints. It would be appropriate for activated metals which undergo corrosion. In this case, a constant release rate may be conservative because the data indicates decreasing corrosion rates in time.

Solubility limited release is modeled by allowing an instantaneous release of radionuclides into solution until the limit is reached. Further, if a solubility limit is specified and other release mechanisms are used to predict release, the amount released is constrained such that the limit is not exceeded. In general, the chemistry that occurs within a disposal facility is complex and changes in time due to the degradation of the containers and waste forms. Obtaining reliable solubility limits in this environment is a difficult task. Any choice of a solubility limit must be justified as conservative under all of the potential conditions.

A detailed description of the mathematics and the models can be found in Ref. 11.

Radionuclide Transport

Radionuclide transport is modeled through one of two models:

A. Mixing-cell Cascade Model

The improved mixing-cell cascade model (11) divides the disposal unit into a number of uniform size mixing cells. It is assumed that each cell is well mixed and therefore, of uniform concentration. Mass is transported from one cell to the next by advection as limited by retardation and radioactive decay. The models used in the DUST code have been generalized to include cell-wise non-uniform sources (i.e., waste form release rates) and exponentially decaying sources while still retaining an exact analytical solution for the concentration in any cell as a function of time.

The governing differential equation for each mixing cell in this model is:

$$\frac{\partial C_i}{\partial t} = -\frac{V_d}{\theta R}\frac{\partial C_i}{\partial x} - \lambda C_i + \frac{q_i}{\theta R} \qquad \text{(Eq. 1)}$$

where:

C = solution concentration;

V_d = Darcy velocity;

θ = the volumetric moisture content of the region (dimensionless);

R = retardation coefficient = $1 + \rho K_d/\theta$

K_d = distribution coefficient

ρ = bulk density

λ = radioactive decay constant;

q = source/sink term used to model release from the waste form.

The resulting analytical solution for an arbitrary number of mixing cells can be found in Ref. 11.

B. One-dimensional Finite Difference Model

The one-dimensional finite difference models the advection-dispersion equation with radioactive decay, non-uniform first order reversible sorption (e.g retardation) and non-uniform sources (waste form release rates). This model is most appropriate when diffusion and dispersion are important transport processes. It is also more appropriate when diffusion controls release from the waste form as the mixing-cell cascade model can not model this case. The governing partial differential equation for this model is:

$$\frac{\partial}{\partial t}(R\theta C) = \frac{\partial}{\partial x}\left(\theta D \frac{\partial C}{\partial x}\right) - \frac{\partial}{\partial x}(V_d C) - \lambda\theta R C + q \qquad \text{(Eq. 2)}$$

where:

D = the diffusion-dispersion coefficient,

$$D = D_{eff} + \frac{a_t |V_d|}{\theta};$$

a_t = transverse dispersivity;

D_{eff} = effective diffusion coefficient.

and all other variables have been previously defined.

VERIFICATION STUDIES

A number of studies have been conducted to verify that the computer code, DUST, correctly calculates the properties of interest (time of container breach, waste form release rates, concentration and flux). Independent testing of the container degradation models (general and local failure) and the waste form release models (diffusion, dissolution, surface rinse with partitioning, and solubility limited) was performed for problems with known analytical solutions.

The finite difference transport model, Eq. (2), was tested for a solute injection at the surface of the media for 5 days. The parameters found in Eq. (2) for this problem are presented in Table I. An analytical solution exists for this problem for a semi-infinite domain (16). This was modeled by taking the domain of the simulation to be 300 cm. and comparing the semi-infinite analytical solution to the numerical solution over the first 100 cm for 10 days.

In test case 1, solute was injected into the media at the boundary x = 0 for 5 days such that the total flux was 1 Curie/cm^2/s. After the fifth day, the flux entering the media is zero. The initial concentration within the simulation domain is zero. At the boundary away from the injection source, the concentration is zero.

Figure 1 presents a comparison of the analytical and numerical solution predicted by the DUST code at 5 and 10 days. The agreement between the two is excellent. The maximum difference between the two solutions is less than 2%.

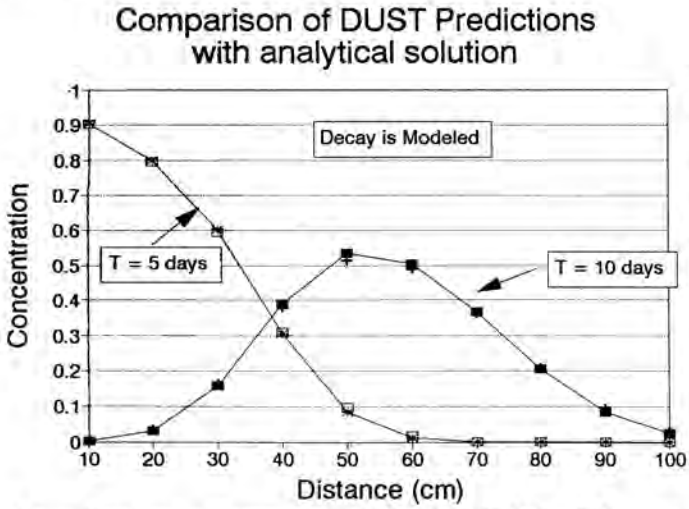

Fig. 1. Test case 1, comparison of the DUST finite difference model predictions with the analytical solution for a pulse source at x = 0 for 5 days. Retardation and decay are modeled. See Table I for the model parameters.

TABLE I

Parameters Used to Verify the Finite Difference Transport Model

	Case 1	Case 2	Case 3
Darcy Velocity (cm/s)	2.89E-4	2.89E-4	2.89E-4
Dispersion/Diffusion (cm^2/s)	4.34E-4	4.34E-4	4.34E-4
Decay constant (1/s)	2.51E-6	0.0	2.51E-6
Source ($Curies/(cm^3-s)$)	0.0	0.0	1.16E-5
Moisture content	0.3	0.3	0.3
Retardation coefficient	3.3	3.3	3.3

Test case 2 was identical to test case 1 with the exception that the decay term was set to zero. This is a slightly more difficult numerical problem because the concentration gradients are larger than in test case 1. Again, the agreement between the analytical and numerical solutions at 5 and 10 days is excellent, Fig. 2.

Test case 3 is similar to cases 1 and 2 except for the following changes. This problem begins with a uniform concentration of 10 $Curies/cm^3$ and a uniform source as given in Table I. The total flux at x = 0 is zero at all times. This problem tests that the external source (i.e., waste form release) term is properly incorporated into the solution procedure.

The results plotted in Fig. 3 at 2, 5, and 10 days indicate that the DUST code is capable of reproducing the analytical solution with a high degree of accuracy.

In the preceding problems, diffusion/dispersion plays an important role in the transport of the radionuclide and therefore, the analytical mixing-cell cascade model can not be used. A number of test cases were run in which the diffusion term was set to zero. This allowed comparison of the 1-D finite

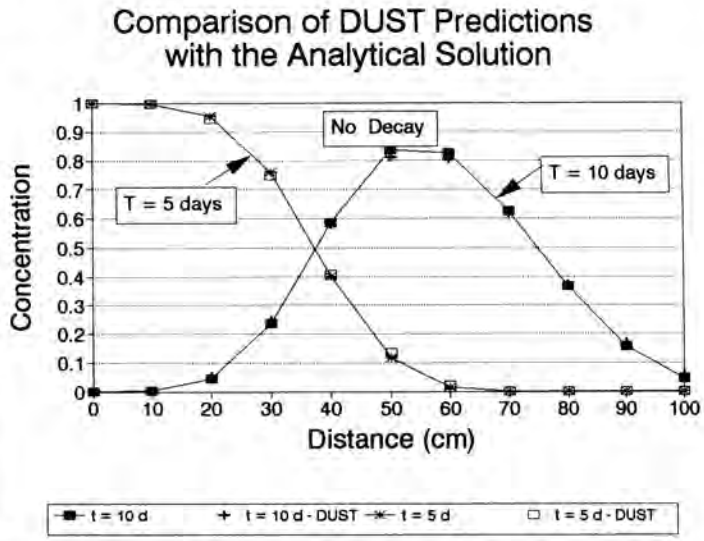

Fig. 2. Test case 2, comparison of the DUST finite difference model predictions with the analytical solution for a pulse source at x = 0 for 5 days. The decay term is zero in this simulation. See Table I for the model parameters.

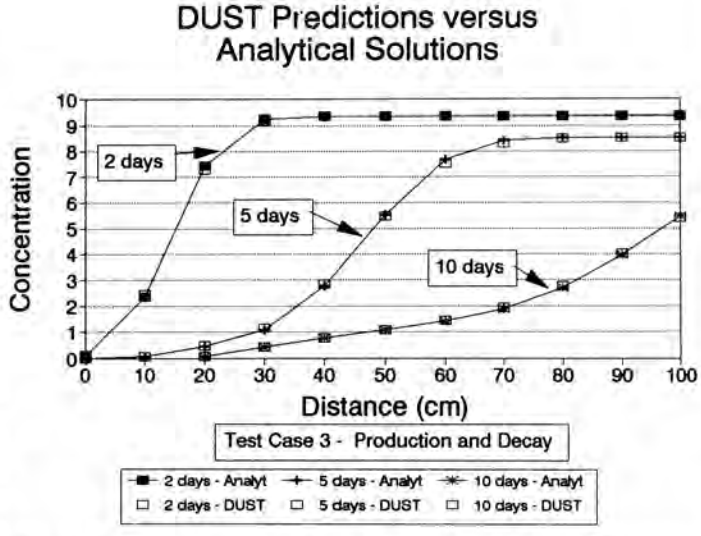

Fig. 3. Test case 3, comparison of the DUST finite difference model predictions with the analytical solution for a uniform initial concentration, zero total flux at the boundary x = 0, and an external uniform source. See Table I for the model parameters.

difference transport model with the mixing-cell cascade model. In addition, the mixing-cell cascade model was compared directly with hand calculations of the analytical solution (11) for several simple problems (few cells, all containers fail simultaneously, constant release rates).

Test case 4 is an example of one of these problems. In test case 4, the domain for the finite difference model is 50 meters. For this simulation, the domain was subdivided into 50 regions each 1 meter in length. The first 10 meters contain only soil. In the next 24 meters there is a waste form every other meter

for a total of twelve waste forms. The container failure times range from 0 to 40 years as presented in Table II. Release from the waste forms is modeled using the dissolution model. The parameters for this model were chosen such that once water contacts the waste form the fractional release rate is 5% per year. The parameters chosen for this test case are presented in Table II. The problem considers radioactive decay, retardation, and advection.

The mixing cell simulation was identical to the finite difference model with the exception that only 40 meters was modeled. For advection driven flow, the 10 meters upstream from the waste form do not receive any contaminant. Thus, the two test cases are identical except that the distance from the top of the simulation domain is offset by 10 meters. The difference is necessary to insure that the boundary condition of the finite difference solution does not influence the results.

Figure 4 presents a comparison of the results of the two simulation techniques at three different locations for a 90 year period. Location 1 is 7 meters from the top of the first waste form, this cell has a container that fails after 30 years. Location 2 is 12 meters from the top of the first waste form. It does not contain a waste form. However, the adjacent upstream cell has a waste container that fails instantly. Location 3 is 23 meters from the top of the first waste form. It has a container that fails after 30 years. Two meters upstream from this waste form is a waste container that fails instantly.

In all locations and at all times, the mixing-cell model and the finite difference model show excellent agreement. At location 1, x = 7 m in Fig. 4, there is a gradual buildup of the concentration in time due to container failures upstream. At 30 years, there is a slight jump in concentration due to the failure of the container in the cell. At location 2, x = 12 m in Fig. 4, there is a rapid rise in concentration at early times due to the failure of the container 1 meter upstream. The concentration peaks after about 10 years and begins to decline for approximately 30 years. At this time, other containers upstream have failed and begun to release contaminants. The concentration shows a local maximum after about 50 years as the containers 3 and 5 meters upstream failed after 40 and 30 years, respectively. At location 3, x = 23 m in Fig. 4, there is a peak after 12 years due to failure of the container 2 m upstream at emplacement. The concentration decreases until 30 years where a sharp increase occurs due to the failure of the container in this cell.

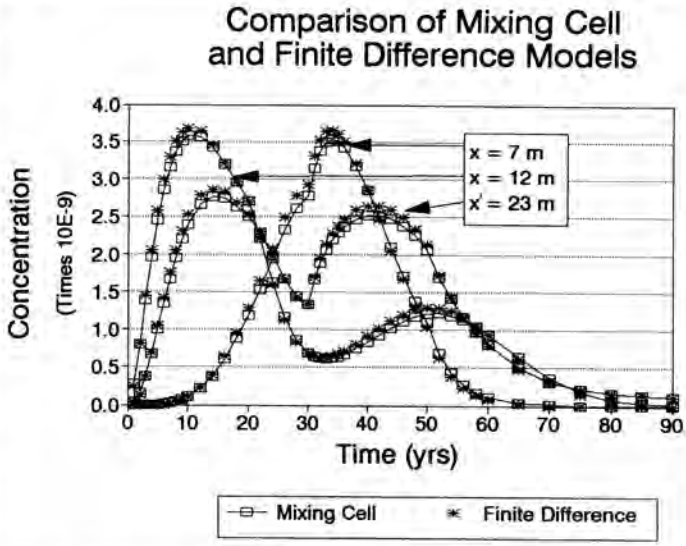

Fig. 4. Test case 4, comparison of the DUST finite difference and mixing cell cascade model at three locations. There are 12 waste forms, each one meter in length separated by one meter of soil beginning at x = 0. The container failure time differs for the waste forms as described in Table II. Release and transport parameters can also be found in Table II.

CONCLUSIONS

A framework for performing estimates of the source term from a LLW disposal facility has been developed. Within this framework, models to predict the processes of water flow, container degradation, waste form leaching, and radionuclide transport have been selected and incorporated into the computer code DUST, (Disposal Unit Source Term).

The DUST code improves upon existing models in that more flexibility is allowed in order to model the various waste form/container systems while still retaining relatively simple models that do not require extensive computer time or provide an undue burden on the code user in terms of input requirements. The DUST code is capable of modeling multiple container failure times and multiple waste form release and contaminant transport properties.

The models in the DUST computer code have been verified through comparison with analytical solutions for problems involving advection, dispersion, decay, retardation, and external sources. The mixing cell cascade model has been

TABLE II

Parameters Used in Test Case 4: Comparison of the Mixing Cell and Finite Difference Model Results.

Container failure times:												
Location (m)	1	3	5	7	9	11	13	15	17	19	21	23
Time (yrs)	0	10	20	30	40	0	10	10	20	20	0	30
Release and Transport Parameters:												
Darcy Velocity (cm/s):	1.59E-6											
Moisture Content:	0.2											
Retardation coefficient	9.0											
Release rate (1/yr)	0.05											
Half-Life (yrs)	12.33											

shown to be accurate through comparison with hand calculations of the analytical solution and through comparison with the finite difference model.

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