PREDICTION OF THE MIGRATION OF RADIONUCLIDES TO THE BOUNDARY OF A SHALLOW LAND BURIAL TRENCH

T. M. Sullivan
Brookhaven National Laboratory
Upton, NY 11973

ABSTRACT

A general model which predicts the "source term," radionuclide release rate, as a function of water flow, container degradation rate, waste form leach rate, and radionuclide migration rate from a low-level waste shallow land burial trench is being developed. This paper discusses modeling radionuclide migration, one component of the "source term." Simulations of radionuclide transport from a generic shallow land burial trench have been performed for a range of water flow rates, dispersivity values, and distribution coefficients. For the modeling assumptions used and the range of parameters tested, the water flow rate plays the major role in redistributing radionuclides within the trench, except in the case of extremely high dispersion. Dispersion was always found to play a significant role in determining transport. This was particularly apparent upstream from the source. Sorption decreased the magnitude of the radionuclide concentration and had the apparent effect of reducing the velocity with which the radionuclides were transported. Diffusion was found to be unimportant in determining radionuclide transport.

INTRODUCTION

A general model which predicts the "source term," radionuclide release rate, as a function of water flow, container degradation rate, waste form leach rate, and radionuclide migration rate from a low-level waste shallow land burial trench is being developed (1). This paper discusses modeling radionuclide migration, one component of the "source term."

The major pathway for release of most radioactive species from a disposal trench will be through the water (tritium and carbon-14 may also be released to the air). Specifically, release will be governed by the processes of advection, dispersion, diffusion, and chemical interactions that change the mobility of the species (e.g. sorption). The relative importance of each of these processes will be a function of the specific radionuclide, the water velocity, and the transport properties within the trench.

This study had two objectives. The first was to determine the relative importance of each of the different processes over a range of expected conditions by conducting a number of numerical simulations in which the transport parameters were varied. Second, any shortages in the data necessary for accurate prediction of radionuclide transport were to be identified.

In this study, the computer code FEMWASTE (2) has been used to predict radionuclide transport (3). FEMWASTE was originally written by Yeh and Ward (2) and has been used by others (4,5) to predict radionuclide transport in shallow land burial sites. We are currently using FEMWASTE-1, the latest version of FEMWASTE. FEMWASTE-1 uses the finite-element method to simulate subsurface transport of radionuclides in the water in both the saturated and unsaturated zones. FEMWASTE-1 is capable of modeling the time-dependent movement of radionuclides in two spatial dimensions due to dispersion, advection, sorption, radioactive decay, sources and sinks within an element, first order decay in the liquid and solid phases, and changes in concentration due to consolidation of the soil.

The remainder of this paper is divided into four major sections. The first section presents the differential equation solved by FEMWASTE-1. The second section, gives a general description of the problems simulated using FEMWASTE-1 and discusses the specification of the geometry of the regions modeled, the source of radionuclides for transport, the initial and boundary conditions, and the material properties. The third section provides the results and a discussion of the influence that advection, dispersion, and sorption have on transport. The fourth section presents conclusions.

GOVERNING EQUATIONS USED TO MODEL CONTAMINANT TRANSPORT

The basic equation is derived from mass balance considerations and takes the form:

$$R \frac{\partial c}{\partial t} = \nabla \cdot \Theta D \nabla C - \nabla \cdot VC + M \qquad (1)$$

where:
$$\theta D_{ij} = a_t |V| \delta_{ij} + (a_\ell - a_t) \frac{V_i V_j}{|V|} + \theta a_m T \delta_{ij} \qquad (2)$$

and
$$R = \left(1 + \frac{\rho k_d}{\theta} \right). \qquad (3)$$

and

C = concentration of the radionuclide in solution (mass of solute/volume of soil);

t = time (second);

S = adsorbed concentration, mass adsorbed per unit mass of soil (mass/mass);

θ = volumetric moisture content, (volume of water/volume of soil);

ρ = bulk density of the soil (mass/volume)

D_{ij} = component of the dispersion tensor D, (area/time);

a_t = transverse dispersivity, (length);

a_l = longitudinal dispersivity, (length);

δij = Kronecker delta;

a_m = molecular diffusion coefficient, (area/time);

T = tortuosity, (dimensionless);

$|V|$ = magnitude of the velocity vector, (volume of flow/area/time);

V_i = i-th component of the velocity vector V, (volume of flow/ area/time);

V_j = j-th component of the velocity vector V, (volume of flow/ area/time);

M = source/sink term for the contaminant, (mass/volume/time);

R = retardation coefficient, ratio of the apparent velocity with which the radionuclide is transported to the velocity with which the water flows.

The term on the left of the equality symbol represents the rate of mass accumulation per unit volume in the liquid and solid phase. The terms on the right of the equality symbol represent the net material accumulation/loss per unit volume due to: dispersion and diffusion, advection, and sources or sinks.

In Eq. (1) it was assumed that the constitutive relationship between the amount of material in the dissolved phase, *to that* adsorbed on the solid phase, was linear, i.e., a linear sorption isotherm was assumed. FEMWASTE-1 can model Freundlich and Langmuir isotherms, however, this was not done for the test problems of this study.

TEST PROBLEM DESCRIPTION

The fundamental problem solved required solution of Eq. (1) for a range of values for dispersion, darcy velocity, and sorption over a region that had material properties and a geometry representative of a shallow land burial trench. The following subsections describe the geometry and material properties used in the simulations.

System Geometry

FEMWASTE-1 models only two spatial dimensions, therefore, we considered a uniform vertical cross-section on an elongated trench, far enough from the ends so that the end effects ideally do not affect the water flow pattern of the section. Thus, we assumed that all flow velocity vectors were essentially in the plane of the cross-section with negligible components directing away from the vertical plane.

This simple model of a shallow land burial trench used for the test runs is shown in Fig. 1. The waste containing area of the trench is taken to be approximately 7 meters deep and 28 meters wide, with the side walls slanting at an angle of approximately 12 degrees from vertical. It is filled with alternating layers of backfill and waste, (the waste region is shaded and labeled as source in Fig. 1). The transport properties are assumed to be uniform within this region. Above the waste region, there is a 1 meter thick clay layer with low hydraulic conductivity to minimize water flow from above. The clay layer is covered by a high conductivity gravel layer, which is 2 meters thick at the center and slants off towards the edge of the trench. Surrounding the trench is undisturbed soil.

Because of assumed complete bilateral symmetry within the trench, only half of a cross-section of the trench is needed to simulate the flow of water and transport of radionuclides. Thus, the right most boundary in this simulation represents the center of the trench. By the method of images, the plane of symmetry can be treated as a zero net flux boundary.

The entire region is discretized into a total of 221 quadrilateral elements and 252 nodes, Fig. 1. The global numbering system starts from bottom to top and from left to right. Therefore, the quadrilateral element in the lower left corner is assigned as element number 1 and the lowest, leftmost, node is node number 1. For the most part, the elements are 100 cm by 100 cm. In Fig. 1, node numbers for various nodes are presented. It should be noted that node 46 is the node at the left corner of the bottom of the trench and node 242 is at the center of the trench (right corner) along the bottom. Nodes 102, 158, and 214 also lie along the bottom of the trench.

Radioactive Source

In general, for the "source term model" the source of radioactive material released and available for transport will be determined by the waste form leaching model (6). However, at this time, the leaching and transport model for the "source term" project are not coupled. Therefore, for the purpose of studying transport processes, a source was assumed. For the problems discussed in this paper, the

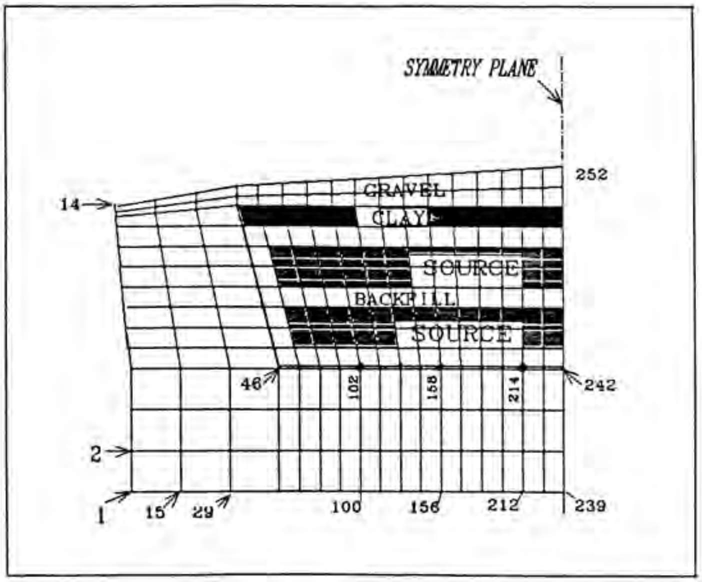

Fig. 1. Finite Element Nodalization Scheme Used by FEMWASTE-1 in the Radionuclide Transport Studies. Numbers in the Figure Refer to Node Numbers. The Bottom of the Trench Begins at Node 46 and Ends at Node 242.

source was located in the shaded region marked source in Fig. 1. The source region covered 52 square meters out of the 98 square meters in the waste region and had a strength of 2.55 10^{-12} g/cm^{3}s. This release rate occurred for the first year of the computer simulation. After this time, the release rate was set to zero. The source strength was chosen such that the total mass per unit volume released corresponded to the source region being loaded to the Class C limit with Cesium, which is 7000 Ci/m^3.

We emphasize that the high release rate and source strength were chosen only to provide numbers for the transport calculation and give an upper bound to release. They are not expected to be representative of an actual burial situation.

Material Properties

For the test problems considered, all four regions (gravel cap, clay layer, waste and backfill in the trench, and undisturbed soil around the trench) were considered to have identical transport properties with the exception of porosity and residual moisture content. The values for these parameters in each region were chosen to be consistent with the value used in the water flow calculations (7) and can be found in Table I.

The reference values for bulk density, dispersivity, diffusion coefficient, tortuosity, and distribution coefficient are presented in Table II. Unless stated otherwise, these were the values used in all calculations. In the problems in which the dispersivity was varied, the ratio between the longitudinal and transverse dispersivity remained equal to 5.

Water Velocities and Soil Moisture Content

Water velocities and soil moisture contents were calculated by FEMWATER-1 (7). The region modeled while calculating water flow was much larger than the one being used for the transport calculation. In the water flow calculations, the boundaries of the simulation were selected such that the

TABLE I

Porosity and Residual Moisture Content for the Different Regions

Region	Porosity	Residual Moisture Content
Undisturbed Soil	0.3	0.024
Backfill and Waste	0.4	0.032
Clay	0.5	0.04
Gravel Cover	0.3	0.0024

TABLE II

Reference Transport Parameters Used in the Test Problem

Property	Value
Bulk Density	1.75 g/cm^3
Longitudinal Dispersivity	213.0 cm
Transverse Dispersivity	42.7 cm
Diffusion Coefficient	1.E-5 cm^2/s
Tortuosity	0.1
Distribution Coefficient	0.0

water table was well beneath the trench and the bottom boundary conditions did not influence flow through the trench region.

Two different steady-state water flow cases were simulated to give an approximate upper and lower bound on flow. The upper bound was calculated assuming rainfall of 127 cm/yr. This value is typical for humid regions within the United States. However, this value for the total amount of precipitation entering the trench is very high due to neglecting evapotranspiration. Water velocities around the trench region ranged from about 10^{-5} (cm/s) to 10^{-6} (cm/s). A flow map is presented in Fig. 2. The direction of flow was predominantly vertically downwards except near the trench cap. Due to the low hydraulic conductivity of he clay layer, very little water flowed vertically down into the trench. However, there was a strong component of flow into the trench along the side of the trench beneath the clay layer. Thus, flow was directed towards the center of the trench in the x-direction and towards the bottom of the trench in the z-direction. The lower bound to water flow was calculated assuming rainfall of only 5 cm/yr. This water infiltration rate is representative of an arid site. The water flow pattern was very similar to the higher rainfall case with the exception that the velocities were approximately 25 times lower, reflecting the difference in the rainfall for the two cases.

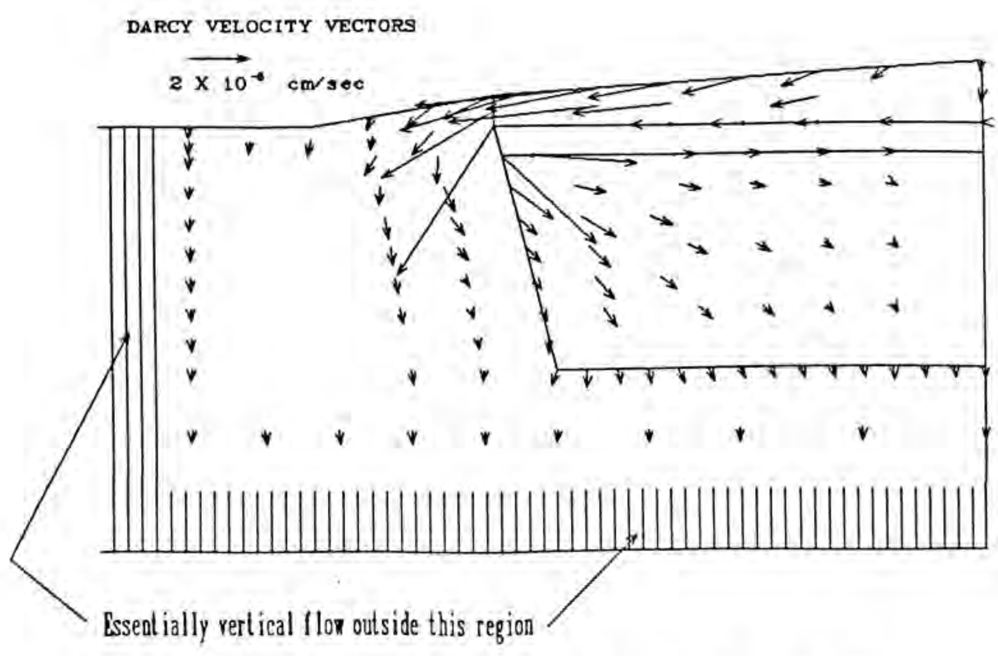

Fig. 2. Darcy Velocity Vectors for the 127 cm/yr Water Influx Case.

In both cases, the volumetric moisture content within the trench is less than 4% and has an average value of about 3.5%. These low values for moisture content are a result of the assumption for this simulation that the trench lies well above the water table. In general, for soils well above the water table, the moisture content is near the residual moisture content of the soil, which is 3.2% in the waste backfill region, as shown in Table II. A residual moisture content of 3.2% is extremely low and would be representative for soils such as course sands or gravels that are well graded and have relatively large pores which allow for excellent drainage. For soils in which the pore size distribution contains many smaller pores (clay or clay/sand mixtures) the minimum volumetric moisture content could be as high as 10 15%. For example, measured residual water content of two clay/sand soil samples from the Savannah River Plant were 12.5 and 16.7% (8).

Initial and Boundary Conditions

In all cases, the concentration at each nodal point is initially set to zero. This assumes that the region is initially free from any radioactive material.

Similarly, in all cases, the net flux through the top, right, and left boundaries is set to zero. At the bottom boundary, the concentration is set to zero. Thus, radionuclides are allowed to leave the system only through the bottom boundary.

Zero flux through the top boundary implies that the rain water entering into the system is not contaminated. Zero flux at the right boundary was chosen because this is a symmetry boundary.

MODELING RESULTS AND DISCUSSION

The influence of water velocity (rainfall rate), dispersion, and sorption on radionuclide transport within a trench were studied and the results are presented in the following sections. Figures 3 through 8 present a graphical description of the log of concentration versus time for the various test cases. In all of these figures, the nodes correspond to the node numbers in Fig. 1. With node 46 being the left corner of the trench bottom and node 242 being the center of the trench bottom. The dashed line at 1 year in Figs. 3 through 6 represents the time when the release rate from the source elements is set to zero.

The Influence of Water Flow Rate-on Radionuclide Transport

The predictions of radionuclide transport for the two different rainfall cases are presented in Figs. 3 and 4. Figure 3 presents the concentrations along the trench bottom for the rainfall rate of 5 cm/yr, and Fig. 4 is for the case of 127 cm/yr rainfall rate. Material properties used in the calculation are listed in Tables I and II.

By examining Fig. 3, it is seen that the concentration along the trench bottom is increasing during the 1 year source release period and continues to increase after the source is stopped. The time at which the concentration peaks along the bottom varies with position and is earliest at the edge of the trench, node 46, and latest at the center, node 242. This is caused by the water flowing towards the center of the trench with a low enough velocity such that species released from the source accumulate in the trench before they can be transported away. During the 1 year release period, all of the nodes along the trench bottom, with the exception of node 46, have almost the same concentration. This is because all of these locations are the same distance from the nearest source region, Fig. 1. In contrast, node 46 is located upstream from any source and is farther from the source region.

For the higher water flow case, slightly different behavior is obtained as seen in Fig. 4. In this case, the concentrations build up very rapidly and remain nearly constant for the one year source period. This implies that the water flow is fast enough to transport away the supply of radionuclides from the source. There is also a difference in the concentrations along the bottom during the source release period with higher values closer to the center, node 242, which reflects that the water flow is towards that region. Once the source has been turned off, the concentrations along the trench bottom begin to decrease immediately. In fact, the concentration along the bottom has decreased by over 3 orders of magnitude within 2.5 years after the source has stopped. This contrasts greatly with the low flow case in which concentrations along the trench bottom decreased less than 1 order of magnitude 9 years after the source had stopped.

It is interesting to note that the peak concentrations for the two rainfall cases differ by roughly a factor of two, whereas, the rainfall rate and, therefore, flow velocity, differ by a factor of 25.

The Influence of Dispersion on Radionuclide Transport

Figures 5 and 6 display the concentration as a function of time for values of longitudinal dispersivity ranging from 71 cm to 2130 cm at two different locations in the trench. Fig. 5 presents data from the left edge, node 46, and Fig. 6 from the center, node 242. These examples were done assuming a rainfall rate of 5 cm/yr and the material properties specified in Tables I and II.

From Fig. 1, it is seen that node 46 is upstream from the source region. Therefore, dispersion is the only mechanism by which radionuclides are transported to this node. Thus, one would expect that the higher the value for the dispersion coefficient, the higher the concentration would be at node 46. Examining Fig. 5, it is seen that this is not quite true. Initially, as expected, the case with the highest

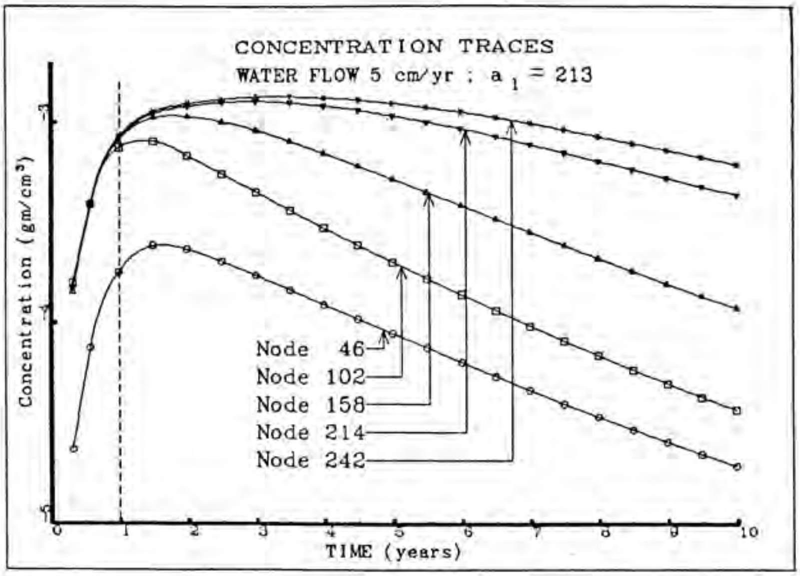

Fig. 3. Plot of Concentration Versus Time Along the Trench Bottom for the Case Where the Water Influx is 5 cm/yr, $k_d = 0$, and a =213.0.

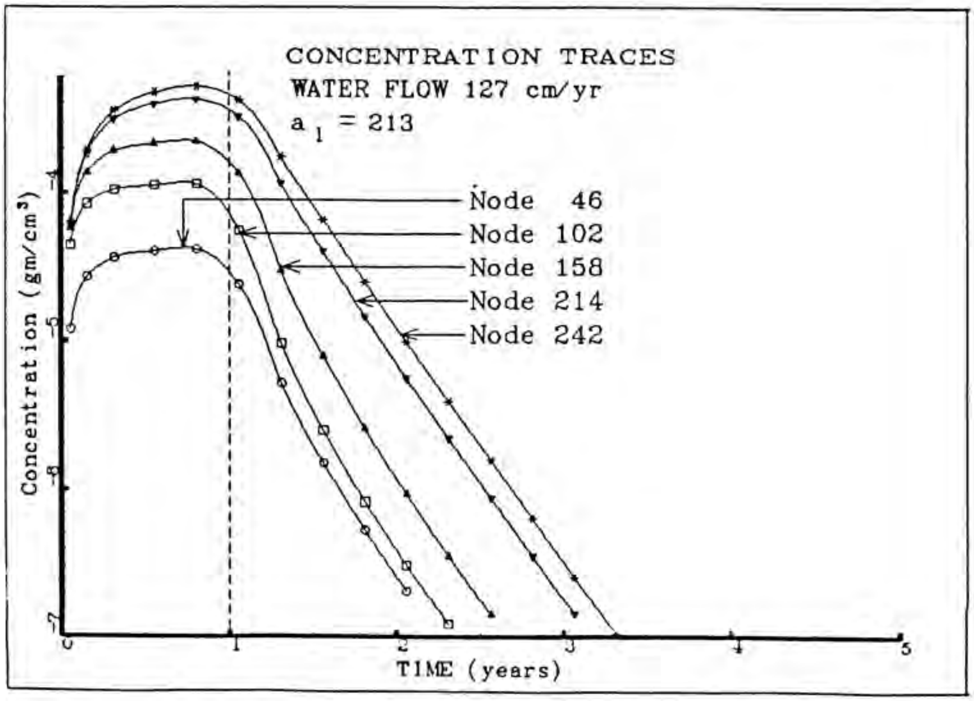

Fig. 4. Plot of Concentration Versus Time Along the Trench Bottom for the Case Where the Water Influx is 127 cm/yr, $k_d = 0$, and a =213.0 cm.

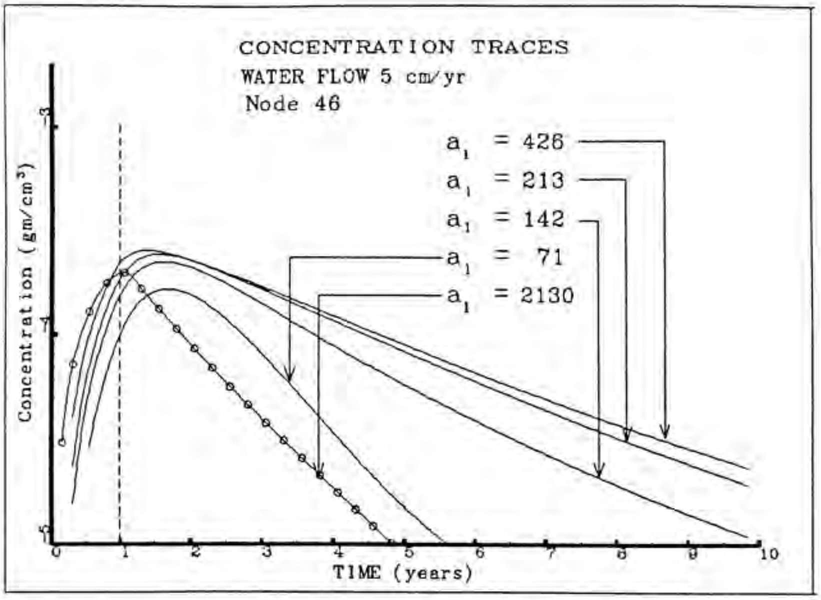

Fig. 5. Plot of Concentration Versus Time at the Lower Left Corner of the Trench, Node 46, for Various Values of the Dispersion Coefficient.

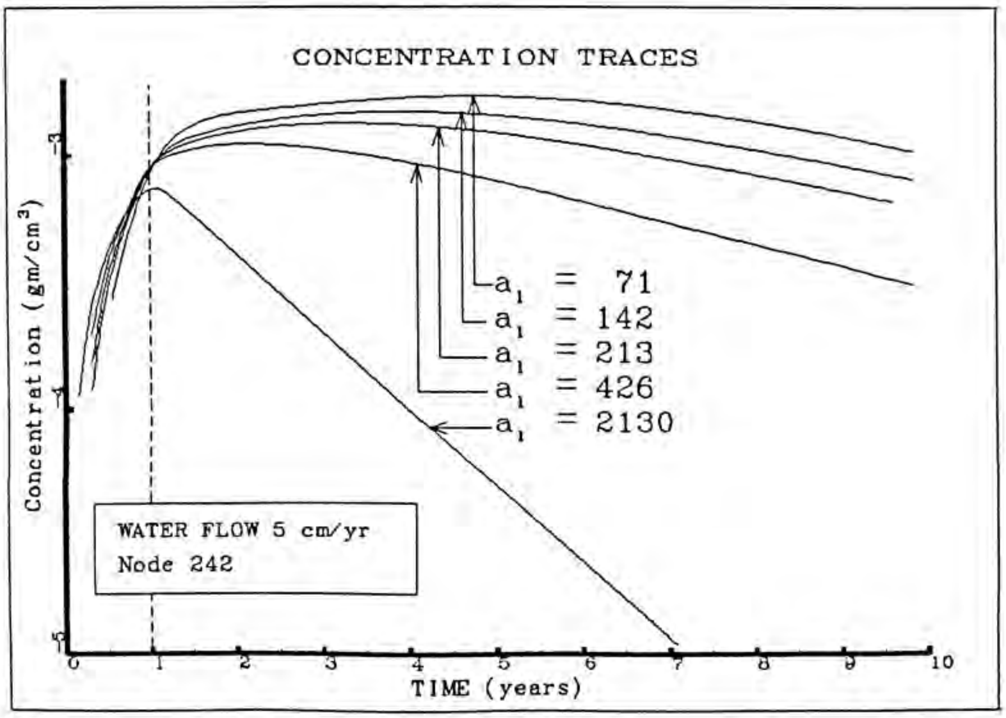

Fig. 6. Plot of Concentration Versus Time at the Center of the Trench, Node 242, for Various Values of the Dispersion Coefficient.

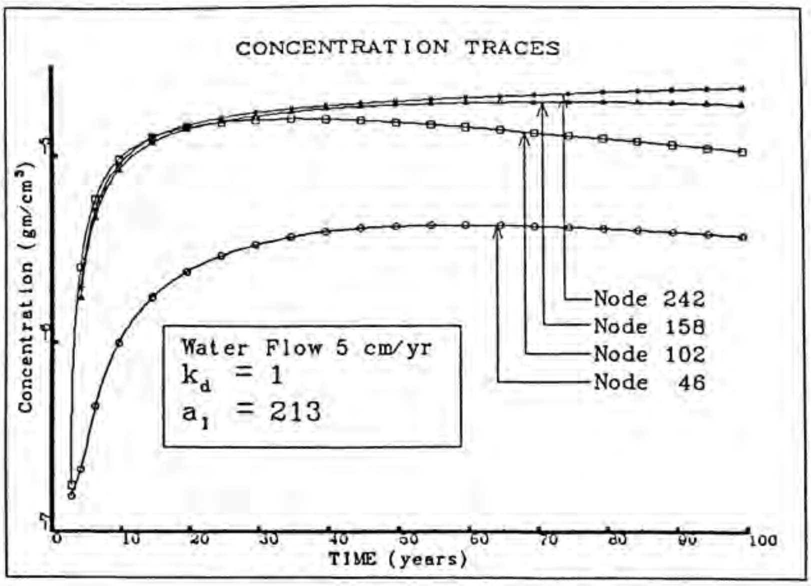

Fig. 7. Plot of Concentration Versus Time Along the Trench Bottom for the Case Where the Water Influx is 5 cm/yr, k_d-1, and a =213.0 cm.

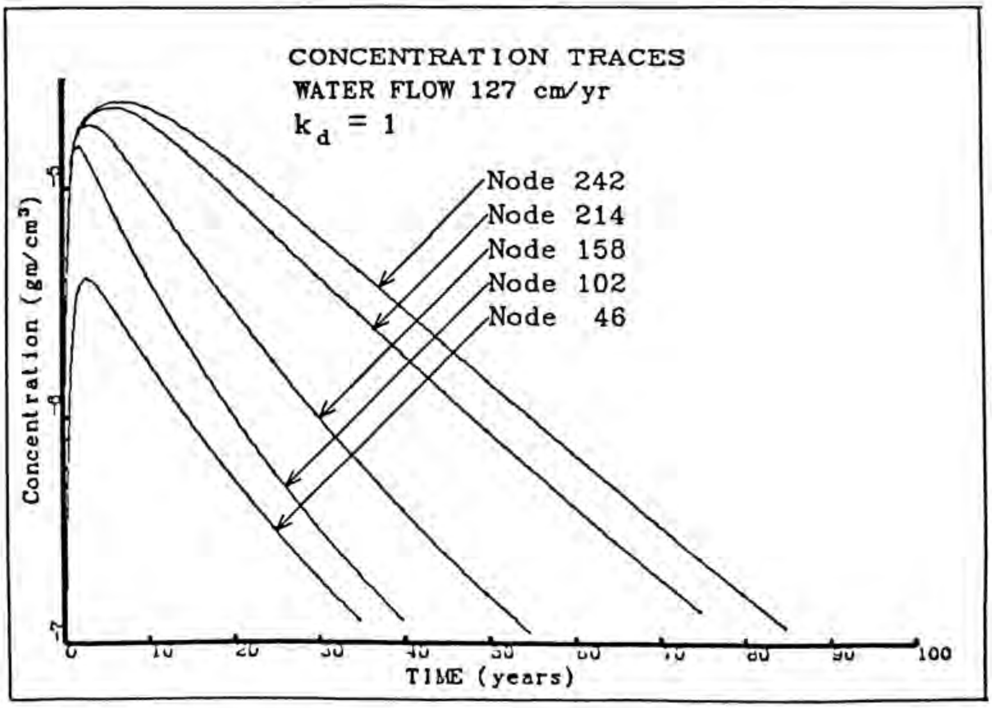

Fig. 8. Plot of Concentration Versus Time Along the Trench; Bottom for the Case Where the Water Influx is 127 cm/yr, k_d = 1, and a =213.0 cm.

dispersion, a = 2130 cm, brings the most material to node 46. However, by the end of the 1 year source period, the case with a = 426 cm, has a higher concentration. Soon after, the concentrations for the other four test cases exceed that of the highest dispersion case. For the cases where the dispersion coefficient ranged from 71 to 426 cm, the behavior was as expected, the higher the dispersion value, the higher the concentration at node 46, and the earlier the peak in concentration.

Through examination of the fluxes and the time-dependent concentration profiles from the different test cases, it was noticed that for the high dispersion case, the dispersive component of the flux always exceeded the advective component and concentrations decreased monotonically from the source region at all times. This indicates that dispersion was the dominant method of transport. For all cases with the dispersion coefficient less than or equal to 426, advection was the dominant transport method and the concentration profiles were characterized by a peak moving away from the source region. It is interesting to notice that for the advection dominated transport cases, once the peak has passed through the bottom of the trench, dispersion acts to bring material to the trench and reduce the rate of decrease in concentration at the trench bottom.

At the center of the trench, node 242, both advection and dispersion are responsible for bringing material to this point. At this point, the higher the dispersion coefficient, the faster material is transported away from the trench and the lower the concentration. This is reflected in Fig. 6 which shows that the case with the lowest dispersion coefficient has the highest concentration at all times after the source has been stopped and reaches its peak value at a later time.

An important point to notice in Fig. 6 is that increasing the dispersion coefficient by a factor of 6 from 71 to 426 cm has only a minor change on the value of the concentration at this location, (the center of the trench). Whereas, increasing it by a factor of 5 from 426 to 2130 cm has a major influence on the concentration profile. This is further evidence that transport is advection dominated at the lower values of dispersivity and dispersion dominated at the highest value of dispersivity.

From Figs. 5 and 6, it is clear that the dispersivity values can play a major role in predicting transport. However, there is no acceptable theory to predict the values for the dispersion coefficient. Further, a literature review (9) found very few experimental values for dispersivity in the unsaturated zone. Also, for saturated flow, it has been shown that the dispersivity is an increasing function of the distance over which the measurement was taken. However, the experimental values for unsaturated dispersivity have been taken over a few meters, at most. Another problem with dispersivity values measured in small scale tests is that they do not account for the waste packages located within the trench. It is known that soils containing large impermeable rocks have much higher dispersivity values than similar soils without rocks (10,11).

Based on the literature review presented in (9) and the scale of the problem, it appears that longitudinal dispersivity values of around a few hundred centimeters are reasonable. This would indicate that transport would be advection dominated. The high value of dispersivity selected from this problem was given by Yeh in the documentation for FEMWASTE (4) and originates from a study of chromium transport on Long Island. If the dispersivity is this large, dispersion would be the dominant transport mechanism.

For the range of dispersivity values and water flow rates tested, diffusion was negligible when compared to dispersion. For diffusion to become an important transport mechanism, the diffusive term would have to be roughly the same size as the dispersive term in Eq. (2). Using the values in Table II and an average moisture content of 3.5% yields a diffusion coefficient of $3.5\ 10^{-8}$ cm^2/s. For the dispersive term to become that small would require that the longitudinal dispersivity would be roughly 0.2 cm, if the magnitude of the velocity vector was $1.6\ 10^{-7}$ cm/s, (5 cm/yr). It is felt that a dispersivity value of 0.2 cm is too small a value for the scale of this problem. At higher rainfall rates, e. g., higher flow velocities, diffusion is less likely to be an important transport mechanism.

The Influence of Sorption on Radionuclide Transport

The predictions of radionuclide transport with chemical sorption for two different rainfall cases are presented in Figs. 7 and 8. These problems are identical to the ones solved in the section on the influence of rainfall on transport with the exception that the distribution coefficient is set to 1. Figure 7 presents the concentrations along the trench bottom for the rainfall rate of 5 cm/yr, and Fig. 8 is for the case of 127 cm/yr rainfall rate. Material properties used in the calculation are listed in Tables I and II.

Comparing Figs. 3 and 7, it is clear that even a relatively small distribution coefficient, $k_d = 1$, has a dramatic impact on the concentration profiles. For the case of $k_d = 1$ with a water flow rate of 5 cm/yr, Fig. 7, the concentrations at the trench bottom increase for the first 10 years and then remain relatively constant for the next 90 years of the simulation. The reason that sorption has such a pronounced effect on transport is because the moisture content of the trench is so low, averaging roughly 3.5% in the trench. Using this value for moisture content and the bulk density from Table II in Eq. (3) gives a retardation factor of 51 when $k_d = 1$.

Figure 8 further demonstrates the importance of sorption on transport. Due to the higher water flow rate, the concentration at the different locations does decrease substantially over the 100 year simulation time. However, without sorption the concentration at all locations along the trench bottom was less than 10^{-7} g/cm^3 after 3.5 years; with sorption the concentrations remained greater than 10^{-7} for 35-85 years depending on the location being considered. Also, because of sorption, peak concentrations were attained after the 1 year source period, with the peak appearing later and with greater magnitude closer to the center of the trench.

The typical scenario in these test cases involved a rapid injection of material into solution, i.e., the assumed source, with concurrent sorption on the soil. Due to the low water content within the trench, most of the mass is initially adsorbed on the soil. At early times when all of the material is in the trench and for $k_d = 1$, there is 50 times as much mass adsorbed onto the soil as compared to that in solution, i.e., 98% of the mass is adsorbed. This accounts for the concentrations having a value roughly 50 times lower in the case of $k_d = 1$, as opposed to $k_d = 0$. As modeled, sorption is a reversible process. Therefore, as the contaminant in solution migrates away from the trench due to advection and dispersion, some of the mass adsorbed on the soil is released into solution. This keeps the concentration in the trench from decreasing as fast as the case without sorption. In this sense, sorption acts to spread the release of contaminant out over time.

One method of defining the retardation coefficient is the ratio of the velocity of non-sorbing species to the velocity of the sorbing species. Therefore, since the water velocity vectors for the 5 cm/yr and 127 cm/yr rainfall rate test cases were essentially identical in direction but different in magnitude, proper choice of the distribution coefficient should allow radionuclide concentration (scaled to account for adsorption) in the 127 cm/yr rainfall rate case to be identical to the 5 cm/yr case without adsorption. This was attempted by choosing $k_d = 0.488$, giving an average retardation factor of 25.4, the ratio of the two rainfall rates. Although not shown, comparison between the two cases over a 10 year period demonstrated substantial agreement when the predicted concentrations of the high water flow rate with retardation case were multiplied by 25.4 to account for the mass adsorbed.

It is important to realize that besides the usual limitations associated with the use of a linear sorption isotherm, this isotherm is independent of soil moisture content. This assumes that for a fixed concentration in solution, the amount adsorbed onto the soil will be the same in unsaturated soils as it is in saturated soils. Thus, for soils with low moisture content this model predicts sorption to be extremely effective in slowing down radionuclide transport. However, there is experimental evidence which suggests that sorption may decrease as the moisture content decreases (8,12). If this is the case, the model used in this paper will underpredict the transport out of the trench.

CONCLUSIONS

Simulations of radionuclide transport from a generic shallow land burial trench have been performed for a range of water flow rates, dispersivity values, and distribution coefficients. For the modeling assumptions used, and the range of parameters tested, the water flow velocity plays the major role in redistributing radionuclides within the trench, except in the case of extremely high dispersion. Dispersion was always found to play a significant role in determining transport. This was particularly apparent upstream from the source. Sorption decreased the magnitude of the radionuclide concentration and flux, and had the apparent effect of reducing the velocity with which the radionuclides were transported. Diffusion was found to be unimportant in determining radionuclide transport.

Two areas were identified in which there is a shortage of relevant modeling data. Little information exists on dispersivity and radionuclide sorption in unsaturated soils. Of the two, sorption is more likely to have a greater influence on transport.

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