The use of dynamic modeling in assessing tritium phytoremediation

Karin T. Rebel, S. J. Riha, J. C. Seaman, and C. D. Barton

ABSTRACT

To minimize movement of tritium into surface waters at the Mixed Waste Management Facility at the Savannah River Site, tritium-contaminated groundwater released to the surface along seeps in the hillside is being retained in a constructed pond and used to irrigate forest acreage that lies over the contaminated groundwater. Management of the application of tritium-contaminated irrigation water needs to be evaluated in the context of the large amount of rainfall relative to evapotranspiration, the strong seasonality in evapotranspiration, and intra-annual and interannual variability in precipitation in this region. A dynamic simulation model of water and tritium fluxes in the soil-plant-atmosphere continuum was developed to assess the efficiency (tritium transpired/tritium applied) of several irrigation management strategies. The model was parameterized using soil-water content data measured at 18 sites for the first year of the project and evaluated using tritium activity measurements made at the same 18 sites over 2.5 yr. The model was then used to evaluate several irrigation strategies. The 25-yr efficiencies (tritium transpired/tritium applied) of the irrigation strategies were related to the quantity of irrigation water applied. There was a strong ($r^2 = 0.99$) negative linear relationship between irrigation water applied and efficiency. When a quasi-steady state has been reached in the system, the annual efficiencies of all the irrigation strategies were negatively correlated with annual rainfall. Quantification of these relationships allows irrigation managers to choose irrigation strategies based on desired long-term system efficiency, which differ with climate and irrigation strategy.

INTRODUCTION

Buried, low-level, solid radioactive waste containing tritium leached following rainfall infiltration, allowing tritiated water (HTO) to enter groundwater at the Savannah River Site. The groundwater
A plume containing HTO is released at the surface along seeps in the hillside and enters a low-order stream, Fourmile Branch, that serves as the single largest source of tritium entering the Savannah River (Arnett, 1997). Without corrective action, tritium release to Fourmile Branch will continue for several decades. Therefore, the overall project objective is to reduce tritium flux to Fourmile Branch. To accomplish this objective, a sheet pile dam was constructed at the base of the watershed to collect the tritium-enriched seeps for use as a source of irrigation water for a mixed hardwood and pine forest located upgradient from the pond (Blake, 1999). Although the irrigation field lies above the contaminated aquifer that serves as the source for the collection pond, a major goal of the irrigation system is to maximize the evapotranspiration of the applied water and minimize leaching below the rooting zone.

Tritium in the irrigation water can be (a) transpired by the vegetation and/or evaporated directly into the atmosphere (Luvall and Murphy, 1982; Kalin et al., 1995; Murphy, 1995), (b) retained in the soil, or (c) leached below the root zone. The amount of tritium that is lost from the soil via evapotranspiration and leaching will depend on the climate, the rate and timing of tritium application, and soil and vegetation properties, but neither of these losses are directly measured. Therefore, a capacitance-based one-dimensional model (Buttler and Riha, 1992; Riha and Rossiter, 1993) of water and tritium movement and storage was parameterized for the site and used to predict how much tritium is transpired by the vegetation versus how much tritium is leached below the rooting zone and is retained in the soil over time. The objective of this modeling study is to determine the efficiency (tritium transpired/tritium applied) of several different irrigation management strategies when assessed over many years with varying weather.

MATERIALS AND METHODS

Model Description

Soil-water movement and storage were simulated using a capacitance approach (Jones and Kiniry, 1986; Buttler and Riha, 1992; Riha and Rossiter, 1993). The soil is conceptualized as consisting of a series of layers, each having a specified capacity to hold water. Water is transferred downward from one layer to the next in the soil profile if the amount entering the layer exceeds the layer’s water-holding (field) capacity (FC). The rate of downward transfer is dependent on a drainage coefficient (SWCON). The capacity of each soil layer to hold water is calculated as the difference between the saturation water content and the current volumetric water content. This method is not iterative; the capacities at the beginning of the time step are used to estimate the redistribution of water during that time step.
Soil-water uptake is simulated using an absorption approach (Riha and Rossiter, 1993). The maximum amount of soil-water uptake is the potential evapotranspiration (ETP) multiplied by a partitioning factor to obtain potential transpiration. Potential evapotranspiration is partitioned into potential transpiration and potential evaporation according to the fraction solar radiation intercepted by the canopy, which is a function of the leaf area index and the extinction coefficient for diffuse ray penetration (Landsberg, 1986; Norman and Campbell, 1989). The potential transpiration is allocated to the soil layers based on the relative root density distribution in the soil profile. As the soil-water content in a layer decreases and approaches a minimal soil-water content, referred to as the permanent wilting point (PWP), the demand for water uptake that is not met in a layer can be transferred to deeper layers. Thus, root densities do not directly limit water uptake in this simple representation but serve solely to partition transpiration demand in the soil profile. When demand can no longer be transferred to deeper layers (because the limit of rooting depth has been reached, or all deeper layers are approaching or are at the PWP), then actual transpiration will be less than the potential.

Tritium fluxes in the soil are simulated using the simple assumption that all tritium entering a soil layer is completely mixed with the water and tritium stored in that layer, and that tritium movement occurs only through advection. In other words, diffusion and dispersion are not simulated in this model (Horton and Ross, 1960). Tritium uptake in a layer is the product of the tritium activity in that layer and the water uptake by roots in that layer. Tritium transpired by the vegetation is the sum of tritium uptake in all soil layers with roots. Tritium that is leached is the flux of tritium out of the deepest soil layer containing roots. This one-dimensional model runs on a daily time step.

Model Evaluation

Field Measurements
Eighteen measurement sites were established in the irrigation area to monitor water content and tritium activity. Time-domain reflectometry (TDR, Trime Inc.) was used to measure volumetric soil-water content. Each site contained two tubes for TDR measurements, which were inserted to a depth of 140–185 cm (55–73 in.), depending on ease of installation. Soil-water content was then measured weekly at a depth of 25, 55, 95, 135 and (when possible) 175 cm (10, 21, 37, 53, and 69 in.). To estimate residual soil tritium, each of the 18 monitoring sites included five suction lysimeters at depths of 50, 100, 150, 200, and 300 cm (20, 39, 59, 78, and 118 in.), and five soil vapor samplers were installed at depths of 25, 55, 135, 205, and 295 cm (10, 21, 53, 80, and 116 in.). Sample collection using the suction lysimeters proved unreliable during periods of drought because of the inability to maintain a sufficient vacuum to yield a sample at higher soil matrix tensions; therefore, lysimeter sampling was suspended for much of the study.

Brian Looney and Joe Rossabi developed the vapor samplers, which consisted of a polyvinyl chloride well pipe screened over a 6-in. (15-cm) span at the base and installed to the sampling depth of interest, then grouted to the surface (B. Looney and J. Rossabi, 2001, personal communication). Soil pore water was collected by pulling a small vacuum on the well pipe at the surface and condensing the water vapor. This technique proved to be effective in yielding sufficient pore water for tritium analysis, regardless of the soil moisture conditions. Water was analyzed for tritium by liquid scintillation analysis with an estimated detection limit of 20 pCi/L and a counting error below 2% for elevated tritium concentrations (Minaxi Tricarb 4000, Packard Instrument Co.).

Parameterization
The model was parameterized to simulate water content and tritium activity at each of the 18 measurement sites. Field capacity for each soil layer at each site was estimated from soil-water content data several days after large rainfall events and from late winter water content values in deeper layers. Permanent wilting point and SWCON were determined from FC using a pedotransfer function. Root distribution and depth were estimated from patterns of water uptake with depth (Bouten et al., 1992; Musters and Bouten, 1999; Zuo and Zhang, 2002). Data from a nearby weather station on the Savannah River Site were used to calculate ETP using the Priestley-Taylor equation (Priestley and Taylor, 1972), and precipitation values were obtained from the weather station and from on-site measurements. The partitioning factor was calculated from seasonal measurements of the probability of diffuse ray penetration using a ceptometer (AccuPar, PAR80, Decagon Devices, Inc.) and the method described by Norman and Campbell (1989). The rate
and timing of irrigation water application and the tritium activity of the irrigation water are described in Hitchcock et al. (2005).

Performance
Model performance was evaluated using 2.5 yr of tritium data. The predicted tritium activities varied more over time in the surface soil layers than in deeper layers, which was generally true of the measured values. Figure 1 presents the mean, plus and minus one standard deviation, of both measured and simulated tritium activity, of the 18 sites over the 3 yr of the study. In general, the model adequately simulated the observed dynamics and tritium levels with time and depth; the simulated values were almost always within one standard deviation from the mean of the measured values.

Figure 1. Mean measured tritium activity (squares) ± S.D. (bars) and mean simulated tritium activity (black line) ± S.D. (gray lines) with soil depth. S.D. = standard deviation.

Simulations
Parameters
To evaluate the long-term efficiency of irrigation management, 25 yr (1974–1999) of daily Augusta, Georgia, weather data were used to drive the model. Daily rainfall was a direct input to the model; net radiation was calculated knowing solar radiation and air temperature. Evapotranspiration was then calculated using the Priestley-Taylor equation.

Average 25-yr precipitation and potential evapotranspiration were 1127 mm/yr (44.3 in./yr) and 1443 mm/yr (56.8 in./yr), respectively. The 25-yr daily average ETP exhibited the expected annual sinusoidal pattern, with a high of about 7 mm (0.27 in.) in the summer and a low of about 1 mm (0.04 in.) in the winter. The
25-yr daily average rainfall did not exhibit seasonal trends. In this type of climate, in winter months, precipitation exceeds ETP, whereas in summer months, precipitation is less than ETP.

The soil profile was divided in nine layers, ranging from 0.1 to 0.4 m (0.3 to 1.3 ft) in thickness with the thin layers at the top, where the system is more dynamic. A single soil profile to a depth of 3.1 m (10.1 ft) of soil was simulated using properties presented in Table 1, which were considered typical of the site. Soil properties vary with depth because the soils at the site are generally sandy at the surface, grading rapidly to more fine-textured, clayey soil layers at depth (Daniels et al., 1984; Rodgers and Herren, 1990; Riha, 2001).

Roots occupy the first six layers of the soil profile to a depth of 1.9 m (6.2 ft). Relative root density decreases with depth (Table 1). A partitioning factor of 0.9 was used for all the simulations and held constant over the year, thus representing a maximum potential transpiration rate for this site.

Irrigation Strategies

Irrigation managers can change both the timing and the rate of application of the pond water. Therefore, simulated irrigation strategies were devised that varied the timing of irrigation and the amount of irrigation water applied. The irrigation strategies evaluated in this simulation study can be grouped into four categories:

1. Irrigate daily as a constant percent of previous-day ETP, or irrigate daily at a higher percent of ETP in summer (April 15–October 15) and lower percent in winter (October 16–April 14).
2. Irrigate daily at a constant rate, or irrigate daily at a higher rate in summer (April 15–October 15) and lower rate in winter (October 16–April 14).
3. Irrigate if 6, 12, or 18 mm (0.23, 0.47, or 0.70 in.) of deficit (ETP minus precipitation) has accrued, restarting deficit accrual accounting after irrigation or after 10, 20, or 40 mm (0.39, 0.78, or 1.57 in.) excess precipitation over ETP has accumulated; deficit accrual has to be rezeroed periodically because precipitation exceeds ETP for much of the year.
4. Irrigate as a constant percent of ETP only if the value of ETP is equal to or less than the decrease in the soil-water content in the root zone on the previous day or at a higher percent of ETP in summer (April 15–October 15) and lower percent in winter (October 16–April 14).

The tritium activity in the irrigation water was assumed to be at a constant level of 16,000 pCi/mL.

Sensitivity Analyses

The sensitivity of predicted 25-yr average efficiency (tritium transpired/tritium applied) to selected model input parameters was evaluated. For the sensitivity analyses, the irrigation strategy was kept constant (0.45 \( /C2 \) PET in winter and 0.90 \( /C2 \) PET in summer if deficit the previous day, e.g., irrigation strategy 4). The partitioning factor was varied from a low of 0.5 to a high of 0.95, which are the minimum and the maximum partitioning factors determined from seasonal forest cover measurements. Tree root density distributions were evaluated: even distribution with depth, linear decline with depth, and exponential decline with depth. Soil physical parameters were varied from –20 to +20% of their value over the complete soil profile. The results are displayed in a tornado diagram (Figure 2). Efficiency is most sensitive to the partitioning factor and least sensitive to SWCON, the drainage coefficient.

RESULTS

Long-Term Efficiency

The various irrigation strategies summarized above were simulated for 30 yr, but the results reported here are from the last 25 yr (1974–1999) of the simulation. This approach was used so that all values reflect the performance of the system when soil tritium values reach a quasi-steady state after 2–5 yr.

Table 1. Soil Variables Used for the 25-Yr Simulations

<table>
<thead>
<tr>
<th>Layer</th>
<th>Thickness (m)</th>
<th>Soil Depth (m)</th>
<th>FC (m³ m⁻³)</th>
<th>PWP (m³ m⁻³)</th>
<th>Relative Root Distribution</th>
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<tr>
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<td>0.1</td>
<td>0.12</td>
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</tr>
<tr>
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<td>0.4</td>
<td>0.15</td>
<td>0.05</td>
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<td>0.75</td>
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<td>0.05</td>
<td>0.2</td>
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<td>1.15</td>
<td>0.26</td>
<td>0.18</td>
<td>0.1</td>
</tr>
<tr>
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<td>0.4</td>
<td>1.55</td>
<td>0.26</td>
<td>0.18</td>
<td>0.1</td>
</tr>
<tr>
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<td>0.35</td>
<td>1.9</td>
<td>0.2</td>
<td>0.13</td>
<td>0.1</td>
</tr>
<tr>
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</table>
most important result of this study is that in general, no matter what irrigation management strategy was simulated, as more irrigation water is applied, more tritium is transpired (e.g., greater phytoremediation occurs), but the system becomes less efficient (tritium transpired/tritium applied). This is because at higher application rates, more tritium is stored in and leached from the soil.

An inverse linear relationship ($r^2 = 0.99$) exists between the amount of irrigation water applied and the long-term efficiency of the tritium phytoremediation project (Figure 3) using the range of irrigation strategies described above. For a given irrigation level, some irrigation schemes are slightly more efficient than others, but over a 25-yr simulation, these differences are small. The irrigation strategies tested resulted in a range of annual average application of irrigation water from 220 to 1370 mm (8.6 to 54 in.) of water and efficiencies of 56–87% (Figure 3). The simulations indicate that, in the long term, increasing the annual amount of irrigation water applied by 100 mm (3.9 in.) will decrease the efficiency of the system by 2.8%, assuming constant tritium activity in the irrigation water. Although efficiency has a linear relationship with the irrigation water applied, the flux of tritium transpired and leached has a nonlinear relationship with irrigation water applied (Figure 4). As more water (and, therefore, more tritium) is applied, a decreasing fraction of the tritium is transpired, and an increasing fraction of the tritium is leached.

**Interannual Variability in Efficiency**

Interannual differences in efficiency between the various irrigation schemes varied by as much as 25%. An analysis of the interannual variability in efficiency for the 25 yr of simulation indicates a high dependency on annual precipitation (Figure 5). As expected, efficiency decreases as annual precipitation increases. The scatter around this general trend is not surprising given that differences in the size and persistence of rainfall events, as well as total rainfall, would be expected to affect system efficiency. The irrigation strategy that included irrigating only when a soil-water deficit developed was less correlated with annual precipitation ($r^2 = 0.59$) than those strategies that were independent of soil-water status ($r^2 = 0.71$ and 0.72) (Figure 5).

**Intra-Annual Variability in Efficiency**

We evaluated several irrigation strategies in which the irrigation rate varied with the season (e.g., higher in summer, lower in winter). In Figure 6, daily average irrigation water applied over the course of 1 yr is

**Figure 2.** Sensitivity of average 25-yr efficiency to input parameters.

**Figure 3.** Average 25-yr efficiency of tritium remediation as a function of average annual irrigation water applied.

**Figure 4.** Tritium transpired (closed diamonds) and leached (open circles) as a function of tritium applied. 1:1 line included.
contrasted for three irrigation strategies: 45% ETP in winter and 90% ETP in summer; 45% ETP in winter and 90% ETP in summer when deficit; 45% ETP in both winter and summer. As expected, the seasonal pattern of tritium application varied considerably, depending on the irrigation strategy. The impact on daily average tritium transpired (Figure 7) shows the expected seasonal trend for the two strategies that were solely dependent on ETP. The irrigation strategy that requires a soil-water deficit on the previous day to irrigate (which increased in summer relative to winter) was highly efficient but resulted in reduced summertime application and transpiration compared to the other irrigation strategies (Figure 7). Although high tritium phytoremediation rates were maintained with the strategies that required no deficit, the scenario that included high summer applications rates (45% ETP in winter, 90% ETP in summer) also resulted in some relatively high summer and early fall rates of tritium leaching (Figure 8).

**Tritium Activity in the Soil Profile**

Tritium activity increased in the soil profile after irrigation was initiated, reaching a quasi-steady state within 2–5 yr. As expected, day-to-day variability in tritium activity decreased with depth. The quasi-steady state values of tritium activity in the soil profile differed greatly among treatments. The strategies with higher efficiencies (e.g., 45% ETP in winter and 90% ETP in summer when there was a soil deficit) had considerably lower tritium activity at depth in the soil profile than the less efficient strategies (e.g., 45% ETP in winter and 90% ETP in summer) and took longer to reach a quasi-steady state.

**CONCLUSIONS**

The irrigation strategies tested resulted in average applications of irrigation water ranging from 220 to 1370 mm (8.6 to 54 in.) of water and efficiencies (percent of tritium applied that is transpired) ranging from 56 to 87%. In this range, a strong ($r^2 = 0.99$) negative linear relationship ($y = -0.039x + 129$) exists between the amount of irrigation water ($x$ in mm) applied and the long-term efficiency ($y$ in percent) of the tritium phytoremediation project using the range of irrigation strategies explored in this study. As more irrigation water is applied, more tritium is transpired.

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*Figure 5.* Dependence of average 25-yr efficiency on annual rainfall for three irrigation strategies: 45%w, 90%s no deficit (circles); 45%w, 90%s deficit (diamonds); 45%w, 45%s, no deficit (triangles). w = winter; s = summer.

*Figure 6.* A 25-yr simulation of the average daily tritium applied ($\mu$Ci m$^{-2}$ day$^{-1}$). 45%ETPw, 90%ETPs (black); 45%ETPw, 90%ETPs when deficit (light gray); 45%ETPw, 45%ETPs (dark gray). w = winter; s = summer.

*Figure 7.* A 25-yr simulation of the average daily tritium transpired ($\mu$Ci m$^{-2}$ day$^{-1}$). 45%ETPw, 90%ETPs (black); 45%ETPw, 90%ETPs when deficit (light gray); 45%ETPw, 45%ETPs (dark gray). w = winter; s = summer.
Arnett, M., 1997, Savannah River Site environmental report for irrigation strategies based on desired long-term system efficiency will allow irrigation managers to choose irrigation strategies to irrigation water applied and system strategies. Quantification of the relationship of tritium storage in soil. The simulations indicated that higher efficiencies in the initial 1–2 yr of system operation can be expected because of the time it takes the system to reach a quasi-steady state with respect to tritium storage in soil. The simulations indicated that higher values of tritium activity at depth in the soil would occur under the less efficient irrigation management strategies. Quantification of the relationship of irrigation strategies to irrigation water applied and system efficiency will allow irrigation managers to choose irrigation strategies based on desired long-term system efficiency.

There was considerable (up to 25%) interannual variation in the various irrigation schemes. An analysis of the interannual variability in efficiency for the last 25 yr of simulation indicates a high dependency of efficiency on annual rainfall for the strategies that are not using soil-water deficits as a criterion for irrigation. An understanding of the dependency of interannual variability in efficiency on annual average rainfall will allow irrigation managers to evaluate irrigation strategies that keep efficiencies within an expected range.

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