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Application of TREECS™ to Strontium-90 for Borschi Watershed near Chernobyl, Ukraine

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ABSTRACT

The Training Range Environmental Evaluation and Characterization System (TREECS™) (http://el.erdc.usace.army.mil/treecs/) is being developed by the U.S. Army Engineer Research and Development Center (ERDC) for the U.S. Army to forecast the fate of munitions constituents (MC) (such as high explosives (HE) and metals) found on firing/training ranges, as well as those subsequently transported to surface water and groundwater. The overall purpose of TREECS™ is to provide environmental specialists with tools to assess the potential for MC migration into surface water and groundwater systems and to assess range management strategies to ensure protection of human health and the environment. The multi-media fate/transport models within TREECS™ are mathematical models of reduced form (e.g., reduced dimensionality) that allow rapid application with less input data requirements compared with more complicated models. Although TREECS™ was developed for the fate of MC from military ranges, it has general applicability to many other situations requiring prediction of contaminant (including radionuclide) fate in multi-media environmental systems.

TREECS™ was applied to the Borschi watershed near the Chernobyl Nuclear Power Plant, Ukraine. At this site, TREECS™ demonstrated its use as a modeling tool to predict the fate of strontium 90 (^{90}Sr). The most sensitive and uncertain input for this application was the soil-water partitioning distribution coefficient (K_d) for ^{90}Sr. The TREECS™ soil model provided reasonable estimates of the surface water export flux of ^{90}Sr from the Borschi watershed when using a K_d for ^{90}Sr of 200 L/kg. The computed export for the year 2000 was 0.18% of the watershed inventory of ^{90}Sr compared to the estimated export flux of 0.14% based on field data collected during 1999–2001. The model indicated that assumptions regarding the form of the inventory, whether dissolved or in solid phase form, did not appreciably affect export rates. Also, the percentage of non-exchangeable adsorbed ^{90}Sr, which is uncertain and affects the amount of ^{90}Sr available for export, was fixed at 20% based on field data measurements. A Monte Carlo uncertainty analysis was conducted treating K_d as an uncertain input variable with a range of 100–300 L/kg. This analysis resulted in a range of 0.13–0.27% of inventory exported to surface water compared to 0.14% based on measured field data.

Based on this model application, it was concluded that the export of ^{90}Sr from the Borschi watershed to surface water is predominantly a result of soil pore water containing dissolved ^{90}Sr being diverted to surface waters that eventually flow out of the watershed. The percentage of non-exchangeable adsorbed ^{90}Sr and the soil-water K_d are the two most sensitive and uncertain factors affecting the amount of export. The 200-year projections of the model showed an exponential decline in ^{90}Sr export fluxes from the watershed that should drop by a factor of 10 by the year 2100.

This presentation will focus on TREECS capabilities and the case study done for the Borschi Watershed.

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1. Introduction

The Training Range Environmental Evaluation and Characterization System (TREECS™) (http://el.erdc.usace.army.mil/TREECS™/) was developed for the Army to predict the fate of munitions constituents (MC), such as high explosives (HE) and metals, released to

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soil and transported from firing/training ranges to surface water and groundwater. The overall objective is to provide environmental specialists with tools to assess the potential for migration of MC into surface water and groundwater systems. The results of these assessments can be used to assess range management strategies to protect human health and the environment from MC exposure in receiving waters. TREECS™ was developed with two levels of capability. Tier 1 consists of screening-level methods that assume highly conservative, steady-state MC loading and fate. Tier 1 requires minimal input data requirements and can be easily and quickly applied by environmental staff to assess the potential for migration into surface water and groundwater. If surface water and/or groundwater MC concentrations predicted with Tier 1 exceed protective health benchmarks at receptor locations, then further evaluation with Tier 2 is recommended to obtain more definitive results.

Tier 2 provides time-varying analyses and solves mass balance equations for both solid and partitioned phase (hereafter referred to as the non-solid phase) MC with dissolution into water. Additionally, MC residue loadings to the range soil can vary from year-to-year based on munitions use. Thus, media concentrations computed with Tier 2 should be closer to those expected under actual conditions and lower than those computed with Tier 1 due to attenuating effects.

Although TREECS™ was developed for the fate of MC from military ranges, it has general applicability to many other situations requiring prediction of contaminant fate in multi-media environmental systems, including the fate of radionuclides in surface water and groundwater. TREECS™ Tier 2 was applied to the Borschi watershed near the Chernobyl Nuclear Power Plant, Ukraine, to demonstrate its use as a modeling tool to predict the fate of radiostrontium-90 that was deposited within the watershed following the Chernobyl reactor accident in 1986. This application also provided an additional validation test case for the TREECS™ Tier 2 soil model. This paper describes TREECS™ Tier 2 and the input data and results of this application to the Borschi watershed.

2. Methods

2.1. TREECS™ Conceptualization

TREECS™ consists of a collection of mass balance-based, mechanistic fate and transport models for environmental media, as well as supporting pre- and post-processing components. Fig. 1 presents a schematic of the conceptual model that TREECS™ Tier 2 addresses. A number of previous models used for assessment of radionuclide fate in watersheds take a more empirical, holistic approach (Garcia-Sanchez, 2008; Monte et al., 2004; Joshi and Shukla, 1991; Smith et al., 2004), whereas TREECS™ uses a

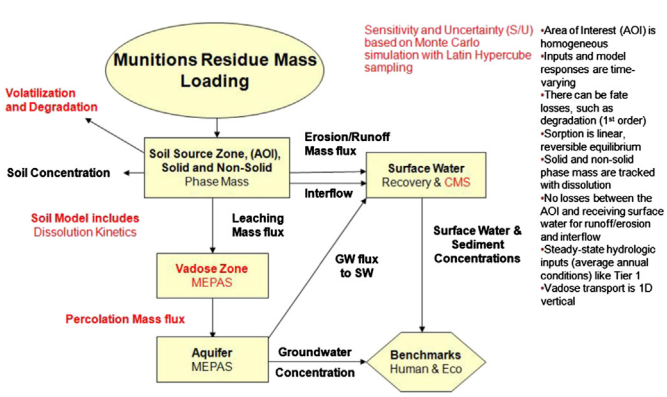

Fig. 1. TREECS™ Conceptualization.

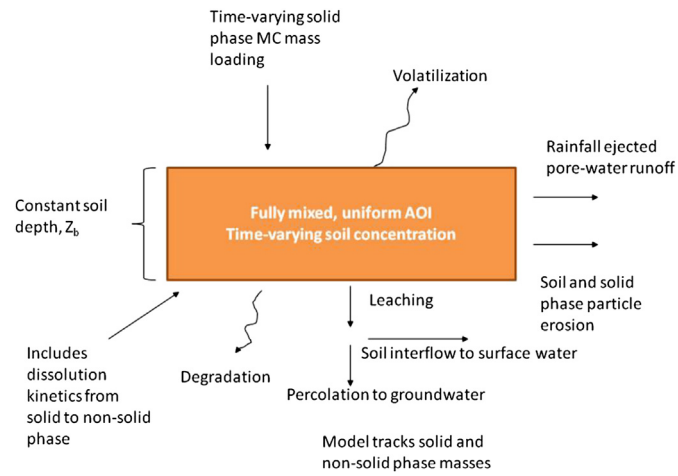

Fig. 2. TREECS™ soil model.

reductionistic approach, which attempts to account for the fundamental physical and chemical processes (Monte et al., 2004). Holistic models require field observations to adjust or fit empirical input parameters. Mechanistic models require estimates or measurement of process-based parameters, and field data are used indirectly to calibrate or adjust those parameters or to validate the model after process-based parameters have been set.

The source zone for MC (or other non-military contaminant) is the surface soil of the area of interest (AOI). The AOI could be the primary impact area of fired munitions for example. The MC residue loading to the AOI must be estimated or specified. Initial MC soil concentrations within the AOI can also be specified if such information is available. In addition to the loading model, fate and transport models are included for the four media of AOI soil, vadose zone beneath the AOI soil layer, groundwater (aquifer), and receiving surface water including sediment. All of the MC fate models within TREECS™ are models of reduced form, meaning that simplifications are imposed (such as spatial dimensionality) to reduce model complexity and facilitate ease-of-use.

Potential environmental and human receptors could be exposed to MC if it has migrated to groundwater wells or receiving surface waters. Thus, the end point metrics are the predicted MC concentrations at target groundwater wells and surface water body downgradient of the AOI. These concentrations are compared to environmental and human health protective benchmarks (conservative screening values) developed by the DoD Range and Munitions Use Subcommittee to determine if more detailed site evaluations are required.

2.2. TREECS™ model descriptions

2.2.1. AOI Tier 2 Soil Model

The Tier 2 soil model, Fig. 2, is described in more detail than the other media models since this is a new model, whereas, the other models are legacy models. The AOI surface soil is treated as a homogenous, fully mixed compartment with a thickness Z_b (m) and a surface area A (m^2), which are both constant over time. Treating a heterogenous AOI as homogenous is not a compromising assumption because the total MC source mass loading and/or inventory are the driving variables for AOI mass export, not MC concentration, since the model is mass balance based. Thus, the mass export from the AOI to other media does not depend on AOI surface area or volume or the AOI MC concentration (Dortch et al., 2009, 2011).

Each constituent can exist in solid and non-solid (water-dissolved from solid) phases. The non-solid phase mass exists in

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equilibrium distributed as dissolved in water within the water filled soil pore spaces, as adsorbed from water to soil particles, and as a vapor in air within the air filled pore spaces. A time-varying mass balance is performed for both the solid and non-solid phases.

The solid phase mass balance is stated as

$$\frac{dM_s}{dt} = L(t) - F_{dis} - F_{es} + F_{precip} \quad (1)$$

where M_s is the solid phase mass (g), t is time (yr), $L(t)$ is time-varying solid phase constituent mass loading (g/yr), F_{dis} is dissolution flux (g/yr), F_{es} is the erosion flux of solid phase constituent particles (g/yr), and F_{precip} is the precipitation flux (g/yr) of constituent due to dissolved pore water concentration exceeding the water solubility limit. The non-solid phase mass balance is stated as

$$\frac{dM_{ns}}{dt} = F_{dis} - F_r - F_e - F_l - F_{decay} - F_{vol} - F_{precip} \quad (2)$$

where M_{ns} is non-solid phase constituent mass (g), F_r is the rain-induced pore water ejection and runoff flux (g/yr), F_e is constituent flux due to soil erosion (g/yr), F_l is leaching flux (g/yr), F_{decay} is degradation flux (g/yr), and F_{vol} is volatilization flux (g/yr). The total (particulate or adsorbed to soil, dissolved, and vapor) non-solid phase constituent concentration within the soil matrix on a total volume basis, C_{tt} (g/m^3) is

$$C_{tt} = \frac{M_{ns}}{V} = \frac{M_{ns}}{AZ_b} = \frac{M_d + M_p + M_a}{AZ_b} \quad (3)$$

where V is the surface soil compartment (AOI) volume (m^3). The terms M_d, M_p, and M_a are the constituent mass (g) dissolved, adsorbed to soil particles, and in air, respectively. The total non-solid concentration can be expressed as the sum of the three media concentrations,

$$C_{tt} = \theta_w C_l + (\phi - \theta_w) C_g + \rho_b C_a \quad (4)$$

where

C_l = concentration dissolved in pore water (liquid) phase, M_d/V_w, g/m^3
C_g = vapor (gas) concentration in air, M_a/V_a, g/m^3
C_a = concentration adsorbed to soil particles, M_p/M_{soil}, mg/kg
θ_w = soil volumetric moisture content (ratio of water volume to total volume), fraction; θ_w can't be greater than soil porosity
φ = soil porosity (ratio of void volume to total volume), fraction
ρ_b = soil dry bulk density, g/ml or kg/L
V_w = volume of water in the AOI soil, m^3
V_a = volume of air in the AOI soil, m^3
M_{soil} = mass of soil in the AOI, g

The vapor and particulate concentrations can be related to the liquid concentration through phase equilibrium partitioning, or $C_a = K_d C_l$ and $C_g = K_H C_l$, where K_d (L/kg) is the distribution coefficient for partitioning a constituent between soil particles and water, and K_H is the dimensionless Henry's constant for partitioning between air and water. Substitutions result in $C_l = \frac{C_{tt}}{\theta_w R}$ where the retardation factor R is defined as

$$R = 1 + \frac{(\phi - \theta_w) K_H + \rho_b K_d}{\theta_w} \quad (5)$$

The leaching flux is computed from $F_l = q_w A C_l$ where q_w is the average annual Darcy water infiltration rate (m/yr). Although the soil model is time-varying, average annual hydrologic forcing is used, which greatly simplifies model input requirements without compromising the long-term fate calculations. A portion or all of the leaching flux can be diverted back to surface water via soil interflow if the low permeability of the vadose zone prevents downward flux.

The degradation flux is computed from $F_{decay} = AZ_b(\lambda_l \theta_w C_l + \lambda_a \rho_b C_a)$ where λ_l and λ_a are the degradation rates (yr^{-1}) for the liquid (aqueous dissolved) and aqueous adsorbed concentrations, respectively. Degradation of the solid phase constituent is not presently included in the model. It is assumed that the vapor phase does not degrade, but it can volatilize or diffuse from the soil into the overlying air.

The volatilization flux is computed from $F_{vol} = K_v A(\phi - \theta_w) C_g$ where K_v is the volatilization rate (m/yr), or vapor escape rate from soil to the overlying air. The erosion flux is computed from $F_e = EAC_{tt}$ where E is the average annual soil erosion rate (m/yr) as computed from the Universal Soil Loss Equation (USLE) (USDA, 1983).

Following the work of Gao et al. (2004), Dortch et al. (2011) present the derivation of the annualized rain-induced pore water ejection and runoff flux, which is computed from

$$F_r = Ad_e\left(1 - e^{-\kappa}\right) C_{tt} N \quad (6)$$

where N is the average number of rainfall events a year, and d_e is the soil exchange layer thickness (m). The variable κ is defined as

$$\kappa = \frac{a\phi F_{dp} P_r}{\rho_b \theta_w d_e N} \quad (7)$$

where a is the soil detachability (kg/L); ϕ is the saturated water content, which is the soil porosity; P_r is the average annual rainfall (m/yr); and F_{dp} is defined as

$$F_{dp} = \frac{\theta_w}{\theta_w + (\phi - \theta_w) K_H + \rho_b K_d} = \frac{1}{R} \quad (8)$$

Default values of $a = 0.4$ kg/L and $d_e = 0.005$ m are typically used.

Starting with the relationship of solid phase particle dissolution into a liquid provided by Cussler (1997), Dortch et al. (2011) derive an annualized dissolution flux,

$$F_{dis} = P_t \alpha M_s C_s \quad (9)$$

where P_t is the average annual total precipitation (m/yr), C_s is the constituent solubility in water (mg/L), and α is the average specific surface area (m^2/g) of the solid phase mass, which depends on the size and shape of the solid phase particles and the constituent solid phase density. For spherical particles, $\alpha = \frac{6}{\rho_{sm} d_i}$, where ρ_{sm} is the solid phase constituent mass density (g/m^3), and d_i is the average particle diameter (m). Dortch et al. (2011) showed that the above dissolution formulation is similar to previously reported dissolution models (Phelan et al., 2004; Lynch et al., 2002; Taylor et al., 2009a,b). Dortch et al. (2011) also showed that dissolution rates computed with Equation (9) compare favorably with both experimental results reported by Taylor et al. (2009b) and their linear model.

The average particle diameter d_i and the specific surface area α vary with time as mass is dissolved from the particles. For a constant particle density, it can be shown (Phelan et al., 2004) that the average particle diameter d_i can be related to total particle mass M_s, which varies over time, with the result

$$d_i(t) = d_i(t-\Delta t)\left\{\frac{M_s(t)}{M_s(t-\Delta t)}\right\}^{1/3} \tag{10}$$

where t is time, and Δt is the time step in the solution procedure. Since M_s is being calculated each time step, d_i and thus α can be computed each time step. Particle size can shrink and expand, but it is not allowed to expand beyond the initial size. The exponent of Equation (10) is a result of assuming spherical particle shape.

Volatilization from soil was evaluated by Jury et al. (1983) by using vertically one-dimensional (1D), soil models where contaminant concentrations vary over soil depth as a result of diffusion within the soil, volatilization across the soil-air interface, and other processes, such as leaching and degradation. A simpler approach was required in this model since the soil is treated as a single homogenous layer, which precludes computing time-varying concentrations that vary with soil depth. The volatilization mass transfer rate K_v (m/yr) is a transport coefficient across the soil – air boundary that can be approximated as the diffusion coefficient of the vapor divided by the diffusion length or thickness for mass transfer. The effective diffusion rate coefficient for a vapor within air spaces of the soil matrix can be estimated from Millington and Quirk (1961),

$$D_{G_{eff}} = D_G^{air}\frac{(\phi-\theta_w)^{10/3}}{\phi^2} \tag{11}$$

where $D_{G_{eff}}$ is the effective diffusion coefficient (m^2/day) for a vapor in soil, and D_G^{air} (m^2/day) is the constituent vapor or gas diffusion coefficient in air. Values for D_G^{air} are available for some of the constituents in the TREECS™ constituent databases, or it can be estimated from molecular weight (Lyman et al., 1982). The volatilization mass transfer rate is then,

$$K_v = 365\frac{D_{G_{eff}}}{d_v} \tag{12}$$

where d_v is the diffusion layer thickness (m) in the top of the soil layer. By comparing results of the present model against reported volatilization flux rates, it was possible to determine an appropriate value of about 0.4 m for d_v. This relatively simple volatilization model was found by Dortch et al. (2011) to provide reasonable volatilization flux for widely ranging Henry's constants by using the same diffusion layer thickness of 0.4 m.

After substituting the above defined flux terms and previously introduced relationships for phase concentrations in terms of C_{tt}, the non-solid phase mass balance (Equation (2)) can be solved in terms of the unknown C_{tt},

$$\frac{dC_{tt}}{dt} = \frac{F_{dis}}{AZ_b} - \left[\frac{d_e}{Z_b}\left(1-e^{-\kappa}\right)N + \frac{E}{Z_b} + \frac{q_w}{\theta_w Z_b}F_{dp} + \right.$$

$$\frac{dC_{tt}}{dt} = \frac{F_{dis}}{AZ_b} - \left[\frac{d_e}{Z_b}\left(1-e^{-\kappa}\right)N + \frac{E}{Z_b} + \frac{q_w}{\theta_w Z_b}F_{dp} + \left(\lambda_l F_{dp} + \lambda_a F_{pp}\right) + F_{ap}\frac{K_v}{Z_b}\right]C_{tt} - \frac{F_{precip}}{AZ_b} \tag{13}$$

where F_{pp} and F_{ap} are media concentration conversion factors defined as

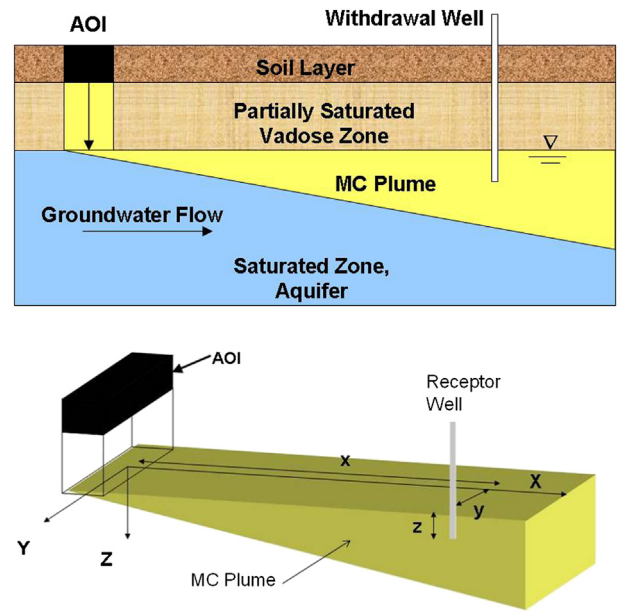

Fig. 3. TREECS™ MEPAS vadose zone and aquifer model.

$$F_{pp} = \frac{\rho_b K_d}{\theta_w + (\phi-\theta_w)K_H + \rho_b K_d} \tag{14}$$

$$F_{ap} = \frac{(\phi-\theta_w)K_H}{\theta_w + (\phi-\theta_w)K_H + \rho_b K_d} \tag{15}$$

Equations 1 and (13) constitute a system of two coupled ordinary differential equations that can be solved for the two unknowns, C_{tt} and M_s, which vary over time. The coupling terms are the dissolution and precipitation fluxes. The precipitation flux term in Equations (1) and (13) is zero when solubility is not limiting. After solving the concentration C_{tt}, all of the various flux terms defined above can be calculated for each time point and output as a time series for use as input loads to the vadose zone and surface water models.

A check is required at the end of each time step update to see if the computed value of the soil pore water concentration C_l exceeds the solubility of the constituent in water, C_s. If the computed value of C_l exceeds C_s, then the precipitation flux F_{precip} is computed from

$$F_{precip} = \frac{AZ_b(C_l - C_s)}{\Delta t} \tag{16}$$

where Δt is the time step for the most recent time update of the solution. If C_l is less than C_s, then F_{precip} is zero for the next time step update. If pore water concentration C_l exceeds solubility, then precipitation flux computed with Equation (16) is used in Equations (1) and (13) for the solution at the next time step. Equations (1) and (13) are solved using the fourth-order-accurate Runge–Kutta time integration method with an option to use either a constant or variable time step that is automatically adapted to maintain stability.

2.2.2. Vadose Zone and Aquifer Models

These two models, Fig. 3, are legacy models and are the same as the ones in the Multimedia Environmental Pollutant Assessment System (MEPAS). MEPAS consists of various models of reduced form for computing multimedia fate and transport, human exposure concentrations, and human receptor doses and health risks (Buck

et al., 1995). The MEPAS groundwater models consist of time-varying contaminant fate/transport models of the vadose zone and aquifer. The MEPAS version 5.0 groundwater models (http://mepas.pnl.gov/mepas/maqu/index.html) are used within Tier 2 of TREECS™ to compute fluxes through the vadose zone and aquifer and resulting aquifer concentrations at specified well locations. The vadose zone model solves the 1D vertical, reactive, transport equation. The aquifer model solves the 1D, reactive, transport equation for longitudinal advection and three-dimensional dispersion. A detailed description of these models is not repeated here since the scientific documentation of the MEPAS groundwater models is provided by Whelan et al. (1996), and descriptions of both models are provided by Dortch et al. (2007, 2011).

2.2.3. Surface water models

Users have two options to select from within TREECS™ Tier 2 for modeling contaminant fate in surface water and sediments: RECOVERY (Ruiz and Gerald, 2001), Fig. 4, and the Contaminant Model for Streams (CMS) (Fant and Dortch, 2007), Fig. 5. Both models can be freely downloaded from http://el.erdc.usace.army.mil/products.cfm?Topic=model&Type=watqual. RECOVERY is best suited for pooled surface water, such as ponds and lakes, while CMS is best suited for streams and rivers. These two models are briefly described here since detailed documentation of the models is given in the above references, and a more detailed overview of each model is provided by Dortch et al. (2011).

Both models solve time-varying, mass balance equations for total (dissolved and particulate, i.e., sediment adsorbed) contaminant mass in surface water and bottom sediments with reversible, equilibrium partitioning between dissolved and adsorbed particulate forms. For the RECOVERY model, the water column is treated as a fully mixed single compartment. The bottom sediments are divided into two types, a single, surficial mixed sediment layer at the sediment–water interface, and deep sediment below the surficial mixed layer. The deep sediments consist of multiple layers with each layer 1-cm thick. This treatment results in three mass balance equations with three unknowns, which apply to the water column, the mixed sediment layer, and the deep sediment layers. Two coupled ordinary differential equations are solved for the surface water and the mixed sediment layer. A partial differential equation is solved for the deep sediment layers. The deep sediment extends below the depth of contamination into clean sediment so that a zero concentration gradient boundary condition can be applied at the bottom of the sediment column. Fate processes include: water column flushing; sorption partitioning in the water column and benthic sediments; degradation in water and sediments; volatilization from water; water column sediment settling

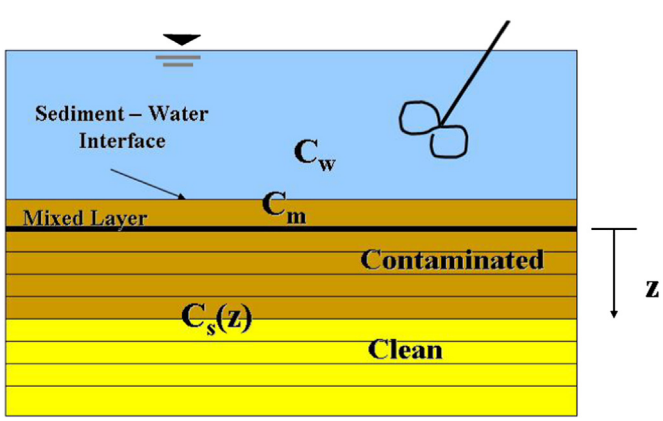

Fig. 4. TREECS™ RECOVERY surface water model.

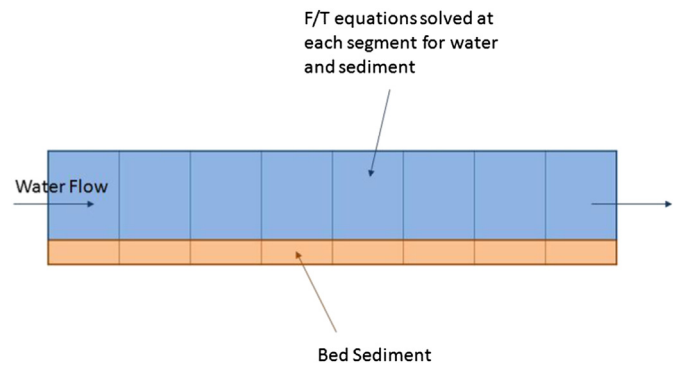

Fig. 5. TREECS™ contaminant model for streams surface water model.

and bottom sediment resuspension; deep sediment burial; mass transfer of dissolved constituent between the water column and mixed sediment layer pore water; bioturbation between the mixed sediment layer and top layer of the deep sediments; and pore water diffusion within the deep sediments. Loading boundary conditions include inflowing contaminant mass due to export from the soil model, which includes rainfall extraction, erosion, and soil interflow fluxes. There is also an option to enter user-specified constant external loadings of contaminant mass, such as those due to wastewater discharges, air deposition, or stream background loadings. Although the dependent variable is total concentration, the model produces output for total and dissolved concentrations in the water column and bed. RECOVERY has been validated against field data for pesticides in water and sediment (Boyer et al., 1994).

The CMS is very similar to RECOVERY with the primary difference being the dimensionality and its orientation. CMS treats the water column as 1D in the longitudinal, or stream-wise direction, thus, representing contaminant fate and transport in streams and rivers. Only a single, fully mixed compartment is used to represent the benthic sediments underneath each 1D water cell. There is exchange between the sediment compartment and the overlying water just as in RECOVERY, but there is no longitudinal exchange between benthic sediment compartments except for that associated with surface water fate and transport. The model solves a partial differential equation for the 1D, advection-diffusion-reaction (mass balance) equation of the surface water cells and an ordinary differential equation for each benthic sediment compartment. The CMS assumes steady, uniform flow. Stream flow can vary over time, but there is no hydraulic or hydrologic routing involved. There are various options for estimating the flow cross-sectional area and depth based on flow rate. The modeled fate processes are the same as those in RECOVERY except that bioturbation is not included since there is only one benthic layer. CMS has been verified against analytical solutions and RECOVERY (Fant and Dortch, 2007).

2.2.4. Model linkages

The exchange of model outputs that are used as inputs to downgradient models, such as soil export to vadose zone and surface water, are handled within TREECS™ via the file specification system used in the Framework for Risk Analysis in Multimedia Environmental Systems (FRAMES, Whelan et al., 1997). FRAMES has been used by several U.S. Federal agencies to facilitate multi-media environmental modeling.

For worst-case conservatism, mass fluxes exported from soil to the target surface water are transferred directly without attenuation, transport time delay, or mass loss. Thus, if the target surface water is a pond downstream of a firing range, and the stream

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connecting the range to the pond is not part of the surface water model, then mass fluxes from the range become input mass loadings to the pond without alteration in route through the connecting stream.

3. Model application

3.1. Site description

The Borschi watershed has a surface area of 8.5 km^2 and is located 3 km south of the Chernobyl Nuclear Power Plant, Fig. 6. Radio-strontium-90 (^{90}Sr) which is a fission product resulting from the accident in 1986, poses a health concern around and downgradient from Chernobyl due to its high specific radio-activity (143 Ci/g) and relatively long half-life (29 years). Near the end of the 20th century, soil and sediment concentrations of ^{90}Sr were surveyed and a total inventory of approximately 1.0×10^{13} (1.0 E13) Bq was determined for the Borschi watershed (Freed et al., 2003). Freed et al. (2003) estimated that the annual export of ^{90}Sr from the Borschi watershed to its outlet varied between 1.27 E10 and 1.62 E10 Bq for the period 1999–2001 with an average of about 1.43 E10 Bq or about 0.14% of the inventory. This export was a result of snow melt, storm events, moderate rains, and low base flows from below-ground discharge.

3.2. Model inputs

The information regarding ^{90}Sr in the Borschi watershed provided by Freed (2002) and Freed et al. (2003, 2004) was used to establish the test conditions and model inputs. The objective of the model application was to determine the annual flux of ^{90}Sr from the Borschi watershed around the year 2000 and to compare that flux to the average flux estimated by Freed et al. (2003) for the period 1999–2001 based on measured field data. This was a relatively straightforward application of TREECS™ since there were no target receiving waters to model downstream of the watershed. Thus, the only model required was the Tier 2 soil model which is used to represent the area of interest, which in this case is the entire Borschi watershed. The output from the Tier 2 soil model is time-varying mass fluxes or exports (g/yr) to surface water and groundwater. This model does not presently output results in radiation units, but this did not preclude use of this model for a radionuclide since the output can be converted from mass to radiation units as described in this paper.

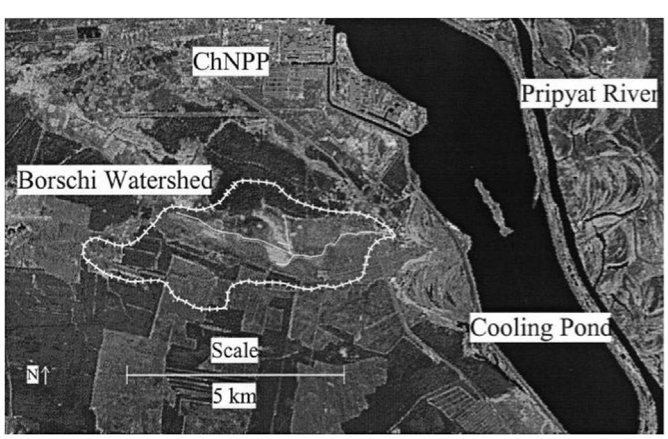

Fig. 6. The Borschi Watershed and the Chernobyl Nuclear Power Plant (ChNPP) seen from a SPOT satellite image, August 1995. (Freed et al., 2003).

Data presented by Freed et al. (2004) indicates that the soil thickness containing most of the ^{90}Sr is about 0.2 m. The average air temperature of the site is 6.7 °C, which results in an average soil temperature of approximately 7.7 °C. The loadings of ^{90}Sr were set to zero. The initial soil concentration was computed based upon the ^{90}Sr inventory of 1.0 E13 Bq (Freed et al., 2003, 2004). This inventory was divided by the specific radio-activity of ^{90}Sr (143 Ci/g), and that result was divided by the conversion from Ci to Bq (3.7 E10 Bq/Ci) to obtain an inventory mass of 2 g. This mass inventory was converted to a soil concentration of 7.9 E−7 mg/kg by dividing by the product of the site volume (8.5 E6 $m^2 \times 0.2$ m) and the soil dry bulk density (1490 kg/m^3). This result was further reduced by 20% to account for the fraction of non-exchangeable ^{90}Sr that is irreversibly bound to soil and sediment. Measurements of non-exchangeable ^{90}Sr (Freed, 2002) varied between 0 and 70%, 10–40%, and 30–90% for Borschi watershed soils, wetland sediments, and channel sediments, respectively. The approximate average non-exchangeable fraction for soils in general is between 10 and 20% (Freed, 2002). Most of the Borschi watershed is abandoned agricultural fields. Since the measured values ranged up to 70% for the agricultural soils, the higher value of 20% for soils in general was used. Thus, the mass concentration of ^{90}Sr that was input to the model was 6.32 E-7 mg/kg. Although this is a very small concentration, it produces a substantial amount of radiation due to the high specific radio-activity of ^{90}Sr.

All of the mass inventory is expected to be in non-solid form (i.e., dissolved) given the long period of time since the mass was deposited, the relative fast oxidation rate of ^{90}Sr for soils exposed to air, the small sizes (approximately 1 micron diameter) of solid phase fuel particles scattered from the accident, and the high solubility of oxidized ^{90}Sr. However, Freed (2002) maintained that a considerable portion of the ^{90}Sr particles were still in solid fuel particle form rather than dissolved. Thus, model runs were conducted where the initial inventory was either in solid phase or non-solid phase form. For solid phase inventory, it was assumed that weathered ^{90}Sr exists as strontium oxide (SrO), which has a specific gravity of 4.7 and solubility of 6900 mg/L, as well as an initial particle diameter of 1.0 micron. These input parameters were used within the soil model to compute the dissolution flux of solid phase ^{90}Sr. It is noted that SrO reacts quickly and exothermally with water to form $Sr(OH)_2$ which is highly soluble. Compounds of Sr, which may exist in the fuel particles, should weather more slowly and should be less soluble. Thus, for one test case, the entire inventory was assumed to be in solid phase form with a much lower solubility of 100 mg/L.

The input for soil dry bulk density of 1.49 g/cm^3 was based on the assumption that the soil is predominantly loamy sand based on the soil description of Freed et al. (2004). This texture also results in a volumetric soil moisture content of approximately 12% and a porosity of 44%, which are required by the model.

The average annual hydrology required by the model was estimated based on information provided by Freed (2002, 2003), which stated that the average annual precipitation is 0.6 m, the watershed stream flow is about 15–20% of precipitation on average, and base flow is about 80% of stream flow. The average annual stream flow rate on a watershed area basis was based on the average for the period 1999–2001, which was 0.097 m/yr and is 16% of precipitation. With 20% of stream flow due to rainfall runoff, the average annual runoff from rainfall was set to 0.0194 m. The average rainfall was estimated based on the reported snow melt, which was 22% of the annual water flux for 1999–2001; thus, rainfall (precipitation less snowfall) is approximately 0.47 m/yr.

Freed et al. (2004) stated that the surficial deposits are underlain by clay marl, which could restrict percolation to groundwater. Thus, the base flow can be caused by soil interflow resulting from

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infiltration that is diverted to surface water rather than percolation to groundwater recharge. Infiltration is defined within TREECS™ as the annual depth of water that penetrates the surficial soil layer after allowing for runoff and evapotranspiration. Thus, infiltration has two possible fates for this model, interflow that re-enters surface waters as base flow or percolation to groundwater. With the base flow (interflow) making up 80% of the stream flow, the interflow is expected to be about 0.0776 m/yr. About 80% of infiltration is believed to be diverted to interflow for the Borschi watershed,[2] leaving about 20% for groundwater recharge, which is lost from the system for the present modeling case since aquifer fate was not considered. There is an input parameter within the TREECS™ soil model to prescribe the percent of infiltration that is diverted to interflow, and this parameter was set to 80% initially. With interflow equal to 80% of infiltration and an interflow rate of 0.0776 m/yr, the infiltration rate, which is a model input, was calculated to be 0.097 m/yr.

The soil model also requires the number of days per year that runoff-producing rainfall occurred. Freed et al. (2003) reported the number of days each year that had storms for 1999–2001. The average of these is 14 days, which was used for the model input. This input parameter was varied by a factor of 6 to evaluate its sensitivity, and was found to have very little effect on model computed export fluxes for this application since most of the stream loading results from interflow or base flow.

Soil erosion was estimated using the USLE that is in the Hydro-Geo-Characteristics Toolkit (HGCT) of TREECS™. The USLE parameters were set as follows: $R = 150$ (assumed based on precipitation); $K = 0.04$ (based on soil texture of loamy sand); $LS = 0.425$ based on slope = 0.02 and runoff length > 400 ft; $C = 0.03$ (based on land use description); and $P = 1$. The sediment delivery ratio (SDR) was turned on. The soil erosion rate was computed to be 2.5 E-6 m/yr, which is quite small.

Based on guidance from the U.S. Environmental Protection Agency (EPA) (EPA, 1999), the soil – water partitioning distribution coefficient (K_d) for strontium should vary between about 15 and 200 L/kg for soil pH between 5 and 8 and clay content between 4 and 20%. The soil clay content of the Borschi watershed is sparse (less than 10%), and the pH is between 6 and 7 (Freed, 2002). Thus, K_d values between about 15 and 200 L/kg are expected for the Borschi watershed soils. Site measurement indicated that K_d for channel bottom sediment was 76 L/kg (Freed, 2002). Although sediment K_d can be quite different than soil values, a value of 76 L/kg was used in this model initially. The input value for K_d was considered uncertain and was varied during an uncertainty analysis as discussed below.

The half-life of ^{90}Sr was set to the TREECS™ database value of 29 years. Volatilization was set to zero. The model start time was year 2000, and the model was run for 200 years. However, the flux at year 2000 was of primary interest for comparison to the export flux estimated by Freed et al. (2003) based on field data for 1999–2001.

4. Results and discussions

4.1. Baseline model results

The TREECS™ soil model was executed, and the export flux to surface water was output in mass flux units (g/yr). The mass flux units were converted to radiation flux units of Bq/yr for ^{90}Sr export. The export flux of ^{90}Sr versus time is plotted in Fig. 7 for the baseline input conditions discussed in the previous section and assuming all of the ^{90}Sr inventory is dissolved (i.e., non-solid phase) and $K_d = 76$ L/kg. These baseline results are reported for the export of dissolved ^{90}Sr since the flux of particulate (i.e., adsorbed to soil) ^{90}Sr was two orders of magnitude lower. The export flux was predominantly dissolved in water, and particulate ^{90}Sr contributed little to the total export flux.

The computed ^{90}Sr export flux at time zero (year 2000) is 4.95 E10 Bq/yr for the baseline conditions, which is 0.5% of the inventory. This is compared to the flux estimated from field data (Freed et al., 2003) of 1.43 E10 Bq/yr, which is 0.14% of the inventory. Thus, the model computes an export flux that is a little more than three times higher than the field-estimated flux.

Freed et al. (2003) reported that the ^{90}Sr concentration in the stream at the watershed outlet varied from 6 to 35 Bq/L during 1999–2001. The model-computed stream concentration was obtained by dividing the computed export flux of 4.95 E10 Bq/yr by the average annual stream flow of 8.5 E8 L/yr (computed from the product of 0.1 m/yr of runoff, 8.5 km^2 surface area, and 1 E9 L/m/km^2), resulting in 58 Bq/L, which is about triple the observed concentrations.

4.2. Sensitivity testing

The baseline test case was repeated with the entire ^{90}Sr inventory in solid phase form with solubility of 6900 mg/L. The results were visually identical to those shown in Fig. 7. Thus, for a high solubility, it does not matter whether or not the inventory is assumed to be dissolved or solid phase since the dissolution rate is so fast. It is recognized that slower dissolution rates can occur when ^{90}Sr is mixed with other compounds, such as the uranium oxide and possibly zirconium compounds that can exist within fuel particles. Thus, another test run was made with the baseline inputs except with the entire inventory in solid phase with a solubility of 100 mg/L. The results of this test were also visually identical to those shown in Fig. 7. Therefore, the dissolution rate is sufficiently fast such that it does not matter what form the inventory is assumed to be. The rapid dissolution rate is probably caused by the small initial particle size of 1 micron, which is consistent with field observations (Freed, 2002). The other tests discussed below were conducted with the entire inventory in dissolved form.

The primary uncertainties of the model inputs are the percent of infiltration that goes to soil interflow, and thus stream base flow, and the value of K_d. Another uncertain input is the estimated soil erosion rate. However, setting the erosion rate to zero had practically no effect on export flux for the conditions of this application since the estimated erosion rate was already quite small compared to the flux of dissolved ^{90}Sr stemming from runoff and interflow.

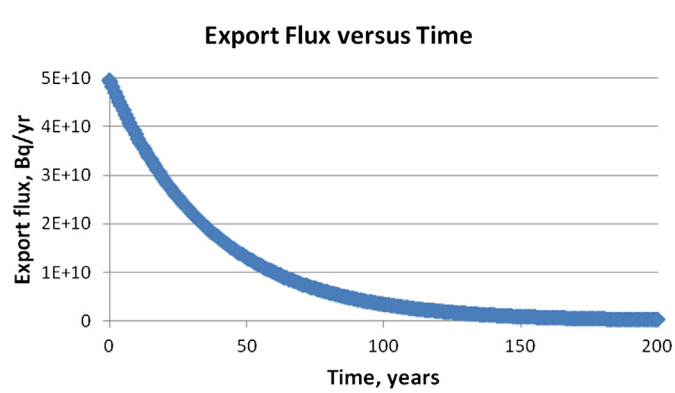

Fig. 7. TREECS™-computed watershed export flux to surface water of ^{90}Sr versus time for baseline conditions (time zero is year 2000).

[2] Personal communication with Boris Faybishenko, Earth Sciences Division, Lawrence Berkeley National Laboratory, April 2012.

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The infiltration rate had to be estimated based upon the estimated base flow (i.e., interflow) and the assumption of 80% of infiltration goes to interflow. The soil annual evapotranspiration *ET* (m/yr) can be calculated from the annual precipitation depth *P* minus the annual runoff and infiltration depths. For a ratio of interflow *IF* to infiltration *I* (*IF/I*) of 0.8, the ratio of *ET/P* is 0.81, which is unrealistically high. With $IF/I = 0.4$, $ET/P = 0.65$, which is more reasonable. Halving the ratio *IF/I* doubles the infiltration rate since the interflow flux is the same. The model was run with double the infiltration rate and 40% of infiltration going to interflow. The initial flux at time zero (year 2000) was identical to that of the baseline case, but the flux decreased slightly faster with time compared to the baseline case. The faster decrease is due to greater leaching losses to groundwater associated with the higher infiltration rate.

The above results leave only the value of K_d as the primary uncertain and sensitive model input. A value of 200 L/kg was tested with all other inputs set to those of the baseline conditions. This change had the effect of reducing the ^{90}Sr export flux from 4.95 to 1.88 Bq/yr, or almost a third, for the year 2000. The lower flux represents 0.19% of the inventory, which is much closer to the field estimate of 0.14%. The revised model-computed stream concentration is 22 Bq/L, which is well within the range of observed concentrations. Increasing K_d of ^{90}Sr has the effect of decreasing the export flux initially and reducing the rate of attenuation of export flux over time, i.e., the slope of export flux versus time is flatter. However, after 200 years, the export flux of ^{90}Sr from the watershed is about the same regardless of whether a value of 76 or 200 L/kg is used for K_d, which is probably due to the over-riding effects of radioactive decay. Increasing K_d has the effect of increasing the watershed residence time of the radionuclide (Joshi and Shukla, 1991), which is the same result found here.

An additional sensitivity test was run to evaluate the effects of radioactive decay versus natural hydrologic attenuation factors associated with runoff, interflow, and infiltration losses coupled with sorption partitioning. A run was made with the same baseline conditions noted above except with the radioactive half life set to 1.0 E20 years, which is essentially no decay. The results of this run are compared with the previous baseline results (Fig. 7) with radioactive decay half life of 29 years. The two results are plotted together in Fig. 9 with a logarithmic scale for export flux so that the magnitude of change is easier to see. These results show that radioactive decay and hydrologic attenuation cause the watershed export flux to decrease by a factor of 209 over the 200 year simulation, whereas hydrologic attenuation alone (without decay) causes the export flux to decrease by only a factor of 3.8 over the 200 years. Thus, radioactive decay amplifies the attenuation by a factor of about 55 compared to hydrologic attenuation alone. It is noted that decay in this model can represent any type of decay process, including radiation, biodegradation, and abiotic degradation (such as photolysis, oxidation, etc.), and the input parameter for all decay processes is half life with values that can vary from very small (on the order of days) to very large (centuries or millennia).

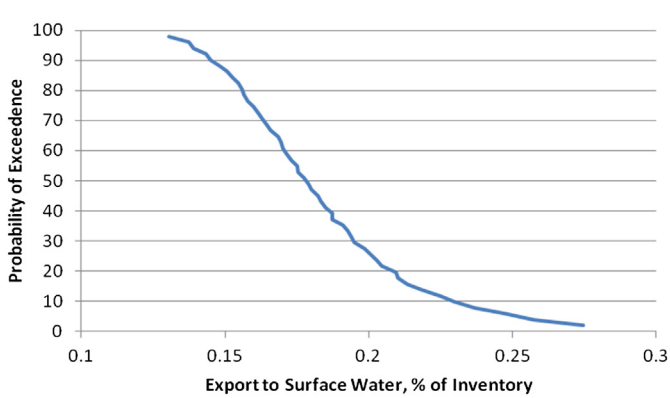

Fig. 8. Probability of exceedence for percent of ^{90}Sr inventory exported to surface water for uncertain K_d.

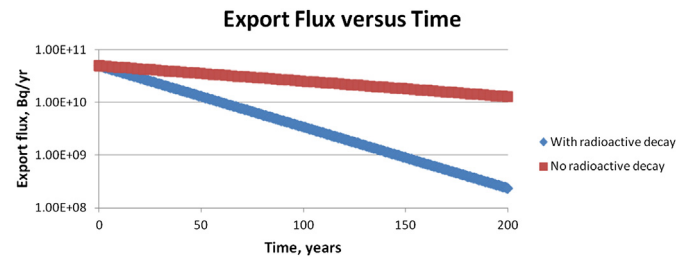

Fig. 9. TREECS™-computed watershed export flux to surface water of ^{90}Sr versus time for baseline conditions (time zero is year 2000) with and without radioactive decay.

4.3. Uncertainty analysis

Since there is uncertainty associated with K_d, an uncertainty analysis was conducted. The Sensitivity/Uncertainty (S/U) feature that is available within TREECS™ was used to evaluate output variability associated with K_d uncertainty. The S/U feature uses Monte Carlo simulation with Latin Hypercube sampling for improved efficiency.

For the uncertainty analysis, the minimum and maximum values for K_d were set to 100 and 300 L/kg, respectively, and a normal distribution was assumed with a mean of 200 and standard deviation of 33 L/kg. The range in K_d was based on results reported by EPA (1999) for a wide variety of soils.

After testing for convergence, the number of Monte Carlo iterations was set to 50 for evaluating output. The exceedance probability for export as percent of inventory in year 2000 is shown in Fig. 8 and is based on the variations in the uncertain value of K_d. The plot in Fig. 8 has a 50% exceedance of 0.18% and a range of about 0.13–0.27% of inventory exported to surface water compared to 0.14% as estimated from field data.

5. Conclusions

The TREECS™ soil model provided reasonable estimates of the surface water export flux of ^{90}Sr from the Borschi watershed when using a soil-water distribution coefficient (K_d) for ^{90}Sr of 200 L/kg. The computed export was 0.18% of ^{90}Sr inventory compared to the estimated export flux 0f 0.14% (±0.02%) based on field data from 1999 to 2001. The model indicated that assumptions regarding the form of the inventory, whether dissolved or in solid phase form, did not affect export rates due to rather rapid dissolution rates associated with the small particle size of solid phase ^{90}Sr. Also, the assumption regarding the percentage of infiltration diverted to soil interflow did not affect watershed export at the start of the simulation (year 2000), but it does affect the rate of decrease of ^{90}Sr export over time. The model shows that radioactive decay affects attenuation of watershed export of ^{90}Sr more than hydrologic factors by several orders of magnitude.

The most sensitive and uncertain input for this application is the soil-water distribution coefficient (K_d) for ^{90}Sr. EPA-recommended K_d values for ^{90}Sr in soils similar to those in the Borschi watershed are between 15 and 200 L/kg. A value of 200 L/kg produced export results that were closer to that estimated from field data for

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the year 2000. Lower values of K_d produced higher export fluxes initially (year 2000) with a faster attenuation rate. An uncertainty analysis was conducted treating K_d as an uncertain input variable with a range of 100–300 L/kg. This analysis resulted in a range of about 0.13–0.27% of inventory exported to surface water during the year 2000 compared to 0.14% based on field data.

This application demonstrates how TREECS™ can be applied for a radionuclide. Such an application is done by modeling the constituent mass as normally done and converting the output mass concentration/flux values to radiation concentrations and fluxes. The specific radioactivity of the radionuclide must be used to make these conversions.

Acknowledgments

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