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MATHEMATICAL SIMULATION OF CONTAMINANT DISTRIBUTION IN AND AROUND THE URANIUM MILL TAILING PILES, RIVERTON, WYOMING

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DISCLAIMER

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INTRODUCTION

Objectives

The ultimate objective of the Uranium Mill Tailings Remedial Action Project (UMTRAP) is to minimize the potential environmental hazards due to the existing inactive uranium mill tailing piles. One of these sites, at Riverton, Wyoming, is located on the flood plain of the Wind River, with the water table lying within a few meters of the bottom of the tailings. Field data (see White et al., 1984, elsewhere in this volume) clearly indicates that contaminants, both radioactive and non-radioactive, are mobile within the tailings as well as in the adjacent ground water system. From the point of view of remedial action, the following important questions arise:

> At what rates and quantities will the contaminants continue to migrate in the ground water system over the next several hundred years?
>
> What will be the soil-water regime in the upper part of the tailings which controls the migration of radon gas to the atmosphere?
>
> In view of the projected system behavior, what are the economically viable and environmentally acceptable engineering solutions for remedy?

The purpose of the mathematical modeling efforts at the Riverton site is to address the question of prediction; the transport of contaminants in the ground water system as well as the dynamic soil-water regime near the upper boundary.

The use of mathematical models for the above purpose is dictated by the following questions: Do adequate computational models exist that can simulate the physico-chemical processes that characterize the mill tailings? Can these models reasonably explain the chemical evolution of the system since the beginning of the tailings emplacement? If so, can the historical behavior be used as the basis for predicting the behavior over the next several hundred years?

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In practice, this modeling approach is bounded by financial and time constraints of the over all project schedule. Apart from establishing that the mathematical tools for prediciton do indeed exist, the engineering question of paramount importance relates to the minimum acceptable quantity of field data that is necessary to form the basis for prediction. How much of such data is site-specific? How much of such data can be transferred from one site to another?

The eventual success of the modeling effort therefore lies in first establishing the ability of the existing mathematical tools to explain the existing physical and chemical states of the system at the Riverton site and establish the system parameters that can be used for predictive purposes. The second step is to predict the behavior of the system for a variety of possible remedial designs. The task of deciding upon the critical quantity of field data required for predictive modeling is intimately related to this predictive effort at Riverton. Thus, the modeling effort for the Riverton site is expected to be of a generic nature providing an approach that will be applicable to other UMTRAP sites.

Scope

As part of the Research and Development phase of the UMTRA program, the Lawrence Berkeley Laboratory (LBL) has set itself the goal of explaining the physico-chemical evolution of the Riverton site on the basis of the already collected field data at the site (Tokunaga and Narasimhan, 1982, Smith and Moed, 1982; White et al., 1984). The predictive aspects as well as addressing the question of critical quantity of field data have to be considered during the design phase of the project as a joint effort between the LBL team and the construction engineers. At the present time, LBL is in the process of completing the Research and Development phase of the work. As of this writing, the development of an appropriate set of mathematical models has been completed. The computations of the soil-water regime at the upper tailings surface, involving climatological factors is nearing completion. Computations of chemical transport are still in progress. This paper is devoted to a description of the key mathematical issues, the mathematical models that are needed to address these issues and a discussion of the model results pertaining to the soil water regime at the tailings-atmoshpere interface. A complete discussion of all the model results will form the subject matter of a report that shall be ready at the time of this meeting.

THE MODELING PROBLEM

For purposes of modeling chemical transport the entire system involving the Riverton tailing piles and its environs can be divided into three subsystems:

Subsystem 1: Fluid flow within the tailings including the interaction of the tailings and their cover with the atmosphere.

Subsystem 2: Vertical migration of contaminants within the tailings and the chemical interactions between the tailings and the water table.

Subsystem 3: The lateral migration of chemical contaminants within the regional ground water system.

Subsystem 1: Infiltration Through Tailings

Seasonal precipitation at the Riverton site, be it in the form of rainfall or snow fall, provides the water source that transports the soluble constituents within the tailings downward towards the water table. The evapotranspiration mechanisms as well as the moisture regime near the tailings surface control the magnitude of radon migration upward to the atmosphere. The first task of the entire modeling problem is to adequately quantify the purely physical process of moisture movement within the tailings. The outcome of this quantification are the volumetric rates of moisture movement within the tailings as a function of time and space. These volumetric rates, often expressed in the form of fluid velocity distributions, govern the transport of contaminants by advection and hydrodynamic dispersion. In addition, the moisture movement problem will also yield the time-dependent distribution of water saturation within the tailings. These saturations in turn determine the magnitude of dilution that can occur as mixing occurs within the tailings.

There is ample field evidence at Riverton to suggest that the tailings are presently in a state of partial water saturation and are likely to remain dominantly so in the future. Because of the very low hydraulic conductivity of partially saturated soils, gravity tends to dominate flow in such materials, leading to more or less vertical moisture migration. For this reason, it is reasonable to model the dynamic movement of moisture within the tailings on the basis of a vertical, one-dimensional column. Computationally this is very desirable because the simulation of the evapotranspiration process at the upper boundary, even in the case of a one-dimensional column requires considerable computational effort. The modeling problem relating to infiltration through the tailings is thus one of saturated-unsaturated fluid flow in a heterogeneous column (including layered tailings and layered covers) with time dependent evapotranspiration processes taking place at the top and with the water table at the bottom that may or may not fluctuate with time. Properties such as hydraulic conductivity and saturation of the partially saturated materials will in general, vary with time.

Subsystem 2: Tailing-Water Table Chemical Interactions

The interstitial water within the Riverton tailings is known to be extremely acidic, rich in sulphuric acid. As rain water, which is nearly neutral or, infiltrates the tailings from above, a dilution front forms at the top and begins to move towards the bottom of the tailings. Towards the bottom of the tailings, where the tailings water moves down to the water table, the highly acidic water come into

contact with the native ground water. At the Riverton site, the ground waters are neutral to alkaline in nature and occur in a aquifer matrix that is rich in calcite. As a result, the ground waters have a significant capacity to neutralize the acid tailings solutions and cause several chemical species to precipitate or coprecipitate. This precipitation is of far reaching significance in the design of remedial measure. For, the precipitation mechanism causes a retardation in the migration of species that would otherwise move far in the regional ground water system. In other words, the amount of the contaminants that move down to the water table less the amount that is fixed by precipitation within the aquifer below the water table, constitutes the contaminant source that is available for long-term transport within the regional ground water system.

The modeling of this neutralization process is at present a chemical problem of considerable complexity The neturalization reactions of interest are strongly governed by pH and redox potentials, which change with time as a function of chemical composition. In order to model the chemical transport process in the presence of neutralization reactions, therefore, one has to model the simultaneous transport of several chemical species, while at the same time consider the local mixing of waters of contrasting chemical compositions, leading to precipitation and dissolution of selected species. Since the movement of water within the partially saturated tailings is dominantly vertical, and since much of the precipitation and dissolution is likely to occur in the region immediately below the tailings, it is reasonable to treat this subsystem as one-dimensional. Such a simplification is in fact necessary because the computational effort needed for the neutralization problem are so large that treating the entire system in more than one dimension will render the computational efforts incompatible with the resouces and time available for the present study

Subsystem 3: Chemical Transport in the Regional Ground Water System

Field data suggest that almost all the contaminants are restricted to the shallow, alluvial aquifer which is separated from the deep aquifers by several tens of meters of compacted, low permeability sediments. In modeling the regional ground water system, therefore, it is necessary only to model the shallow aquifer, which is fully saturated with water. Within this sytem, the contaminants are transported by advection, hydrodynamic dispersion and molecular diffusion. Within the portion of the aquifer which immediately underlies the tailings, the net addition of the contaminants after accounting for precipitation will form the chemical source term for the regional ground water system.

THE MODELS

The mathematical models required to simulate Subsystems 1 and 3 are available at LBL and need very little modification before application. As of this writing, a mathematical model for simulating chemical transport with precipitation-dissolution is not readily available in the

literature. A new computer program was therefore developed as part of this work to handle Subsystem 2.

The models used for the three subsystems are, respectively, TRUST, DYNAMIX and TRUMP. All of these programs are based on the Integral Finite Difference Method (IFDM: Narasimhan and Witherspoon, 1976). Broadly stated, the approach consists in dividing the flow region into discrete volume elements and evaluating the flux of water or the solute from one element to another using gradients approximated by the finite difference concept. Average properties such as concentration, saturation, fluid potential and porosty over each volume element are associated with a nodal point within the element. Because of the integral nature of the formulation and because of the manner in which geometric data are handled in the IFDM, all the aforesaid programs are potentially capable of handling complex geometries of the flow region in one-, two- or three-dimensions. In addition, these programs can also handle heterogeneities, anisotropy, time dependent boundary conditions or sources and arbitrarily prescribed initial conditions.

Program TRUST

This program solves for fluid flow in partially saturated deformable media. This program has been validated against field and laboratory data (Narasimhan and Witherspoon, 1978) and has been applied to uranium mill tailings by Reissenauer et al. (1982). For the specific problem of climate-tailings interaction at Riverton, the handling of evapotranspiration boundary conditions along with cyclic rainfall events need particular consideration. The handling of cyclic rainfall is fairly straightforward since boundary conditions and sources can be tabulated as a function of time in the TRUST program.

In general the evapotranspiration process can be treated as a prescribed flux boundary condition, with the magnitude of flux dependent on the fluid potential at the soil surface. In addition, one could impose a lower limit to the potential at the soil surface below which the evapotranspirative flux is essentially zero. The mechanics of handling this type of boundary condition within the TRUST code is available. However, the functional dependence between the fluid potential at the surface and the boundary flux is much more complicated to define. Investigators in the fields of soil physics and hydrology have developed empirical equations to relate evapotranspiration to the combined effects of monthly-averaged climatological data such as temperature, humidity and radiative heat transfer at the surface. As examples, one may cite the work of Staple (1974) and Morton (1978). The application of these techniques in conjunction with the TRUST program are discussed later.

Program TRUMP

Program TRUMP (Edwards, 1972) solves for steady state and transient temperature distributions in multidimensional systems. It considers advection, diffusion and time-dependent sources. If one recognizes the

similarities between the heat transport problem and the chemical transport problem, TRUMP can be used directly to solve chemical transport problems (e.g. Rasmuson et al., 1983). Thus, one may identify temperature with chemical concentration, energy with the amount of solute, radioactive decay with concentration dependent source term and heat capacity of the water-saturated rock with the retardation factor of an adsorbing soil.

For handling advection, TRUMP requires an a priori knowledge of the fluid fluxes. In other words, one has to first solve the fluid flow problem with a program such as TRUST and obtain the spatial distribution of fluid fluxes, which shall then form the input to TRUMP. If fluid fluxes vary with time, one could account for such temporal variation in TRUMP by tabulating fluid fluxes as a function of time at various locations within the flow region. This approach of decoupling the fluid flow problem from the chemical transport problem is reasonable and advantageous as long as the concentration changes do not significantly influence the density of water and hence provide a feed back to the fluid flow problem. The actual chemical concentrations measured at the Riverton site are small enough to take advantage of this simple decoupled procedure.

Program DYNAMIX

This is a new program that has been developed as part of the present work to handle precipitation and dissolution processes that occur when waters of contrasting chemical characteristics mix during chemical transport. To handle the transport on the one hand and the mixing on the other in a rational fashion, one has to couple two different computational algorithms, one dealing with transport and the other, with chemical mixing. In the present work, TRUMP as chosen as the basis for implementing the transport process and a program PHREEQE (Parkhurst et al., 1981) was chosen to handle the chemical mixing.

PHREEQE is a model to simulate geochemical reactions. Based on an ion-pairing aqueous model, this program can calculate pH, redox potential and mass transfer as a function of reaction progress. The composition of solutions in equilibrium with multiple phases can be calculated. Thus, given two different solutions of prescribed chemical compositions and volumes, and the mineral phases that are present in the solids, PHREEQE can compute the composition of the resulting mixture as well as the amounts of any species that may be dissolved or precipitated. To carry out these calculations PHREEQE has its own geochemical data base, particularly suited for low temperature reactions. This data base can be extended with ease.

The coupling of TRUMP and PHREEQE obviously required additional effort. PHREEQE requires as input the concentration of each chemical species of interest in the aqueous phase. In general, therefore, each of these species has to be transported separately. TRUMP, however, can handle the transport of only one species as it stands now. Therefore, TRUMP was extended to transport 10 species. As these species are

being transported, the electrolyte that enters the volume element mixes with the solution that resides in the element. This mixing may result in the dissolution or precipitation of certain species, depending on the pH, the redox potential and the mineral phases in equilibrium. Thus the process of coupling TRUMP and PHREEQE consists in calling the latter algorithm whenever mixing is to be performed in an elemental volume. Essentially, the algorithm carries out mixing in a hydrodynamic system. For this reason, the algorithm has been called DYNAMIX, an acronymn for DYNamic MIXing.

RESULTS

As of this writing computations are in progress to simulate the observed fluid potential distributions and contaminant distributions in the field by parametric studies using material properties measured in the laboratory. The calculations are carried out in the following sequence. First, infiltration calculations are made using TRUST, to obtain the distribution of fluid fluxes and water saturations within the tailings. These fluxes and saturations are used as input to the DYNAMIX code to obtain the migration of the dilution front in the upper part of the tailings and the mixing of the tailings water with the native ground water at the lower interface. The DYNAMIX calculations will provide the net input of the different contaminants from the tailings to the shallow aquifer. These inputs constitute the source terms for the simulation of contaminant migration in the regional ground water system using the TRUMP code. To date, considerable effort has been spent in studying the fluid flow problem within the tailings, while preliminary consideration has been given to the other two aspects. As such, the following discussions focus attention primarily on the fluid flow calculations.

Simulation of Fluid Flow within the Riverton Tailings

Earlier observations by Tokunaga and Narasimhan (1982) gave clear evidence of the existence of relatively stable downward directed hydraulic gradients within the Riverton tailings indicating quasi-steady, downward directed fluid fluxes. The only exception to this pattern is the reversal that occurs in the upper meter or so of the tailings, governed by fluid fluxes immediately following rainfall and upward directed fluxes during non-rainfall periods. It was estimated by Tokunaga and Narasimhan (1982) that the quasi-steady downward flux was in the range of 1 to 5 cm per year. The purpose of the numerical simulation in this study has been to match the observed potential distributions within the tailings using laboratory-determined material properties as input data and climatological data as boundary conditions. The numerical tool used for this purpose is the program TRUST (Narasimhan et al., 1978).

As already mentioned, the system was idealized as a one dimensional column located in the vicinity of site RB2, where the tailings thickness is approximately 6 m. The infiltration studies have

been carried out in three phases. The first preliminary model involved cycling precipitation in the form of time-dependent sources and handling evapotraspiration as prescribed changes in matric potentials at the tailings surface. The second method utilized a method developed by Staple (1974) which couples fluid potentials at the surface of the tailings with the Penman potential evaporation model. The third phase involved consideration of the Complementary Relationship Areal Evaporation (CRAE) model of Morton (1978).

The preliminary model consisted of a vertical set of volume elements with the lowest node (volume element) connected to a fixed potential surface (the water table) and the uppermost node connected to a time-dependent surface potential. The latter boundary potential alternated between -6.0 m and 0.001 m of water to simulate evaporation and rainfall events respectively. The evaporation and rainfall periods were, respectively, 0.1 year and 6 hours. Nine and one half of these cycles comprised each simulated year. The duration of the rain period was selected to be with the Riverton mean annual precipitation of 0.25 m. By extending this simulation to 10 years, a quasi steady state hydraulic head profile was generated through the bulk of the tailings. The simulated profile is very similar to the one observed in the field (Fig.1). The steady state recharge of the ground water in the tailings simulation is 0.03 m per year, which is consistent with the earlier estimates ofTokunaga and Narasimhan (1982). Although this model has yielded credible results, its chief deficiency is that it is based on arbitrarily chosen magnitudes and durations of the surface potentials. The two succeeding approaches aim to avoid this deficiency by utilising available climatological data from the vicinity of the Riverton tailings. For this purpose, climatological data for the Riverton and the Lander, Wyoming weather stations wer compiled for the ten-year period 1972-1981 and compiled into monthly averages.

The method of Staple (1974) effectively substitutes the surface soil water vapor pressure in place of the saturation vapor pressure in the Penman Potential Evaporation Equation.

Evaporaation functions were generated for surface sinks dependent on potentials. Rainfall was concent·ated into 0.0051 m events via 2-hour durations of a 0.0025 m per hour surface source. The rainfall distribution nominally followed monthly averaged rainfall patterns of Riverton. The simulation was carried out for three years following an initail condition of full saturation.

The outcome of using the method of Staple is that a net loss of fluid is calculated at the surface. This net loss can only be accounted for by depleting the moisture stored within the tailings. This is reflected in the hydraulic head profiles depicted in Fig. 2. The zero-flux internal boundary continues to migrate well into the body of the tailings, well below its position observed in the field. Several attempts using alternate selections of rainfall intensities and magnitudes did not appreciably change the net moisture loss at the surface. These observations are contradictory to the field observations, which clearly show net surface gains of rain water.

The Complementary Relationship Areal Evaporation model (CRAE) of Morton (1978) was the third approach used to estimate evaporation losses form the Riverton tailings. Monthly averaged climatic data were again used. Because the method involves a large set of equations, only the results of the calculations are discussed in the interest of saving space. Figure 3 shows evaporative losses in cm as a function of monthly averaged precipitation. Results of Staple's method are included for comparison. Although the CRAE model too predicts net evaporative losses, the magnitudes of such losses are clearly smaller than those of the method of Staple. It should be pointed out here that the CRAE is a model for regional evaporation. Therefore the CRAE results presented in Fig. 3 more likely reflect the Riverton region as a whole. The actual evaporative losses at thge Riverton site may be much smaller than this average. Therefore, with reasonable estimates of the deviation of the tailings surface from the typical surface of the Riverton region, the CRAE approach may prove to be useful management tool for the Riverton tailings. It seems reasonable to expect that the sparsely vegetated, cobbly tailings cover at the Riverton site would evapotranspire at about 50 per cent of the regional rate. This reduction would be sufficient to make the CRAE method yield results consistent with field observations.

ACKNOWLEDGEMENTS

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RB2 SIMULATIONS WITH
STEP-CHANGED SURFACE POTENTIALS

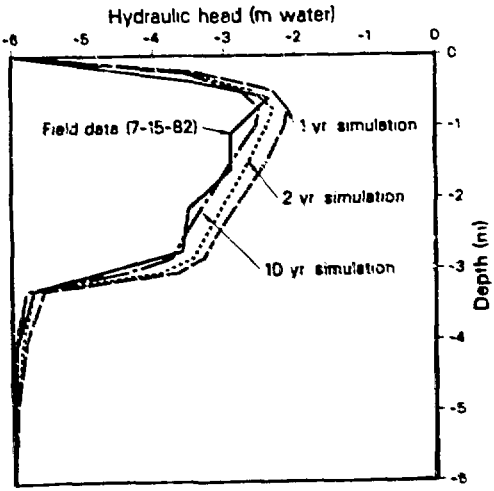

Figure 1

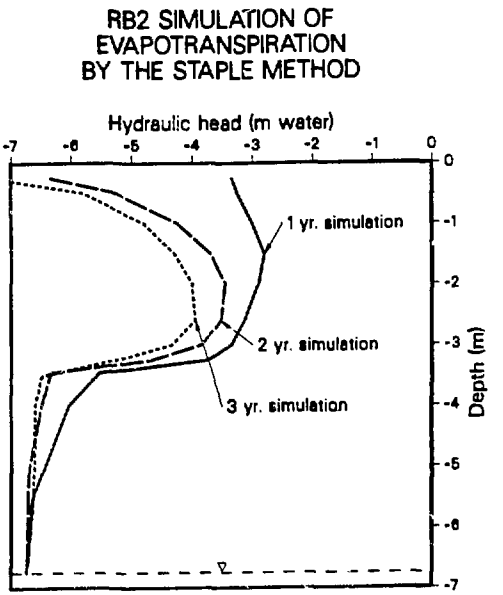

Figure 2

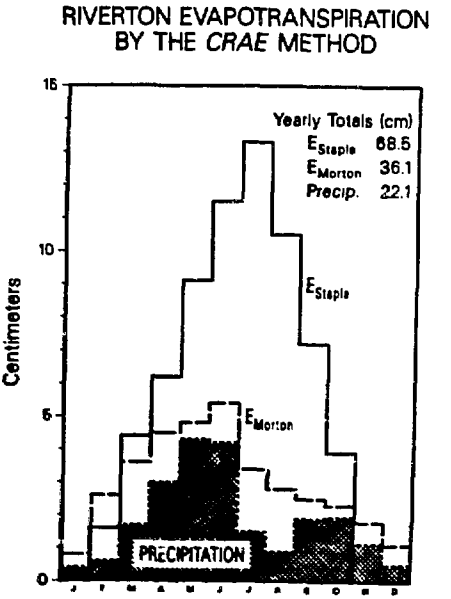

Figure 3