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MECHANISMS OF CONTAMINANT MIGRATION FROM GROUTED WASTE

by

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INTRODUCTION

Low-level radioactive decontaminated salt solution is generated at the Savannah River Site (SRS) from the In-Tank Precipitation process. The solution is mixed with cement, slag, and fly ash, to form a grout, termed "Saltstone", that will be disposed in concrete vaults at the Saltstone Disposal Facility (SDF) [1]. Of the contaminants in the Saltstone, the greatest concern to SRS is the potential release of nitrate to the groundwater because of the high initial nitrate concentration (0.25 g/cm^3) in the Saltstone and the low Safe Drinking Water Act (SDWA) maximum contaminant level (MCL) of 44 mg/L. The SDF is designed to allow a slow, controlled release over thousands of years. The release is limited or controlled by the low Saltstone hydraulic conductivity and low diffusivity. The facility performance objective is to comply with the SDWA MCL of 10 mg/L of $NaNO_3$ as nitrogen [2], or 44 mg/L as $NaNO_3$, in the groundwater at the SDF boundary.

This paper addresses a modeling study of nitrate migration from intact non-degraded concrete vaults in the unsaturated zone for the Radiological Performance Assessment (PA) of the SRS Saltstone Disposal Facility [3]. The PA addresses the performance requirements mandated by DOE Order 5820.2A [4]. Simulations of degraded concrete vaults were also performed but are not part of this paper.

THE SALTSTONE DISPOSAL FACILITY

The SDF will consist of 15 concrete vaults. The first vault, completed in 1990, is 100-feet wide by 600-feet long by 25-feet high. The other 14 vaults will be twice as wide. The bottom of the vault is at ground level which is at least 20 feet above the historical high water table. The vault sides are 1.5-feet thick and the vault floor is 2-feet thick. The vault will be filled with 24 feet of contaminated grout with one foot of uncontaminated grout at the top. A sloped concrete roof approximately 3-feet thick at the center and 2-feet thick at the edge will complete the vault.

After the vaults are filled, the SDF will undergo closure. Although the closure plan has not been completed, the current closure concept includes backfilling around the vault, backfilling two more feet above the vault, and then placing two feet of clay, one foot of gravel, three feet of soil, and two feet of top soil above the backfill. The purpose of the closure is to reduce infiltration, to reduce the likelihood of exposure to an inadvertent intruder, and to minimize the probability of waste exposure by erosion. A schematic diagram of the SDF is shown in Figure 1.

CONCEPTUAL MODEL

Modeling Domain

A two-dimensional vertical cross-section of a Saltstone vault was modeled (Figure 2). The top of the simulation domain was the backfill soil below the clay cap. The closure cap was excluded to reduce the model complexity. However, the effect of the closure cap was represented by a reduced infiltration rate at the upper boundary. The bottom of the simulation domain was the water table. Only half of a vault was simulated to take advantage of symmetry. The Saltstone grout monolith was placed at the upper left portion of the model and was surrounded by the concrete vault. The vault design included a sloping concrete roof; however, this was not included in the simulation and can be considered conservative since in increases the amount of perched water on the roof.

Initial and Boundary Conditions

An initial nitrate pore water concentration of 0.25 g/cm^3 was calculated based on the inventories to be disposed and the grouting process. There was assumed to be no nitrate in the pore fluid of the concrete vault or in the backfill soil. The concrete vault and the Saltstone were assumed to be at 100% water saturation. The backfill soil water saturation was calculated from the soil characteristics (relative conductivity and capillary pressure) with a 40 cm/yr infiltration, the accepted rate at SRS based on lysimeter tests and water balance studies.

The boundary conditions used for the simulation were: 1) constant water influx of 2 cm/yr at the top of the domain which accounts for the effect of the closure cap; 2) the bottom of the simulation domain was represented by the water table which was modeled with a prescribed head of zero cm; 3) a zero nitrate concentration was assigned to the top and the bottom boundaries assuming nitrate was quickly removed at the aquifer; and, 4) no convective or diffusive flux was allowed at the vertical boundaries due to symmetry. One vertical boundary represented the centerline of a vault while the other boundary was midway between vaults.

Transport Mechanism

The dominant mechanisms for nitrate release from the Saltstone were convection and diffusion. Convection results as a small amount of perched water flows through the vault and leaches nitrate from the Saltstone. Diffusion occurs as a result of the concentration gradient between the Saltstone and the model boundaries. As a consequence of these two processes, simultaneous transient flow and transport were simulated. This option was more realistic than using a steady-state flow assumption because it took approximately 2000 years for the flow field to reach steady-state.

The relative importance between convection and diffusion was governed by the flow and transport properties of the porous media. It was assumed that the properties of the materials remained unchanged during the entire period modeled. The hydraulic conductivities and the diffusivities for Saltstone, concrete, and the backfill used in this study were:

Material	Conductivity (cm/sec)	Diffusivity (cm^2/sec)
Saltstone	1.0×10^{-11}	5.0×10^{-9}
Concrete Vault	1.0×10^{-10}	1.0×10^{-8}
Backfill Soil	1.0×10^{-5}	5.0×10^{-6}

The Saltstone diffusivity has been measured extensively in leaching tests [6]. The measured slag-formulated saltstone conductivity data ranges between 1×10^{-12} and 1×10^{-8} cm/sec. Further validation tests to better determine the saltstone conductivity are underway.

Other mechanisms that typically affect contaminant transport are adsorption, decay, chemical reactions, and solubility limits. These were unimportant in this study because nitrate is non-sorbing, non-decaying, non-reactive and highly soluble.

MODELING METHODOLOGY

The PORFLOW 2.40 [5] simulation code was used for the study. A 56 x 97 computational grid which was non-uniformly spaced was used for the simulation and is shown in Figure 3. The vertical grid dimensions were optimized by one-dimensional modeling. Small grid blocks at the material interfaces were used to minimize numerical dispersion. Harmonic averages of flow and transport properties were used for the calculation.

Because of the huge contrast in conductivity and diffusivity between the backfill and the cementitious materials, most of the infiltrating water was diverted through the backfill. Only a small fraction of water penetrated the vault. Since all of the nitrate was initially in the Saltstone, the amount of water penetration was crucial to nitrate release. It was found that a double-precision code was necessary to account for the minute changes in water penetration.

A valuable lesson learned from this study was to use small time steps and a large number of iterations, especially in the beginning of the run. Earlier runs using relatively large time steps resulted in a mass balance error of more than 100%. A run with a starting time step of 0.0001 year which gradually increased to 1 year by 10000 years reduced the mass balance error to less than 0.01%. Using small time steps also reduced the number of iterations per time step.

The alternate direction implicit (ADI) solver was used for this application. Because the problem was basically vertical flow, three vertical (y-direction) sweeps were followed by one horizontal (x-direction) sweep. An abrupt change was imposed on the system when the vault was superimposed and the infiltration was reduced from 40 to 2 cm/yr. These impulses resulted in a significant mass balance error even when very small time steps were used. A solution to the mass balance problem was to solve only the pressure equation for the first year and then solve for pressure and mass simultaneously. The pressure solution was discontinued once steady-state conditions prevailed which occurred by 2000 years.

RESULTS

Based on conservative assumptions and the best available data, a Base Run (Run 1) was conducted to predict nitrate release from the Saltstone vault. The resulting steady-state water saturations and pressure heads are shown in Figures 4 and 5. Due to the small pore structure of the concrete and saltstone, both of these were completely saturated. The 2 cm/yr infiltration rate resulted in water perching above the vault and being diverted around the side of the vault through the backfill. The area beneath the vault was relatively dry and had the highest capillary pressure. Figure 6 shows the flow streamlines for steady-state conditions with arrowheads to indicate travel times. Due to the dramatic differences in water velocities in separate regions of the simulation, three different time interval markers were used to illustrate water movement. Above and beside the vault, the travel time markers indicate 10 years. Below the vaults, the time markers indicate 5000 years. Within the saltstone itself, the travel time between arrowheads is 50000 years.

Nitrate could either migrate upward against infiltration or downward to the aquifer. With 2 cm/yr infiltration, the amount of nitrate that diffused through the top boundary was negligible. More than 99.99% of the released nitrate discharged to the aquifer. The rate of nitrate discharge was obtained by integrating the product of concentration and flow rate across the bottom of the simulation domain to obtain a release rate in grams per year for the modeling domain. Since the modeling domain was a prototype for the SDF, the release rate was divided by the initial inventory to obtain the fractional yearly release. This predicted fractional yearly release for the Base Run is shown in Figure 7. This release history was then used as a key input to the saturated zone model. The saturated zone model consisted of a three-dimensional steady-state flow field and transient contaminant migration. The predicted peak concentration at the site boundary resulting from the intact vault simulation was 8 mg/L, about 1/5 of the SDWA MCL. This saturated transport simulation is outside the scope of this paper.

DISCUSSION

The bottleneck to nitrate release was diffusion and convection in the Saltstone and concrete. Once nitrate was released into the soil, it was transported to the water table by convection at the top and side of the vault and mainly by diffusion under the vault. Two hypothetical cases were tested to verify the relative importance of the transport mechanisms. In Run 2, the conductivity of the vault was assumed to be zero. In Run 3, the diffusivity of the vault was assumed to be zero. The fractional release curves for Runs 2 and 3 are compared to that of the Base Run in Figure 8. Under the assumptions used for the runs, diffusion and convection appear to be about equally important. The peak fractional release of 3.0×10^{-6}/yr for the Base Run was slightly lower than 3.4

x 10^{-6}/yr, the combined peak releases from Runs 2 and 3. This probably resulted from an offset of upward diffusion by downward convection at the top of the vault.

Three more runs were conducted to verify the key mechanisms. In Run 4, the conductivities of both Saltstone and concrete were increased by a factor of 10 while the diffusivities were kept the same as the Base Run. In Run 5, the conductivities were the same as the Base Run, whereas the diffusivities were increased by a factor of 10. In Run 6, both conductivities and diffusivities were increased by a factor of 10. The peak fractional releases increased to 1.7 x 10^{-5}/yr for Run 4, 2.0 x 10^{-5}/yr for Run 5, and 2.6 x 10^{-5}/yr for Run 6, compared to 3.0 x 10^{-6}/yr for the Base Run (Figure 9).

From a mass transport point of view, the Saltstone and the concrete were in series. Low convective flux was obtained when either or both of the Saltstone and the concrete conductivities were low. Low diffusive flux was obtained when either or both of the diffusivities were low. The effect of secondary parameters such as dispersivity, infiltration rate and soil characteristics were also modeled but will not be covered in this paper.

CONCLUSIONS

There are two primary conclusions resulting from this simulation study. They are:

The flow and transport phenomena in the unsaturated zone for the intact SDF were successfully modeled by PORFLOW.

Modeling results indicated that the intact SDF will meet DOE Order 5820.2A performance objectives.

ACKNOWLEDGEMENT

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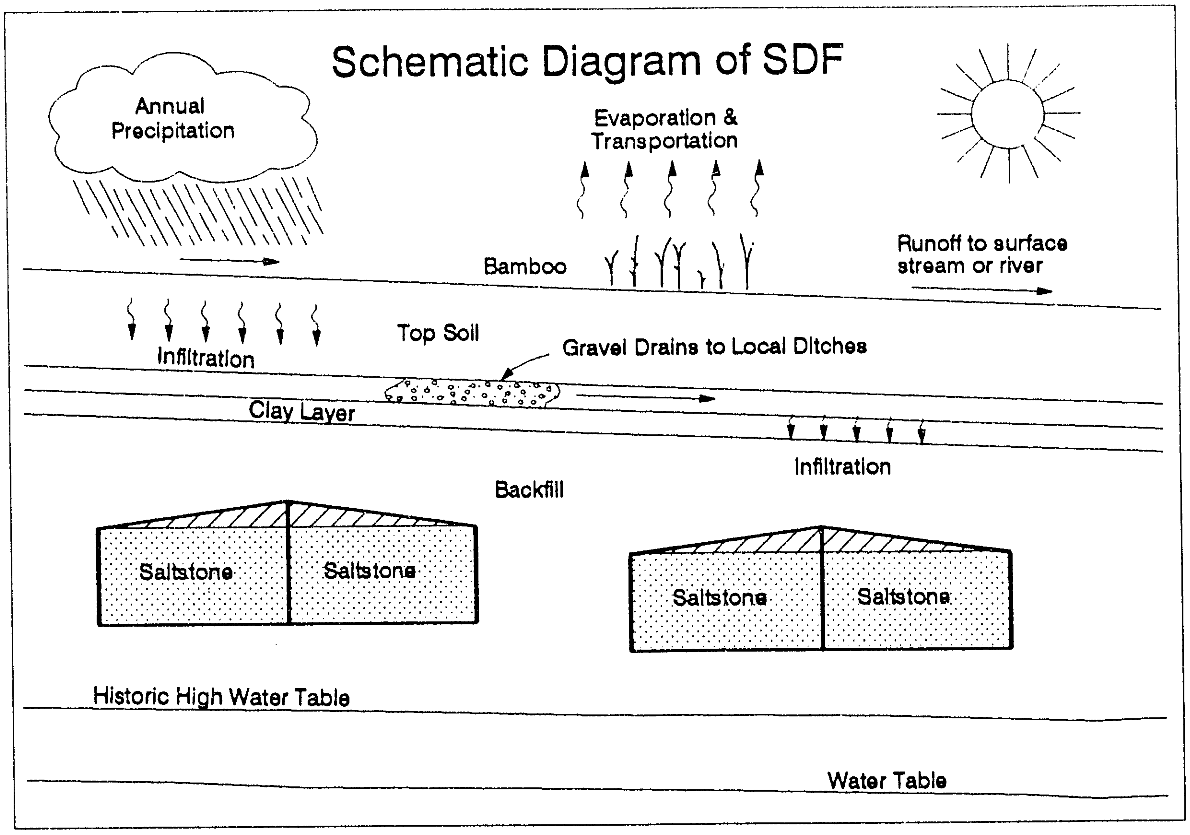

Figure 1

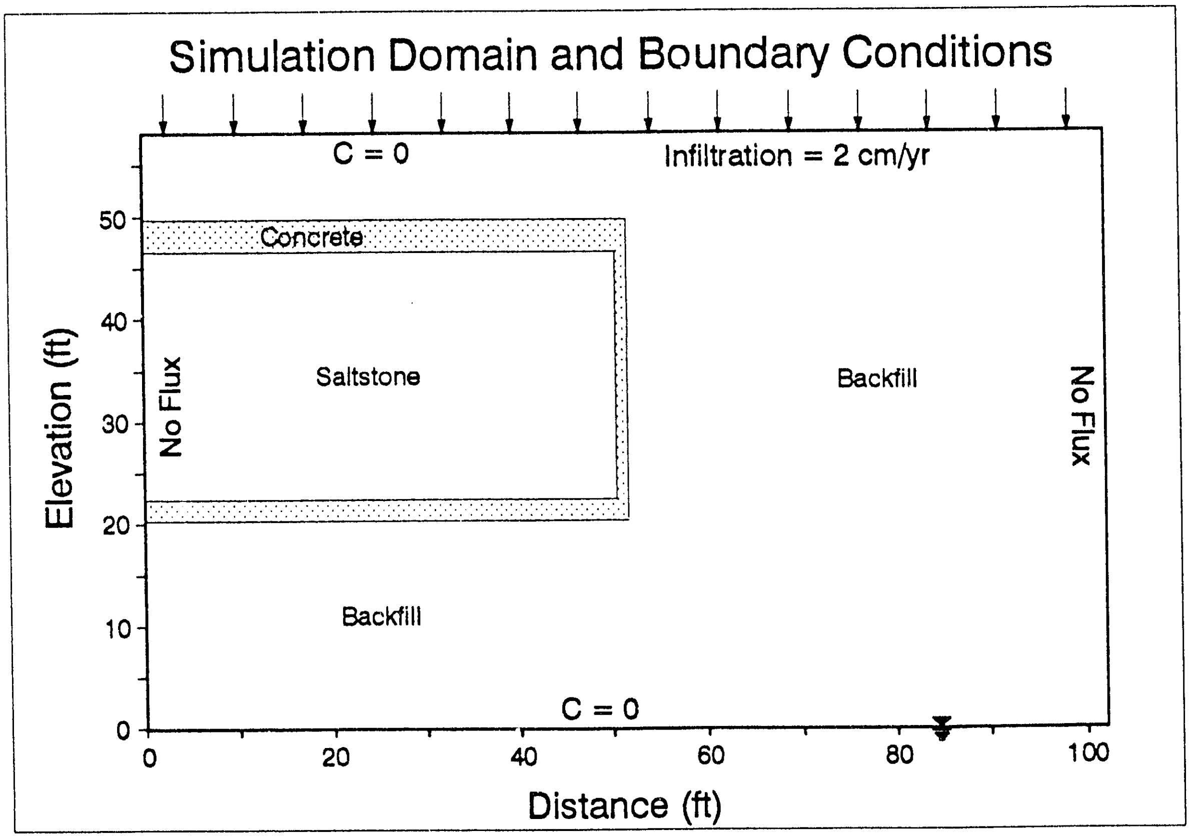

Figure 2

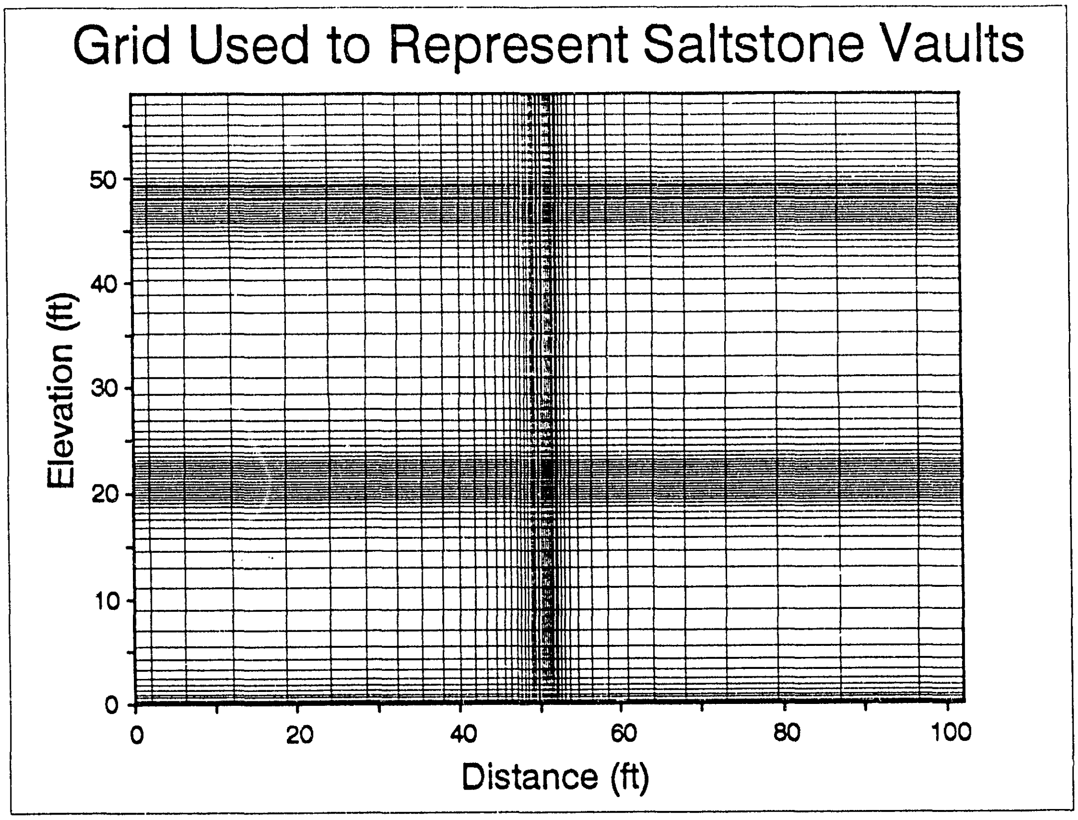

Figure 3

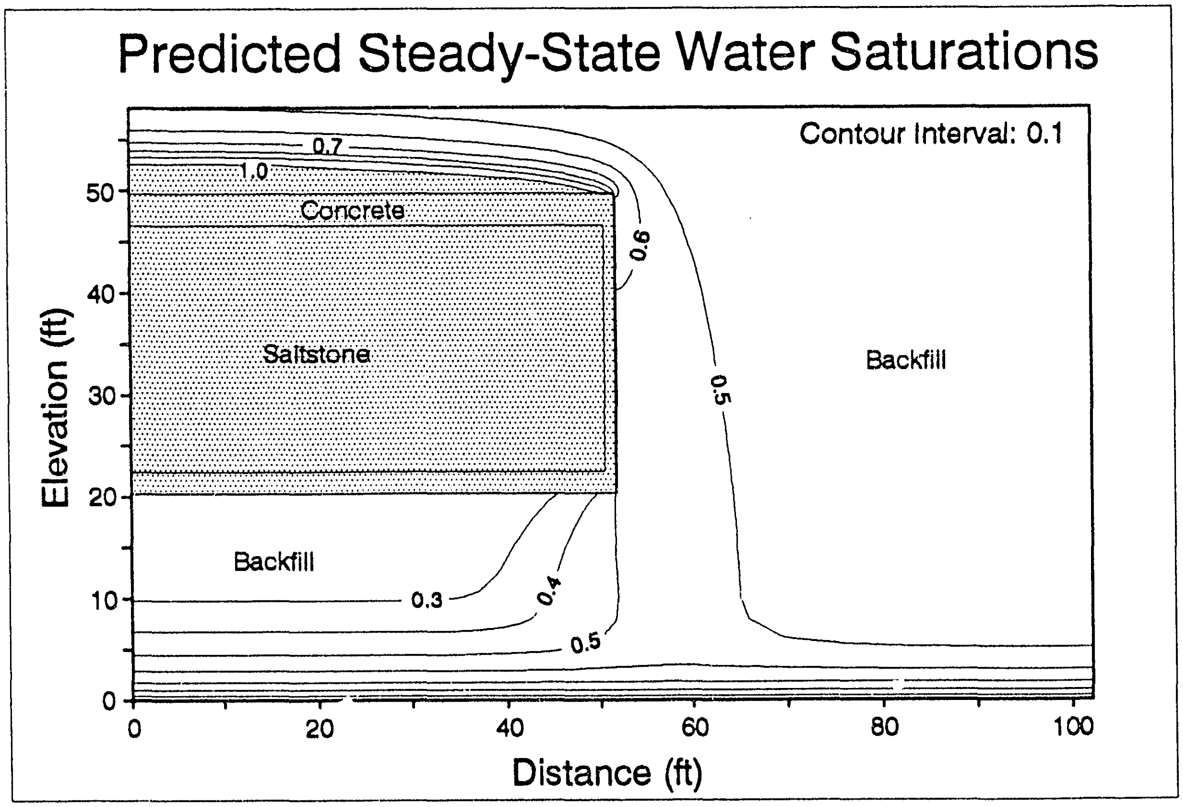

Figure 4

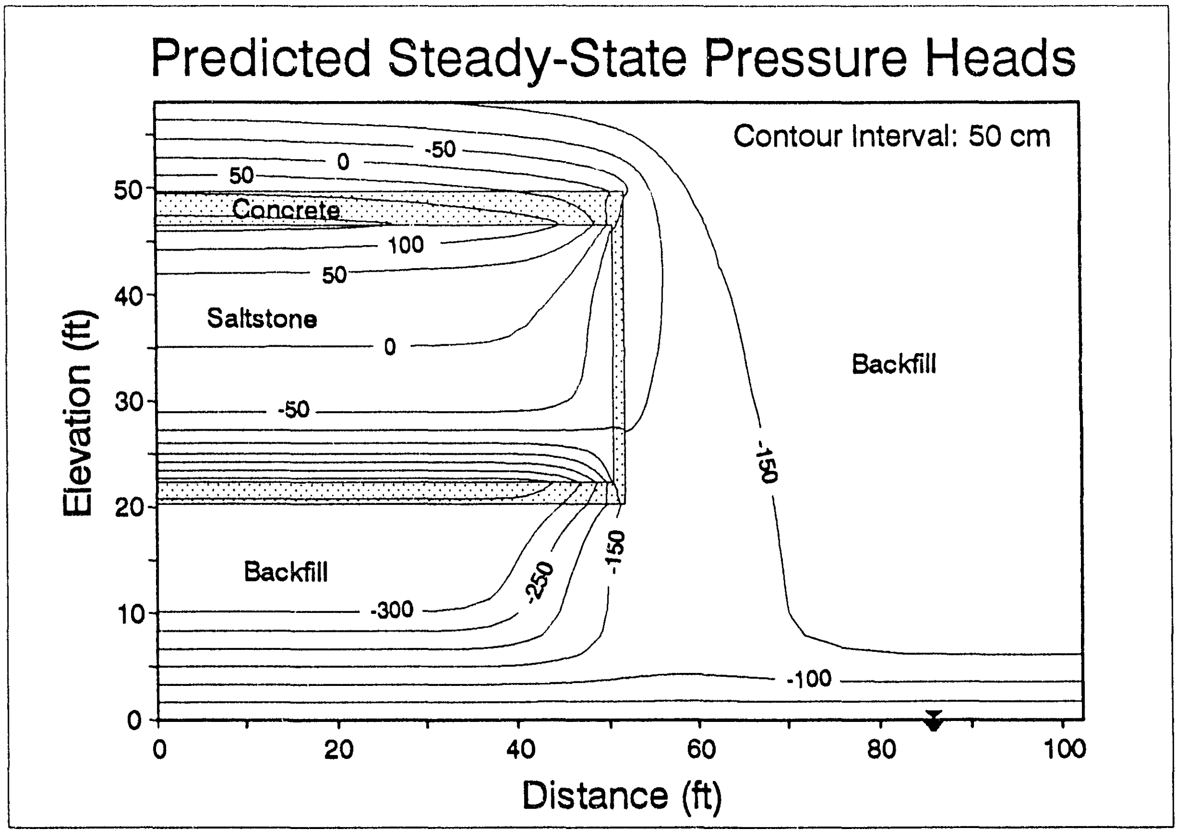

Figure 5

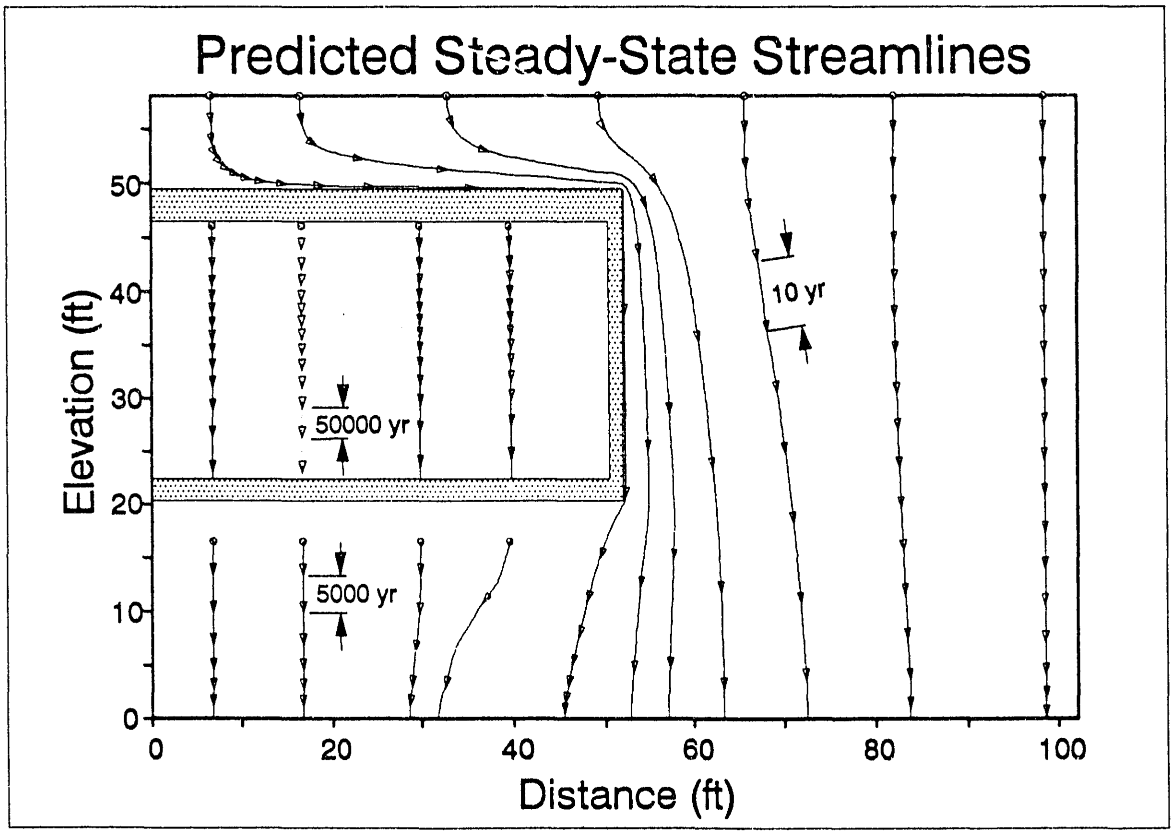

Figure 6

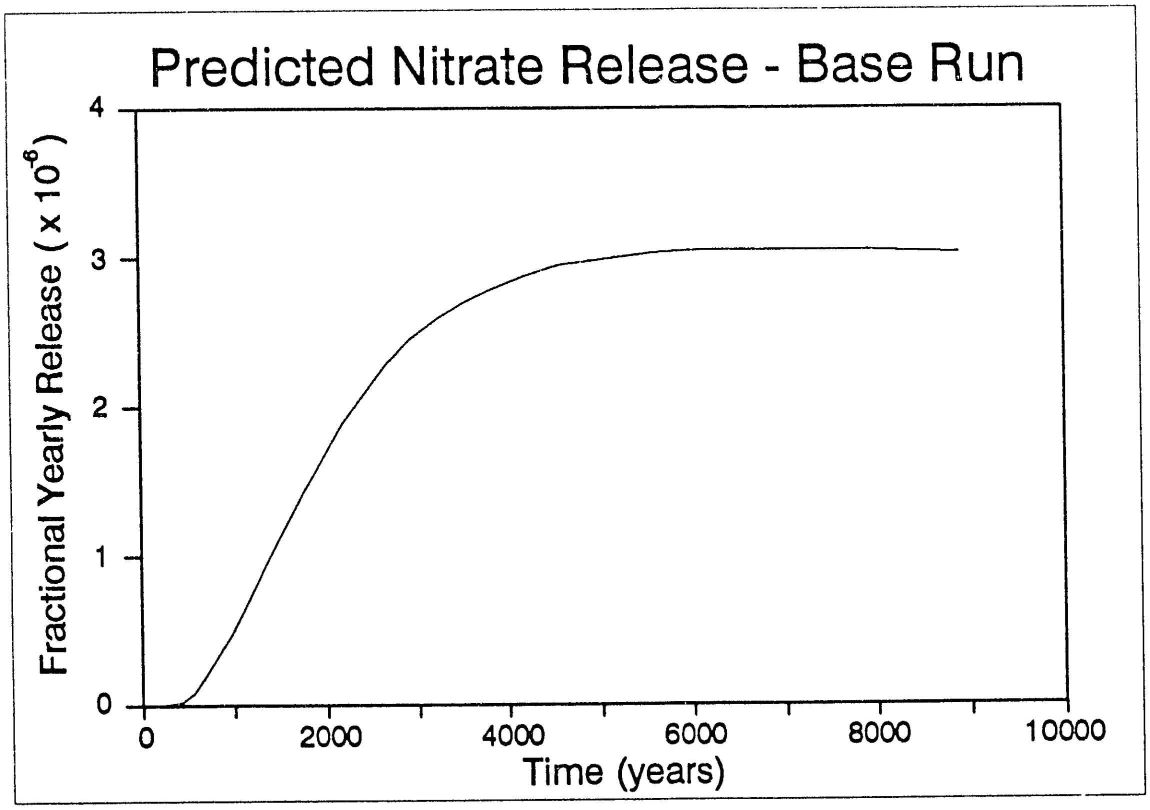

Figure 7

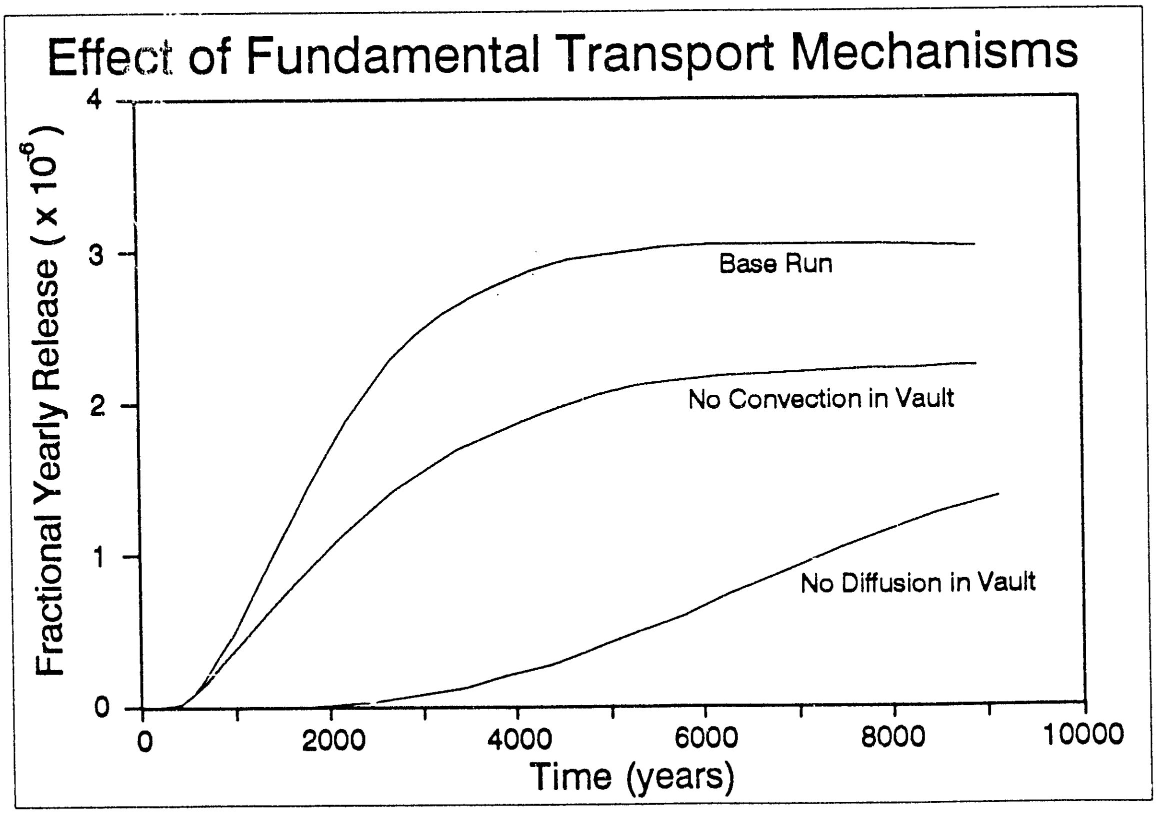

Figure 8

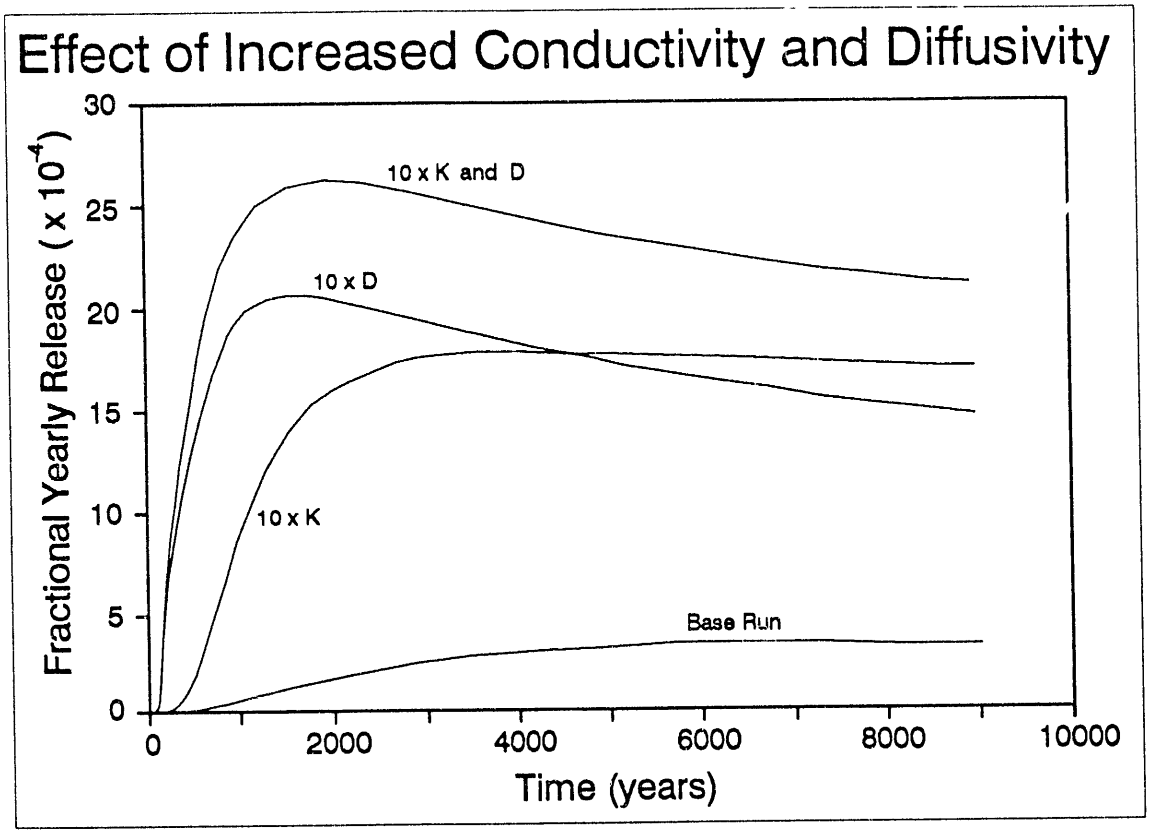
Effect of Increased Conductivity and Diffusivity
Fractional Yearly Release (x 10^{-4})
Time (years)
10 x K and D
10 x D
10 x K
Base Run
30
25
20
15
10
5
0
0
2000
4000
6000
8000
10000

Figure 9

END

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