

GROUNDWATER MODELING OF SOURCE TERMS AND CONTAMINANT PLUMES FOR DOE LOW-LEVEL WASTE PERFORMANCE ASSESSMENTS

Laura M. McDowell-Boyer and John E. Wilson

Oak Ridge National Laboratory
Grand Junction, Colorado

ABSTRACT

Under U.S. Department of Energy (DOE) Order 5820.2A, all sites within the DOE complex must analyze the performance of planned radioactive waste disposal facilities before disposal takes place through the radiological performance assessment process. These assessments consider both exposures to the public from radionuclides potentially released from disposal facilities and protection of groundwater resources. Compliance with requirements for groundwater protection is often the most difficult to demonstrate as these requirements are generally more restrictive than those for other pathways. Modeling of subsurface unsaturated and saturated flow and transport was conducted for two such assessments for the Savannah River site. The computer code PORFLOW was used to evaluate release and transport of radionuclides from different types of disposal unit configurations: vault disposal and trench disposal. The effectiveness of engineered barriers was evaluated in terms of compliance with groundwater protection requirements. The findings suggest that, due to the limited lifetime of engineered barriers, overdesign of facilities for long-lived radionuclides is likely to occur if compliance must be realized for thousands of years.

INTRODUCTION

According to U.S. Department of Energy (DOE) Order 5820.2A (1988), the potential impact of disposal of radioactive waste at sites within the DOE complex must be assessed before DOE will approve disposal. The potential impact is evaluated in terms of potential human exposure to radionuclides considering the federal, state, and local exposure limits and requirements for groundwater resource protection. Compliance with groundwater resource protection requirements is generally interpreted as meeting the Environmental Protection Agency's public drinking water standards. The radiological performance assessment addresses

Research sponsored by U.S. Department of Energy under contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

"The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. DE-AC05-84OR1400. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of the contribution, or allow others to do so, for U.S. Government purposes."

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

the potential impact of each proposed facility, providing an complete pathway analysis of release and transport of radionuclides in the environment surrounding the disposal site. Under DOE Order 5820.2A, no time limit exists for which the facility must comply with all pertinent requirements, although a 10,000-year limit is being considered in a revision to that Order.

Performance assessments were recently completed by Oak Ridge National Laboratory for two low-level waste facilities at the Savannah River site, in South Carolina. As part of these assessments, models were developed to describe both release of radionuclides from several different types of waste forms and engineered barriers and transport through the vadose zone and in groundwater. The computer code PORFLOW (ACRI 1993) was used to simulate critical processes driving release and transport in both unsaturated and saturated media and to ultimately predict groundwater concentrations over time resulting from disposal of radioactive waste at the Savannah River site.

Modeling release of radionuclides from waste forms requires characterization of both the hydrologic and geochemical environment posed by the waste, the engineered barriers, and the subsurface environment immediately surrounding the facilities. Several different configurations were analyzed in this work. A sampling of the types of configurations considered is shown in Fig. 1. In one case, performance of a monolithic concrete waste form was assessed; in another, simple trench disposal was considered. In a third case, disposal of contaminated solid waste in metal boxes placed in concrete vaults was addressed.

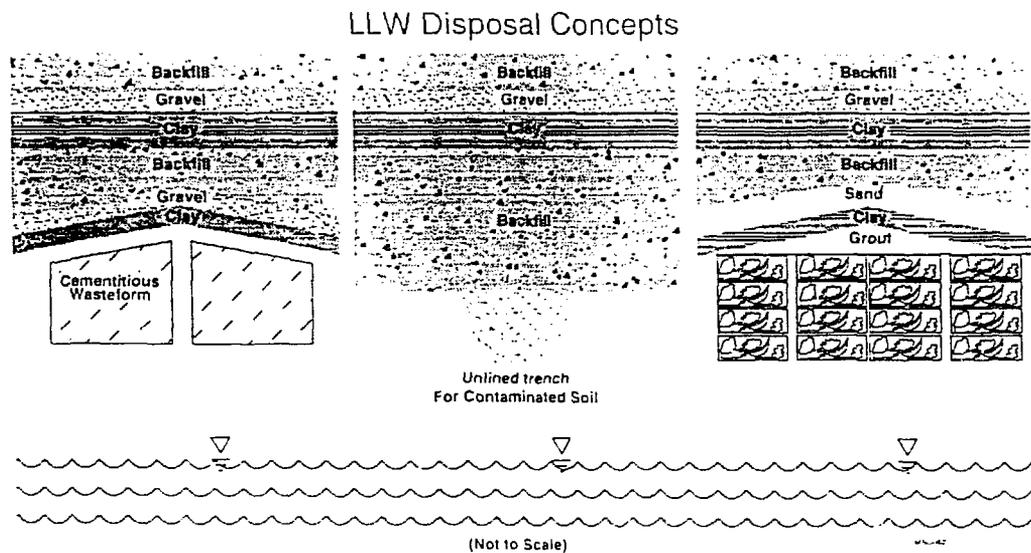


Figure 1. Types of low-level radioactive waste disposal configurations for which unsaturated and saturated zone modeling was carried out.

Transport of radionuclides in groundwater from the waste forms to the point of compliance required analysis of transport in both the unsaturated and saturated zones. Site-specific data were used to describe the hydrologic environment at the Savannah River Site. Local streams greatly influence the gradients and directions of groundwater flow for the facilities evaluated and serve as natural boundaries for potential contaminant plumes. A natural lower boundary for transport exists in the areas of interest, across which upward-directed flow is indicated by available well data.

MODELING STUDY

Release of Radionuclides from Waste Disposal Facilities

In modeling release of radionuclides from the various waste forms considered, a two-dimensional flow field was first established using PORFLOW to represent the undisturbed hydrologic environment in the unsaturated zone, based on site-specific moisture characteristic curves. The engineered barriers, including clay zones, gravel drainage layers, and concrete vaults, were superimposed on this flow field by defining zones within the model domain with different hydrologic properties. The waste form was simulated as a homogeneous porous material in all cases. For practical reasons, a workstation, rather than personal computer (PC), is required for this type of simulation because of the large number of grid elements needed to adequately pose this simulation problem. Idaho National Engineering Laboratory conducted modeling of some of the waste forms in these performance assessments that required a workstation.

Degradation of hydraulic properties of the engineered barriers over time is expected, but considerable uncertainty is associated with prediction of when, and to what extent, this will occur. Cracking is expected to occur in concrete structures and waste forms as a result of chemical attack on the cement matrix or of physical mechanisms such as ground motion. For some of the concrete vaults, roof collapse is also expected. In all cases, clay and gravel layers are assumed to eventually fail. During modeling, degradation was addressed by changing hydraulic properties of the affected materials over time.

In addition to hydraulic properties, release of radionuclides is also controlled by geochemical properties. Geochemical mechanisms were simulated by assigning equilibrium partitioning coefficients to radionuclides in the waste forms to represent the partitioning of radionuclides onto the solid matrix as a result of sorption or other processes resulting in reversible immobilization. Some species are expected to be very insoluble in concrete materials in which the pH is very high; solubility limits were prescribed for these species.

Transport of Radionuclides in the Subsurface Environment

Transport in the unsaturated zone to the water table was simulated in these performance assessments concurrently with simulation of release from the waste forms: that is, a separate

source term model, characterizing release mechanisms from the waste, was not implemented. Rather, the source term was conceptualized as geochemical and hydrodynamic processes that can be represented with sorption coefficients and hydraulic properties in a subzone within the unsaturated domain. This two dimensional domain provided fluxes of water and contaminants to the water table as a function of time. Because initial inventories of radionuclides were very uncertain, the predicted contaminant fluxes to the water table were normalized to a unit initial inventory in the waste.

Flow in the saturated zone was simulated with PORFLOW using a five-layer, three-dimensional domain to account for the different hydraulic properties of the strata underlying the waste disposal facilities and the differential influence of local creeks on each of the strata. In Fig. 2, a potentiometric surface derived from well data is compared to the modeled potentiometric surface of the water table unit in the region of interest for one of the waste disposal units at the Savannah River site. To gain confidence that the model results were representative of the groundwater flow in the region of interest, modeled fluxes at the streams were compared to base flow measurements for these streams. With the exception of a few minor deviations at the head of the streams, the fluxes compared well to base flow measurements.

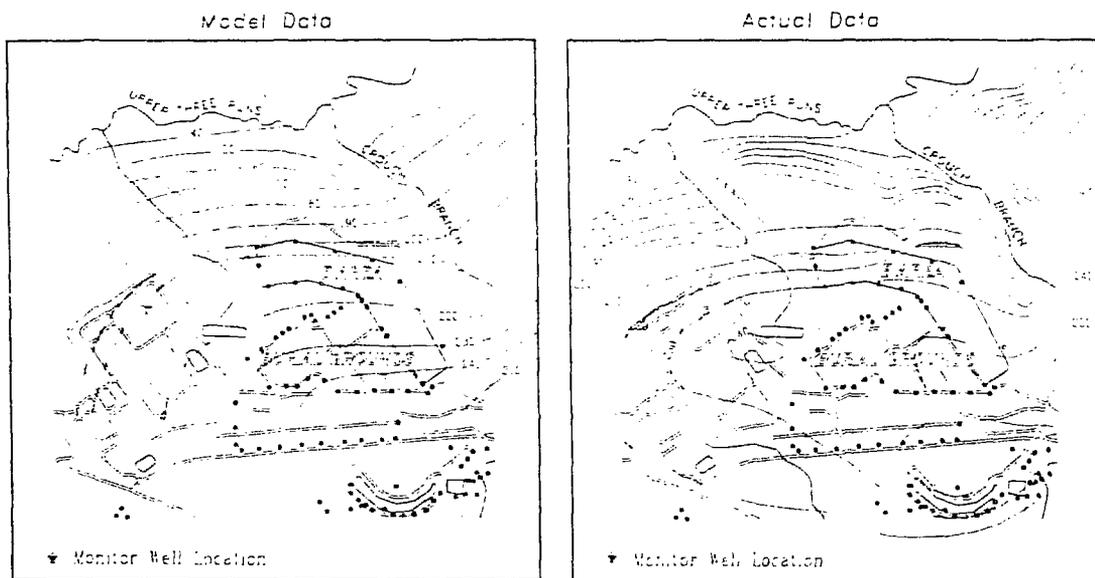


Figure 2. Comparison of potentiometric surfaces derived from modeled and actual water-level data.

A steady-state flow field, based on an estimate of annual average infiltration, was used to simulate transport of radionuclides reaching the water table from waste disposal units. Transport simulations were carried out for hundreds of thousands of years in order to evaluate peak groundwater concentration at the compliance point. The compliance point is located at the

point of maximum groundwater concentration within the simulation domain, at a point at least 100 m from the edge of any disposal unit. Normalized fluxes of radionuclides to the water table, generated in the unsaturated zone simulations, were specified as the time-dependent source to the saturated zone in the PORFLOW simulations. Potentially important radioactive decay products were addressed in the simulations. PORFLOW rigorously tracks the ingrowth of decay products during transit, taking into account the potentially different transport properties of the decay products. Figure 3 shows a three-dimensional view of the simulated H-3 plume originating from vault-disposed waste at 110 years after closure of the facility. During this time, degradation of engineered features is not assumed to have occurred. The points of compliance are shown in this figure, along with the surface water local to the disposal site.

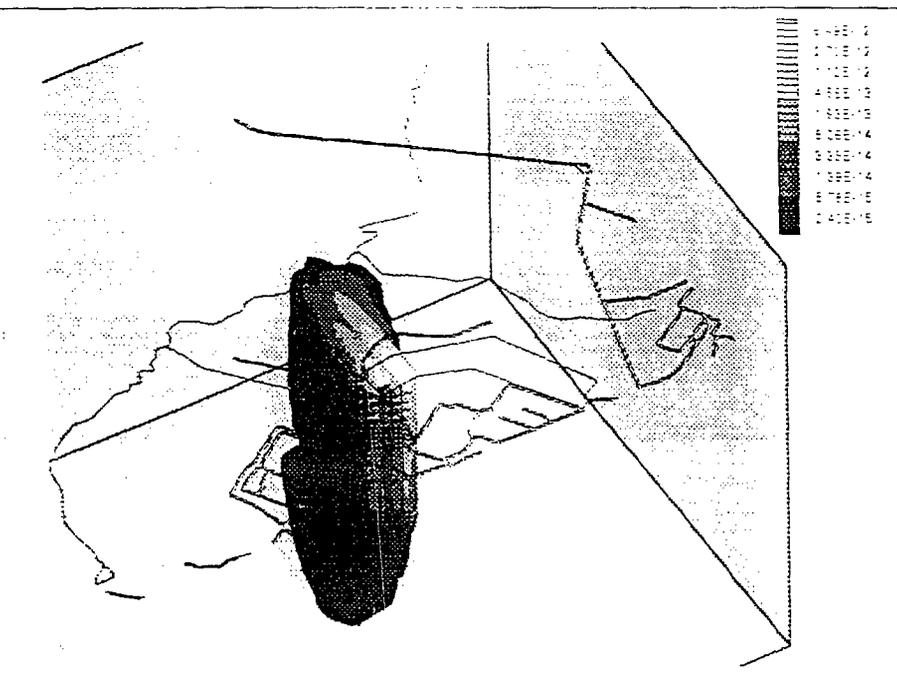


Figure 3. Simulated H-3 plume for vault-disposed waste at 110 years after disposal.

SIMULATION RESULTS

The results of the unsaturated and saturated flow and transport simulations were used 1) to propose waste acceptance criteria, in Ci, for each radioactive species expected and for each waste disposal facility designated and 2) to evaluate the relative importance of engineered features of the proposed facilities. Waste acceptance criteria based on potential groundwater contamination were developed by comparing peak groundwater concentrations of radionuclides, normalized to a unit activity in the disposal facility, at the point of compliance with public drinking water standards. Acceptance criteria were specific for each type of disposal unit

considered. The relative importance of engineered features was evaluated by comparing the simulation results before and after degradation processes have changed the hydraulic properties of the vaults and by comparing trench-disposal results with vault-disposal results.

Figure 4 shows the fractional flux to the water table of Tc-99 and I-129 from vault-disposed waste as a function of time. At approximately 1100 years, when vault-roof collapse was assumed to be accompanied by a decrease in hydraulic conductivity of the vault floor, a spike in the flux of these radionuclides to the water table is predicted to occur. This spike represents the pulse of radionuclides that is expected for radionuclides with low sorption potential shortly after a catastrophic failure of the concrete vaults. The transport of more strongly sorbing radionuclides is retarded, and simulation results show that these radionuclides may not peak for tens of thousands of years after vault failure.

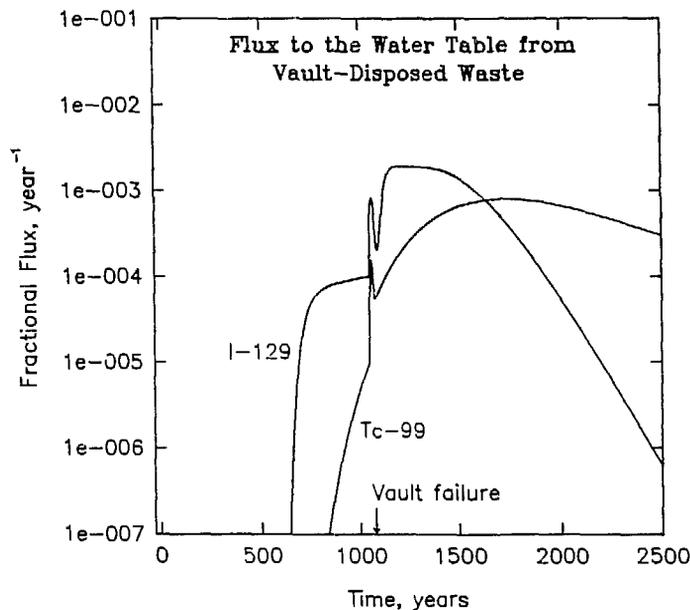


Figure 4. Predicted fraction of initial inventory of radionuclide reaching the water table each year from vault-disposed waste.

Figure 5 compares predicted groundwater concentrations, over time, of selected radionuclides from vault-disposed waste and trench-disposed waste. For the two relatively short-lived radionuclides, H-3 and Sr-90, the peak groundwater concentration for trench disposal is many orders of magnitude above that for vault-disposed waste. In these cases, the engineered barriers are very effective in reducing potential groundwater contamination. The absolute magnitude of the peak groundwater concentrations for H-3 and Sr-90 differ according to the

sorption coefficients (K_d 's). The more highly sorbing Sr-90 has a lower peak groundwater concentration per Ci disposed in either trench or vault because the initial pore concentration in the waste form is lower due to sorption.

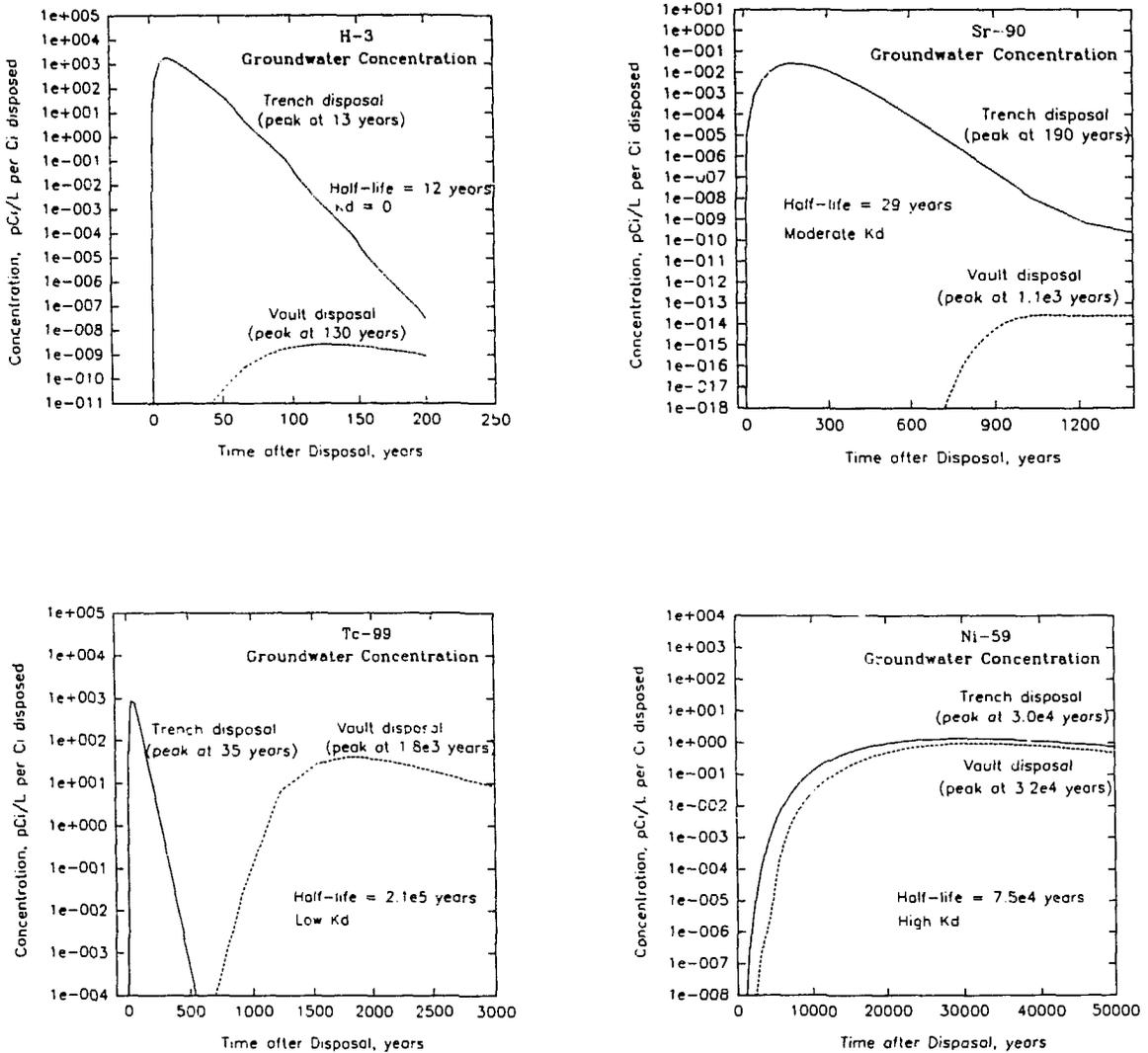


Figure 5. Comparison of predicted groundwater concentrations as a function of time for trench-disposed and vault-disposed waste containing H-3, Sr-90, Tc-99, and Ni-59.

For Tc-99, a weakly sorbing radionuclide, the timing of the peak groundwater concentrations is quite different for trench-disposed and vault-disposed waste. However, the magnitudes of the peaks are not as divergent as for the short-lived radionuclides. The difference

in the magnitudes of the peak groundwater concentrations for the two methods of disposal can be attributed to the somewhat higher Tc-99 Kd in the vault-disposed waste relative to the trench-disposed waste. For long-lived Tc-99, the effectiveness of the engineered barriers appears significantly diminished.

For Ni-59, a highly sorbing radionuclide, the apparent effectiveness of engineered barriers is even lower than it appears to be for Tc-99. In this case, however, the times at which peak groundwater concentrations peak do not differ significantly for the two disposal methods. Because the movement of Ni-59 is so greatly retarded by its strong sorption potential, the catastrophic failure of the vaults at 1100 years is relatively inconsequential to the movement of this radionuclide from the source region to the compliance point in groundwater.

CONCLUSIONS

The results of this modeling study provide insight into the effectiveness of engineered barriers in designing low-level radioactive waste disposal facilities. In general, for radionuclides with half-lives significantly less than the expected lifetime of the engineered barriers, reducing contact of the waste with water can improve the performance of a waste disposal facility in terms of peak concentration realized at the compliance point in groundwater. However, for long-lived radionuclides, for which radioactive decay is not significant during the lifetime of the engineered barriers, the early reduced contact with water is ultimately ineffectual in terms of compliance.

An implication of these observations is that unless integrity of engineered barriers can be defensibly assumed to be in excess of 10,000 years, the utility of these barriers for long-lived radionuclides is limited with respect to the groundwater pathway. It follows that the design of disposal facilities for long-lived radionuclides should deemphasize features that are cost-intensive, and ultimately degradable, in favor of features that rely on the natural capacity of the environment for which a facility is planned to divert water or enhance dilution processes.

In closing, it should be noted that the PORFLOW computer code performed well in simulating the unsaturated and saturated flow and transport problems posed in this analysis. For saturated flow modeling and simple unsaturated flow modeling, a PC environment was a practical tool for carrying out the simulations required.

REFERENCES

- ACRI, 1993, "PORFLOW: A Model for Fluid Flow, Heat and Mass Transport in Multifluid Multiphase Fractured or Porous Media, Version 2.50", Analytic and Computational Research, Inc., Bel Air, California.
- DOE, 1988, "Radioactive Waste Management, Order 5820.2A", U. S. Department of Energy, Washington, DC.