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COMPARISON OF UNSATURATED FLOW AND TRANSPORT MODELS WITH VOLUME AND NITRATE MEASUREMENTS FROM A PILOT-SCALE, IN-SITU LYSIMETER WITH DIFFERENT GEOMETRY LOW LEVEL RADIOACTIVE WASTE DESIGNS*

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ABSTRACT

Results from a large-scale field lysimeter experiment containing three thirty-ton monoliths formed from actual decontaminated nuclear waste have been used to validate flow and transport models. The models used are a two-dimensional, finite difference, air and water flow model and a two-dimensional, finite difference transport and diffusion model. The best match of nitrate concentrations calculated to be released from the monoliths with values observed from the lysimeter occurred with monolith diffusivities in the range of 5×10^{-9} to 2×10^{-8} cm^2/sec and dispersivities ranging from 15 to 65 cm. The validated models have been used to predict the performance of a low-level waste disposal facility at the Savannah River Site.

INTRODUCTION

The Savannah River Site (SRS) is a Department of Energy (DOE) facility for producing defense nuclear materials. At SRS, a process has been developed to dispose of low-level, liquid radioactive wastes by stabilization with blast furnace slag, flyash, and cement[1,2]. The resulting mixture, called saltstone, is poured into large concrete forms where it solidifies. The solid saltstone monolith will then be externally covered with concrete. The filled and covered concrete forms will be mounded over with earth and a clay cap. Prediction of the performance of the resulting landfill design required validation of mathematical models by comparison with field data. Design optimization required both the collection of laboratory and field data as well as requiring the use of mathematical models to extrapolate these data to full-scale and to longer time periods.

Previous calculations compared an unsaturated zone flow model with SRS lysimeter observations. This was done in a steady-state fashion assuming the lysimeter had reached a relatively steady condition. The lysimeter experiment used for this previous work was termed a 1/10 scale lysimeter test. This lysimeter contained a waste form called saltcrete which used a different formulation than the present saltstone mix. There are now five years of data available from another field lysimeter experiment. This test is termed the Tank 24 lysimeter. The Tank 24 lysimeter consists of three separate saltstone monoliths each with a different cap placed within individual sections of the same overall lysimeter. The acquisition of these data provided a better understanding of the transient, unsaturated flow mechanisms and the importance of different design waste forms.

Lysimeter Construction and Data

The in-situ field experiment in a lysimeter with unconfined monoliths of saltstone waste has been in progress at SRS over the last five years. A schematic of the lysimeter is shown in Figure 1. Two of the three different lysimeters, the no cap and gravel cap designs, have provided excellent quality data. The third, the clay cap design, has been shown to have a leak thought to be at the liner-concrete sump seal and as a consequence, the clay cap results are not discussed further.

The Tank 24 lysimeters consisted of three separate lysimeters each being trapezoidal in shape having a top surface area of 35 x 45 feet and a bottom area 7 x 17 feet. Figure 1 presents transverse and longitudinal sections through the lysimeter installation. From the transverse section at the top of Figure 1, it can be seen that all three lysimeters contain a saltstone monolith—two of which are covered with cap materials, gravel, and clay. The top of each monolith is located approximately 8 feet below the original grade while the bottom of the lysimeter lies about 22 feet from the ground-surface. From Figure 1, it can be noted that the bottom 14 feet of the lysimeter is lined with an impermeable membrane (hypalon). Each lysimeter contains a pump (not illustrated) which is located in a concrete sump

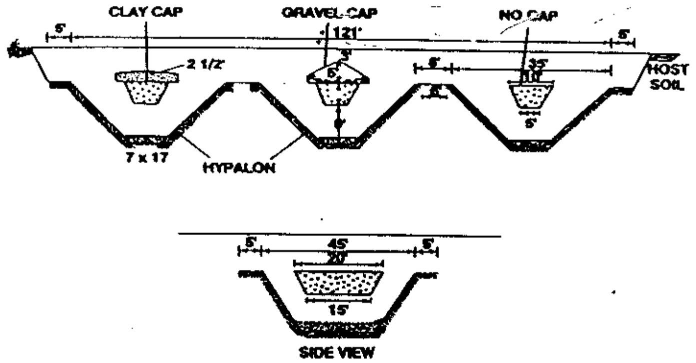

FIGURE 1. SCHEMATIC DIAGRAM OF SALTSTONE LYSIMETERS

at the bottom of the lysimeter. The facilitate drainage, a gravel layer (on the order of one foot thick) lines the bottom of each lysimeter.

The sides of the top 8 feet of the lysimeters are unlined as well as are horizontal sections (6 feet long) between each of the lysimeters. The transverse section at the top of Figure 1 and the side view longitudinal section below illustrate that the monoliths are each 5 feet thick with a top and bottom dimension of 10 feet and 5 feet, respectively, and the longitudinal dimension is 20 feet on top and 15 feet on the bottom.

The main data collected at the Tank 24 lysimeters are the pumped volumes and the results of chemical analyses performed on a sample of the pumped water. In addition, suction cups were placed around the saltstone waste forms and chemical analyses on this fluid were also performed. Pumping was first performed on May 29, 1984 approximately 3 1/2 months after the lysimeters were exposed to natural precipitation. Flow records from this first pumping period are probably not as reliable as the subsequent pumped volumes. The lysimeters were pumped for the second time on August 10, 1984 and subsequently on a two week basis. Flow volumes after August 10, 1984 were more accurately recorded than for the initial pumping period. Samples were taken from the volumes removed from each lysimeter and analyzed for nitrate and other constituent concentrations.

Rainfall data were available from various locations around the SRS plant site. Daily rainfall data from gauges at 200-F and 200-H Areas were available over the entire history of the site. These gauges are both located approximately one mile from the lysimeter site. In addition, weekly data were available from the SRS Burial Ground rain gauge located approximately 1/4 mile from the site. Data were available from the Tank 24 lysimeter site beginning in February 1985. A tipping bucket rain gauge was installed at the site in June 1985. These data are useful since they indicate the duration of each rainfall event as well as the total volume of rain. The composite precipitation record is shown in Figure 2(a). In the figure, the precipitation has been accumulated over the periods between pumping the lysimeter and is shown as a cumulative precipitation rate bar graph over the two week periods. Also shown in Figure 2 as 2(b) and 2(c) are the collected sump volumes for the gravel and no cap monolith lysimeters, respectively. As can be noted in Figure 2, the correlation between precipitation and collected sump volumes appears to be low. Even the correlation between the no cap and gravel cap collected volumes appears

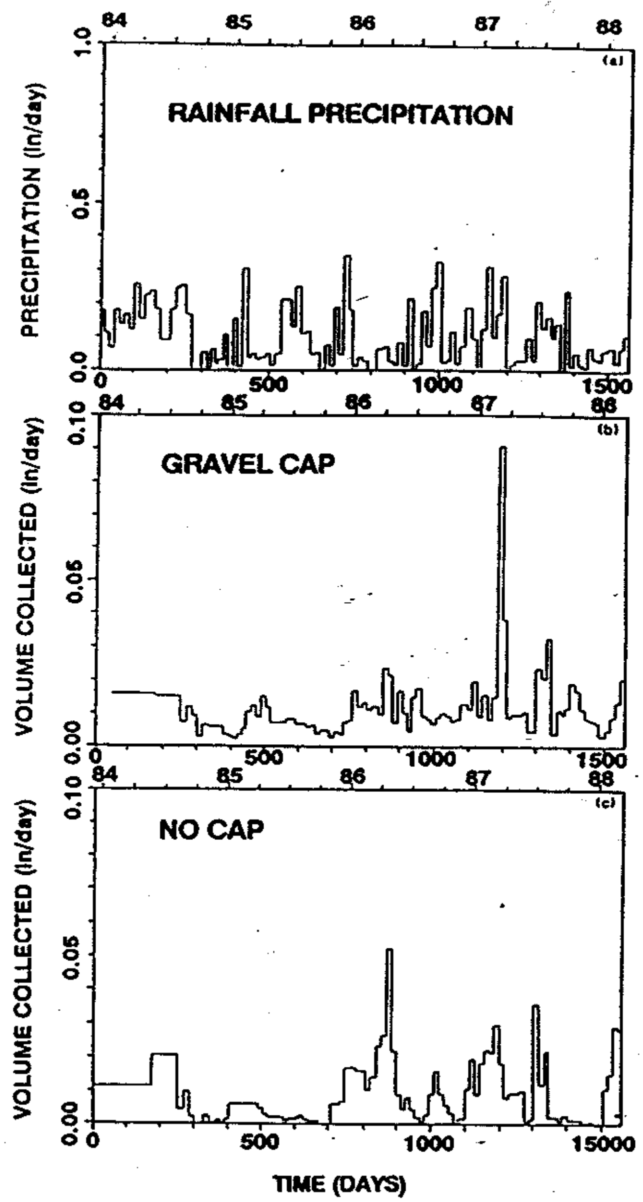

FIGURE 2. MEASURED PRECIPITATION AND VOLUME-COLLECTION RATES AT TANK 24 LYSIMETERS

to be low. The models used with a range of unsaturated zone properties were intended to help explain the differences in the observed results.

In addition to the volume rates being different, the measured nitrate concentrations reaching the sump have been substantially different for the no cap versus the gravel cap lysimeters. These differences are illustrated in Figure 3.

The gravel-cap saltstone lysimeter gave nitrate breakthrough almost 2.5 years later than the no cap saltstone. This difference in nitrate leaching could be caused by either unsaturated flow property differences along with the "wick effect" of the gravel cap or it could be caused by either variations in leaching within the saltstone or dispersion with the backfill. It is important to determine whether the unsaturated models along with nitrate transport models can explain this difference.

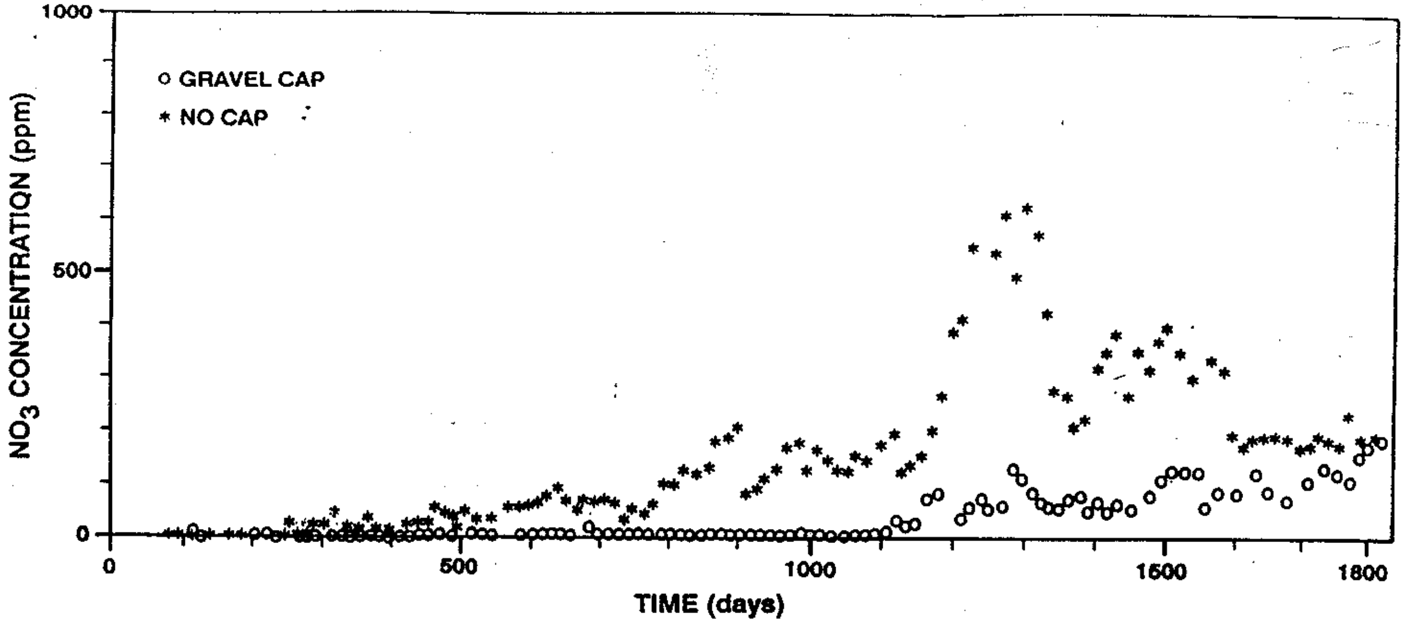

FIGURE 3. OBSERVED NITRATE CONCENTRATION FROM LYSIMETERS

Models Used

This paper presents the comparison of mathematical model calculated results with the collected field data. A combination of mathematical models was used. A one-dimensional finite difference solution of the Richards Equation was used to determine the net infiltration/evaporation algorithm for the lysimeter. Measured daily precipitation and average historical monthly pan evaporation rates were input to the model. Comparison of the calculated flow to that pumped from the lysimeter sump was used as a criterion for the adequacy of the net infiltration/evaporation algorithm.

Although a number of both saturated and unsaturated measured soil properties were available, these measurements define a fairly wide range in properties. Saturated conductivities varied two orders of magnitude. Measured unsaturated relative conductivity and capillary suction varied significantly as well. As a consequence, a stochastic version of the finite difference solution of the Richards Equation was used to determine an uncertainty range in net infiltration/evaporation which could result from the measured range in saturated and unsaturated properties. This stochastic model used Latin Hypercube sampling of nine different input property vectors to provide a set of parameters for each Richards Equation solution. Two separate analyses of one hundred samples each were performed. Some of these solutions indicated the bottom part of the lysimeter would become nearly saturated. If a saturated condition exists, the incompressible Richards Equation approach is not adequate for predicting the observed unsteady drainage rates.

A finite difference two-dimensional, two-phase air and water flow model was used to represent the complex flow through the lysimeters. A finite difference transport and diffusion model was coupled to the two-phase flow model. The transport and diffusion model includes leaching and diffusion of nitrates out of the very low permeability saltstone. A non-orthogonal calculational grid was initially used for the flow model to provide assurance that grid effects were not impacting the calculated results. A refined Cartesian grid was then used to perform both the two-phase flow and transport calculations. This 46 x 42 grid was chosen to minimize numerical truncation errors by getting high grid refinement near the saltstone/soil interface where the diffusion gradient was large. The grid is illustrated in Figure 4 for the gravel-cap lysimeter and used grid sizes ranging from 0.125 feet to 1.5 feet. The same grid was used for the no-cap lysimeter with backfill soil properties replacing those of the gravel cap. The model used an automatic time stepping feature but maximum time steps were limited to five days.

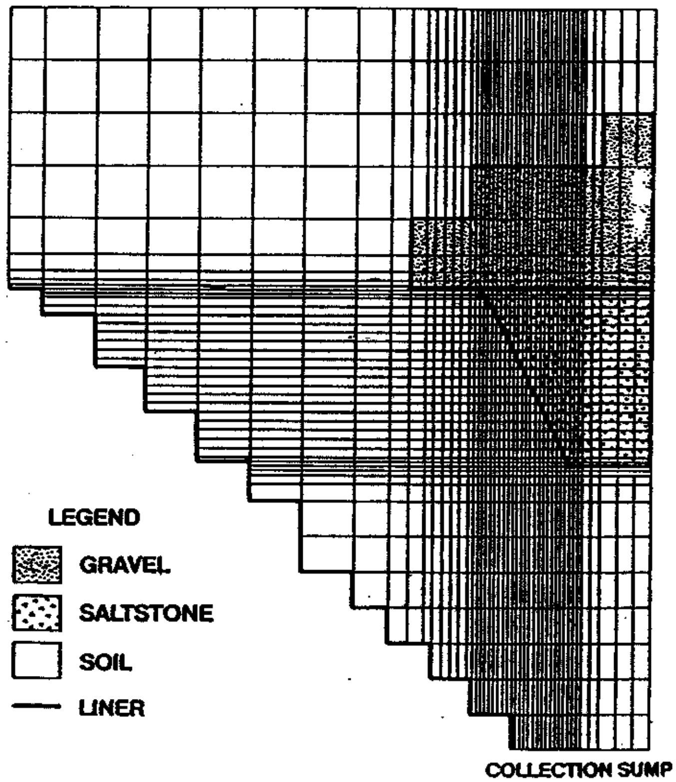

FIGURE 4. SCHEMATIC OF REFINED CARTESIAN GRID USED IN ECLIPSE

Lysimeter Property Data

The two lysimeters used for this study are comprised of soil backfill, the cap area above the monolith (either gravel or soil backfill material was used), saltstone, the gravel sump and the hypalon liner. There is a small area of undisturbed host soil in contact with soil backfill between lysimeters but this area was not modeled. All man-placed materials, backfill soil, gravel and saltstone were assumed to be isotropic. Because of weathering and vegetation growth, a near surface layer might be expected over time to envelop an increased hydraulic conductivity. Use of an increased near-surface conductivity did appear to give better comparison of calculated and observed results. The saturated properties used for each material are summarized in the table below:

Saturated Hydraulic Material Properties for Lysimeter

Material	Hydraulic Conductivity (cm/s)	Porosity (%)
Backfill Soil	10^{-4} (range 10^{-5} to 10^{-3})	33 - 45
Saltstone	2.5×10^{-9}	46
Gravel Cap	0.5 (range 0.1 to 1.0)	33 - 45
Hypalon	Assumed 0	0.01

If host soil properties were needed, a range of 10^{-5} to 2×10^{-4} was used with a horizontal/vertical anisotropy of about two.

The unsaturated soil properties for soil/backfill were taken from the range of laboratory measured values. The range over which the stochastic sampling was done and the midpoint value is illustrated in Figure 5.

Three sets of gravel cap unsaturated properties were used. In the first, the "best" set of properties which could be extrapolated from the literature[4] were used to define a "recommended" gravel. In the second set, an upper bound on properties which cause the most water take-up by the gravel cap were used. These properties corresponded to the gravel being equivalent to a "stony soil". The final set of properties used for the gravel was to assume it was totally impermeable. The two phase flow model showed that the "recommended" gravel performed essentially as if the gravel were impermeable. The "stony soil" gravel took in a volume of water over four times greater than that of the "recommended" gravel. However the calculated cumulative flow to the lysimeter sump differed by only a few percent. The unsaturated properties for the "recommended gravel" and the "stony" soil ware illustrated in Figure 6.

Comparison of Model Calculations and Lysimeter Data

The best case infiltration/evaporation algorithm determined by stochastic sampling in a one-dimensional Richards Equation was used as input to the two-phase flow model. The soil conductivity for the no cap lysimeter was 3.9×10^{-5} cm/sec and that for the gravel cap was 3.4×10^{-5} cm/sec. A 35% porosity was used for the soil in both

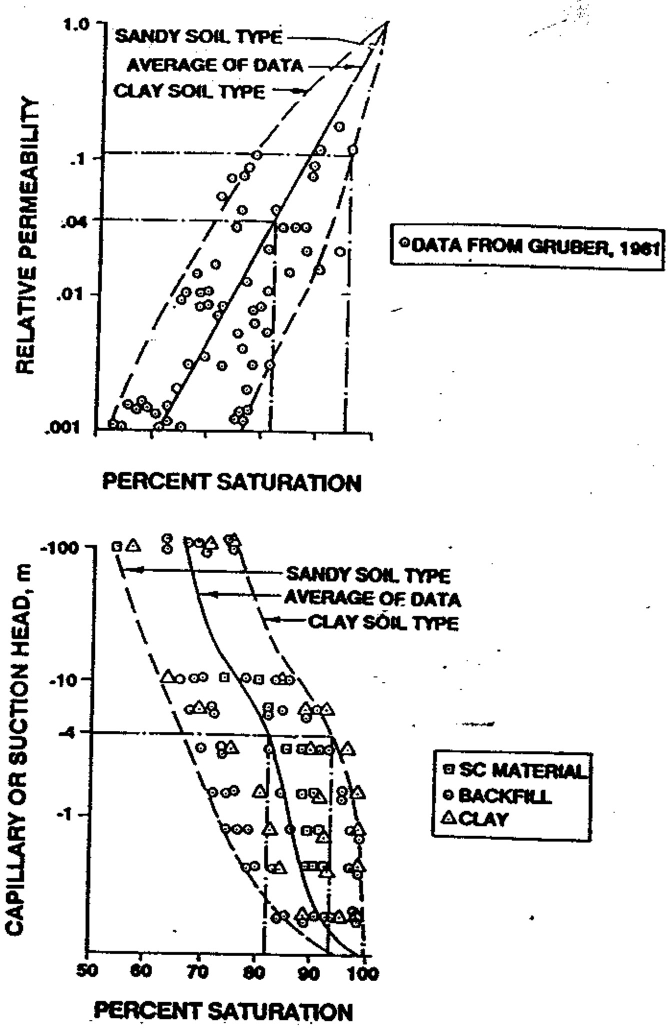

FIGURE 5. MEASURED UNSATURATED PROPERTIES FOR SRP SOILS

lysimeters. The capillary pressure and relative conductivity curves as a function of saturation were the same for both lysimeters. The capillary pressure curve was essentially the same as the midpoint curve representing the laboratory data (see Figure 5). The relative conductivity curve was essentially the low end conductivity from the laboratory data. Ninety one (91) percent of the average observed SRS pan evaporation rate was found by the stochastic sampling to best represent the net infiltration.

The calculated collection results for the no cap lysimeter are compared to the observed sump rate and the cumulative collection in Figure 7. The calculated values are solid lines and are compared to the observed values represented by dashed lines. The comparison of cumulative collections are quite good while the calculated rates seem to be correct on average but miss some observed peak rates as well as overestimate the low observed collection rates.

The calculated collection results (solid lines) for the gravel cup lysimeter using "recommended" gravel are compared with the observed values (dashed lines) in Figure 8. Again the calculated cumulatives are good but the calculated rates seem averaged compared to the observed collection rates. Results for the "stony" gravel and the

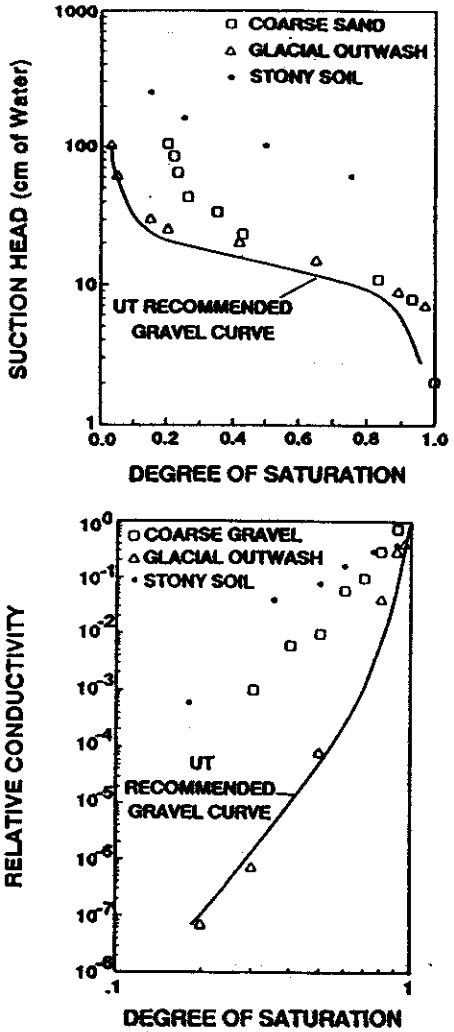

FIGURE 6. ILLUSTRATION OF GRAVEL UNSATURATED PROPERTIES

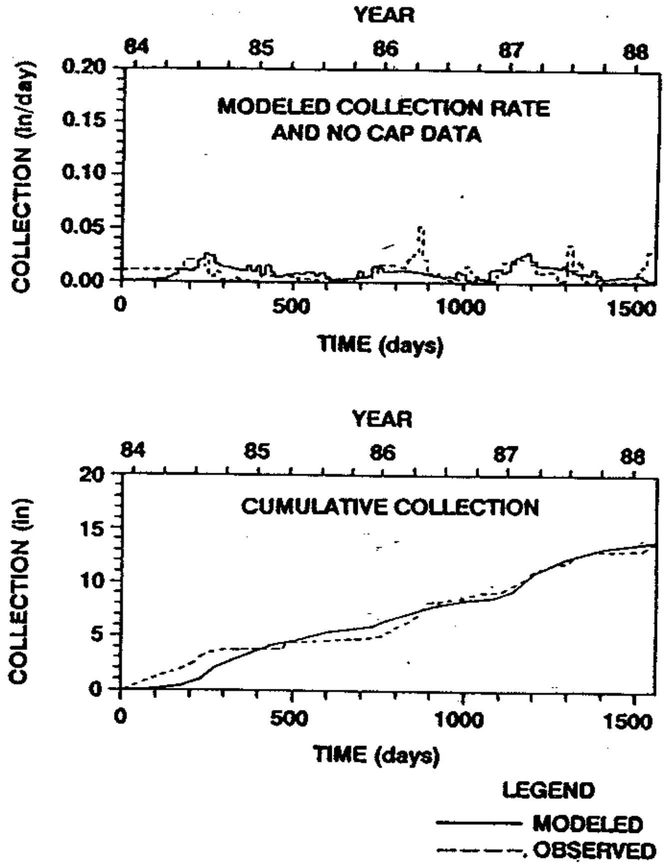

FIGURE 7. CALCULATED vs OBSERVED VOLUMES FOR NO-CAP LYSIMETER

impermeable gravel differed little from the "recommended" gravel.

The nitrate results were calculated using a range of dispersivity coefficients in the soil and a range in diffusivities for nitrate exiting the saltstone. The following table summarizes the range examined.

Range of Saltstone Diffusivities and Soil Dispersivities

Lysimeter Design	Saltstone Diffusivity, (cm^2/sec)	Soil Dispersivity, (cm)
No Cap	1×10^{-9} to 1×10^{-7}	30 - 150
Gravel cap	5×10^{-9} to 1×10^{-8}	10 - 20

A plot of the calculated results for the best set of soil dispersivity and saltstone diffusivity parameters is shown in Figure 9.

As can be noted, the calculated NO_3 concentrations for the no cap lysimeter underestimate the peak observed concentrations and do not appear to have reached a maximum concentration. The calculated NO_3 concentrations for the gravel cap lysimeter also appear to underestimate the peak observed concentration to date occurring at about 1,300 days. Data available in the latter part of 1988 and not plotted in the figures show the no cap lysimeter concentrations have continued to decline to around 200 ppm (mg/L) while the gravel cap NO_3 levels have continued to increase to about 200 ppm.

The impact of not reproducing the observed lysimeter sump volumetric rates was examined by specifying the produced water in the flow model applied to the no cap lysimeter. this, of course, does not mean the saturation and flows within the lysimeter also match the actual ones. The calculated NO_3 concentrations using these rates, however, did illustrate a peak concentration at about 1,300 days corresponding more closely with the observed NO_3 results. It should be noted that the differences in calculated concentrations caused by soil and saltstone parameters was significantly greater than that caused by differences between the calculated no cap and gravel cap produced volumes.

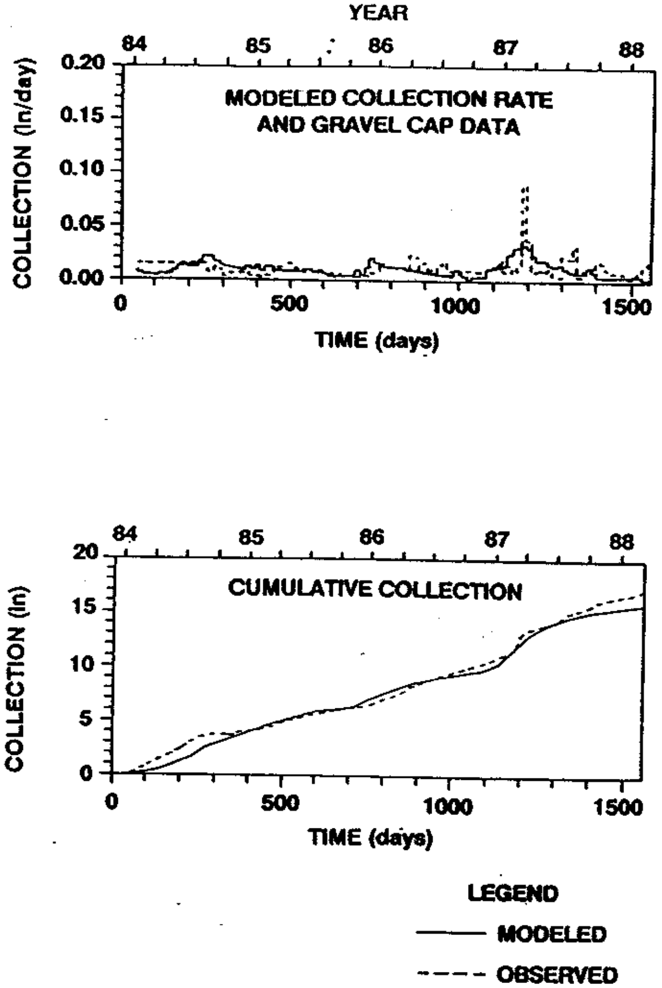

FIGURE 8. CALCULATED vs OBSERVED VOLUMES FOR GRAVEL-CAP

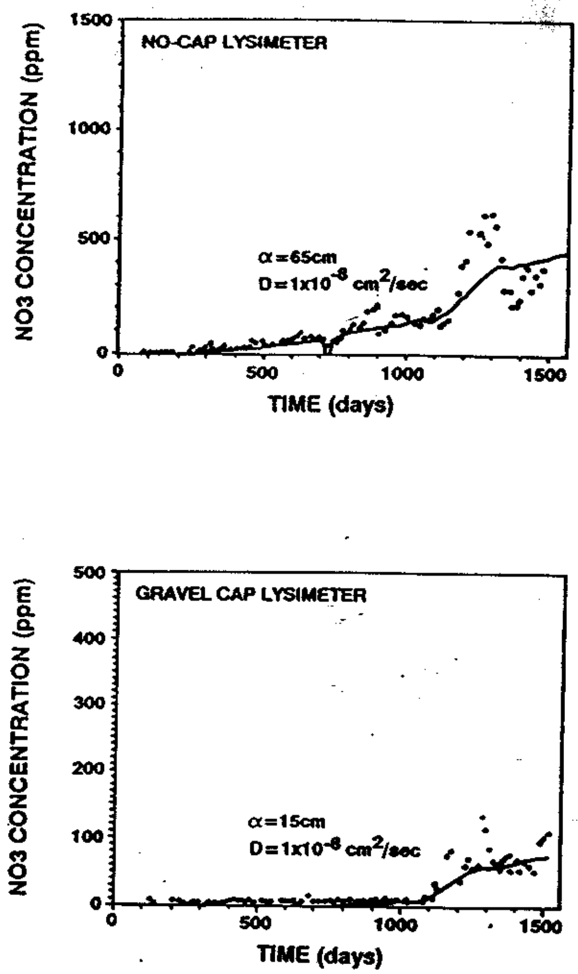

FIGURE 9. CALCULATED vs OBSERVED NITRATE NO_3 FOR LYSIMETERS.

A number of combinations of soil dispersivity and saltstone diffusivity can be used to give about the same quality of match of the observed concentrations. Larger soil dispersivities can be compensated by using lower saltstone diffusivities and lower dispersivities by higher diffusivities. The compensations are not precisely proportional, however. The range of saltstone diffusivities which best matched the gravel and no cap observed NO_3 concentration data provide a range from 5×10^{-9} up to about 2 x 10-8 m²/sec. This range is very consistent with laboratory measurements on saltstone samples poured during the construction of the lysimeter. The range in best fit soil dispersivities differs by a factor of about four - 15 cm to 65 cm. Again this range in dispersivities is consistent with what would be expected from literature values.

It should be noted that there were suction cups installed around the saltstone monolith in each lysimeter. Although there was difficulty in getting reliable NO_3 data from these suction cups, the general observed concentration levels around the monolith were in the 10,000-20,000 mg/L range. Calculated results adjacent to the monolith were in the range of 6,000-21,000 mg/L NO_3 thus indicating good agreement with the observations.

A logical question to ask is—are these variabilities in diffusivity and dispersivity reasonable considering the saltstone was poured to be the same insofar as possible and the soil in the two lysimeters was a homogeneous and uniformly packed as could be expected? It is our opinion that the answer to the above question is yes. It would be unreasonable to expect large block saltstone pours to have better reproducibility than probably an order of magnitude in saltstone diffusivity—especially in the range of 10^{-8} cm²/sec. Soil dispersivities within a factor of five might be expected. However, considering the range in both unsaturated and saturated properties measured on soil cores taken from the same area, a factor of 3-5 in dispersivity should not be too surprising. The soil variability undoubtedly could cause this factor of difference. It is our opinion, that the use of two phase plan and transport multi-dimensional models has been shown to explain the observed differences in the two lysimeters and can be used reliably to extrapolate these results into the future.

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