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

by Mark S. Dortch

PURPOSE: 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 Army. TREECS™ will have varying levels of capability 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. Although TREECS™ was developed for the fate of MC from ranges, it has general applicability to many other situations requiring prediction of contaminant fate in multi-media environmental systems.

TREECS™ was applied to the Borschi watershed near the Chernobyl Nuclear Power Plant, Ukraine. At this site, TREECS™ was used as a modeling tool to predict the fate of radionuclides. This application also provided an additional validation test case for the TREECS™ Tier 2 soil model. This Technical Note (TN) describes the input data and results of this application.

BACKGROUND: As of this writing, there are two levels of capability in TREECS™. Tier 1 consists of screening-level methods that assume highly conservative, steady-state contaminant loading and fate. Tier 1 requires minimal input data requirements and can be easily and quickly applied. Tier 2 provides time-varying analyses and solves mass balance equations for both solid and non-solid phase contaminant mass with dissolution from the solid phase. Additionally, contaminant residue loadings to soil can vary from year-to-year based on munitions use or other loading factors. Thus, media concentrations computed with Tier 2 should be lower than those computed with Tier 1 and should be closer to those expected under actual field conditions. Although Tier 2 is more comprehensive than Tier 1, the Tier 2 models are of reduced form (e.g., limited spatial dimensionality), which greatly reduces input data requirements and expedites ease-of-application. Such models allow multi-century simulation periods that can be executed in seconds. Technical reports describing the model formulations, initial proof-of-concept applications, and field validations can be obtained from the Web site noted above.

Borschi watershed is located 3 km south of the Chernobyl Nuclear Power Plant (Figure 1). Radio-strontium-90, ^{90}Sr, which is a fission product resulting from the accident in 1986, poses a health concern around and down-gradient from Chernobyl due to its high specific radioactivity (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 1.0E13 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 1.62E10 Bq/yr for the period 1999 – 2001,

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with an average of about 1.43E10 Bq/yr or about 0.14 % of the remaining inventory as of 2000. This export was a result of snow melt, storm events, moderate rains, and low base flows from below-ground discharge.
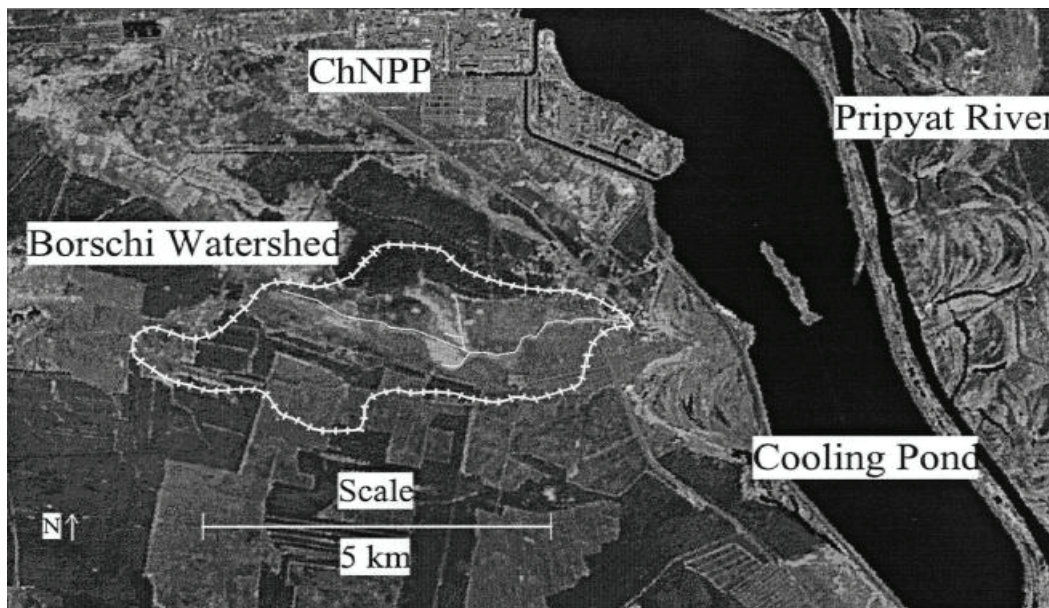

Figure 1. The Borschi watershed and the Chernobyl Nuclear Power Plant (ChNPP) seen from a SPOT satellite image, August 1995 (from Freed et al. 2003).

MODEL INPUTS: The information regarding ^{90}Sr in the Borschi watershed provided by Freed (2002) and Freed et al. (2003 and 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: in this case, 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 as described in this TN.

The surface area of Borschi watershed is 8.5 km^2. 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 deg C, which results in an average soil temperature of approximately 7.7 deg C. The loadings of ^{90}Sr were set to zero. The initial soil concentration was computed based upon the ^{90}Sr inventory of 1.0E13 Bq (Freed et al. 2003 and 2004). This inventory was divided by the specific radioactivity of ^{90}Sr (143 Ci/g), and that result was divided by the conversion from Ci to Bq (3.7E10 Bq/Ci) to obtain an inventory mass of 2 g. This mass inventory was converted to a soil concentration of 7.9E-7 mg/kg by dividing by the product of the site volume (8.5E6 m^2 x 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 adsorbed strontium that is irreversibly bound to soil and sediment. Measurements of non-exchangeable

strontium (Freed 2002) varied between 0 to 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 to 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 strontium that was input to the model was 6.32E-7 mg/kg. Although this is a very small concentration, it produces a substantial amount of radiation due to the high specific radioactivity 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 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 Sr. However, Freed (2002) maintained that a considerable portion of the 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 Sr exists as strontium oxide (SrO), which has a specific gravity of 4.7 and solubility of 6,900 mg/L, as well as an initial particle diameter of 1.0 micron. These input parameters are used within the soil model to compute the dissolution flux of solid phase 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 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 et al. (2002 and 2003); this information stated that the average annual precipitation is 0.6 m, the watershed stream flow is about 15 to 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 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 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 reenters 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[1], leaving about 20 % for groundwater recharge. This 20% is lost from the system for the present modeling case

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

since Sr fate in the aquifer 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 Universal Soil Loss Equation (USLE) that is in the Hydro-Geo-Characteristics Toolkit (HGCT) of TREECS™. The USLE parameters were set as follows: R = 150 (assumed); K = 0.04 (based on soil texture of loamy sand); slope = 0.02 (assumed) 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 2.5E-6 m/yr, which is quite small.

Based on guidance from the U.S. Environmental Protection Agency ((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 from 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, only 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.

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 Figure 2 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 contributes little to the total export flux.

The computed ^{90}Sr export flux at time zero (year 2000) is 4.95E10 Bq/yr for the baseline conditions, which is 0.5 % of the inventory. This should be compared to the flux estimated from field data (Freed et al. 2003) of 1.43E10 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
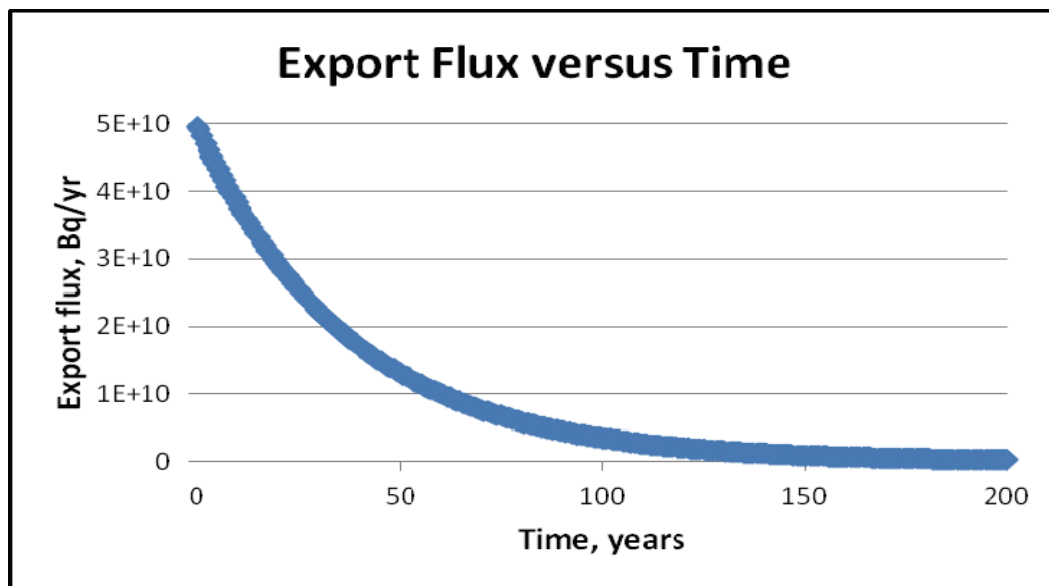
Figure 2. TREECS™-computed watershed export flux to surface water of ^{90}Sr versus time for baseline conditions (time zero is year 2000)

the computed export flux of 4.95E10 Bq/yr by the average annual stream flow of 8.5E8 L/yr (computed from the product of 0.1 m/yr of runoff, 8.5 km^2 surface area, and 1E9 L/m/km^2), resulting in 58 Bq/L, which is about triple the observed concentrations.

SENSITIVIY TESTING: The baseline test case of Figure 2 was repeated with the entire ^{90}Sr inventory in solid phase form with solubility of 6,900 mg/L. The results were practically identical to those shown in Figure 2. For a high solubility, therefore, 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 Sr is mixed with other compounds, such as the uranium oxide and possibly zirconium compounds that can exist within fuel particles. Consequently, 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 practically identical to those shown in Figure 2. Thus, 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.

The infiltration rate had to be estimated based upon the estimated base flow (i.e., interflow) and the assumption that 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. 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.

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 Figure 3 and is based on the variations in the uncertain value of K_d. The plot in Figure 3 has a 50 % exceedance of 0.18 % and a range of about 0.13 to 0.27 % of inventory exported to surface water compared to 0.14 % as estimated from field data.

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 Sr of 200 L/kg. The computed export was 0.18 % of ^{90}Sr inventory compared to the estimated export flux 0f 0.14 % based on field data from 1999 – 2001. The model indicated that assumptions regarding the form of the inventory, whether dissolved or in solid phase form, did not affect export rates. As long as the soil interflow is the same (0.078 m/yr), the assumption regarding the infiltration rate and percentages of infiltration diverted to soil interflow and groundwater recharge 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. Also, the percentage of non-exchangeable adsorbed Sr is uncertain and variable. Although this percentage does affect the amount of Sr available for export, which linearly and inversely affects the export as a percentage of initial inventory, it was kept fixed at 20 %.

The most sensitive and uncertain input for this application, besides the percent of non-exchangeable Sr, is the soil-water distribution coefficient (K_d) for Sr. EPA-recommended K_d values for 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. Lower values of K_d produce higher export fluxes. An uncertainty analysis was conducted treating K_d as an uncertain input variable with a range of 100 to 300 L/kg. This analysis resulted in a range of about 0.13 to 0.27 % of inventory exported to surface water compared to 0.14 % based on field data.
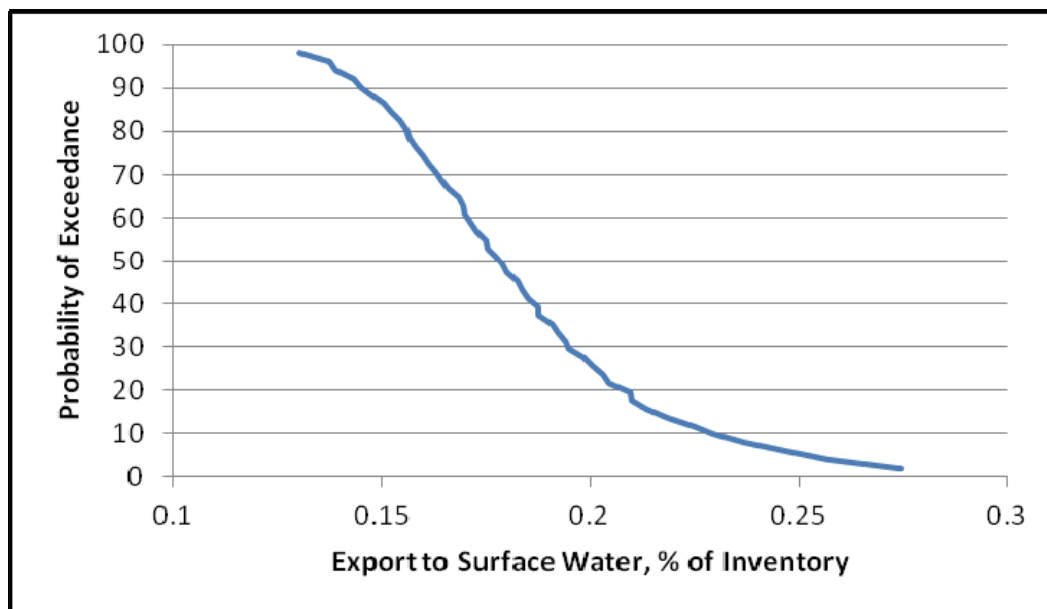
Figure 3. Probability of exceedance for percent of ^{90}Sr inventory exported to surface water for uncertain K_d

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 Sr being diverted to surface waters that eventually flow out of the watershed. The percentage of non-exchangeable adsorbed Sr and the soil-water K_d are the two most sensitive and uncertain factors affecting the amount of export.

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

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