

# MODELING OF FIELD LYSIMETER RELEASE DATA USING THE COMPUTER CODE DUST

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## ABSTRACT

In this study, we attempted to match the experimentally measured mass release data collected over a period of seven years by investigators from Idaho National Engineering Laboratory from the lysimeters at Oak Ridge National Laboratory and Argonne National Laboratory using the computer code DUST. The influence of the dispersion coefficient and distribution coefficient on mass release were investigated. Both were found to significantly influence mass release over the seven year period. It is recommended that these parameters be measured on a site specific basis to enhance the understanding of the system.

## INTRODUCTION

Researchers at the Idaho National Engineering Laboratory (INEL) have been directing the Nuclear Regulatory Commission (NRC) sponsored lysimeter program. In this program there is a set of five lysimeters located at Argonne National Laboratory (ANL) and another set of five at Oak Ridge National Laboratory (ORNL). Releases from these lysimeters have been measured every three months for the last seven years.

The lysimeters are right cylinders 91 cm ID and 312 cm in height. At each site four of the lysimeters are filled with soil while the 5<sup>th</sup> control lysimeter is filled with unimin silica oxide sand. At ORNL the soil used is from the C horizon of a Fuquay sandy loam from the Savannah River Plant (SRP) adjacent to the Barnwell facility in South Carolina. ANL lysimeters are filled with a local soil which represents a typical Midwestern type. It is a morley silt loam with the surface layer removed.

Each lysimeter is filled with seven cylindrical wasteforms measuring 4.8 cm ID and 7.6 cm in height. They are stacked one on top of the other forming a height of 53.2 cm and a volume of 1 L. The wasteforms were solidified in either vinyl ester-styrene or Portland type I-II cement. The waste streams included two resin types. Type I was a mixture of synthetic organic ion exchange resins (phenolic cation, strong acid cation, and strong base anion) while the type II resin was a mixture of synthetic ion exchange resins (strong acid cation and strong base anion resins) with a mixture of inorganic zeolite (1).

Each lysimeter is equipped with 5 moisture collecting cups, soil moisture/temperature probes which are located at various places throughout the lysimeter and a leachate container located at the bottom of the lysimeter, Fig. 1 (1). Below the fill material a layer of filter fabric was placed between the soil or sand and the gravel bed. The filter promotes drainage by preventing clogging of the leachate container. A 10-20 cm gravel bed is located below the filter fabric. The gravel bed promotes drainage and acts as a filter to further prevent sand or soil particles from entering the leachate container.

Measurable releases are being collected from moisture cup 3 which is located approximately 23 cm from the bottom of the wasteforms and the lysimeter leachate collector system (including the gravel) located 51 cm below the bottom of the wasteforms. The radionuclides found in the leachate collector to date are Cs-137, Sr-90, Cs-134, Sb-125.

In this study, we attempted to match the experimental data collected over a period of seven years by INEL from the lysimeters at ORNL and ANL using the computer code DUST. The DUST code is designed to calculate radionuclide release from the wasteform and the subsequent transport in a LLW disposal facility.

## DUST CODE

The code used to model the release of the radionuclides from the lysimeters is DUST (Disposal Unit Source Term). The code is a one dimensional code which models wasteform release and contaminant transport. The DUST code has the ability to simultaneously model three different types of wasteform release mechanisms: diffusion, dissolution, and surface rinse. Transport is modeled by a finite difference method or by a mixing cell cascade approach. The mixing cell model is limited in that it does not take diffusional wasteform release into consideration nor does it model dispersion during transport. Therefore, for our simulation we selected the finite difference model. More detailed descriptions of the models in the code are given (2,3).

## DATA

After review of the data, we chose to model the release of Cs-137 and Sr-90 from Portland types I-II cements located in lysimeter 5 at ORNL and ANL because releases from the lysimeters filled with soil were so low that meaningful comparisons between the computer code and the experimental data could not be performed. Releases from the control lysimeters for cesium, Cs, were less than 2 parts in  $10^7$ , while releases of strontium, Sr, were less than 5 parts in  $10^4$  of their respective initial inventories.

At ANL, lysimeter 5 contained resin type I solidified in Portland type I-II cement, while lysimeter 5 at ORNL contained resin type II also solidified in Portland type I-II cement. Experimental measurements of release from these wasteforms indicate diffusion is the rate controlling mechanism (1). Measured diffusion coefficient values for the resins under study solidified in Portland type I-II cements are  $9.6\text{E-}10\text{ cm}^2/\text{s}$  (4) for Sr-90 and  $5\text{E-}11\text{ cm}^2/\text{s}$  (1) for Cs-137. These values will be used as our base case values. The darcy velocities ranged from  $2.59\text{E-}6\text{ cm/s}$  at ANL to  $3.6\text{E-}6\text{ cm/s}$  at ORNL (5). The soil bulk density values were  $1.55\text{ g/cm}^3$  at ANL and  $1.60\text{ g/cm}^3$  at ORNL (6). Moisture content values were

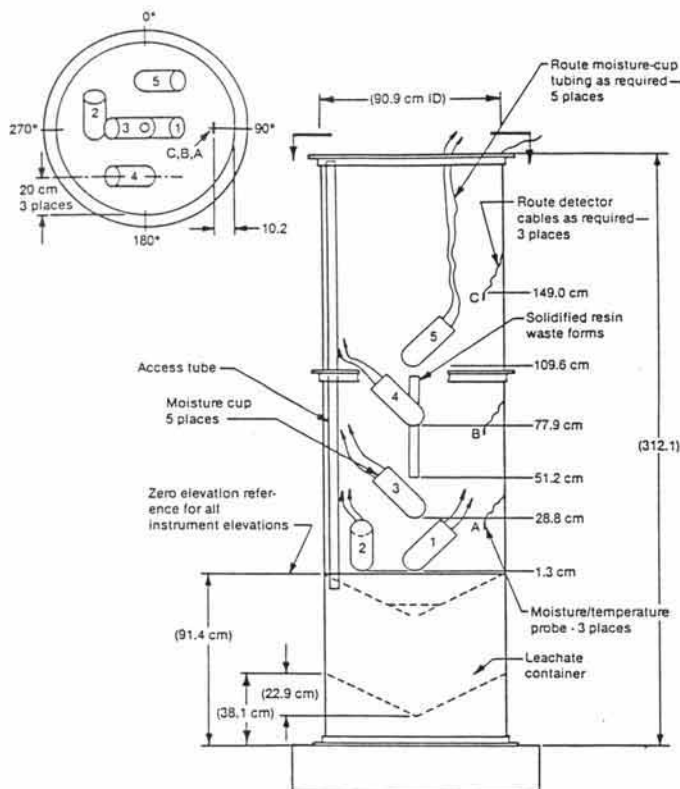


Fig. 1. Cross-section of EPICOR-II field lysimeter.

calculated using the effective soil porosity and the fraction of saturation values found in Rogers, 1988 (1). In lysimeter 5 at both sites the moisture content was calculated as 21%. The dispersion and distribution coefficients were not measured for Sr-90 or Cs-137. As such, they were estimated by fitting the model predictions to the data. The released mass collected from the lysimeters over the first seven years, which was used to make comparisons to the DUST code predictions, represented less than 0.05% of the total inventory of Sr-90 in lysimeter 5 at ORNL and ANL. At ORNL the released amount represented less than 1.7E-5% of the Cs-137 inventory in lysimeter 5.

## MODEL

The base geometry for our model placed 111 nodes with equal spacing of 1 cm. The 7 wasteforms were broken into 53 separate forms of 1 cm height. However, diffusion controlled release from the wasteform was modeled using the total dimensions of the combined wasteforms, that is, a cylinder of 2.4 cm in radius and a height of 53 cm. We took concentration and flux traces to match moisture cup 3 and the lysimeter leachate collector. The concentration and flux traces were taken at the points 23 and 51 cm below the wasteforms. In this paper, we use the total mass released 51 cm beneath the wasteform as our performance measure. Outputs were stored every quarter year and the actual data was interpolated so the model and actual times coincided with one another. The soil diffusion coefficient had a minor role on the release from the bottom of the lysimeter and it was set to  $1\text{E-}7\text{ cm}^2/\text{s}$  at both ORNL and ANL. Zero concentration was specified at the top boundary for the ANL and ORNL trials. Zero dispersive flux was specified at the bottom boundary in both cases. This boundary condition simulates release through drainage. Ini-

tial amounts of Cs-137 and Sr-90 varied at ORNL and ANL due to the fact that they were solidified with two different resin types. In ORNL lysimeter 5, the type I wasteform, had a total initial inventory of 3.29E-3 Ci of Sr-90 and 1.432 Ci of Cs-137 (6). The type II wasteform at ANL had a total initial inventory of 1.84E-2 Ci of Sr-90 (6). Cs-137 was not modeled at ANL for lack of abundant releases.

Three parameters are known to strongly influence release through the lysimeter soil column. They are  $K_d$  (the distribution coefficient) and the dispersion coefficient, which together control transport from the wasteform through the soil column, and the wasteform diffusion coefficient which controls wasteform release rates. Several cases were modeled where either  $K_d$ , the dispersion coefficient, or the wasteform diffusion coefficient were varied to best match the actual release data from the lysimeters.

## RESULTS AND DISCUSSION

During several trials it was proven that altering the time step size from 1.56E-3 to 0.5 years caused the predicted integrated mass flux to vary by a factor of 2-8 during the first 10 years. At these times the fractional release at the lysimeter drain is so small ( $<0.05\%$ ) that numerical truncation errors are important. After 10 years, however, the predicted integrated mass fluxes become independent of time step size over the range tested. Based on these results a time step size of 0.03125 yrs was chosen for problems simulated less than 10 years and 0.125 yrs for trials greater than 10 years.

The results of varying the finite difference mesh space size on predicted mass release at the lysimeter bottom indicated that a one cm. mesh provided sufficient accuracy. Calculating identical problems on a one-half cm. mesh resulted in a difference in predicted cumulative release of less than 20% over 7 years.

A number of trials were run by varying one of the three key parameters (distribution coefficient, dispersion coefficient, or wasteform diffusion coefficient) effecting release and attempting to match the measured release data.

### Effect of $K_d$

In general, increasing the value for  $K_d$  increases the time it takes the contaminant to travel to the lysimeter drain. Therefore, releases are lowered as  $K_d$  increases. Figure 2 shows this effect for Sr-90 using a wasteform diffusion coefficient of  $9.6\text{E-}10\text{ cm}^2/\text{s}$  (5) and an assumed dispersion coefficient of 8.5 at ORNL lysimeter 5. The assumed  $K_d$  values ranged from 22 - 28. The graph indicates how sensitive the release rates are to the  $K_d$  values. Releases at 7 years decrease by a factor of 5 when  $K_d$  changes from 22 to 28.

### Effect of Dispersion Coefficient

In contrast, increasing the value for the dispersion coefficient initially causes mass to reach the lysimeter drain at earlier times. In Fig. 3, the sensitivity of the predicted release to the dispersion coefficient is presented. The  $K_d$  value is held constant at 24.0 and the wasteform diffusion coefficient is held at  $9.6\text{E-}10\text{ cm}^2/\text{s}$ . As a rule of thumb, the dispersion coefficient is typically 0.01-0.1 times the scale of the experiment for unsaturated systems (7). In these lysimeters, the bottom of the wasteform is approximately one-half meter from the measurement point and the top of the wasteform is one meter. Hence, we expect the dispersion coefficient value to range from 0.5 to 10. In our studies we changed the dispersion coefficient from

## Effects of $K_d$ Values On DUST Code Predictions

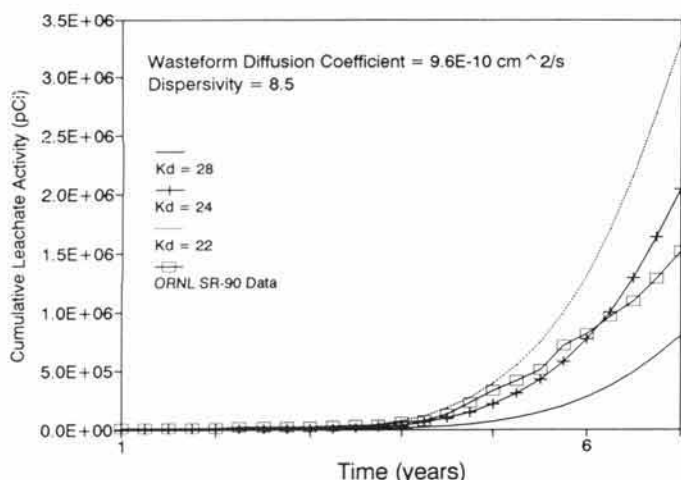


Fig. 2. Effect of  $K_d$  value on predicted Sr-90.

## Effects of Dispersivity On 50yr DUST Code Predictions

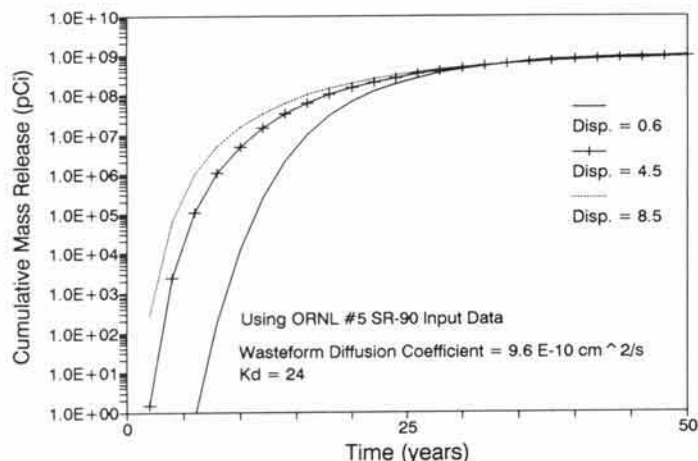


Fig. 3. Effect of dispersion coefficient on predicted release of Sr-90 release from ORNL lysimeter 5.

0.6 to 8.5. In doing so, the predicted mass release varied by six orders of magnitude over the experimental time frame, 7 years.

However, although the mass release is very sensitive to the value of the dispersion coefficient at early times, after 30 years the value of the dispersion coefficient alters the predicted release by less than 10% (Fig. 3). This indicates that ultimately release at the lysimeter drain is controlled by the distribution coefficient.

### Effect of Wasteform Diffusion Coefficient

Figure 4 depicts the effects of wasteform diffusion coefficient on the predicted releases from Sr-90 at ORNL. The  $K_d$  value is held at a constant of 24 and the dispersion coefficient is held at 8.5. Figure 4 shows that if one changes the wasteform diffusion coefficient by two orders of magnitude (from  $9.6E-9$

## Effects of Wasteform Diffusion on DUST Predictions

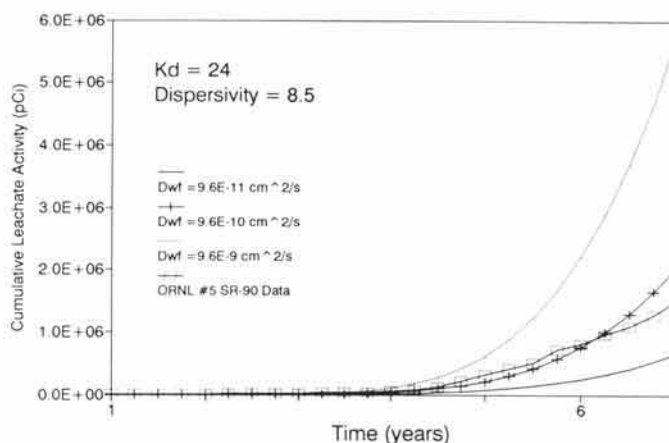


Fig. 4. Effect of wasteform diffusion coefficient on predicted release of Sr-90 from ORNL lysimeter 5.

-  $9.6E-11$  cm<sup>2</sup>/s) the lysimeter release curve increases by slightly less than one order of magnitude. It is interesting to note that the DUST predictions using the measured diffusion coefficient is very close to the ORNL Sr-90 data.

Ancillary calculations of the release from the wasteform indicate that for diffusion release after 7 years approximately 89% of the mass would be released if  $D_{wf} = 9.6E-9$  cm<sup>2</sup>/s; 39% if  $D_{wf} = 9.6E-10$  cm<sup>2</sup>/s; and 14% if  $D_{wf} = 9.6E-11$  cm<sup>2</sup>/s. The experimentally measured values for Sr-90 at the bottom of the lysimeter are less than 0.05% of the total inventory. Therefore, either the wasteform release rates are much lower than those measured in Ref. 5 or transport processes are controlling release through the soil column.

For diffusion controlled release, with fractional releases less than 20%, release is proportional to the square root of the diffusion coefficient. Consequently, if release from the wasteform were controlling releases at the bottom of the soil column, diffusion coefficients would have to be approximately  $1E-17$  cm<sup>2</sup>/s. This is almost eight orders of magnitude less than the measured value. It is very unlikely that the value is this small.

### Best Estimates for Distribution and Dispersion Coefficients

Using the measured wasteform diffusion coefficients, the other two key parameters ( $K_d$  and the dispersion coefficient) were varied to obtain the best fit to the entire release curve. By adjusting these parameters simultaneously, roughly, the same total release can be achieved at a given time. However, the time dependence of mass release varies substantially based on the choice of these parameters. Therefore, we attempted to match the measured concentrations at all times.

Lysimeter leachate concentrations were calculated from the actual data using the amount of mass collected per volume of leachate sampled over a given time period. The actual data was again interpolated so that the time of collection equaled the DUST code output times of every quarter year. The volume of leachate and released amounts were found in Refs. 5 & 8.



Figure 5 contains actual data along with the DUST predicted concentrations for Sr-90 at ORNL lysimeter 5. After several trials we determined our best estimated parameters to be  $K_d = 24$ , dispersion coefficient = 8.5, and the measured wasteform diffusion coefficient of  $9.6E-10 \text{ cm}^2/\text{s}$ . Although the measured release data varies substantially between sampling periods, the general trend of increasing concentration can be seen in Fig. 5. After 6 years, the predicted concentrations are within 25% of the measured concentration. During the 7<sup>th</sup> year, the concentration in solution decreases slightly while the predicted concentration increased leaving a factor of 3 difference between the two. The decrease in measured concentration reflects the year to year variability in the data.

### LYSIMETER #5 ORNL SR-90 CONCENTRATION COMPARISON

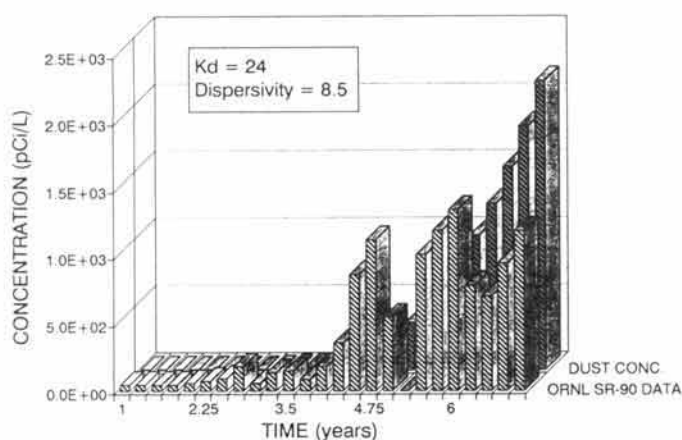


Fig. 5. Predicted versus measured release concentrations of Sr-90 for ORNL lysimeter 5.

Figure 6 contains actual data along with the DUST predicted concentrations for Sr-90 at ANL lysimeter 5. Our best estimated parameters used by DUST in these trials were  $K_d = 24.5$ , dispersion coefficient = 8.5, and the measured wasteform diffusion coefficient of  $9.6E-10 \text{ cm}^2/\text{s}$ . Figure 6 shows predicted concentrations that again show a reasonable fit to the data. This simulation was extended over a 20 year period and the results are seen in Fig. 7. If the model parameters are correct, the concentrations will increase slowly from 1000 pCi/L at 7 years to a value close to  $1E5 \text{ pCi/L}$  at 20 years. This prediction implies that the amount of mass collected in lysimeter bottom will significantly increase over the next 15 years.

The lack of measured dispersion coefficient and  $K_d$  further necessitate the need to obtain longer term data. Figure 8 shows two predicted Sr-90 release curves for lysimeter 5 at ORNL plotted along with the actual data. The first case has a dispersion coefficient = 0.6 and  $K_d = 10$  (case 1). The second has a dispersion coefficient = 8.5 and  $K_d = 24$  (case 2). After 7 years, the model predicts essentially the same total release. At early times, case 2 has the higher releases. However, over 20 years, case 1 will have released 33% of the total Sr-90 inventory, whereas case 2 will have released 3.3% of the inventory. This is caused by the lower value of the dispersion coefficient (which minimizes early releases) and lower  $K_d$

### LYSIMETER #5 ANL SR-90 CONCENTRATION COMPARISON

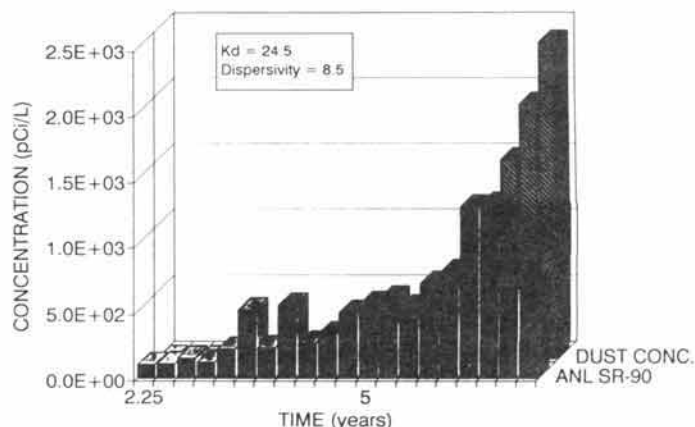


Fig. 6. Predicted versus measured release concentrations of Sr-90 for ANL lysimeter 5.

### LYSIMETER #5 ANL 20YR SR-90 CONCENTRATION COMPARISON

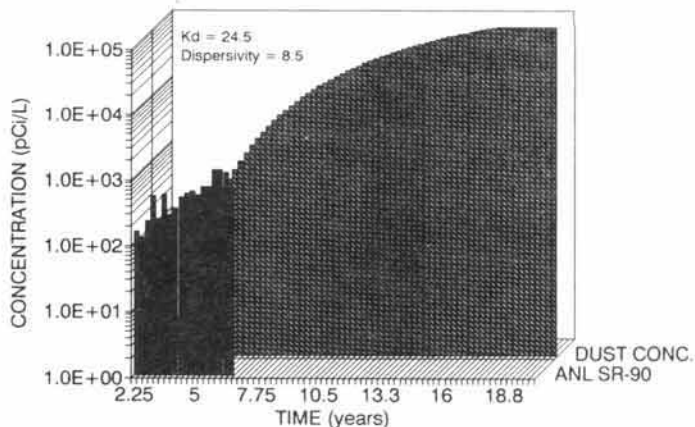


Fig. 7. Predicted concentrations of Sr-90 over 20 years for ANL lysimeter 5.

value (which reduces the travel time causing higher releases) in case 1.

## CONCLUSIONS

This numerical study was hampered by the lack of soil specific data. Due to the high sensitivity of predicted releases to the model parameters ( $K_d$  and dispersion coefficient), it is essential that site specific data be collected for these parameters. In addition, longer term tests are needed to better quantify model parameters, i.e., greater fractional releases are needed to better model the release patterns from the lysimeters. In fact, because less than 0.045% of Sr-90 and  $1.7E-5\%$  of Cs-137 total inventory has been recovered in the lysimeter bottoms, any parameter values estimated in this study are subject to large uncertainties.

The numerical studies of this system show that dispersion is an important factor in determining release over the first 7 years. Using experimentally determined wasteform release

## DUST Predicted 20yr Cumulative Release SR-90 ORNL LYSIMETER #5

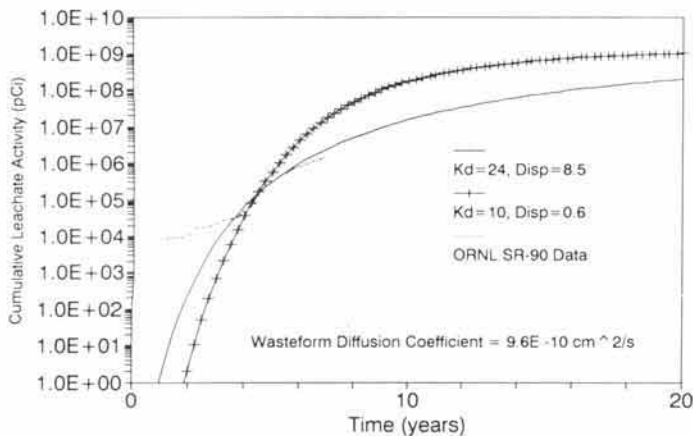


Fig. 8. Predicted cumulative release of Sr-90 over 20 years from ORNL lysimeter 5. Actual releases for the first 7 years are presented for comparison.

rate and the best fit  $K_d$  values, a change in dispersion coefficient from 0.6 to 8.5 altered the mass release by 6 orders of magnitude. After 50 years, however, model predictions indicate that release at the bottom of the lysimeter is independent of the dispersion coefficient to within 10%. This indicates that transport in this system is advection dominated for these values of dispersion coefficient.

Wasteform release rates are not controlling the release measured at the lysimeter drain. Our studies on wasteform release rates indicate that over 39% of the inventory will be released in the first 7 years if the diffusion coefficient is  $9.6E-10 \text{ cm}^2/\text{s}$ , as measured. Since the total release at the lysimeter drain at both sites is less than 0.045%, it appears that transport parameters are controlling release to the drain and solute concentrations.

### RECOMMENDATIONS

- It is strongly recommended that soil distribution coefficients ( $K_d$ 's) be measured for the radionuclides of interest.
- If possible, measurements of the dispersion coefficients should be obtained. Interpretation of dispersion data

may be difficult as dispersion is a function of the pore size distribution, scale of the experiment, and the presence of impurities, e.g., small stones. Therefore, the exact conditions of the measurement sample may not match those in the lysimeters.

- These lysimeter experiments should be continued. After seven years, release was less than 0.045% of the inventory for any radionuclide. This is far too little for obtaining a reliable, quantitative, understanding of the system.

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