# Part 3: UNSATURATED ZONE MODELS FOR RADIONUCLIDE FATE AND TRANSPORT

In an effort to provide useful information for model application, EPA's NRMRL/SPRD in Ada, Oklahoma, conducted an evaluation of five unsaturated zone fate and transport models for radionuclides. The results of this effort follow. The models reviewed are only a subset of the potentially appropriate models available to the public and are not meant to be construed as having received EPA approval. Other models also may be applicable to SSL development, depending on site-specific circumstances.

Each of the unsaturated zone models selected for evaluation are capable, to varying degrees, of simulating the transport and transformation of chemicals in the subsurface. Even the most unique site conditions can be simulated by either a single model or a combination of models. However, the intended uses and the required input parameters of these models vary. The models evaluated include:

- HYDRUS
- MULTIMED_DP 1.0
- FECTUZ
- CHAIN
- CHAIN 2D

The applications, assumptions, and input requirements for the five models evaluated are described in this section. The model descriptions include model solution method (i.e., analytical, numerical), purpose of model, and methods used by the model to simulate water flow, decay reaction, and radionuclide transport. Each description is accompanied by a table of required input parameters. Input parameters discussed include soil properties, chemical properties, meteorological data, and other site information. In addition, certain input control parameters may be required, such as time stepping, grid discritization information, and output format. Information on determining the general applicability of the models to subsurface conditions is provided, followed by an assessment of each model's potential applicability to the soil screening process.

**HYDRUS**. Information on the HYDRUS model (Version 6.0) was obtained from Šimůnek et al. (1998). HYDRUS is a finite-element model for one-dimensional solute and heat transport simulations in variably saturated media. The flow equation incorporates a sink term to account for water uptake by plant roots. The boundary conditions for flow, and heat and solute transport can vary with time. A finite source also can be modeled. Soil parameters are described by the van Genuchten parameters. The model also considers hysteresis in the water movement. Solute transport and transformation incorporates molecular diffusion, hydrodynamic dispersion, linear equilibrium reactions between the liquid and gaseous phases, nonlinear nonequilibrium partitioning (sorption) between the solid and liquid phases, zero-order production, and first-order decay/degradation reaction. The input parameters required by HYDRUS are presented in Table 3.1.

HYDRUS considers up to six solutes/radionuclides in the transport equation. For decay reaction, the radionuclides can be either coupled in a unidirectional chain or may move independently of each other. In the former case, two or more radionuclides are coupled in a sequential chain reaction. In the later case, each decay reaction is independent of each other. A specific distribution coefficient (or soil-water partition coefficient, a $K_d$ value) as well as a specific decay rate for each radionuclide is required for the HYDRUS model simulation.

## Table 3.1. Input Parameters Required for HYDRUS

| **Soil properties** | **Site characteristics** | **Pollutant properties** | **Root uptake parameters** |
|---|---|---|---|
| Number of soil materials | Uniform or stepwise rainfall intensity | Molecular diffusion coefficient | Potential transpiration rate |
| Depth of soil layers | Volumetric fraction of solid phase | Dispersivity | Osmotic coefficient |
| Saturated water content | Volumetric fraction of organic matter | Freundlich isotherm coefficients‡ | Pressure head where transpiration is reduced by 50% |
| Residual water content | Thermal dispersivity | Freundlich isotherm exponents‡ | Root density as a function of depth |
| Saturated hydraulic conductivity | Empirical parameters for thermal conductivity calculations | First order rate constants (dissolved) | Power function in stress-response function (van Genuchten or Feddes) |
| Soil bulk density | Volumetric heat capacities of solid phase, organic matter, and liquid phase | First order rate constants (adsorbed) | -- |
| van Genuchten retention parameter, α | Number of solutes | Decay coefficient (dissolved) | -- |
| van Genuchten retention parameter, β | Contaminant concentrations in soil | Decay coefficient (adsorbed) | -- |
| Rescaling factors for hydraulic properties | -- | -- | -- |
| **Heat transport properties** | | | |
| Volumetric solid phase fraction | Coefficients of the thermal conductivity functions | Volumetric heat capacity of liquid phase for each soil | Longitudinal and transverse thermal dispersivities for each soil |
| Volumetric organic fraction of each soil | Volumetric heat capacity of solid phase for each soil | Volumetric heat capacity of organic matter for each soil | Thermal initial and boundary conditions |

‡ Freundlich isotherm becomes a linear isotherm and Freundlich isotherm coefficient becomes a distribution coefficient (Kd value) when Freundlich isotherm exponent is 1.

**MULTIMED_DP.** Information on the MULTIMED_DP 1.0 (Multimedia exposure assessment model, including fate and transformation products) model was obtained from Liu et al. (1995), Salhotra et al. (1995) and Sharp-Hansen et al. (1995). MULTIMED_DP was initially developed as a multimedia fate and transport model to simulate contaminant migration from a waste disposal unit (MULTIMED) through different pathways in air, surface water, soil, and ground water. It has been modified to simulate the transport and fate of first and second-generation transformation products through the unsaturated and saturated zones. In MULTIMED_DP Version 1.0, the model now has an option to allow it to be used for unsaturated zone transport alone. The capacity for Monte Carlo simulation has been extended to the unsaturated zone, as well as to the saturated zone. Contamination of a surface stream due to the complete interception of a steady-state saturated zone plume is simulated by the surface water module. The air emissions and the atmosphere dispersion modules simulate the movement of chemicals into the atmosphere. For this review, only the fate and transport of pollutants from the soil to the ground water pathway is considered in detail. In MULTIMED_DP, infiltration of waste into the unsaturated or saturated zones can be simulated using a landfill module or by direct infiltration to the unsaturated or saturated zones. The unsaturated flow model simulates a one-dimensional steady flow with a semi-analytical solution and includes the option to consider seasonal variability in precipitation and evapotranspiration while retaining the assumption of steady-state. Transport in the unsaturated zone considers the effects of advection, dispersion, linear or nonlinear sorption, volatilization, hydrolysis, biodegradation, and first-order chemical decay. It can address steady or time variable infiltration and a finite or infinite source. A one-dimensional uniform steady flow in the saturated zone is assumed. The saturated transport module is also one-dimensional, but considers three-dimensional dispersion, linear adsorption, first-order decay, and dilution due to recharge. Mixing in the underlying saturated zone is based on a specified vertical dispersivity, the length of the disposal facility parallel to the flow direction, the thickness of the saturated zone, the ground water velocity, and the infiltration rate. The parameters required for the unsaturated and saturated zone transport in MULTIMED_DP are presented in Table 3.2.

The MULTIMED_DP model can handle parent, daughter, and granddaughter species chain decay. The chain decay reactions, for example, can be either one parent with one or two daughters or one parent with one daughter and two granddaughters. Effective/overall decay rate, individual decay rates , and distribution coefficient are required for the parent, daughter, and granddaughter species.

**Table 3.2. Input Parameters Required for MULTIMED**

| **Site characteristics/Source characteristics** | | |
|---|---|---|
| Recharge rate | Length scale of facility | Initial parent concentration at landfill |
| Infiltration rate | Width scale of facility | Initial daughter concentration at landfill |
| Depth of unsaturated zone | Duration of pulse | Initial granddaughter concentration at landfill |
| Area of waste disposal unit | Source decay constant | -- |
| | **Unsaturated zone parameters** | |
| Number of physical flow layers | Residual water content | Distribution coefficient |
| Thickness of each layer | pH of layer | Temperature of layer |
| Number of porous materials | Soil bulk density | Brooks and Corey exponent |
| Saturated hydraulic conductivity | Longitudinal dispersivity | Air entry pressure head |
| van Genuchten retention parameter, α | Reference temperature for air diffusion | -- |
| van Genuchten retention parameter, β | – | – |

### Table 3.2. Input Parameters Required for MULTIMED

| Saturated zone parameters | | |
|---|---|---|
| Hydraulic gradient | Organic carbon content | Parent retardation coefficients |
| Hydraulic conductivity | Longitudinal dispersivity | Daughter retardation coefficients |
| Mixing zone depth | Transverse dispersivity | Granddaughter retardation coefficient |
| Aquifer thickness | Vertical dispersivity | Well distance from site |
| Aquifer porosity | Temperature of aquifer | Angle off-center of well |
| Bulk density | pH | Well Vertical distance |

## Table 3.2. Input Parameters Required for MULTIMED

### Parent/Daughter/Granddaughter Decay Module

- Parent to daughter 1 dissolved Phase decay rate constant
- Parent to daughter 2 dissolved phase decay rate constant
- General parent dissolved phase decay rate constant (decay not contributing to either daughter product)
- Parent to daughter 1 sorbed phase decay rate constant
- Parent to daughter 2 sorbed phase decay rate constant
- General parent sorbed phase decay rate constant (decay not contributing to either daughter product)
- Daughter to granddaughter j, j+1 dissolved phase decay rate constant
- Daughter **I** general dissolved phase sorbed phase decay rate constant
- Daughter **I** to granddaughter j, j+1 sorbed phase decay rate constant
- Daughter **I** general sorbed phase decay rate constant
- Daughter second-order acid-catalysis hydrolysis rate constants
- Granddaughter k dissolved phase decay rate constant
- Granddaughter k sorbed phase decay rate constant
- Parent general term second-order acid-catalysis hydrolysis rate constant at reference temperature
- Parent general term neutral hydrolysis rate constant at reference temperature
- Parent general term second-order base-catalysis hydrolysis rate constant at reference temperature
- Parent to daughter 1 second-order acid catalysis hydrolysis rate constant at reference temperature
- Parent to daughter 1 neutral hydrolysis rate constant at reference temperature
- Parent to daughter 1 base-catalysis hydrolysis rate constant at Reference Temperature
- Parent general term neutral hydrolysis rate constant at reference temperature
- Parent general term second-order base-catalysis hydrolysis rate constant at reference temperature
- Parent general term second-order base-catalysis hydrolysis rate constant at reference temperature
- Daughter second-order acid-catalysis hydrolysis rate constants
- Daughter neutral catalysis hydrolysis rate constants
- Daughter second-order base-catalysis hydrolysis rate constant
- Granddaughter second-order acid catalysis hydrolysis rate constant
- Granddaughter neutral hydrolysis rate constant
- Granddaughter second-order base-catalysis hydrolysis rate constant
- Parent to daughter **I** dissolved and sorbed phase stoichiometric coefficients
- Daughter to granddaughter m dissolved and sorbed phase stoichiometric coefficients
- pH of the unsaturated zone or saturated zone
- Reference temperature
- Temperature of the unsaturated or saturated zone

**FECTUZ.** FECTUZ is a one-dimensional fate and transport model for the unsaturated zone which is incorporated into the EPACMTP program (EPA 1995b, 1995c). It is an extension of the VADOFT code (Huyakorn and Buckley, 1987) which is also incorporated into the RUSTIC and PRZM codes. FECTUZ simulates migration of contaminants from a landfill (or a surface impoundment), through the unsaturated zone to an unconfined aquifer with a water table present at some depth. The model allows for finite or infinite sources which can undergo decay using Bateman's equation (EPA, 1995b). The model can simulate linear and nonlinear adsorption and first order decay. FECTUZ is limited to the simulation of simple hydrogeological sites.

FECTUZ is capable of chain decay with up to seven radionuclide species (one parent with up to six daughter products). The chain decay path can be either straight or branched. A daughter product (a species) can have one or more immediate parent species. The sum of the chemical and biological transformation coefficients gives the overall decay coefficient. The fraction of each parent that decays into the same daughter product has to be specified. A distribution coefficient and a decay rate are specified for each individual radionuclide. A radionuclide species with a zero decay rate indicates it is the end product in the decay chain.

FECTUZ assumes steady state flow in the unsaturated zone which can be composed of one or more uniform soil layers. It simulates the transport of contaminants in soil using a advective-dispersive equation. FECTUZ employs three solution options for the transport equation. An analytical solution is used for steady state single species decay with linear adsorption. A semi-analytical solution is used for transient and steady state chain decay and linear sorption. A finite element solution is used for chain decay with nonlinear sorption. The input parameters required for FECTUZ are presented in Table 3.3.

**Table 3.3. Input Parameters Required for FECTUZ Module within EPACMTP**

| Soil properties | Site characteristics | Pollutant properties |
|---|---|---|
| Soil bulk density | Thickness of unsaturated zone | Organic carbon partition coefficient |
| Saturated water content | Uniform thickness for discretized soil layers | Freundlich isotherm coefficients |
| Saturated hydraulic conductivity | Uniform infiltration rate except for surface impoundments | Dispersivity |
| Residual water content | Constant source or Decaying source or finite pulsed source | Decay coefficient (dissolved) |
| van Genuchten retention parameter, α | -- | Decay coefficient (adsorbed) |
| van Genuchten retention parameter, β | -- | Parent and daughter species decay reaction stoichiometry |
| Fraction of organic carbon | -- | -- |

‡ Freundlich isotherm becomes a linear isotherm and Freundlich isotherm coefficient becomes a distribution coefficient (Kd value) when Freundlich isotherm exponent is 1.

**CHAIN.** CHAIN (van Genuchten, 1985) is a simple program which uses analytical solutions to solve for the simultaneous one-dimensional advective-dispersive transport of contaminants for up to four members of a sequential (first-order) decay chain. The model assumes that the soil system is a homogeneous soil system and that the moisture content and infiltration are constant in time (steady state flow). The adsorbed concentrations of the contaminant are represented by linear reversible isotherms. The transport contaminant boundary condition is either a constant concentration or a source decay given an arbitrary general release mechanism or the specific release mechanism defined by the Bateman equations (van Genuchten, 1985). The analytical solutions provide the solution in terms of concentration vs. distance (depth) at selected times as well as concentrations vs. time at selected depths. The input parameters required for the CHAIN model are presented in Table 3.4.

The consecutive chain reaction included in the transport model can involve up to four species. For example, a reaction could have parent and daughter products (two species) or a reaction could involve parent, daughter, and granddaughter products (three species). There is a decay rate associated with each pair of parent-daughter (or daughter-granddaughter) products. For the last product in the consecutive chain, a decay rate of zero is specified.

**Table 3.4. Input Parameters Required for CHAIN Model**

| Soil properties | Site characteristics | Pollutant properties |
|---|---|---|
| Soil Bulk density | Source initial concentrations | Retardation values for decay chain members |
| Volumetric water content | Bateman coefficients for source decay | First order decay coefficients for decay chain members |
| Pore velocity (flux/ water content) | General release parameters for source decay | Pulse time |
| -- | Dispersion coefficient | -- |

**CHAIN 2D.** CHAIN 2D is a two-dimensional model for the simulation of variably saturated flow, contaminant transport, and heat transport developed by Šimůnek and van Genuchten (1994**)**. The water flow is represented by Richards equation for saturated and unsaturated flow. It contains a sink term for water uptake by plant roots. The flow region can be composed of non-uniform soils to incorporate the effects of anisotropy. The flow model boundary conditions can include prescribed head, gradient, flux boundaries, or free drainage and a simplified representation of nodal drains. Heat and contaminant transport are modeled by the advective-dispersive equation which includes conduction and convection processes. The contaminant transport simulation can be modified for nonlinear nonequilibrium reactions between solid and liquid phases and for linear equilibrium reactions between liquid and gaseous phases. The contaminant transport simulation also includes zero order production and two first-order decay reactions: one which is independent of other solutes, and one which solutes are in sequential chain decay reactions. CHAIN 2D can simulate up to six species independent of one another or in a unidirectional chain decay. The boundary conditions for the contaminant transport could be constant concentration or constant flux. Flow and transport can be simulated in a vertical or horizontal plane or in an axisymmetrical cylindrical system. The flow and transport equations are solved using the Galerkin finite element method. The parameters required for CHAIN 2D model are presented in Table 3.5.

Similar to the HYDRUS model, CHAIN 2D considers up to six solutes/radionuclides in the transport equation, The radionuclide can be either coupled in a unidirectional chain or may move independently of each other. In the former case, two or more radionuclides are coupled in a sequential chain reaction. In the latter case, each decay reaction is independent of each other. A specific distribution coefficient as well as a specific decay rate for each radionuclide is required for the model simulation.

## Table 3.5. Input Parameters Required for CHAIN 2D

| **Soil properties** | **Site characteristics** | **Pollutant properties** | **Root uptake parameters** |
|---|---|---|---|
| 2D cell discritization | Transpiration rate | Ionic or molecular diffusion coefficient in free water for each species | Root density as a function of depth |
| Saturated water content | Evaporation/infiltration rates | Ionic or molecular diffusion coefficient in gas phase for each species | Power function in stress-response function |
| Saturated hydraulic conductivity | Initial contaminant concentrations in soil | Longitudinal and transverse dispersivities for each species | Pressure head where transpiration is reduced by 50% |
| Soil bulk density | Contaminant species initial and boundary conditions | First order decay coefficient for each species in liquid, solid or gas phase | -- |
| van Genuchten retention parameter, α | Initial head conditions | Zero order rate constant for each species in liquid, solid or gas phase | -- |
| van Genuchten retention parameter, β | Location and rates of pumping/ injection wells | Adsorption (Freundlich)‡ isotherm coefficients for each species | -- |
| Residual water content | Seepage faces, tile drains | Source Decay | -- |
| | | **Heat transport properties** | |
| Volumetric solid phase fraction | Coefficients of the thermal conductivity functions | Volumetric heat capacity of liquid phase for each soil | Longitudinal and transverse thermal dispersivities for each soil |
| Volumetric organic fraction of each soil | Volumetric heat capacity of solid phase for each soil | Volumetric heat capacity of organic matter for each soil | Thermal initial and boundary conditions |

‡ Freundlich isotherm becomes a linear isotherm and Freundlich isotherm coefficient becomes a distribution coefficient (Kd value) when Freundlich isotherm exponent is 1.

**3.1 Considerations for Unsaturated Zone Model Selection.** The accuracy of a model in a site-specific application depends on simplifications and assumptions implicit in the model and their relationship to site-specific conditions. Errors may be introduced from assumptions made when deriving input parameters. Although each of the five models evaluated has been tested and validated for simulation of water and radionuclide movement in the unsaturated zone, they are different in purpose and complexity, with certain models designed to simulate very specific scenarios. A model should be selected to accommodate a site-specific scenario as closely as possible. For example, if contaminant chain decay is of concern, the model should consider chain decay reaction. After a model is determined to be appropriate for a site, the contaminant(s) and the conditions to be modeled, the site-specific information available (or potentially available) should be compared to the input requirements for the model to ensure that adequate inputs can be developed. The unsaturated zone models addressed in this study use either analytical, semianalytical, or numerical solution methods. Analytical models represent the simplest models, requiring the least number of input parameters. They use a closed-form solution for the pertinent equations. In analytical models, certain assumptions have to be made with respect to the geometry of the system and external stresses. For this reason, there are few analytical flow models (van der Heijde, 1994). Analytical solutions are common for fate and transport problems which arise from the solutions of advective-dispersive equations. Analytical models (e.g. CHAIN) may require the assumption of uniform flow conditions, both spatially and temporally. Semianalytical models (e.g., MULTIMED_DP) approximate complex analytical solutions using numerical techniques (van der Heijde, 1994). Transient or steady-state conditions can be approximated using a semianalytical model. However, spatial variability in soil or aquifer conditions are usually not accommodated. Numerical models (e.g., HYDRUS or CHAIN-2D) use approximations of pertinent partial differential equations, such as finite-difference or finite-element methods. The resolution of the area and time of simulation is defined by the modeler. Numerical models may be used when simulating time-dependent scenarios, spatially variable soil conditions, and unsteady flow (van der Heijde, 1994).

**3.2 Model Applicability to SSLs for Radionuclides**. Evaluation of model applicability to SSLs is based on the following considerations:

1. Whether a model can be used in simulating fate and transport of the five selected radionuclides -- uranium, strontium, technetium, plutonium, and tritium?

2. Whether a model can simulate fate and transport of the selected radionuclides for a test case (the Las Cruces Trench Site, New Mexico)?

3. Is a model capable for use in the soil screening process?

4. What are the limitations for a specific model?

To facilitate the evaluation, the characteristics and capabilities of all five models are summarized in Table 3.6. These characteristics/capabilities include flow and transport processes, site-specific conditions, solution methods, assumptions, and model outputs. Each characteristic/capability is evaluated and is check marked when that characteristic/capacity is implemented in the model. Note that Table 3.6 addresses only unsaturated zone fate and transport model components in spite of the HYDRUS, MULTIMED_DP, and FECTUZ models having saturated zone flow and transport capabilities. The following text highlights a general description of the evaluation process, discusses some of the differences between the models, outlines their advantages and disadvantages, and describes appropriate scenarios for model application.

<u>Model Applicability to the Five Selected Radionuclides</u>: As provided in Table 3.6, all five models address four essential processes that predominately control the migration of radionuclides in the unsaturated zone -- advection (derived from infiltration), dispersion, sorption and radionuclide (straight chain and/or branched) decay. Therefore, **any of the five models can be used in simulating fate and transport for the five selected radionuclides --** uranium, strontium, technetium, plutonium, and tritium under the flow and site-specific conditions (e.g., steady-flow

in uniform soil, or transient flow in a layered soil) which have been implemented in the model. These five radionuclides exhibit a broad range of physical properties in soil -- distribution coefficients ($K_d$) of a radionuclide between water solution and soil, and chain decay rates. The simulation results for all five radionuclides using any of the five models would be similar to the results in the sensitivity analysis for $K_d$ when the simulation time is much smaller than the magnitude of the half-life of any radionuclide.

Model Applicability to Site-Specific Conditions at the Las Cruces Trench Site: At the Las Cruces Trench Site, soil properties and solute transport parameters are available (Wierenga et al. 1991; Hills et al. 1991). They are reproduced in Tables 3.7 and 3.8. Other site-specific conditions are given in Table 3.9. As indicated in Table 3.6, the HYDRUS, CHAIN 2D, FECTUZ, and MULTIMED_DP models can simulate layered soils. However, **when soil at the site is treated as a uniform soil and the averaged soil properties are used, all five models can simulate the transport of radionuclides under steady-state water flow conditions at the site (assuming net recharge rate is known)**. Only HYDRUS and the CHAIN 2D model can handle transient flow at the site. Among the five models evaluated, CHAIN does not have a water flow module, FECTUZ considers water flow with a known infiltration rate, and the HYDRUS, CHAIN 2D and MULTIMED_DP models can calculate infiltration rates from rainfall and potential evaporation data. Note that the average annual infiltration rate at a site is difficult to measure in the field, yet it is required for estimating a dilution factor or DAF in the simplified SSL method. Furthermore, it is worthy to note that the processes/characteristics listed in Table 3.6 are not inclusive enough to accurately simulate the fate and transport of radionuclides in unsaturated zone. Other processes such as colloid-facilitated transport, pH-facilitated transport, cation-exchange, or complexation, are not considered. These processes might significantly influence the radionuclide movement in clay-rich soil, but they are not likely at the Las Cruces Trench Site, where the soils are predominantly classified as sands, sandy loams, loamy sands and sandy clay loams (Wierenga et al. 1991). For the purpose of SSL processes, all five models can simulate the transport of a radionuclide to a reasonable extent because these models consider the four essential processes that probably control the migration of radionulides in the unsaturated zone -- advection (derived from infiltration), dispersion, sorption, and radionuclide (straight and/or branched) decay. This will be further confirmed in the sensitivity analysis.

Model Capability for the Use in the Soil Screening Process: All five unsaturated models for radionuclides evaluated herein are PC-based, public domain models. These five models can calculate the leachate radionuclide concentrations entering ground water (Table 3.10). The leachate concentrations are needed in the SSL process/comparison. The leachate concentrations as a function of time are used to estimate ground water concentrations at the receptor well. These receptor point concentrations are then compared with the acceptable ground-water concentration (e.g. MCL) to determine if a site's soil exceed SSLs. If they do not exceed the acceptable ground-water concentrations, it might be believed that there is no concern for human health and ecology at the site. The conclusion of such a comparison is based on the assumptions that --1) the conceptual site model (CSM) is reasonably developed; 2) site-specific data required by the model are properly collected and meet data quality standards; and 3) the unsaturated zone model for radionuclide migration is properly used. Therefore, all five selected unsaturated zone models for radionuclides -- CHAIN, HYDRUS, CHAIN 2D, MULTIMED_DP, and FECTUZ, are basically **capable for use in the soil screening levels process**. When there is a concern for the uncertainty of the SSL estimates due to variability of the input parameters, the built-in Monte Carlo simulation in the MULTIMED_DP and FECTUZ (EPACMTP version) can be employed. For CHAIN, HYDRUS, and CHAIN 2D models, multiple runs using a set of variable input parameters (either obtained from the data survey or data generation for a know distribution of a parameter.) is needed.

Using any of these five models for the SSL calculation requires more site-specific data and modeling effort than using the simple site-specific SSL calculation (see Section 2 in TBD). However, these five models (and other potential models) can take into account more complex site conditions and can provide more accurate SSL calculation if data collection is sufficient and model application is properly done. In other words, using models in the SSL process require less assumptions than using the simple site-specific SSL calculation. However, for both the simple approach and the more detailed modeling approach the receptor well is assumed to be located at the edge of the waste source. The SSLs are intended to correspond to levels of radionuclides in soil such that MCLs will not be exceeded in the underlying ground water beyond the edge of the waste area. Among those simplifying assumptions listed in the Highlight 2 of TBD Section 2, only the assumptions of ignoring facilitated transport and complexation of nuclides with other constituents in soil solution still apply to the model SSL calculation.

Note that two methods can be used to calculate the receptor point concentration using the leachate radionuclide concentrations to ground water provided by the unsaturated model. One method uses a dilution/attenuation factor (DAF) to account for the mixing of leachate with ambient ground water. Another uses a saturated zone flow and transport model to calculate the resulting concentration considering the processes of advective, dispersive, decay, and biodegradation of the radionuclide in ground water (the MULTIMED_DP and FECTUZ models have a saturated zone flow and transport component.) When the former method is used, the receptor well is at the edge of the source (i.e., there is no dilution from recharge downgradient of the site) and is screened within the plume. A default value of 20 for DAF as proposed in the technical background document (TBD) for SSLs for radionuclide can be used. Alternatively, the dilution models in the section 2.6.4 in TBD can also be used.

It is worthy to note that in using the simple soil/water partition equation, the SSLs are directly calculated from an acceptable radionuclide level at the receptor well. The existing total soil radionuclide concentrations are then compared with the SSLs (TBD, Section 2.6). However, the unsaturated zone models do not perform this direct calculation. Instead, the existing site radionuclide-contamination is simulated, and the resulting radionuclide concentrations are then compared to the acceptable radionuclide level at the receptor well.

As described above, all five models can simulate the migration of radionulides in the unsaturated zone to some degree of satisfaction, depending on the complexity of the site-specific conditions. The final aspect for model applicability considered here is the complexity of the model and its ease-of-use. As given in Table 3.6, CHAIN is the simplest and easiest model while the rest are approximately the same in model complexity and ease-of-use. The pre- and post-processors for EPACMTP (FECTUZ) and MULTIMED (MULTIMED_DP) would provide some degree of help in the use of the model. Lastly, usability of the model user's manuals is also very important for model applicability, but it is not evaluated here. The following sections provide the description of the model applicability in the SSL processes. A summary table is also provided to highlight the key points in the model application to the SSL processes (Table 3.10).

**HYDRUS**. The HYDRUS model can simulate chemical movement in layered soils from a finite source. It may be useful in settings where low-permeability clay layers may attenuate contaminants through adsorption. The model also considers root zone water uptake and evapotranspiration so that infiltration (net recharge) into the soil can be obtained from rainfall data. The net infiltration amount needs to be provided by the user for the models CHAIN, FECTUZ, and MULTIMED-DP. HYDRUS outputs the radionuclide concentration in the soil water as a function of time and depth (including at the water table), which can be used in the SSLs comparison, along with the amount of chemical remaining in the soil. The model also outputs cumulative solute flux across the bottom of the soil profile (water table). Because it can estimate infiltration from rainfall intensities, HYDRUS may be useful in SSL applications. Grid discretization for HYDRUS version 6.0 requires extra effort. Availability of root water uptake parameters and potential evaporation might be limited at a site if the root water uptake process is considered.

**MULTIMED-DP**. MULTIMED-DP simulates simple vertical water movement in the unsaturated zone. Because an initial soil concentration cannot be specified, either the soil/water partition equation or a leaching test (SPLP) must be used to estimate soil leachate contaminant concentrations. MULTIMED_DP is appropriate for simulating contaminant migration in soil and can be used to model vadose zone attenuation of leachate concentrations derived from a partition equation. MULTIMED_DP outputs the leachate radionuclide concentration at the water table which is needed for the SSLs comparison. In addition, because it links the output from the unsaturated zone transport module with a saturated zone module, it can be used to determine the concentration of a radionuclide in a receptor well. MULTIMED_DP is appropriate for early-stage site simulations because the input parameters required are typically available (a database of chemical properties is provided in MULTIMED_DP). This model is the only model which considers runoff. Therefore, when runoff is an important hydrological process at a site, selection of MULTIMED_DP might be considered. Uncertainty analyses can be performed using Monte Carlo simulations for those parameters for which reliable values are not known. Because of the complexity of the model and requirement of a great amount of input data, expertise in properly using the model for SSLs is essential.

**FECTUZ.** FECTUZ at one time was a stand-alone unsaturated zone model, but, in recent years the model is only available as one of the coupled modules in the EPACMTP program (EPA 1995b, 1995c). FECTUZ simulates migration of contaminants from a landfill (or a surface impoundment), with finite or infinite sources, through the unsaturated zone and into ground water. The model allows time varying precipitation data for inputs, but a steady-state water flow is assumed during each precipitation event. The concentrations of radionuclides at a specific time and space are part of the model outputs and can be used in the SSL process. The implementation of Monte Carlo simulation in the EPACMTP program also provides the capacity of uncertainty analysis for the FECTUZ model. It is worthy to note that mixed units for the model inputs (e.g. cm/hr, m/year, cm, m) are used in FECTUZ. Therefore, caution should be taken for the use of correct values and units in FECTUZ.

**CHAIN.** The CHAIN model is simple and easy to use in simulating fate and transport of radionuclides in a uniform soil under steady unsaturated flow conditions. The model outputs the leachate radionuclide concentrations at the specified time and depth (including at the water table) and these concentrations can be used in the SSL process. It is not adequate to use CHAIN for fate and transport of a radionuclide in layered soil. However, the CHAIN model might be used in a layered soil by using representative uniform soil properties as an approximation of heterogeneous soil properties. The CHAIN model could be the first choice as a preliminary assessment tool in SSLs evaluating for radionuclides.

**CHAIN 2D**. Basically, CHAIN 2D is very similar to HYDRUS 6.0. The major difference is it is a two-dimensional model. When the assumption of vertical flow in the unsaturated zone is in question, CHAIN 2D may be the choice to simulate leaching of a radionuclide from the disposal facility to ground water. This is especially true when the leaching area is small and horizontal flow becomes significant in a highly stratified soil. The CHAIN 2D model outputs the radionuclide concentration in the soil water as a function of time and depth, which can be used in the SSLs comparison, along with the amount of chemical remaining in the soil. The model also outputs cumulative solute flux across the bottom of the soil profile (water table). Because it can estimate infiltration from rainfall, the model may be useful in SSL applications. Both the CHAIN 2D and HYDRUS models are capable of simulating water uptake by plant roots. Both models can be used for estimating net recharge when potential evaporation and water uptake parameters are available.

## Examples of Model Application in the Soil Screening Processes

To demonstrate how to apply the five models for radionuclides evaluated herein in the SSL estimation, a conceptual site model was developed at the Las Cruces Trench Site and the base case simulations were performed. It is assumed that the Site had been used as a waste disposal/storage facility where radionuclides from tank leaks or improper waste disposal were released to the soil surface for 1000 days with a total amount for $3 \times 10^{-4}$ mg /cm$^2$ $^{99}$Tc ($^{99}$Tc concentration from the waste source is $1.25 \times 10^{-2}$ mg/L)**.** Rainfall infiltration (with a net annual recharge rate of 87 mm/y) is the driving force for the downward migration of radionuclide to the water table beneath. Base values of the input parameter are given in Table 3.11. It is assumed that the steady-state uniform water flow occurs at the site.

Time-varied leachate concentrations of $^{99}$Tc entering ground water predicted by the CHAIN, HYDRUS, MULTIMED_DP, FECTUZ, and CHAIN 2D models are presented in Figures 3.1 and 3.2. The results indicate that the five models provide similar breakthrough curves except that numerical dispersion is observed using MULTIMED_DP. Figures 1 and 2 also show how variability of the distribution coefficient ($K_d$) influence the migration of $^{99}$Tc. Increasing Kd values would reduce migration of radionuclides. The breakthrough curves for the decay (daughter) product $^{99}$Ru, using the CHAIN and FECTUZ (EPACMTP), are presented in Figure 3.3. The breakthrough curves predicted by the CHAIN and FECTUZ models are not distinguishable. Using MULTIMED_DP, a large numerical dispersion was observed (not shown). It was also found that the breakthrough curves predicted by the CHAIN model will exhibit a great oscillation if single precision is used in the CHAIN code. Figure 3.3 is also used to demonstrate the decay reaction implemented in the five models. No attempt was made in using the HYDRUS and CHAIN 2D models for obtaining the breakthrough curves of the decay products since simulation times of up to 800 years are required.

For the use of models in the SSL process, the task is to simulate the conceptual site scenario and examine whether the leachate concentration entering the ground water will result in an exceedance of the radionuclide concentration over the MCL at the receptor well. The results shown in Figure 3.4 indicate that the calculated concentrations at the receptor well exceed the MCL (5.3E-5 mg/L) for $^{99}$Tc. In other words, the site conditions exhibit a soil radionuclide contamination of $^{99}$Tc exceeding the SSL and thus has imposed a potential risk to human health and the environment. Note a few things about Figure 3.4: 1) three receptor well locations (0 m, 10 m and 30 m from the waste source) were examined; 2) two methods were employed to calculate the dilution effect of the radionuclides in the saturated zone once they enter the water table. One was the use of the simple dilution factor. Another method was the use of a saturated zone model (SZM) such as FECTUZ/EPACMTP to simulate the radionuclide transport. Using the dilution factor, a default DAF value of 20 as well as the dilution factors, at three ground-water velocities (5 m/y, 10 m/y, and 25 m/y), obtained from equations 7 and 8 in the section 2.5.2 of the SSG user's guide were used. The parameters used for the dilution factor calculation and the ground water flow and transport simulation using FECTUZ/EPACMTP are given in Table 3.12.

The details of the model application in the SSL process and the sensitivity analysis are provided in the report entitled "*Evaluation of Computer Models for Simulating Radionuclide Transport in the Unsaturated Zone*"

## Table 3.6. Summary Comparisons of the Vadose Zone Model for Radionuclides in the SSL Process

| Model component | HYDRUS | MULTIMED-DP | FECTUZ | CHAIN | CHAIN 2D |
|---|---|---|---|---|---|
| Contaminants | | | | | |
| Organics | ! | ! | ! | ! | ! |
| Metals | ! | ! | ! | ! | ! |
| Radionuclides (parent) | ! | ! | ! | ! | ! |
| Radionuclides (progeny) | ! | ! | ! | ! | ! |
| Non-aqueous phase liquids | -- | -- | -- | -- | -- |
| Site characteristics | | | | | |
| Vadose zone | | | | | |
| Layered soil | ! | ! | ! | -- | ! |
| Sources types | | | | | |
| Contaminated soil | ! | ! | ! | ! | ! |
| Landfill | ! | ! | ! | -- | ! |
| Surface impoundment | -- | -- | ! | -- | -- |
| Waste piles | -- | -- | ! | -- | -- |
| Source term characteristics | | | | | |
| Mass balance | ! | ! | ! | ! | ! |
| Multimedia partitioning | ! | ! | ! | -- | ! |
| Source decay | -- | ! | ! | ! | -- |
| Source ingrowth (radionuclides) | -- | -- | -- | -- | -- |
| Multiple contaminants per simulations | ! | ! | ! | ! | ! |
| Source release mechanisms | | | | | |
| Leaching | ! | ! | ! | ! | ! |
| Direct release to: | | | | | |
| Vadose zone | ! | ! | ! | ! | ! |
| Groundwater | -- | ! | ! | -- | -- |
| Surface water | -- | ! | -- | -- | -- |
| Air | -- | ! | -- | -- | -- |
| Medium-specific flow | | | | | |
| Air Box model (0-D, complete mixing) | -- | -- | -- | -- | -- |
| Surface Hydrology | | | | | |
| Precipitation | -- | ! | -- | -- | -- |
| Runoff | -- | ! | -- | -- | -- |
| Infiltration | ! | ! | ! | ! | ! |
| ET | ! | ! | -- | -- | ! |
| Surface Water (Stream discharge) | -- | ! | -- | -- | -- |
| Vadose Zone | | | | | |
| Vadose zone (Steady-state infiltration -->soil | ! | ! | ! | ! | ! |
| Vadose zone (n-D dynamic) | ! | -- | -- | -- | ! |
| Groundwater | -- | ! | ! | -- | -- |
| Medium-specific contaminant transport | | | | | |
| Atmosphere (emission through diffusion) | -- | ! | -- | -- | -- |
| Surface water (stream interception and mixing) | -- | ! | -- | -- | -- |
| Vadose zone (1-D advection and dispersion) | ! | ! | ! | ! | ! |
| Vadose zone (2-D advection and dispersion) | -- | -- | -- | -- | ! |
| Groundwater | | | | | |
| Homogeneous aquifer (1-D advection and dispersion) | -- | ! | ! | -- | -- |
| Homogeneous aquifer (2-D advection and dispersion) | -- | ! | ! | -- | -- |
| Homogeneous aquifer (3-D advection and dispersion) | -- | ! | ! | -- | -- |
| Medium-specific heat transport | ! | -- | -- | -- | ! |

| | | | | | |
|---|---|---|---|---|---|
| Contaminant transformations and fate processes | | | | | |
| 1st order decay (not decay products) | ! | ! | ! | ! | ! |
| 1st order decay (with chained daughter and granddaughter decay products) --straight chain | ! | ! | ! | ! | ! |
| 1st order decay -- branch chain | ! | -- | ! | -- | ! |
| Non-1st order decay | ! | -- | -- | -- | ! |
| Linear partitioning (water/soil) | ! | ! | ! | ! | ! |
| Nonlinear partitioning (water/soil) | ! | -- | ! | -- | ! |
| Hydrolysis | -- | ! | ! | -- | -- |
| Chemical reactions/speciation | – | ! | ! | -- | -- |
| | | | | | |
| Intermedia contaminant fluxes | | | | | |
| Surface soil --> Air (volatilization) | ! | ! | -- | -- | ! |
| Surface soil --> Vadose zone (leaching) | ! | ! | ! | ! | ! |
| Surface soil --> Overland (erosion, runoff) | -- | ! | -- | -- | -- |
| Surface water -->Sediment (sedimentation) | -- | -- | -- | -- | -- |
| Vadose zone --> groundwater (percolation) | ! | ! | ! | ! | ! |
| Vadose zone --> Air (volatilization) | ! | ! | -- | -- | ! |
| Groundwater --> Surface water (deposition) | -- | -- | -- | -- | -- |
| Air --> Surface soil (deposition) | -- | -- | -- | -- | -- |
| Air --> Surface water (deposition) | -- | -- | -- | -- | -- |
| | | | | | |
| Exposure pathways | -- | -- | -- | -- | -- |
| | | | | | |
| Human health endpoints | -- | -- | -- | -- | -- |

! denotes component is included in model; -- denotes component is not included in model.

## Table 3.7. Soil Hydraulic Properties at the Las Cruces Trench Site for SSG Model Evaluation Study (Modified from Wierenga et al., 1991)

| Layers | Depth (cm) | Saturated water content ($cm^3/cm^3$) | Residual water content ($cm^3/cm^3$) | van Genuchten alpha coefficient, $\alpha$, ($cm^{-1}$) | van Genuchten beta coefficient, $\beta$ (--) | Saturated hydraulic conductivity, $K_s$, (cm/d) |
|---|---|---|---|---|---|---|
| Uniform Soil Model | | | | | | |
| all | 0-600 | 0.321 | 0.083 | 0.055 | 1.509 | 270 |
| Layered Soil Model | | | | | | |
| 1 | 0-15 | 0.348 | 0.095 | 0.042 | 1.903 | 539 |
| 2 | 15-140 | 0.343 | 0.091 | 0.062 | 1.528 | 250 |
| 3 | 140-205 | 0.336 | 0.085 | 0.060 | 1.574 | 267 |
| 4 | 205-250 | 0.313 | 0.071 | 0.068 | 1.537 | 300 |
| 5 | 250-305 | 0.302 | 0.072 | 0.040 | 1.550 | 250 |
| 6 | 305-370 | 0.294 | 0.090 | 0.070 | 1.711 | 334 |
| 7 | 370-460 | 0.310 | 0.073 | 0.027 | 1.418 | 221 |
| 8 | 460-540 | 0.325 | 0.083 | 0.041 | 1.383 | 172 |
| 9 | 540-600 | 0.306 | 0.078 | 0.047 | 1.432 | 226 |

## Table 3.8. Solute Transport Properties at the Las Cruces Trench Site for SSG Model Evaluation Study (Modified from Porro and Wierenga, 1993)

| Layers | Depth (cm) | Pore velocity, v, (cm/d) | Dispersion coefficient, D, ($cm^2/d$) | Dispersivity, ε, (cm) |
|---|---|---|---|---|
| Uniform Soil Model | | | | |
| all | 0-500 | 14.7 | 62.1 | 4.53 |
| Layered Soil Model | | | | |
| 1 | 82 | 18.4 | 40.5 | 2.20 |
| 2 | 125 | 16.5 | 66.1 | 4.00 |
| 3 | 220 | 17.1 | 52.5 | 3.06 |
| 4 | 310 | 13.2 | 67.3 | 5.09 |
| 5 | 400 | 10.9 | 84.6 | 7.80 |
| 6 | 500 | 12.3 | 61.9 | 5.04 |

## Table 3.9. Characteristics of the Las Cruces Trench Site for SSG Model Evaluation Study (modified from Gee et al., 1994)

| Annual Precipitation (cm/y) | Annual Potential (Pan) Evaporation (cm/y) | Annual Potential Recharge (cm/y) | Average Daily Max. Air Temperature ($^{O}$C) | Average Daily Min. Air Temperature ($^{O}$C) | Elevation (m) | Depth to Water Table (m) | Geology | Typical Soil Type | Typical Vegetation |
|---|---|---|---|---|---|---|---|---|---|
| 33.8 | 239 | 8.7 | 28 | 13 | 1357 | 60 | Alluvial | Berino fine loamy sand | Creosote bush |

# Table 3.10. Summary of the Use of the Unsaturated Zone Models for Radionuclides in the SSL Process

| Model | Processes, outputs, components |
|---|---|
| HYDRUS | - provides the leachate radionuclide concentrations entering the ground water so whether the resulting concentration of the radionuclide at the receptor well would exceed the acceptable level or not, can be examined<br>- calculates infiltration which can be used as inputs in the SSL calculation<br>- considers soil heterogeneity, time-varying infiltration and evapotranspiration<br>- outputs radionuclide concentration in soil, cumulative flux across water table<br>- grid discretization for HYDRUS version 6.0 requires extra effort |
| MULTIMED_DP | - provides the leachate radionuclide concentrations entering the ground water so whether the resulting concentration of the radionuclide at the receptor well would exceed the acceptable level or not, can be examined<br>- uncertainty of model outputs can be examined<br>- considers runoff, evapotranspiration<br>- linked with a saturated flow and transport model<br>- requires a great amount of input data, expertise because of model complexity |
| FECTUZ | - provides the leachate radionuclide concentrations entering the ground water so whether the resulting concentration of the radionuclide at the receptor well would exceed the acceptable level or not, can be examined<br>- uncertainty of model outputs can be examined<br>- linked with a saturated flow and transport model<br>- uses mixed units for the input data |
| CHAIN | - provides the leachate radionuclide concentrations entering the ground water so whether the resulting concentration of the radionuclide at the receptor well would exceed the acceptable level or not, can be examined<br>- used for simplified radionuclide-contaminated site scenario<br>- simple-to-use, less input data requirement<br>- as a preliminary assessment tool in SSL estimation |
| CHAIN 2D | - provides the leachate radionuclide concentrations entering the ground water so whether the resulting concentration of the radionuclide at the receptor well would exceed the acceptable level or not, can be examined<br>- calculates infiltration which can be used as inputs in the SSL calculation<br>- considers soil heterogeneity, time-varying infiltration and evapotranspiration<br>- outputs radionuclide concentration in soil, cumulative flux across water table<br>- considers two-dimensional soil heterogeneity |

## Table 3.11. Base Values of Input Parameters for the Unsaturated Zone Models

| Parameters | Values |
|---|---|
| **Source-Specific Parameters** | |
| Area of disposal facility ($m^2$) | 400 |
| Width of disposal facility (m) | 20 |
| Length of disposal facility (m) | 20 |
| Mass release of (parent) radionuclide $^{99}$TC ($mg/cm^2$) | $3x10^{-4}$ |
| Concentration of $^{99}$Tc in recharge water from waste source (mg/L) | $1.25x10^{-2}$ |
| Duration of waste source being completely released (days) | 1000 |
| Net recharge rate (mm/y) | 87 |
| Water content ($cm^3/cm^3$) | 0.16 |
| Source decay constant | -- |
| **Soil Properties in Unsaturated Zone** (Uniform Soil in Table 3-7) | |
| Saturated hydraulic conductivity, $K_s$, (cm/d) | 270.1 |
| Porosity (--) | 0.358 |
| Residual water content ($cm^3/cm^3$) | 0.083 |
| Saturated water content ($cm^3/cm^3$) | 0.321 |
| Bulk density | 1.70 |
| van Genuchten alpha coefficient, $\alpha$, ($cm^{-1}$) | 0.055 |
| van Genuchten beta coefficient, $\beta$ (--) | 1.509 |
| Depth to water table (m) | 6 |
| **Solute Transport Parameters** | |
| Decay coefficient for parent ($^{99}$TC) (1/y) | $3.3x10^{-6}$ |
| Decay coefficient for daughter ($^{99}$Ru) (1/y) | $7.9x10^{-9}$ |
| Distribution coefficient for parent ($^{99}$TC) (ml/g) | 0.007 |
| Distribution coefficient for daughter ($^{99}$Ru) (ml/g) | 5.0 |
| Dispersion Coefficient ($cm^2/d$ ) | 1.0 |
| Dispersivity (cm) | 4.53 |

**Table 3.12. Input Parameters Used for the Dilution Factor Calculation and Transport Simulation of Radonuclide in the Saturated Zone using FECTUZ/EPACMTP Model**

| Parameters | Values |
|---|---|
| **Hydraulic Properties in Saturated Zone** | |
| Aquifer hydraulic conductivity (m/y) | 500 |
| Hydraulic gradient (m/m) | 0.005 |
| Porosity | 0.32 |
| Bulk density (g/cm$^3$) | 1.70 |
| Longitudinal dispersivity (m) | 1.0 |
| Transverse dispersivitiy (m) | 0.2 |
| Vertical dispersivity (m) | 0.04 |
| **Aquifer Characteristics** | |
| Aquifer (m) | 10 |
| **Computed Dilution Factor Using Equations 7 & 8 in the Section 2.5.2 of the SSG User's Guide** | |
| Dilution factor for ground-water velocity = 5 m/y | 8.1 |
| Dilution factor for ground-water velocity = 10 m/y | 14.2 |
| Dilution factor for ground-water velocity = 25 m/y | 32.4 |

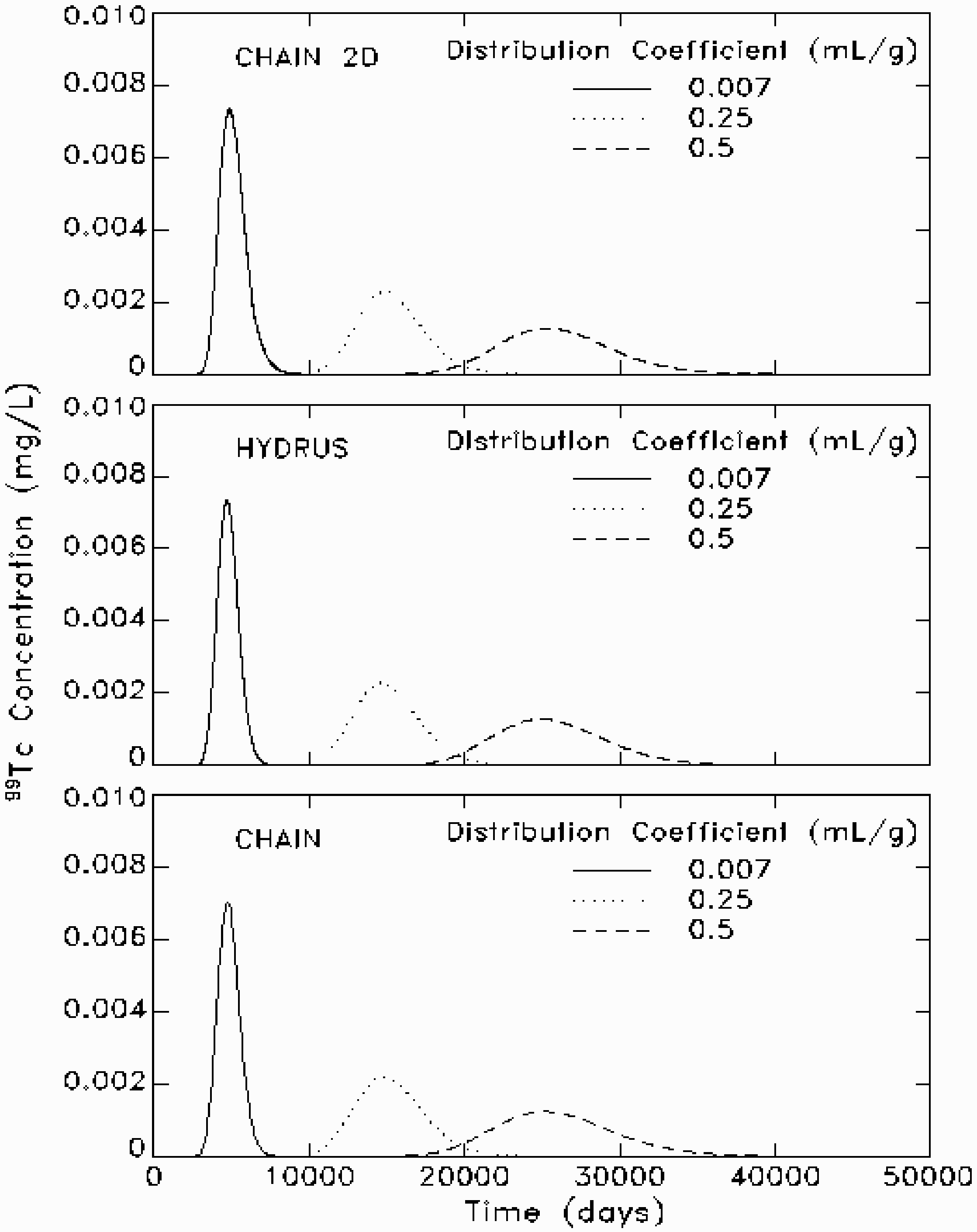

Figure 3.1. Base case simulations of $^{99}$TC breakthrough (through the unsaturated zone) with three distribution coefficients using the CHAIN, HYDRUS, and CHAIN 2D models.

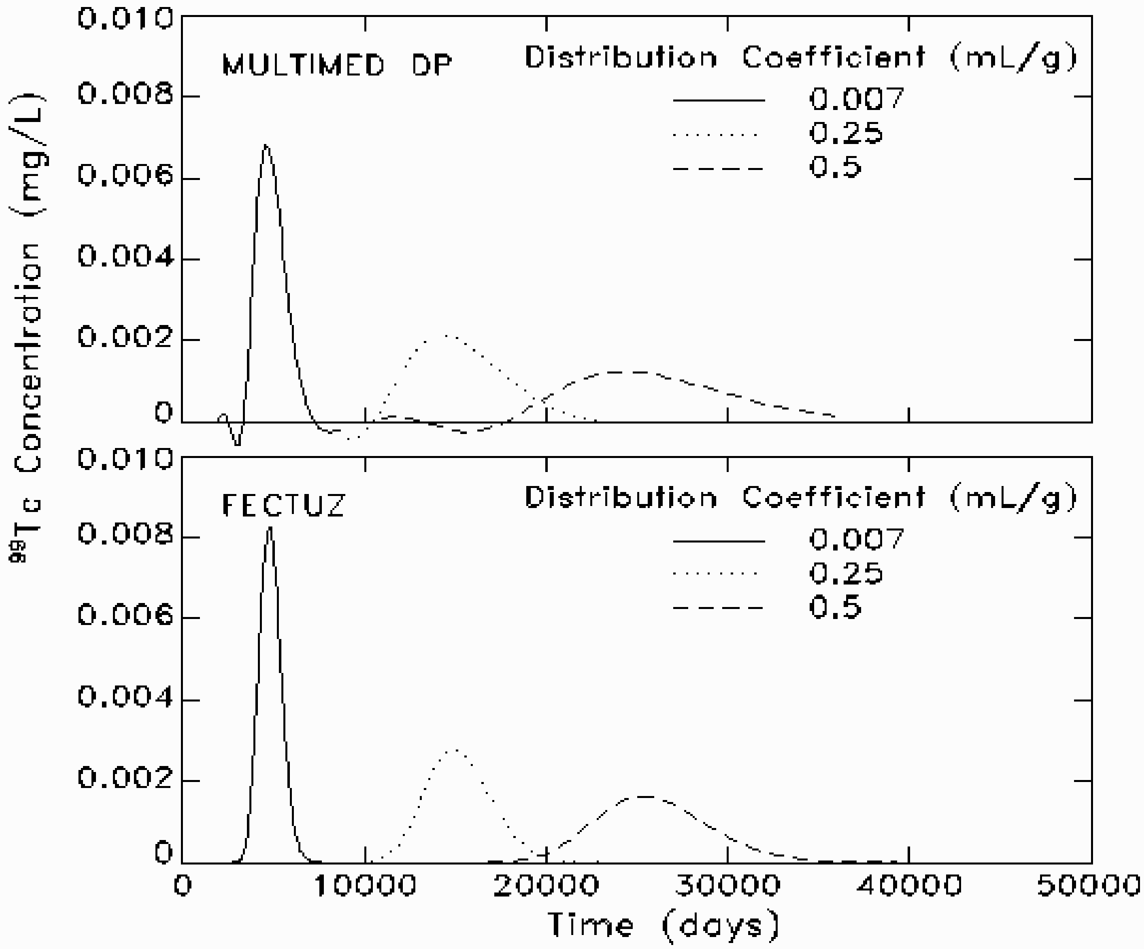

Figure 3.2. Base case simulations of $^{99}$TC breakthrough (through the unsaturated zone) with three different distribution coefficients using the FECTUZ, and MULTIMED_DP models.

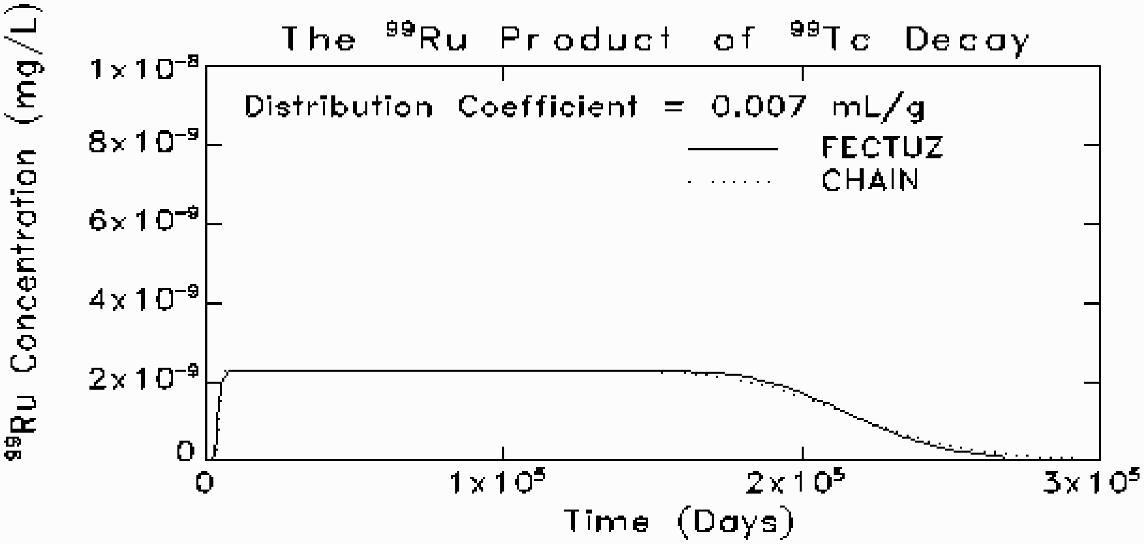

Figure 3.3. Base case simulations of the daughter product -- $^{99}$Ru of $^{99}$TC breakthrough (through the unsaturated zone) with a distribution coefficient of 0.007 mL/g using FECTUZ, and CHAIN models.

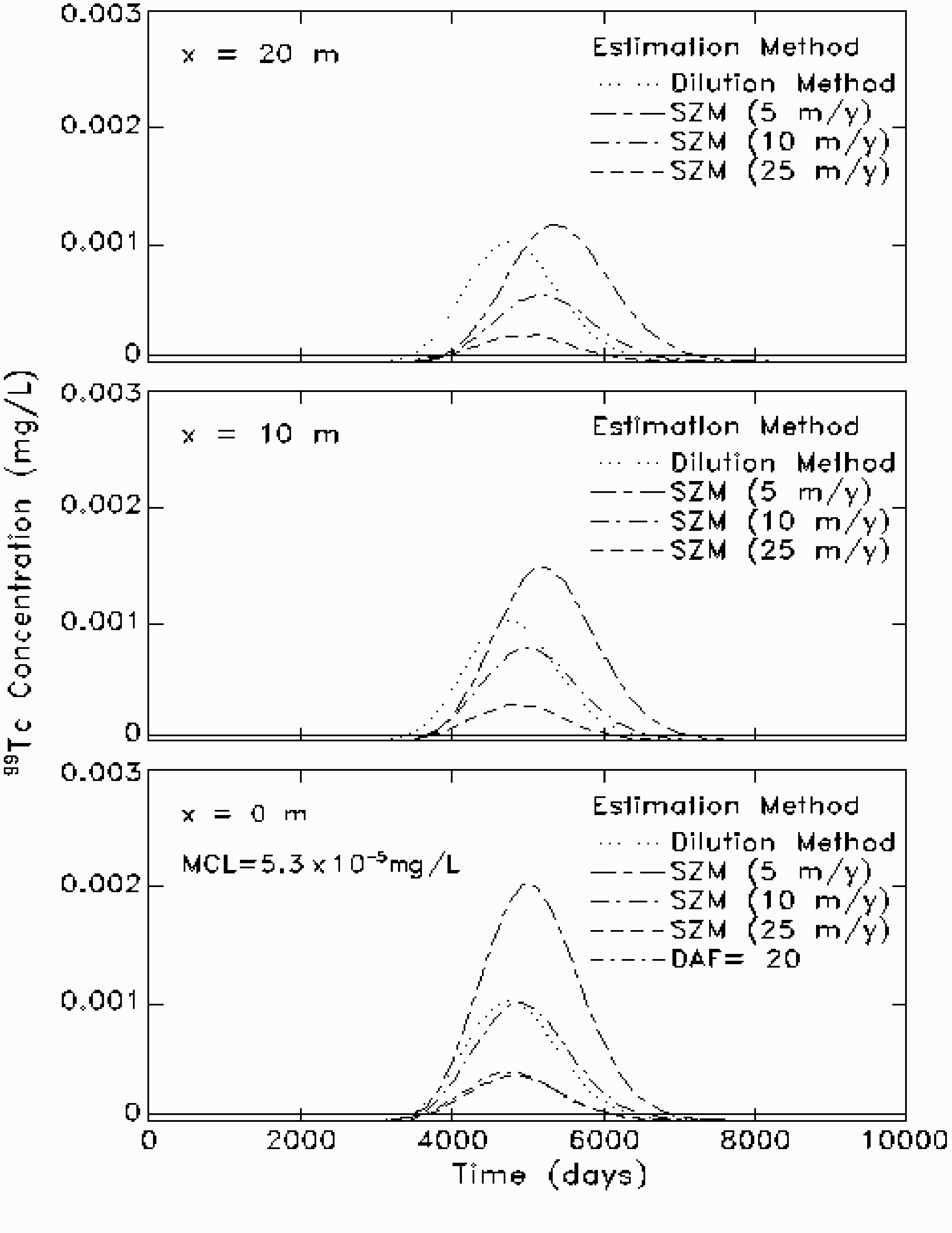

Figure 3.4. Base case predication of $^{99}$TC concentration at the receptor well by employing dilution factor method and using saturated zone model (SZM) -- FECTUZ. The numbers in the parenthes2s are ground water velocity. The solid lines are used to represent the MCL ($5.3 \times 10^{-5}$ ml/L) level.