BENCHMARKING BLT - A COMPUTER CODE FOR LLW SOURCE TERM EVALUATION

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ABSTRACT

A computer code, called BLT, has been specially developed for calculating the release of radionuclides as a function of time (source term) from low-level waste disposal facilities. To provide a reasonable level of confidence in the model predictions, the BLT code was benchmarked against real experimental data. In this study, the release curves for nitrate, technetium-99 and tritium from the *saltstone* lysimeters operated by Savannah River Laboratory were used. The model results are observed to be in good agreement with the field data, within the acceptable limits of uncertainty. The results also show that for soluble species, such as those under study, diffusion is the controlling release mechanism from large porous monolithic waste forms. Furthermore, the results demonstrate that 2-D approximations can be used to simulate adequately real release in three dimensions.

INTRODUCTION

Brookhaven National Laboratory has developed a general computer model - BLT (Breach-Leach-Transport) - to calculate the release of radionuclides from low-level waste shallow land burial facilities (1,2). Central to this code is the Leach model, which has been specifically developed to accommodate three separate physical processes, namely, the diffusion of chemical species through the pore structure within the waste form, the dissolution of the waste form, and the surface rinse, by which chemical species on the surface are "washed off" (2). Diffusion within the waste form is modeled using analytical solutions of the diffusion equation with the effective diffusivity approach. For the dissolution of the waste form, a first-order kinetics is assumed, i.e., the rate varies linearly with the degree of saturation. For the surface rinse process, the chemical species on the surface are "washed off" instantaneously, limited by only the solubility of the chemical species in water. In the original version of BLT, all three sources are calculated independently and summed, but the total release is scaled down linearly by the amount of container corrosion if the container has not yet been completely corroded away. The final release is coupled to the mass transport equation through the source and sink term.

Several significant improvements have been made to the Leach model, since our previous reports on the BLT code (1,2). These changes were described in detail in a paper presented in Waste Management '91 (3). In summary, they are: (a) The three release mechanisms are now treated simultaneously to allow interplay among them, such as solution feedback effects on diffusional release. (b) A constant partitioning between the solid (surface of the waste form) and the liquid (leachate) immediately associated with the waste form is applied in place of the total wash off in the original rinse model. (c) Instead of relying on analytical solutions, the finite difference method is used to solve numerically for the diffusional transport within the waste form in one dimension. The immediately surrounding volume of liquid is modeled as a well-mixed fluid with a uniform concentration (the "mixing bath") through which solution feedback and partitioning effects are coupled.

OBJECTIVE

The objective of the this study is to provide a reasonable level of confidence in the model predictions. This was achieved by demonstrating that the model predictions do agree with real experimental data within acceptable limits of uncertainty.

In this study, the data from *saltstone* lysimeter experiments conducted by Savannah River Laboratory (SRL) were used. These lysimeters containing large monoliths of *saltstone* have been in operation since 1984 (5). Although the trapezoidal cross-section and the large size of these monoliths make them slightly cumbersome to model, the SRL experiments provide a well controlled field study data set over a time frame of several years. Therefore, benchmarking BLT against these experimental data can demonstrate the capability of the BLT code to make reasonably accurate predictions.

MODEL CONDITION AND PROCEDURE

The experiments, designated as Tank 24 lysimeters, were started in January of 1984 (5). They consist of three *saltstone* lysimeters one was covered with a gravel cap, another with a clay cap, and the third without any cover. In this study, the benchmarking work focused on using the data from the latter two lysimeters.

The SRL *saltstone* monoliths were made from a low-level "decontaminated" salt solution and cement-fly ash mixture (6). The salt solution contained sodium nitrate as the main constituent and other radioactive contaminants, the most abundant of which being tritium and technetium-99. The trapezoidal monoliths were cast in situ in a trapezoidal shaped trench, which was backfilled with native soil and compacted to the original grade. Natural rainfall was allowed to percolate and the leachate was collected from a sump constructed at the bottom.

In this study, BLT was used for the two-dimensional simulations of the trapezoidal trench containing a trapezoidal cross-section of the monolithic waste form. Only one half of the vertical cross-section perpendicular to the longitudinal axis of the trench was used, because of bilateral symmetry. The simulation domain was divided into a finite element mesh of 102 nodes and 80 elements (Fig. 1). The top width of the mesh

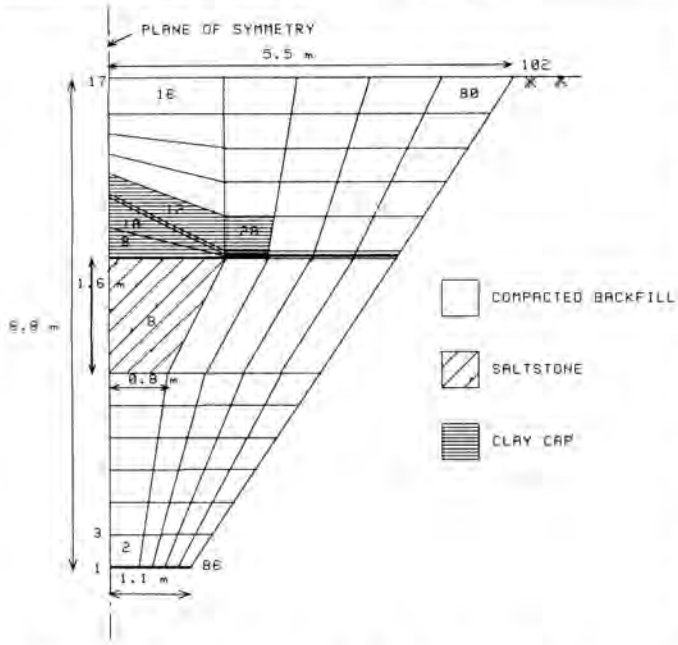

Fig. 1. Finite element mesh used for benchmarking. Because of symmetry, only the right half of the vertical cross-section was used. Half of the *saltstone* monolith is represented by Element No. 8. For the case of the capped lysimeter, the clay cap is represented by Elements No. 9, 10, 11, 12, 25, 26, 27 and 28.

is 5.5 m, the bottom width is 1.1 m, and the overall depth is 6.8 m. Element No. 8 represents the trapezoidal-shaped saltstone monolith, with its bottom at a distance of 2.7 m from the bottom of the trench. The waste form has a top (half) width of 1.6 m, bottom (half) width of 0.8 m and a height of 1.6 m. Elements 9, 10, 11, 25, 26 and 27 were used to represent the clay cap.

The hydraulic and transport properties of the soil, the saltstone waste form and the clay cap material were partly based on measurements made by SRL's consultants (7,8), and the input values are summarized in Table I.

TABLE I

Hydraulic and Transport Properties of the Materials

	Compacted Soil	Saltstone	Clay Cap
Porosity (%)	37	50	27.2
Sat. Hydraul. Conduct. (cm/s)	2.3×10^{-5}	1.0×10^{-8}	3.4×10^{-9}
Bulk Density (g/cm^3)	1.67	1.542	1.7
Longitudinal Dispersivity (cm)	10	10	10
Lateral Dispersivity (cm)	1	1	1
Mol. Diffusion Coeff. (cm^2/s)	1×10^{-5}	1×10^{-5}	1×10^{-5}
Distribution Coefficient	0	0	0
Tortuosity	0.001	0.001	0.001

To obtain the water flow field and the moisture content distribution in the finite element domain, steady state simulations were carried out using the modified FEMWATER code (9). The top boundary is a prescribed flux boundary with a constant flux equivalent to 38 cm (15 inches) of net infiltration per year based on the average rainfall minus evapotranspiration (5). The bottom boundary is a prescribed head boundary at which the pressure head equals zero. The left and the right boundaries are zero flux boundaries. Two simulations were performed: first, for an uncapped lysimeter, and second, for a lysimeter with a clay cap. The results of the water flow simulation were used as a portion of the required input data for the BLT code.

The release of nitrate, technetium-99 and tritium were calculated by BLT using the same water flow field and moisture content data. Time dependent simulations were performed with constant time steps of 30 days up to a total of 1,500 days (about 4.1 years) using the BLT code. The top boundary and the two side boundaries were set to be zero flux boundaries, while at the bottom, a Dirichlet boundary condition with zero concentration was applied at all times. In the field, leachate was collected daily from the sump at the bottom of the trench. The model simulates this bottom by creating a row of nodes (No. 2, 19, 36, 53, 70 and 87) 1 cm above the bottom (zero-concentration) boundary of the simulation domain. In these benchmark runs, this row of nodes were used as the concentration trace nodes, and the area-weighted average of their computed concentrations was taken to be the average concentration of the leachate in the sump.

Containers were not used in the lysimeter experiments, and therefore the BREACH routine was not activated. This was done by simply setting the container thickness to a very low value of 10^{-8} m. In all cases, none of the available mass was assigned to dissolution release. Only diffusion and rinse releases were considered to be significant. The LEACHFD (finite difference model) routine was used for computing the releases.

To benchmark against the experimental data, input (leaching) parameters for rinse and diffusion, such as the distribution of the total available mass and the waste form diffusion coefficient etc., were adjusted within reasonable ranges to "best fit" the observed data. The best fitting was performed only by manual estimation without using any quantitative criteria or predetermined algorithms. The key input parameters for BLT can be separated into three types:

Type 1: Hydrologic parameters calculated by the water flow module (FEMWATER) based on measured hydraulic properties of the media and experimental conditions, e.g. infiltration velocity and moisture content. These parameters were fixed unless the experimental conditions change.

Type 2: Transport parameters of the media, e.g. dispersivity, and some of the leaching parameters of the waste form, e.g. diffusion length and initial mass or activity. These parameters are either calculated or estimated based on measured data and/or experimental conditions. Their values were fixed for all runs and were not adjusted for the purpose of fitting.

Type 3: Leaching parameters of the waste form, viz. available mass distribution between rinse and diffusion, partition coefficient and waste form diffusion coefficient, which were adjusted to "best fit" the experimental data.

The values of the key input parameters are summarized in Table II. Others can also be found in Table I.

RESULTS AND DISCUSSIONS

The output data generated by the BLT code were compared with the observed data of nitrate, technetium-99 and tritium releases taken from published graphs and data provided directly by SRL (10,11,12). In this paper, only the results of the "best fit" cases will be presented.

Nitrate Release

Simulations of nitrate release were carried out under two different conditions: the first for the lysimeter without any covering except backfill (with native soil), and the second for the one with a clay cap. The approach was to adjust the (Type 3) leaching parameters to match the observed release data (6,12). The adjusted parameters are the partition coefficient, the waste form diffusion coefficient and the available mass distribution between rinse and diffusion. The "best fit" benchmark values are shown in Table II.

The total inventory of nitrate was calculated based on the measured concentration of 130 g/liter in the salt solution (6), and on a reported total of 9,500 liters of the solution for each monolith (10).

Sodium nitrate is highly soluble, and the release of soluble species such as nitrate are likely controlled by diffusion and/or rinse mechanisms. Laboratory leaching experiments (ANS-16.1) on samples of saltstone conducted by SRL (13) have shown that the leach rate after the second day for soluble species decreased linearly with the square root of time, indicating a diffusion-controlled release mechanism. Accordingly, 80% of the available mass was distributed for diffusion release, and the remaining 20% for rinse. Dissolution of the waste form is expected to be insignificant for the short time span, and therefore, no dissolution release was considered.

The laboratory experiments by SRL had yielded a value for the effective waste form diffusion coefficient (D_{wf}) of nitrate in the *saltstone* in the order of 1×10^{-8} cm^2/s (13). The effective diffusion length (L) for the three-dimensional trapezoidal waste form was estimated by using the volume to surface ratio. It was calculated to be 38.8 cm. The sensitivity of this parameter, L, was tested, and the results show this value to be satisfactory. A sensitivity study on the parameter, NFDP (number of finite difference points for the 1-D diffusion calculation in the LEACHFD routine), was also conducted. Based on this study, an optimum value of NFDP = 101 was chosen for these benchmark conditions.

Figure 2 shows the BLT model results for the uncapped lysimeter in relations to the lysimeter data up to 1,500 days. Other model predictions from Intera and SRL are also included for comparison. The BLT results compare reasonably well for the first 1,100 days (about 3 years) into the experiment. The BLT simulation correctly predicts breakthrough at the bottom at about 150 days, followed by a gradual increase in concentration. The BLT model prediction also compares closely with those made by the SRL and Intera models. However, all three models fail to match the concentration peaks which occurred after the third year. This discrepancy could be caused by seasonal and climatic variations because our model (and probably also the other two models) relies on a state steady water flow simulation with a constant infiltration rate.

The experimental data on nitrate release from the lysimeter with a clay cap was significantly lower than the release from the one without any cap as discussed above. The concentration of the leachate increased slightly with time, but the average concentration never exceeded 10 ppm as shown in Fig. 3.

The model data for the capped lysimeter also show a significant reduction of advection rate, and the concentration in the leachate is significantly (a factor of three) lower as compared to the leachate from the uncapped lysimeter (Fig. 3). However, the model data peak at about 40 ppm at approximately 900 days about a factor of four more than what the

TABLE II

Key Input Parameters For BLT

	Nitrate	Nitrate	Technetium-99	Tritium
TYPE 1: (Fixed)				
Hydrologic Condition	Capped	Uncapped	Uncapped	Uncapped
Darcy Velocity (cm/s)	5.18×10^{-7}	1.13×10^{-6}	1.13×10^{-6}	1.13×10^{-6}
Moisture Content (%)	40.2	40.2	40.2	40.2
TYPE 2: (Fixed)				
Solubility Limit (g/cm^3)	0.9	0.9	1.0	1.0
NFDP	101	101	101	101
Diffusion Length (cm)	38.8	38.8	38.8	38.8
Initial Mass or Activity	1,235 Kg	1,235 Kg	0.43 Ci	0.114 Ci
TYPE 3: (Fitted)				
Partition Coefficient	0.0	0.0	0.0	0.0
WF Diff. Coeff. (cm^2/s)	1×10^{-8}	1×10^{-8}	1×10^{-9}	5×10^{-7}
Mass Distribution (Rinse:Diffus:Dissol)	2:8:0	2:8:0	0.5:9.5:0	2:8:0
(Other fixed parameters given in Table I)				

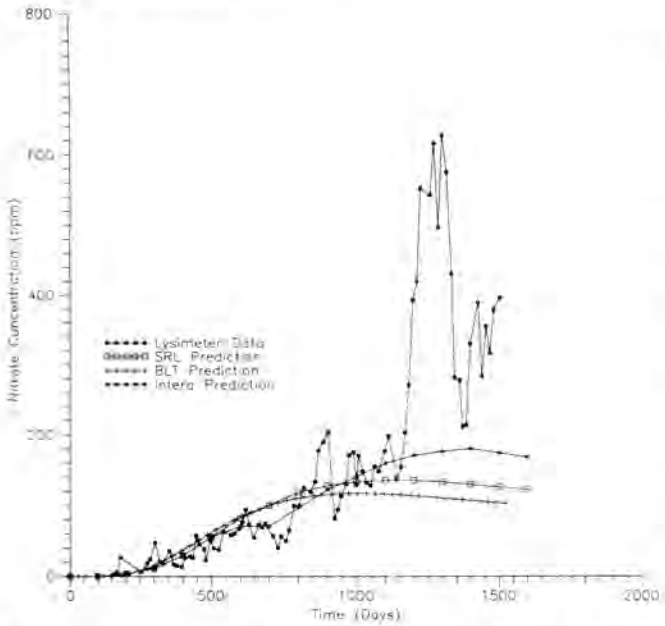

Fig. 2. Concentration of nitrate (ppm) in the leachate vs. time, for the uncapped lysimeter, showing how the BLT prediction compares with the actual lysimeter data and also other model predictions made by SRL and Intera.

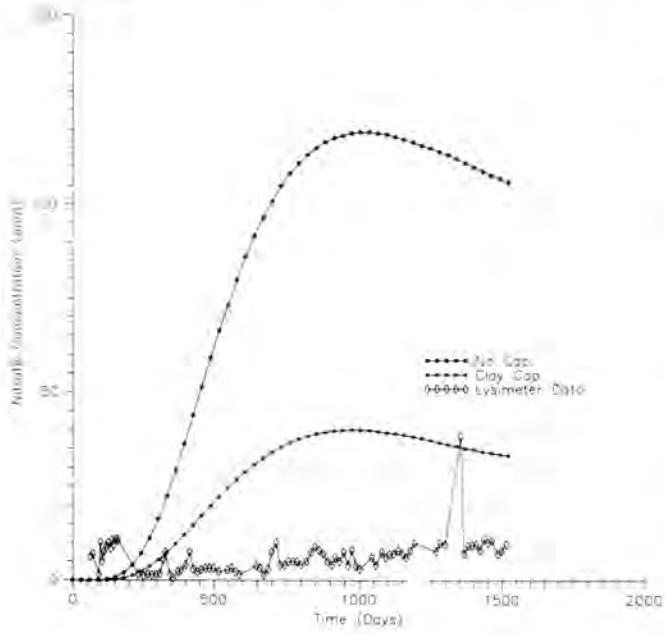

Fig. 3. Concentration of nitrate (ppm) in the leachate vs. time, for the capped lysimeter, showing how the BLT prediction compares with the actual lysimeter data. The prediction made for the uncapped lysimeter is also included to show the effectiveness of the cap on reducing release.

experimental data show. This discrepancy may be due to one of the two possible factors: a) The input hydraulic conductivity value for the clay cap may be too high, because unsaturated hydraulic properties measured under laboratory conditions may not accurately reflect the actual field conditions. b) The clay cap in our model "leaked" numerically because of the large size of the element representing the waste form. Large size elements are not suitable for situations with abrupt spatial variations (e.g. a sharp contrast in moisture content and in flow rate) because of possible numerical oscillation and dispersion.

Technetium-99 Release

For the simulation of the release of technetium-99 from the uncapped lysimeter, the same water flow and moisture content data as for the nitrate release were used. A similar approach was used to "best fit" the experimental data obtained by SRL (10) for the uncapped lysimeter (Fig. 4). The fitted parameters are the Type 3 leaching parameters as described earlier. The benchmark values are shown in Table II.

The total inventory of technetium-99 was determined based on: a) the reported concentration of technetium-99 in the decontaminated salt solution (35 nCi/g) (10), b) the amount of the solution used for making each monolith (9,500 liters) (10), and c) the approximate density for the solution (1.3 g/ml) (6). It was calculated to be about 0.43 Ci (shown in Table II).

The measured concentrations of technetium-99 in the leachate are three orders of magnitude lower than the concentration in the original decontaminated salt solution, and therefore no solubility limit was imposed (C_{sat} = 1 g/ml).

Technetium has been reported to have a soil-water partition coefficient in the order of 10^{-3} (14), and therefore, the partition coefficient (between waste form and leachate) was assumed to be also low. Consequently, no partitioning effect was simulated.

Results from earlier laboratory leaching experiments (ANS-16.1) by SRL have indicated that the release of soluble species from *saltstone* is mainly diffusion controlled (13). In these benchmark runs, the available mass for release was distributed 1 to 9 between rinse and diffusion mechanisms, causing diffusion to be the dominant process.

The effective waste form diffusion coefficient (D_{wf}) for technetium in *saltstone* has not been measured. The "best fit" value used in our benchmark run is 1.0 x1 0^{-9} cm^2/s, which is within one order of magnitude from the measured D_{wf} value for nitrate. The fact, that the observed release curve for technetium-99 from the uncapped monolith is very similar to the release curve of nitrate (10) (Fig. 2), not only suggests a similar controlled mechanism, but also a relatively high effective diffusion rate as discussed earlier for nitrate release. Therefore, our benchmark value for D_{wf} appears to be within an acceptable range.

Figure 4 shows a reasonable match between the BLT model data and SRL's experimental data up to a period of 1,500 days, during which approximately 0.37% of the total inventory (according to model calculation) was released. Because the model depends on a steady state simulation for water flow based on the average annual net infiltration rate, there are no seasonal or month-to-month fluctuations in the model data. Nevertheless, the model results show the initial breakthrough of technetium-99 in the leachate at the bottom sump at about 200 days into the experiment, then followed by a gradual increase in concentration, and finally, a leveling-off after 3 years (approx. 1,000 days). As presented in Fig. 4, BLT model results agree reasonably well with the experimental data during this initial period.

Tritium Release

Apart from the seasonal fluctuations, the experimental tritium (cumulative) release data from the uncapped lysimeter indicate a relatively constant release rate (i.e. a linear cumulative release curve) after a initial period of about 500 days. Similar observations have been made earlier from other lysimeter experiments conducted by Pacific Northwest Laboratory (4).

The key parameters for the tritium release model from the uncapped monolith are presented in Table II. Similar to nitrate and technetium-99, a diffusion-dominant model was used. The "best fit" benchmark value for the effective waste form diffusion coefficient (D_{wf}) for tritium was in the order of 5.0×10^{-7} cm^2/s, which is reasonably close to, but slightly higher than, the measured range of D in various cement materials (15). Nevertheless, it is about two orders of magnitude lower than the molecular diffusion coefficient for H^+ in pure water. Since tritium is in the form of water, no partitioning and solubility limiting effects were applied. The total inventory of tritium was calculated based on the reported concentration in the salt solution of 12 nCi/g (10) similar to what was done for nitrate and technetium.

Figure 5 compares the lysimeter data (11,12) and the BLT model results. As in the case of technetium-99, because our water flow model was based on only the steady state solution, our model release curve is smooth, and without any seasonal variations exhibited by the lysimeter data. Nonetheless, it matches the actual release reasonably well.

Although in the BLT model, the cumulative release curve is almost linear, we can also observe an initial growth of the curve after the initial period of zero release for about 300 days from the start of the experiment. This represents the dispersive front of the breakthrough curve. A corresponding observation can also be made from the lysimeter data at about the same time period, in good agreement with the model data.

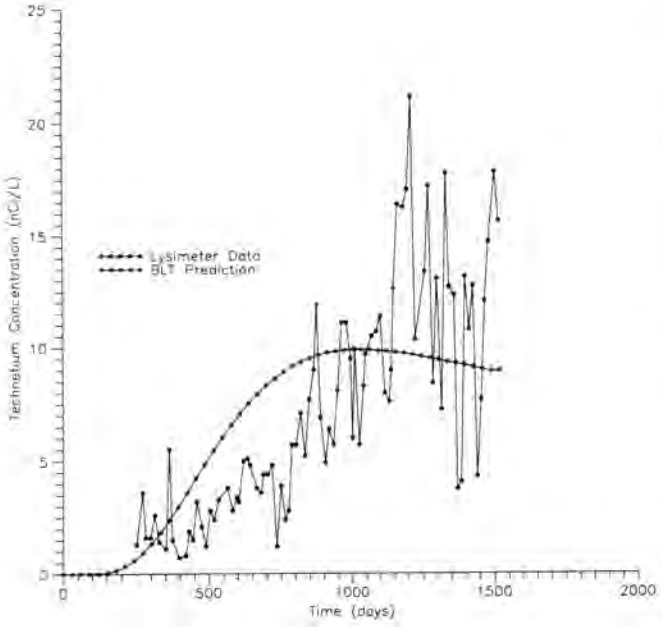

Fig. 4. Concentration of technetium (nCi/l) in the leachate vs. time, for the uncapped monolith, showing how the BLT prediction compares with the actual lysimeter data.

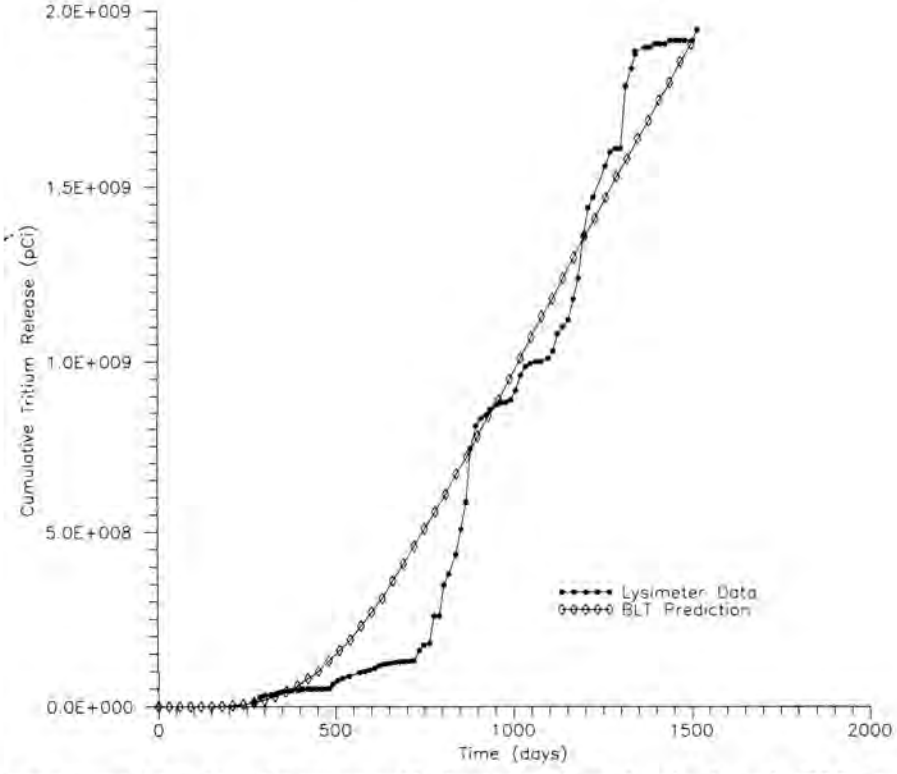

Fig. 5. Cumulative tritium release (pCi) from the uncapped lysimeter vs. time, showing how the BLT prediction compares with the actual lysimeter data.

CONCLUSIONS

Applying the new Leach routine of the BLT code, we have matched the BLT model results, within reasonable limits of uncertainty, with the release curves for nitrate, technetium-99 and tritium from the saltstone lysimeter experiments. Based on the results of these benchmarking runs, the following conclusions and recommendations can be made:

1. For soluble species such as nitrate, technetium-99 and tritium, diffusion appears to be the controlling release mechanism from large porous monolithic waste forms, such as large cement blocks. However, for small or non-porous waste forms the other two release mechanisms rinse and dissolution may be more effective. For less soluble species, dissolution (of the waste form) could be the controlling process.
2. The cumulative tritium release has been observed to be almost linear with time. This linear behavior is due to the fact that only a small percentage of the total tritium inventory has been released (only about 1.67% in 1,500 days for SRL's experiment). Within this small percentage range, the diffusion flux out of the waste form is nearly constant. However, dispersion during transport through the soil does impose a slight curvature on the initial portion of the cumulative release curve.
3. Although in reality the leach and transport processes occur in three dimensions, computation using 1-D (for leach) and 2-D (for transport) approximations appears to be adequate. However, this type of conceptual errors can be minimized if, whenever possible, the values of certain input parameters (e.g. diffusion length for the waste form) are estimated based on the 3-D geometry.
4. A clay cap is an effective moisture barrier. However, it may be difficult to simulate its effect accurately because of the abrupt spatial variations of flow rate and moisture content, not well handled with common numerical techniques.

In summary, the BLT code has been shown capable of predicting the release from lysimeter experiments within reasonable limits of uncertainty. Because of the inherent complexity, correct values of the numerous input parameters and the proper choice of approach are the crucial factors essential for obtaining accurate results. Therefore, the user of the code must ensure that the code is implemented correctly with accurate and realistic input parameters based on sound experimental data and detailed technical considerations, and the results are interpreted carefully and judiciously.

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